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Microscopic Charge Density Wave Transport

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Microscopic Charge Density Wave Transport

Proefschrift

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door

Erwin Slot

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To Aase and Duanne

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1. Introduction

The main interest in one-dimensional (1D) electron systems is their dramatically different behavior compared to bulk electron systems. 1D electron systems can be found in systems like quantum Hall edge states and in molecular wires. Related systems are materials with a very anisotropic crystal and electronic structure. They are often called “quasi-one-dimensional” or “low-dimensional”.

Metals with a chain-like structure often have an electron spectrum that is close to one-dimensional or quasi one-dimensional. In such metals a phase transition to a Charge-Density Wave (CDW) state can occur, in which the charge density is periodically modulated in space. The mechanism for CDW formation was first described by Peierls in 1930 [1] and the phase transition is named after him.

CDWs can move with respect to the underlying lattice and thereby transporting charge. The interaction of the CDW with impurities and crystal defects has a large influence on its transport properties. The CDW is pinned by impurities and a moderate electric field is needed for CDW motion. After Peierls description of the CDW state, it took more than forty years for the experimental observation of CDW motion.

The force applied by the electric field to the CDW and the pinning force of impurities cause it to deform. CDWs belong to the broad class of systems that encompass elastic media in the presence of disorder. Other systems in this class include flux line lattices (FLL) or vortex lattices, Wigner crystals and magnetic bubble arrays. Depending on the elasticity and the disorder strength, length scales for phase coherence can be defined. These length scales are on the order of $1 - 10 \mu\text{m}$ in CDW systems. Finite-size effects can be expected in transport measurements when the sizes of CDW wires are reduced below the phase coherence length scale.

This thesis discusses experimental work on CDWs in inorganic materials with microscopic dimensions. This regime is largely unexplored and new fabrication techniques have been developed to make microscopic CDW devices. Transport measurements reveal expected finite-size effects, but also unpredicted behavior, which has called for the development of new models for microscopic CDWs.

1.1 Charge Density Waves

The Fermi surface of a quasi one-dimensional material consists of only two sheets at $k = \pm k_F$, where k_F is the Fermi wave vector. Peierls showed in 1930 that because of the divergent electron response in low-dimensional systems, such a system is unstable towards external perturbations of wave vectors close to $2k_F$ [2, 3]. He predicted that these materials would undergo a second order phase transition to a CDW state when cooled below a certain temperature, now called the Peierls transition temperature T_P [1, 4]. Below T_P , the lattice is periodically modulated in space with the wave vector $2k_F$, as is the charge density

$$n(x, t) = n_0 + \delta n \cos(2k_F x + \phi(x, t)), \quad (1.1)$$

where n_0 is the charge density without modulation and δn is the amplitude of the charge modulation. The periodic modulation of the charge density is called a Charge Density Wave (CDW). The phase $\phi(x, t)$ is used to describe the deformations and motion of the CDW. The modulation of the lattice is typically less than 1%. Peierls also showed that the new periodicity in the potential creates a gap at the Fermi energy [1]. The electronic energy is lowered and overcomes the elastic energy cost of the lattice deformation, see Fig. 1.1. For a 1D chain with lattice constant a , the Fermi wavelength is $k_F = \pi N_e/a$, with N_e the number of electrons in a unit cell of size a . When the CDW wavelength $\lambda_{CDW} = \pi/k_F$ is an integer multiple of the lattice constant, thus when N_e is an integer number, the CDW is commensurate with the lattice. For more complex Fermi surfaces, the CDW can be incommensurate. In most situations, the CDW is commensurate with the underlying lattice, but in this thesis only incommensurate CDWs are investigated.

The ground state of the CDW can be described by an order parameter analogous to that of the superconductor ground state [3]

$$\Delta = |\Delta| e^{i\varphi}. \quad (1.2)$$

The amplitude of the order parameter $|\Delta|$ is defined as the single-particle gap, see Fig. 1.1. In superconductors this is the well-known superconducting gap, while in CDWs this is the Peierls gap. The appearance of a gap in superconductors leads to a finite amplitude coherence length [5, 3]

$$\xi_0 \propto \frac{\hbar v_F}{|\Delta|}, \quad (1.3)$$

with $\hbar$ Planck's constant and v_F the Fermi velocity. Using this definition for the amplitude coherence length, the large CDW gaps results in very short coherence length on the order of $\xi_0 = 1$ nm. This is of the same order as the CDW wavelength λ_{CDW} .

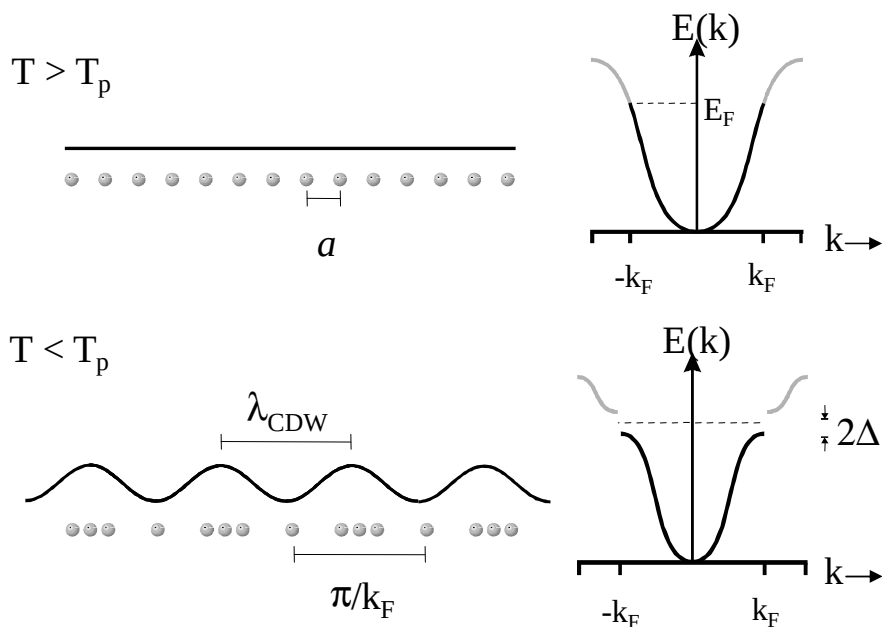


Fig. 1.1: Simplified representation of an electron spectrum of a one-dimensional metal that undergoes a Peierls transition. For temperatures above the Peierls temperature T_P , the charge density is uniform, indicated by the drawn line. The lattice, indicated by the “dots”, with spacing a is also uniform. Below T_P , the lattice is modulated and a Charge-Density Wave (CDW) forms. At the Fermi energy, a gap 2Δ opens. This figure shows the lattice modulation for a commensurate CDW.

For both superconductors and CDWs, the time and spatial derivatives of the order parameter are important for the dynamics of the collective states. However, coupling to applied electric fields is different for both collective states. In superconductors, the time derivative of the phase is proportional to the voltage. When comparing superconductors to CDWs, the roles of current and voltage are exchanged. The time derivative of the CDW phase $\delta\phi(x,t)/\delta t$ is proportional to the CDW current. The energy of incommensurate CDWs is independent of the phase $\phi(x,t)$ and application of a small dc electric field results in motion of the CDW. In the absence of impurities, this would lead to a Fröhlich supercurrent [6].

Fröhlich superconductivity has not been found experimentally, because CDW materials contain defects and impurities with which the CDW interacts. This interaction of the CDW with impurities leads to CDW pinning and a threshold electric field E_T is needed for CDW motion. The onset of CDW motion can be seen in transport measurements as a drop in the differential resistance at E_T , see Fig. 1.2. The linear resistance at zero bias is due to normal charge carriers and/or quasi-particles excited over the Peierls gap. Above E_T , the CDW's contribution to the current I_{CDW} can be found in Fig. 1.2 as the difference between the total current I and the normal current I_N : $I_{CDW} = I - I_N$.

1.2 Single-particle model

A simple model to describe the occurrence of a threshold field is the single particle model. This model treats the CDW condensate as a rigid object (a particle) in a periodic washboard potential. The washboard potential is caused by the overall effect of impurities and is modelled as

$$U(x) = \frac{m^* \omega_0^2}{4k_F^2} (1 - \cos(2k_F x)) \quad (1.4)$$

where m^* is the effective mass, ω_0 the characteristic pinning frequency and x the position of the particle. The driving force of the CDW is the electrical field. The CDW is damped by interaction with quasi-particles. These forces yield the equation of motion for the single particle

$$\frac{d^2 x}{dt^2} + \frac{\gamma_0}{m^*} \frac{dx}{dt} - \frac{\omega_0^2}{2k_F} \sin(2k_F x) = \frac{e}{m^*} E_x \quad (1.5)$$

where γ_0 is the damping coefficient and E_x is the electrical field applied in the chain direction. Because the effective mass m^* of the CDW is large, typically $m^* = 100 - 2000 m_e$, with m_e the electron mass, the inertial term is small compared to the other terms and is usually neglected. Then, the equation is analogous to that of an overdamped pendulum. Applying an electrical field effectively tilts the washboard potential, see Fig. 1.3. For

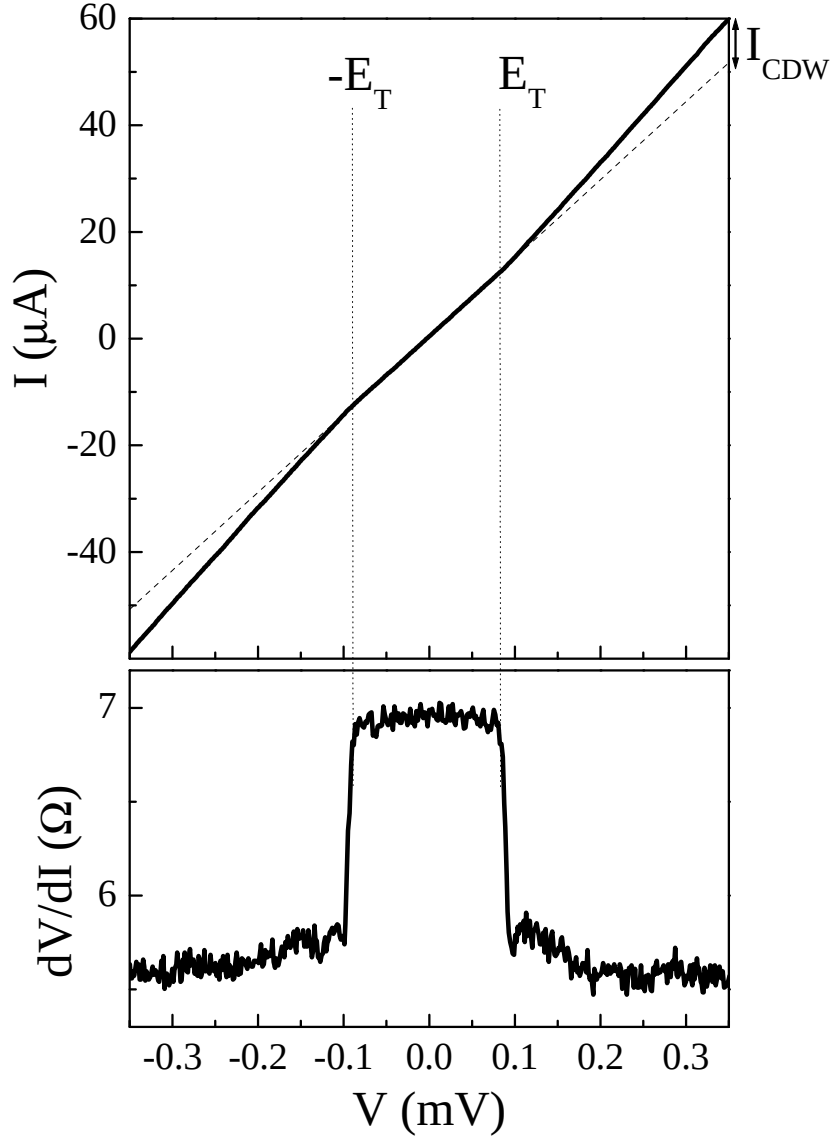


Fig. 1.2: Typical current-voltage IV characteristic of the CDW conductor NbSe_3 at $T = 120$ K and its corresponding differential resistance curve dV/dI . The dashed line depicts the linear resistance due to normal charge carriers and/or quasi-particles excited over the Peierls gap. The dotted lines show where non-linear conduction sets in at the threshold field E_T . This onset of non-linear conduction is clearly seen in the dV/dI as a sharp drop of the differential resistance.

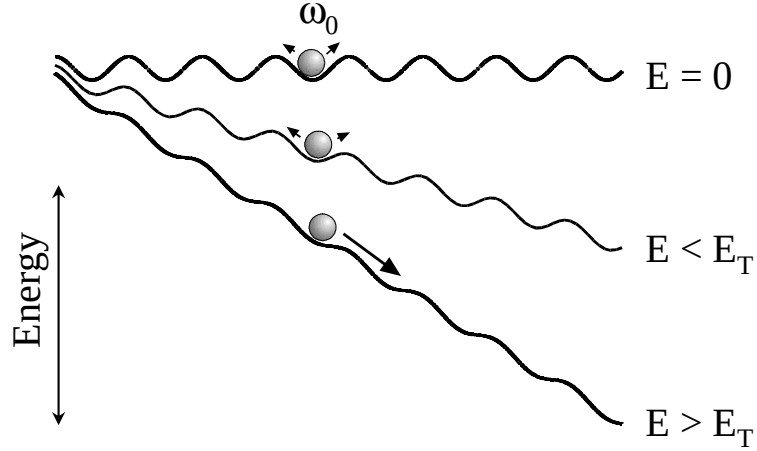


Fig. 1.3: Schematic drawing of the single particle model for CDW conduction. The CDW is represented by the ball. The ball oscillates with frequency ω_0 in the periodic potential. An electric field tilts the potential landscape. Above the threshold field, the particle gets out of a trench and is depinned. The ball rolls down like rolling down a staircase. The frequency with which the particle rolls down the slope is the narrow band noise frequency.

small fields, the particle remains in a trench and the CDW is pinned. Above a threshold field $E_T = m^* \omega_0^2 / 2ek_F$, the tilt is big enough to get the particle out of the trench. The CDW slides through the potential as if rolling down a staircase. The particle moves from step to step in this staircase and a particular frequency can be associated with this periodic motion, known as the “narrow band noise” (NBN) frequency $f_{NBN} = (1/\lambda_{CDW})dx/dt$.

1.3 CDW pinning

The single-particle model considers the CDW as a rigid medium and as such the CDW is completely coherent. A more elaborate model for CDW motion is proposed by Fukuyama, Lee and Rice (FLR) [7, 8]. The model is in essence very similar to the Larkin model for pinning of flux line lattices by weak point disorder [9]. In the FLR model, the CDW is an elastic medium that can adjust its phase ϕ (Eq. 1.1) in the vicinity of impurities and defects. CDW motion is possible when a force is applied large enough to overcome a threshold value, which results from the balance of elastic and impurity pinning energy.

The FLR model distinguishes two regimes; strong and weak pinning. In the strong pinning regime, the impurity pinning force is larger than the elastic force in the CDW. The CDW phase is fully adjusted at each impurity.

In the weak pinning regime, the elastic forces are larger than the impurity pinning force. The CDW phase is adjusted to many randomly distributed impurity sites with a length scale over which the phase varies by a wave length. This length scale is known as the phase coherence length l_ϕ . l_ϕ depends on the impurity concentration and the pinning properties of the type of impurity. Numerical estimates of the phase coherence lengths are on the order of micrometers, which have been corroborated by experiments. In Chapter 5, measurements of finite-size effects are described on samples with sizes smaller than l_ϕ .

1.4 CDW deformation, strain and phase slip

The elastic properties of the CDW naturally lead to deformations of the CDW when a force is applied to it. Such deformations locally change the CDW wave vector by δq with respect to the non-deformed CDW wave vector $q = 2k_F$. A change of the wave vector results in a relative change of the charge density by $\delta q/q$. Electron neutrality dictates a balance between $\delta q/q$ and the number of quasi-particles excited over the Peierls gap. Hence, CDW deformations lead to a change in the quasi-particle conductivity. Especially in CDW materials with a fully gapped Fermi surface, a small deformation can lead to a considerable change in conductivity. This effect is sensitive to temperature, because the number of excited charge carriers decreases strongly with decreasing temperature. Therefore, the largest changes in conductivity are found at low temperatures. The effect of deformations on the conductivity of a fully gapped CDW material is similar to the effect of doping on the conductivity of ordinary semiconductors. In analogy with semiconductors, this effect of CDW deformations is sometimes called “strain-induced doping”.

Elastic forces in the CDW also play an important role in the process of current injection, not considered in the FLR model. Current conversion must occur near electrical current contacts, where electrons from a metallic contact are transferred to the CDW condensate. Ramakrishna *et al.* [10, 11] modelled CDW current conversion as a strain-induced phase-slip process, similar to phase slip in narrow superconducting channels [12] and superfluids [13]. Motion of the elastic CDW causes strain in the CDW; compression near one current contact and stretching near the other. Phase slip processes remove this strain by creating a local amplitude defect, thereby removing or adding a complete CDW wave front. The rate at which wave fronts are added and removed is determined by the magnitude of the strain in the CDW. The strain needed for phase slip processes can be detected in transport measurements as an additional voltage V_{ps} , the phase slip voltage, between the current contacts.

1.5 The Charge-Density wave materials NbSe₃ and o-TaS₃

The experiments presented in this thesis are carried out on two different types of CDW conductors, namely NbSe₃ and orthorhombic TaS₃ (o-TaS₃). The NbSe₃ and o-TaS₃ wires used in our experiments have been synthesized by R.E. Thorne at Cornell University and they are the purest available (residual resistance ratios of up to 400 for NbSe₃). These materials grow in ribbon-like whiskers, NbSe₃ with typical dimensions of 10 mm $\times$ 1 μ m $\times$ 10 μ m and o-TaS₃ with dimensions of 10 mm $\times$ 10 μ m $\times$ 10 μ m. The crystal structure consists of weakly coupled chains, resulting in a quasi one-dimensional electron spectrum. The elasticity and resistivity are anisotropic, where the electrical resistivity is lowest parallel to the chains.

1.5.1 NbSe₃

NbSe₃ has two Peierls transitions, one at $T_{P1} = 142$ K and one at $T_{P2} = 59$ K. NbSe₃ is metallic at room temperature and stays metallic at low temperatures since part of the Fermi surface remains ungapped, see Fig. 1.4. The Peierls transitions can be clearly seen in Fig. 1.4 as dips in the temperature derivative of $\ln R(T)$. The metallic behavior of NbSe₃ at liquid He temperature is used to estimate the impurity concentration with the use of Mathiessen's rule [14]. Typically for pure bulk crystals, the residual resistance ratio $r_R = R(T = 295 \text{ K})/R(T = 4.2 \text{ K}) = 200 - 400$ [15].

The unit cell of NbSe₃ is monoclinic and contains six chains, divided into three types. The chains are parallel to the crystallographic b-axis. The three types can be distinguished by the strength of the outer Se-Se bonds and the spacing between Se atoms in each prism, indicated in Fig. 1.5. Chain III has the closest chalcogen spacing and is associated with the first transition at $T_{P1} = 142$ K. Chain I, with intermediate chalcogen spacing, is associated with the second transition at $T_{P2} = 59$ K. Chain II has the largest chalcogen spacing and does not form a CDW. NbSe₃ exhibits a half-unit cell staggering of the two CDW chains [16]. The dimensions of the unit cell are: $a=10.009 \text{ \AA}$, $b=3.4805 \text{ \AA}$, $c=15.629 \text{ \AA}$ and $\beta=109.47^\circ$. Typical threshold fields for NbSe₃ are 5-50 mV/cm. The threshold field for the second transition is an order of magnitude lower than for the first transition. At temperatures below T_{P2} , the CDW of the first transition remains pinned and only the CDW of the second transition slides. The room temperature resistivity along the b-axis is $1.86 \text{ } \Omega\mu\text{m}$ [15]. Perpendicular to the b-axis the room-temperature resistivity of NbSe₃ is 10 to 100 times higher.

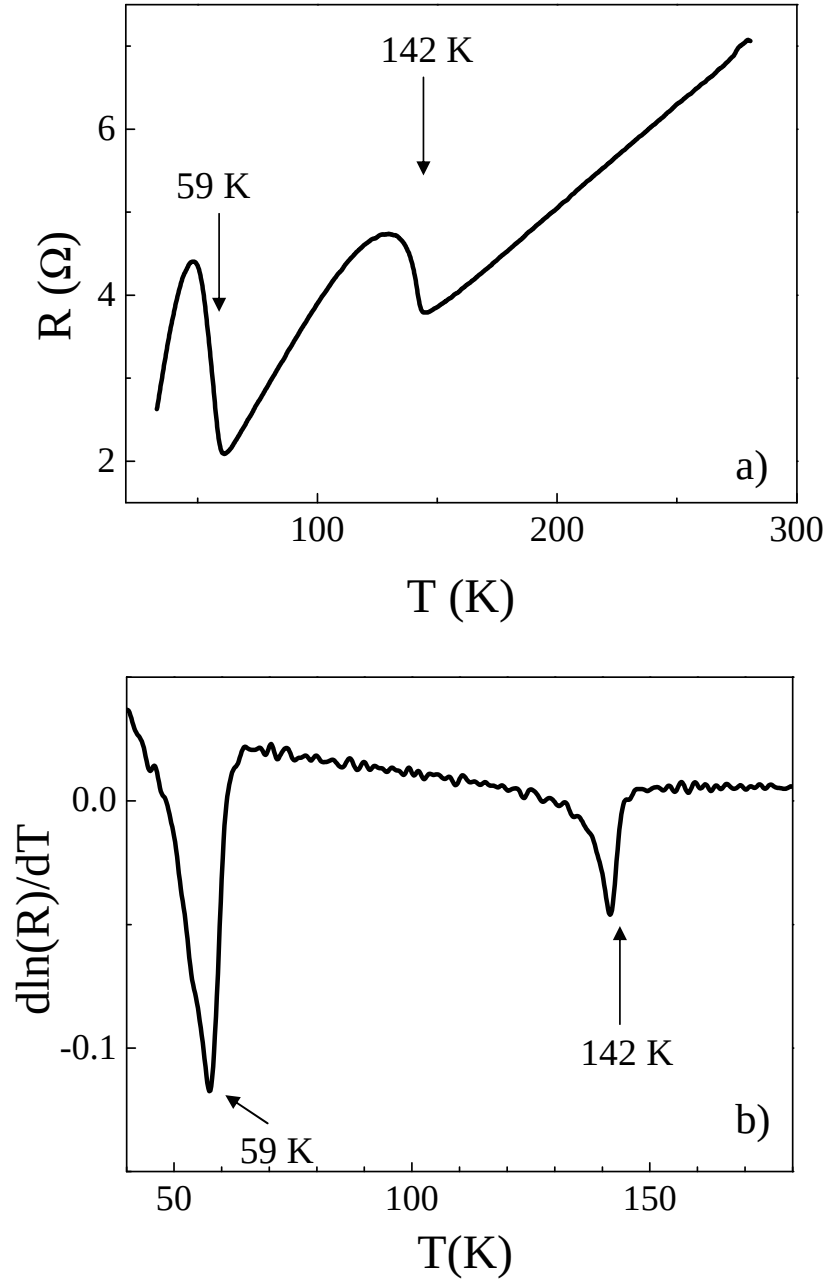


Fig. 1.4: a) Resistance versus temperature curve of a NbSe₃ sample. b) The $d\ln(R)/dT$ shows two clear dips at both Peierls transitions.

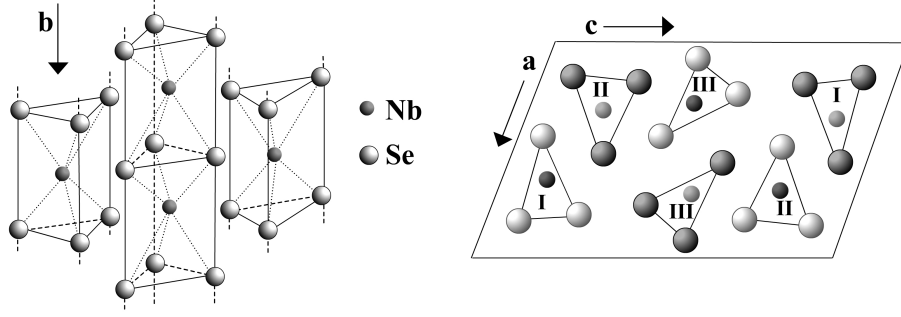


Fig. 1.5: The chain structure of NbSe₃. The unit cell of NbSe₃ has three types of chains (I, II, III). The darker circles denote the atoms in plane and the brighter circles denote the atoms out of plane.

1.5.2 TaS₃

TaS₃ can be grown in two different crystal structures, monoclinic and orthorhombic. The orthorhombic type is used in the research described in this thesis. Orthorhombic TaS₃ (o-TaS₃) is metallic at room temperature and has a single CDW transition at $T_P = 218$ K. The Fermi surface is fully gapped below the transition temperature, and the resistance versus temperature shows semiconducting behavior below the Peierls transition, as shown in Fig. 1.6. The threshold electric field for sliding is on the order of 1 V/cm at $T = 120$ K.

The unit cell of o-TaS₃ contains 24 chains, see Fig. 1.7. The chains are parallel to the c-axis. The dimensions of the unit cell are $a=36.80$ Å, $b=15.18$ Å, $c=3.34$ Å. In contrast to NbSe₃ and monoclinic TaS₃, o-TaS₃ does not exhibit half-unit cell staggering; the Ta atoms are all in plane [17]. The CDW can not be attributed to particular chains in the unit cell as has been suggested for NbSe₃ [18]. The room temperature resistivity of o-TaS₃ is $3 \Omega\mu\text{m}$ along the chains [19]. Perpendicular to the c-axis the resistivity at room temperature is about two orders of magnitude higher and about three orders higher at $T = 80$ K [20].

1.6 Mode-locking

The occurrence of NBN in CDW conductors is introduced in Section 1.1 by the single particle model. The NBN frequency is proportional to the CDW current:

$$f_{NBN} = \frac{j_{CDW}}{n_c e \lambda_{CDW}} \quad (1.6)$$

with j_{CDW} the CDW current density, n_c the concentration of condensed carriers. NBN can be measured by applying a dc current to a CDW conductor

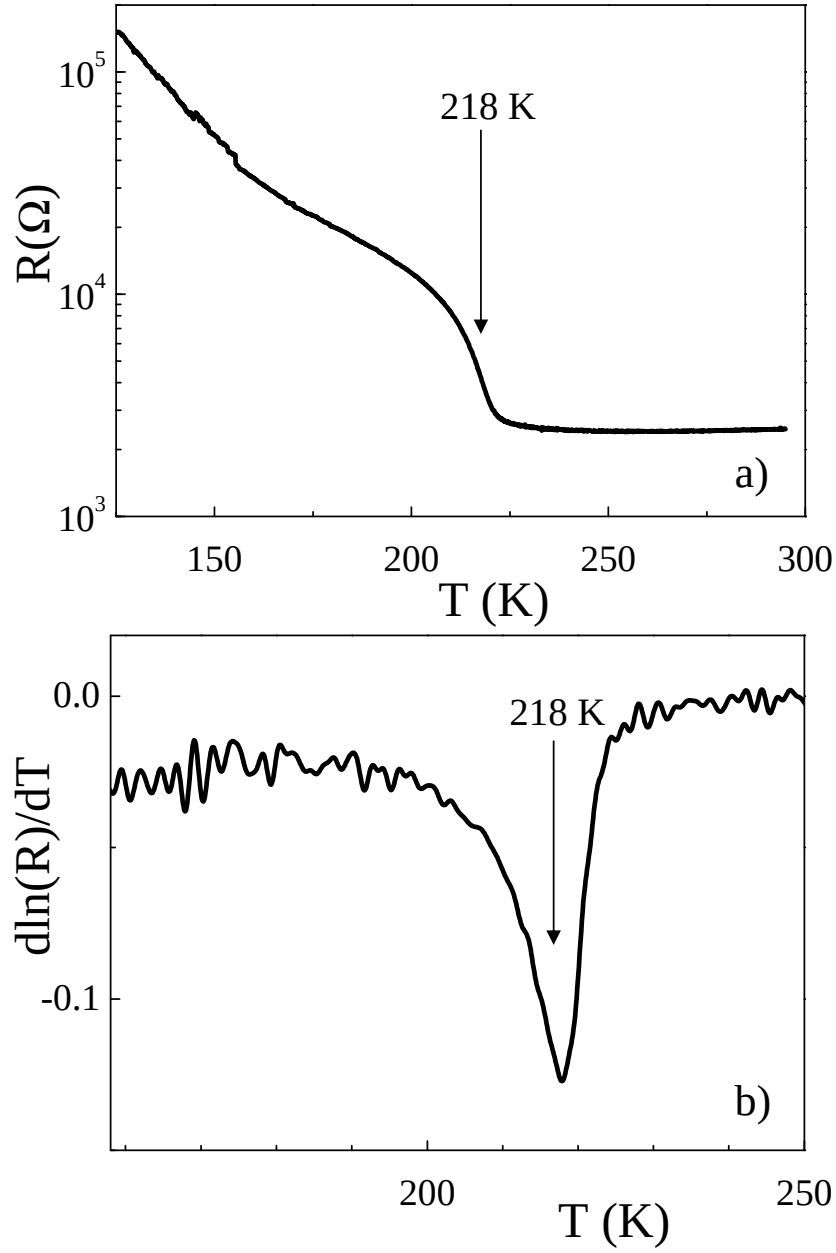


Fig. 1.6: a) Resistance versus temperature curve of a α -TaS₃ sample on a log-linear scale. b) The $d\ln(R)/dT$ shows a clear dip at the Peierls transition.

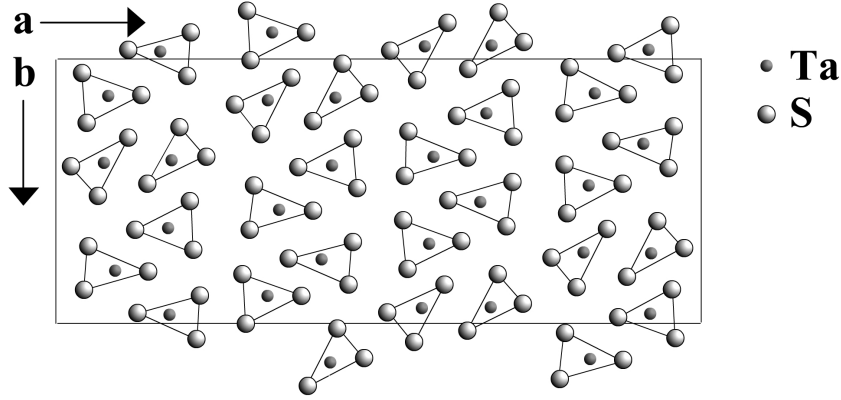


Fig. 1.7: The unit cell of o-TaS₃. The chains are along the crystallographic *c*-axis. The sulphur atoms are in plane and the tantalum atom are out of plane.

large enough for the CDW to slide. NBN will show up in a spectral analysis of the voltage as a sharp peak at the NBN frequency, usually between 1-100 MHz. The narrower the peak, the more coherent the CDW slides.

Transport measurements where a dc current is applied simultaneously with an ac signal on the order of the NBN frequency are called “Shapiro step measurements”. Such measurements have first been performed on Josephson junctions, and later on vortex lattices and CDW conductors. All these systems can be crudely described by the single-particle model. The steps in the velocity-force ($dx/dt - F$) characteristic come from the solution of the differential Eq. 1.5, with $eE_x = F + \delta F \sin(2\pi f \cdot t)$. The ac signal with frequency f oscillates the tilt of the washboard potential and the marble gets trapped in a trench when the frequency of the oscillation is close to the NBN frequency.

Figure 1.8 shows a Shapiro step measurement on a CDW conductor. The CDW mode-locks to the external ac signal when its NBN frequency is close to the frequency f of the ac signal. The CDW velocity is fixed by the applied frequency and thus CDW mode-locking appears as voltage steps in the current-voltage characteristic and peaks in the differential resistance. When the voltage steps are fully horizontal (and equivalently the peaks in the differential resistance reach the same level as for zero-bias), the CDW is completely mode-locked to the external frequency. This means that the CDW moves coherently throughout the entire cross section of the sample.

Shapiro step measurements can be used to determine the number of parallel chains in the cross section of a sample. To do this, a linear fit is made to the height of the voltage step ΔI_{CDW} , shown in Fig. 1.8, as a function of the applied frequency. The slope of the fit is $0.238 \mu\text{A}/\text{MHz}$. For NbSe₃ at $T = 120$ K, two chains per unit cell each contribute two electrons

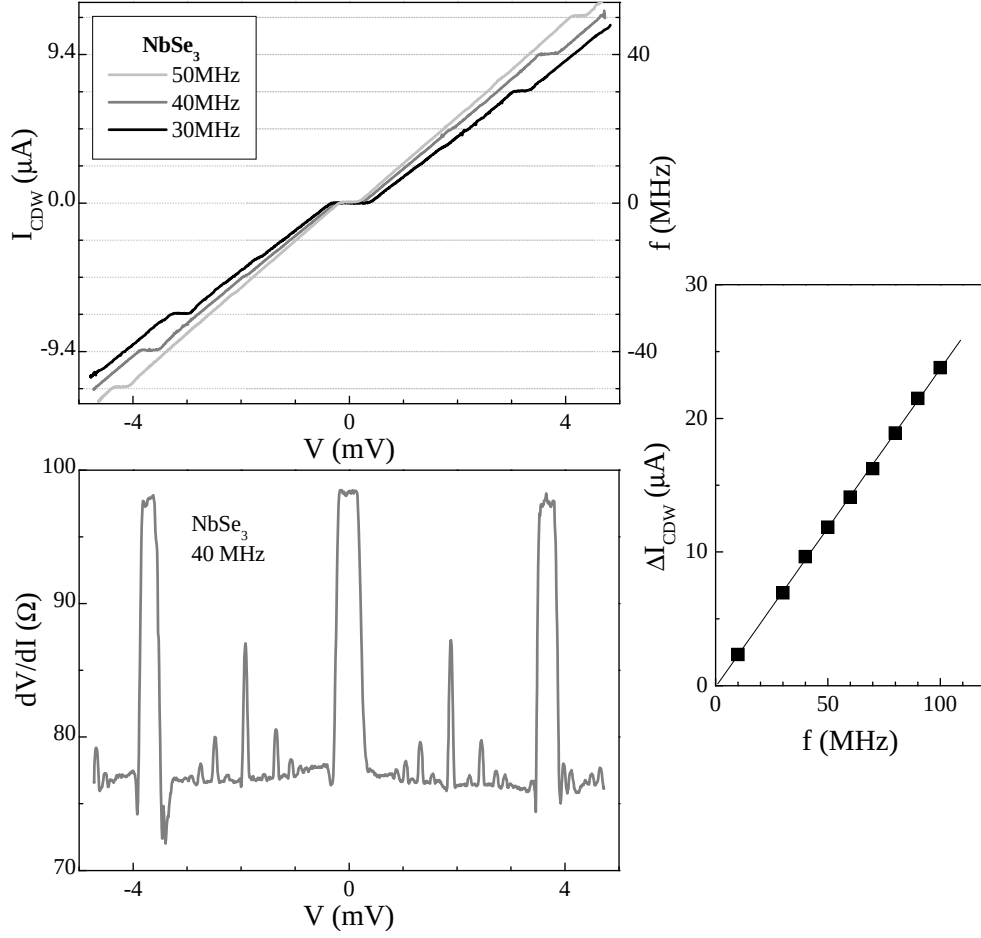


Fig. 1.8: Shapiro step measurement on NbSe₃ at $T = 120$ K. The current-voltage characteristics show clear voltage steps where the CDW mode-locks to the external frequency. These voltage steps correspond to the peaks in the differential resistance. The step height ΔI_{CDW} is proportional to the applied frequency f .

to the CDW current and therefore

$$\frac{\Delta I_{CDW}}{f} = 2eN \quad (1.7)$$

with N the number of chains in the cross section of the sample. The cross sectional area A can be calculated from the number of chains; two out of six chains in NbSe₃'s unit cell $A_0 = 1.48 \text{ nm}^2$ contribute to the CDW current and thus $A = A_0 \times N/2$. Using $\Delta I_{CDW}/f = 0.238 \text{ } \mu\text{A}/\text{MHz}$, $N = 0.744 \times 10^6$ and $A = 0.55 \text{ } \mu\text{m}^2$ for the sample shown in Fig. 1.8.

1.7 Fabrication of microscopic Charge-Density Wave devices

Reduction of sizes in superconductor devices has revealed many interesting phenomena, such as the Josephson effect, the even-odd effect in superconducting Coulomb islands, Cooper pair tunneling, and Andreev reflection. These effects were studied in detail, because reliable fabrication techniques have been developed to make small superconductor devices. One important contribution to the research of the microscopic regime of superconductors was the development of its thin film growth.

This thesis is about the effects that occur when the sizes of CDW systems are systematically decreased to microscopic dimensions. In order to reveal these finite-size effects, new fabrication need to be developed. Previous attempts by others have been undertaken to grow thin film of the CDW conductors NbSe₃ [21] and Rb_{0.30}MoO₃ [22]. A major problem of the film growth is the granularity of the films. The grain size is too small to systematically study microscopic effects in these films. Recent advances in technology, however, made it possible to access the microscopic regime in CDW conductors using single crystals instead of thin films.

This Section describes several fabrication techniques that have been developed in this thesis to make microscopic CDW devices using single crystals.

1.7.1 Cleaving of samples by hand

CDW conductors that grow in strands, such as the NbSe₃ strands shown in Fig. 1.9, can be cleaved by hand using a knife or a pair of tweezers. A large wire is put on a glass plate with a pair of tweezers and crushed at one of the ends with a knife. The end of the wire frays and splits into many small wires and one of these wire is selected by pulling on it with a pair of tweezers. This technique of selecting the right sample requires practise and experience for successful results. The advantage of this technique is that the small samples are cleaved from a large wire and have a clean surface to make electrical contact to. Typical widths of the cleaved wires are $1 \text{ } \mu\text{m}$ and thicknesses of

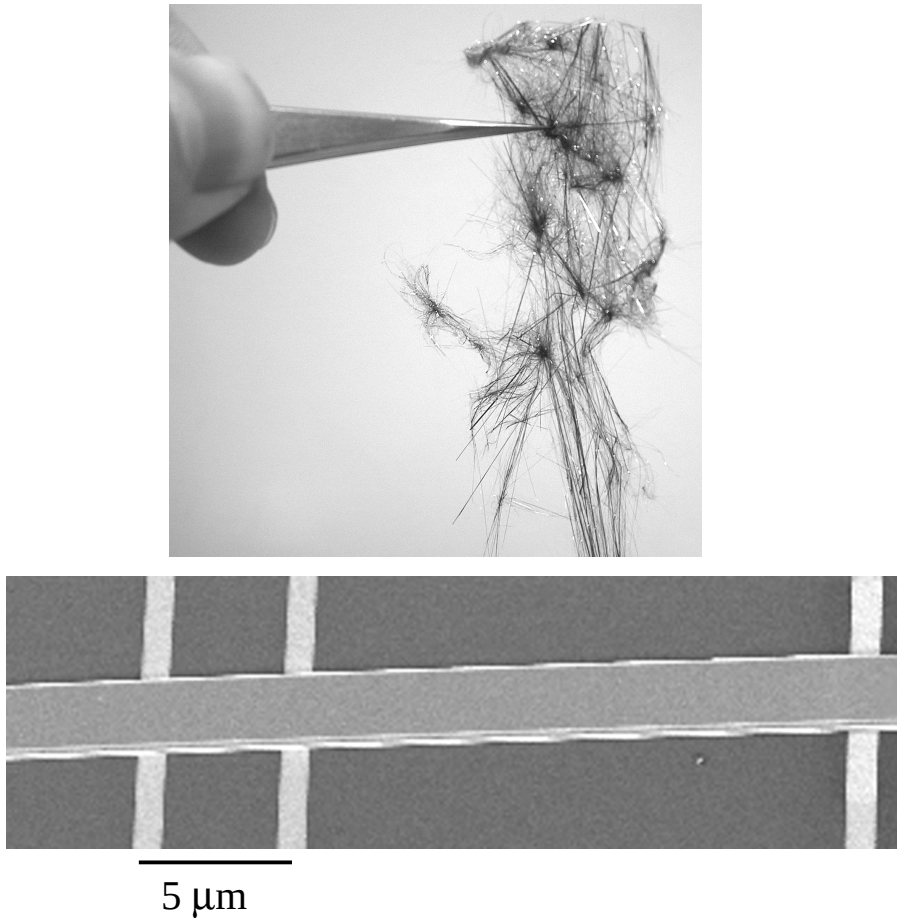


Fig. 1.9: Strand of NbSe₃ held with a pair of tweezers. The Scanning Electron Microscope image shows a NbSe₃ wire cleaved from a large strand and put on top of three Au contacts.

several 100 nm. The surface of these small wires rarely contain steps. The wire is put on a substrate with predefined metal contacts, as shown in the Scanning Electron Microscope image in Fig. 1.9. This technique is used in all Chapters except Chapter 6

1.7.2 Small probe spacing with electron beam lithography

Electrical contact can be made by putting a small crystal onto a contact pattern on a flat substrate. Small wires stick to the substrate, presumably by electrostatic forces, when they are flat on the surface. In this thesis, electron beam lithography (EBL) has been used to define a contact pattern on the substrate. Then a metal is evaporated on the substrate. Many metals, including superconductors, can be used for evaporation, but Au is a proven choice for making good electrical contact to CDW wires. The resolution of this technique allows for Au contacts down to 100 nm wide, with a pitch of 300 nm, see Chapter 4. The small width and pitch of the contacts enables the study of the microscopic dynamics of CDW conductors.

1.7.3 Focused-Ion Beam etching

A Focused-Ion Beam (FIB) can be also used to etch structures in CDW materials on a submicron scale. An ion beam is scanned in a user-defined pattern over a CDW crystal. The ion beam bombards the CDW material with Ga ions, which remove the CDW material by vaporization. The resulting structure can be imaged with a low-current ion beam or electron beam *in situ*. The fabrication technique is very flexible and many different geometries can be fabricated with dimensions down to 100 nm. The required time for FIB fabricated structures is very short, making this a very convenient fabrication technique. A disadvantage is the local implantation of Ga ions in the material, resulting in damage of the surface of the etched structure. This fabrication technique has been used for the research described in Chapters 3, 5 and 7 to fabricate narrow channels, CDW junctions and constrictions. The FIB can also be used for local deposition of metals. A precursor gas of a metal complex is cracked by the Ga ion beam. This local deposition is used for creating Pt-CDW heterostructures [23].

1.7.4 Ultra-sonic cleaving of wires

We have developed a new fabrication method for creating very small CDW crystals on the order of tens of nanometers. The fabrication method starts with selecting a number of wires, a few millimeters long, from a batch of CDW crystals. These are put in a bottle with pyridine or chloroform to create a suspension of small crystals. Pyridine is chemically inert and is commonly used to create a suspension of carbon nanotubes. The bottle is put in an ultra-sonic bath. The ultra-sonic treatment produces a suspension

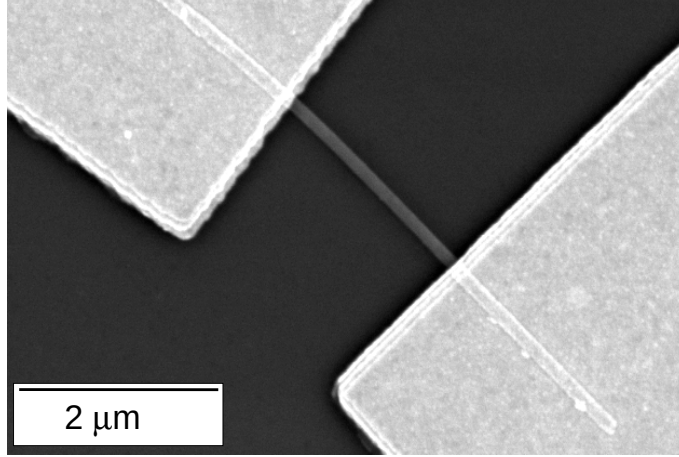


Fig. 1.10: A Scanning Electron Microscope image of a NbSe_3 nanowire with two gold contacts on top of it.

of small crystals. The pyridine ensures that the small crystals do not cluster and stay dispersed. The larger crystals gradually sink to the bottom of the bottle and the smallest crystal remain at the top of the suspension. A drop of the top of the suspension is put on a degenerately doped Si-O substrate with pre-defined markers. The pyridine is carefully blow-dried leaving behind the CDW nanowires. The nanowires are imaged with a microscope to determine their location with respect to the pre-defined markers. EBL is used to define the electrical contacts to the nanowires, see Fig. 1.10. This fabrication technique has been used in Chapters 5 and 6 to fabricate CDW nanowires with dimensions down to 20 nm.

1.7.5 Mechanically controlled Break-junctions of Charge-Density Wave wires

Mechanically controlled break-junctions (MCBJ) have been used to make atomic wires of metals [24]. The metal wire is put on a phosphor-bronze substrate, which is clamped in a three-point configuration. A moveable rod is pushed against the middle of the substrate to bend the metal wire up to the point where the contact consists of a single atom. In this thesis, the same technique is used with CDW conductors to create CDW junctions in analogy with superconducting weak links [5]. The CDW wire is stretched in the MCBJ configuration and constrictions are formed in the wire. The resistance of the CDW constrictions is mechanically controlled with the moveable rod, see Chapter 7.

1.8 Microscopic models for Charge-Density Waves

Experimental studies on small CDW crystals have stimulated the development of new models describing the microscopic aspect of CDW motion. Observed finite-size effects include the dependence of the phase slip voltage on current contact separation [25], the Aharonov Bohm effect in CDW sliding through columnar defects in NbSe₃ [20] and mesoscopic oscillations of the threshold field [26]. A recent review of these and other finite-size effects in CDW conductors can be found in Ref. [27].

In Chapter 4 of this thesis, measurements of local CDW dynamics have led to a semi-conductor model for quasi-1D conductors. It uses the electrical potential to describe CDW motion and the chemical potential to describe the motion of quasi-particles. These individual potentials are needed to describe the measured transport properties on a microscopic scale. The influence of local variations in defect or impurity densities is washed out over large scales, but very prominent on a small scale.

For samples with small probe spacing, geometrical effects are an important consideration. The potential distribution close to a current contact is inhomogeneous and this is particularly pronounced for anisotropic materials. In Chapter 2, experimental verification of a previously proposed model for the potential distribution close to a lateral current contact is given. Measurements of the voltage distribution close to current contacts are in good agreement with the proposed model.

The reduction of sizes also has a strong effect on the threshold field for sliding, which is the subject of Chapter 5. The sizes of CDW wires are reduced such that the wires are in the 1D weak pinning limit. The measurements are discussed in the framework of the FLR model for pinning.

Additionally, the reduction of sizes has revealed a new effect on the low-bias conductivity at low temperatures as well. A systematic study of the low-bias conductivity on the sizes is described in Chapter 6. The conductivity follows a power-law on temperature and voltage characteristic for 1D electron systems. Several models are proposed to describe the measurements including Environmental Coulomb Blockade and Wigner crystallization.

Transport measurements of CDW junctions in Chapter 7, made using newly developed fabrication techniques, required a new description of tunneling in CDW. The new description uses the semiconductor model for electron tunneling in superconductors. To describe the observed features, the particular band structure of the used CDW crystals is incorporated in this model.

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2. Electric-field distribution near current contacts

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Electric-field distribution near current contacts of anisotropic materials
Physical Review B **65**, 033403 (2002)

Abstract

We have measured the non-uniformity of the electric field near lateral current contacts of the Charge-Density wave materials NbSe₃ and o-TaS₃. In this contact geometry, the electric field increases considerably near a current contact. Fitting our data to an existing model yields values for the conduction anisotropy and a characteristic longitudinal length scale. This length scale is on the same order as the phase-coherence length in Charge-Density wave devices.

2.1 Geometrical effects in anisotropic conductors

The Charge-Density wave materials used in the experiments reported in this thesis are anisotropic conductors with a chain-like structure. The experimental study of the CDW dynamics usually involves transport measurement along the chain direction. Electrical contact is most often made by connecting metal electrodes to the crystal at various positions on the top or bottom side (face) of the crystal [1] or, more recently, by etching side contacts in the crystal itself [2]. Most transport studies have been performed on samples with contact spacings of $10\text{ }\mu\text{m}$ and larger.

Samples used in studies of the mesoscopic regime of CDW dynamics [2, 3] have small current contact spacings (smaller than $10\text{ }\mu\text{m}$). Non-uniformity of the electric field near current contacts should be taken into account when reducing the contact spacings. This has been largely neglected in experiments so far, because contact separations were large compared to the typical length scale for non-uniformity of the electric field near current contacts. As the length scale for mesoscopic phenomena is on the order of micrometers in the longitudinal direction, it is essential for studying the mesoscopic regime to reduce contact spacings to this scale. Since both types of lateral contacts mentioned above apply current in the transverse direction, a region of non-uniform electric field exists in the vicinity of current contacts. This non-uniformity is known under the name “fringing effects”. In previous experiments so far, measurements of fringing effects were limited by perturbing contacts and large contact separation.

In case of anisotropic materials, fringing effects are particularly pronounced. In such materials, the length scale over which fringing effects are important is $\sqrt{A}$ times larger than in the isotropic case. Here, $A = \sigma_{\parallel}/\sigma_{\perp}$ the conductivity anisotropy, which is the ratio of the conductivity along $\sigma_{\parallel}$ and perpendicular $\sigma_{\perp}$ to the chains along the crystal length.

2.2 Sample characterization

We have measured the electric field distribution on submicron length scales in the longitudinal direction. We find good agreement with existing models which indicate that fringing effects are important up to distances of $t\sqrt{A}$ from the current contact, where t is the crystal thickness. From our data, we can deduce the corresponding A . We find it to be ~ 100 for NbSe_3 along the a^* -axis and ~ 1000 for o-TaS_3 perpendicular to the c -axis at $T = 120\text{ K}$; see Chapter 1 for a description of the crystal structure of both materials. Our measurements are performed in the pinned state, so that our data concerns geometrical effects only, and does not explore the complicated current dependent field profiles that develop when the CDW depins [4, 5].

Experiments are performed on crystals of NbSe_3 and o-TaS_3 . Both mate-

Tab. 2.1: Sample characteristics at $T = 120$ K. The cross sections S are deduced from room temperature resistance measurements; those from Shapiro step measurements are in brackets. No Shapiro step measurements have been performed on sample TaS₃-A. The values of $t\sqrt{A}$ are deduced from the fit parameter Y in Eq. 2.3. From the thickness t of the crystals, values of the anisotropy A have been calculated. The error margins of A are also listed.

	S (μm^2)	Y (10^{-3})	$t\sqrt{A}$ (μm)	t (μm)	A
NbSe ₃ -A	0.54 (0.54)	20	3.9	0.3	170 ± 50
NbSe ₃ -B	0.20 (0.21)	45	1.7	0.2	70 ± 40
TaS ₃ -A	1.52 (–)	2.8	28	0.7	1600 ± 320
TaS ₃ -B	0.48 (0.55)	12	6.5	0.3	470 ± 235

rials have an anisotropic chain-like structure and exhibit CDW states at low temperatures. NbSe₃ exhibits CDW transitions along the crystallographic b-axis at $T_{P1} = 145$ K and $T_{P2} = 59$ K, while part of the conduction electrons remains uncondensed providing a metallic single-particle channel down to the lowest temperatures. From literature, the anisotropy of NbSe₃ is ~ 10 -20 along the c-axis [6] and estimated to be about 100 along the a*-axis. o-TaS₃ has a single transition at $T = 220$ K below which all conduction electrons are condensed. The literature value of the anisotropy of o-TaS₃ is typically 1000 and increases as the temperature is lowered [7].

Electrical contact is made by placing the crystals on arrays of 50 nm thick gold strips defined with electron-beam-lithography. The width of the strips is 100 nm and the smallest separation is 300 nm. The narrow gold strips are used to inject current and to measure voltage. The position of the crystal is fixed by putting a drop of glue (ethyl-cellulose dissolved in ethyl-acetate) on top of it. In case of o-TaS₃, reliable ohmic contacts are obtained by heating the substrate to 120-130 °C up to an hour before putting the glue down. In case of NbSe₃ crystals, heating the substrate to 80 °C prevents the tiny crystals from floating in the glue solvent. The contact resistances at $T = 120$ K are on the order of 2 k Ω for the NbSe₃ samples and on the order of 100 k Ω for the o-TaS₃ samples.

Cross sections are deduced from measurements of the resistance R for different voltage probe spacings L at room temperature using resistivity values of 3 $\Omega\mu\text{m}$ for o-TaS₃ [8] and 2 $\Omega\mu\text{m}$ for NbSe₃ [9]. We have also used Shapiro step measurements to determine the cross section, as described in Chapter 1. These measurements involve a combined dc and ac current, such that the narrow band noise frequency produced by CDW sliding mode-locks to the external ac frequency. At $T = 120$ K, we found complete mode-

locking on three samples, indicating high quality samples with flat surfaces. The cross sections deduced from Shapiro step measurement agree very well with those deduced from the resistance measurements at room temperature, see Table 2.1. The width of the crystals is determined under an optical microscope, from which we can deduce the thickness of the crystals. The error in t is estimated to be up to 25%. Such a high error is because the width of the crystals was determined with an optical microscope. A scanning electron microscope could not be used to measure the width because non-conducting ethyl-cellulose was put on top of the crystals.

2.3 Measurements of the electric field distribution

Fringing effects were measured in two different o-TaS₃ and two different NbSe₃ samples. These measurements regard the pinned state of the CDW and are performed in a four-probe current-biased configuration. The normal carrier conductivity of o-TaS₃ is very low at low temperatures so that only measurements above 90 K are performed. NbSe₃ has a metallic channel down to liquid helium temperature and measurements have been performed for $T > 25$ K.

To measure fringing effects, we have deduced the linear resistance from current-voltage (IV) characteristics for several different current contact pairs. The current is injected from one of the narrow leads to a big gold pad on the side, 250 μm away to minimize the influence of fringing effects of the other contact. The IV -characteristics are measured by slowly sweeping the current and measuring voltage at several distances from the current contact. The distance x is the distance from the middle of the current contact to the middle of the voltage-probe pair (see Fig. 2.1a). We have measured the voltage with probe pairs between current contacts, as well as beyond contacts. When the voltage probes are in the vicinity of one of the current contacts, the linear resistance is larger than the linear resistance when current is injected far away. This higher resistance due to the non-uniform electric field near the current contact is called the “spreading resistance”.

Measurements of the linear resistance of segments R_S between voltage probes of samples NbSe₃-A and TaS₃-A at $T = 120$ K are plotted in Fig. 2.1b. A negative sign of the distance x represents the distance of the voltage probes beyond the current contacts. The linear resistance R_S is normalized to the linear resistance R_0 . R_0 is the resistance of a segment when current is injected far ($> 200 \mu\text{m}$) from the voltage probes and fringing effects are thus negligible.

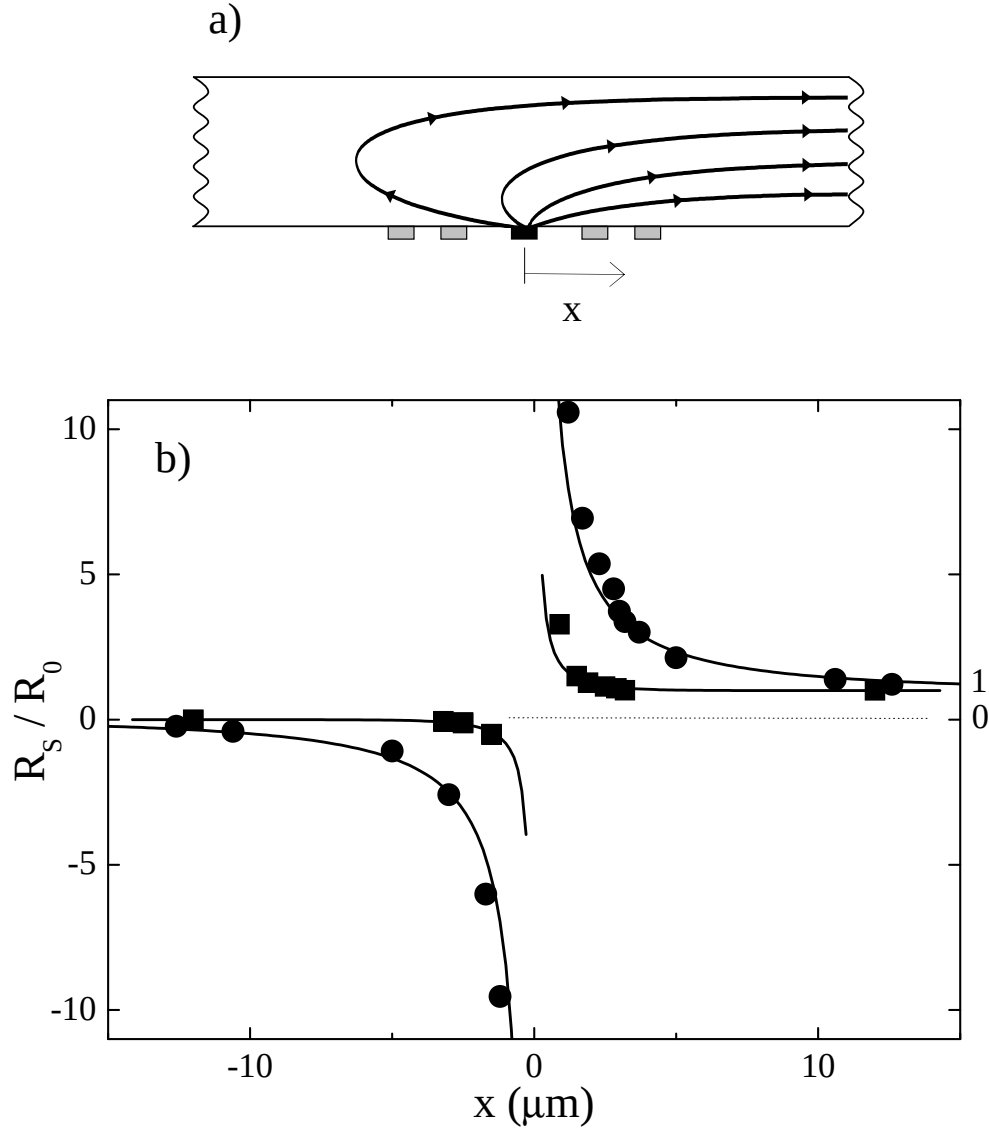


Fig. 2.1: a) Side-view drawing of part of a crystal with contacts at the bottom. The black contact is the current contact. Lines with arrows depict the numerically calculated paths of the current. The current is uniform at the right side of the drawing. Beyond the current contact on the left side, there is no net current, but at the bottom current flows in the opposite direction. b) The linear resistance R_S of segments of the crystals as a function of the distance from the current contacts x . The circles denote data of sample TaS₃-A and the squares denote data of sample NbSe₃-A taken at $T = 120$ K. The solid lines are fits of Eq. 2.3 to the data.

2.4 Analysis of the electric field distribution

We have fitted our data to analytical expressions obtained from Borodin *et al.* [10] who discuss the potential profile on the face of the crystal with lateral current contacts. They considered a metal electrode of width l with its middle at $x = 0$. The other contact is at $x = \infty$. The potential $U(x)$ on the bottom face of the crystal ($y = 0$) is

$$U(x) = -E \frac{t\sqrt{A}}{\pi} \operatorname{arccosh} \left| \frac{\cosh\left(\frac{\pi l}{2t\sqrt{A}}\right) - \exp\left(\frac{\pi x}{t\sqrt{A}}\right)}{\sinh\left(\frac{\pi l}{2t\sqrt{A}}\right)} \right| \quad \text{for } |x| > \frac{l}{2}, \quad (2.1)$$

where E is the electric field far from the current contact ($x \rightarrow \infty$). A negative x denotes positions beyond current contacts.

We have performed numerical calculations of the Laplace equation, which agree with the analytical potential profile on the bottom face of the crystal given by Eq. 2.1. Fig. 2.1a shows the current distribution near a current contact as determined from numerically solving the Laplace equation in two dimensions

$$\frac{d^2 U(x, y)}{dx^2} + \frac{d^2 U(x, y)}{dy^2} = 0, \quad (2.2)$$

where $U(x, y)$ is the potential distribution. The direction of x is indicated in Fig. 2.1; y is directed in the thickness direction. The boundary conditions are:

- $U(x, 0) = 0$ for $|x| \leq l/2$
- $\frac{dU(x, 0)}{dy} = 0$ for $|x| > l/2$
- $\frac{dU(x, t)}{dy} = 0$
- $\frac{dU(x, y)}{dx} = E$ for $x \rightarrow \infty$ and $0 \leq y \leq t$

The current is injected perpendicular to the length of the crystal. The electric field just above the contact is very high and the current even flows beyond the contact. As a consequence, the voltage probes beyond the current contacts detect a voltage of opposite sign and therefore the spreading resistance is negative beyond the contacts. However, the net current through the entire cross section is zero beyond current contacts.

We use Eq. 2.1 to derive an estimate for the resistance ratio $\frac{R_S}{R_0}$, which is the quantity obtained from our measurements. Suppose the potential difference between two voltage probes equals ΔU . Then, for small probe spacing L , the potential difference $\Delta U \approx \frac{dU}{dx} L$, where $\frac{dU}{dx}$ is the local electric field taken in the middle of two adjacent voltage probes and the resistance

ratio $\frac{R_S}{R_0}$ equals $\frac{dU/dx \cdot L}{E \cdot L}$. We introduce a dimensionless contact width $Y = \frac{\pi l}{4t\sqrt{A}}$. For narrow contacts ($Y \ll 1$), $\frac{R_S}{R_0}$ can be expressed as

$$\left| \frac{R_S}{R_0} \right| = \frac{\exp(4Yx/l)}{2Y \sqrt{\left(\frac{1 - \exp(4Yx/l)}{2Y} \right)^2 - 1}} \quad \text{for } |x| > \frac{l}{2}. \quad (2.3)$$

The spreading resistance R_S approximates R_0 when measuring far from the current contact, so that $\frac{R_S}{R_0}$ goes to unity for $x \rightarrow \infty$. Beyond the current contact the spreading resistance is negative and goes to zero for $x \rightarrow -\infty$. The assumption that L is small is valid for all our samples. This has been checked by fitting our data to Eq. 2.1 using the exact ΔU and comparing them to the fits to Eq. 2.3.

We have fitted our data with $l = 100$ nm and using Y as the single fit parameter within and beyond contacts. Good fits were obtained on all samples in the temperature range studied, see the solid lines in Fig. 2.1b. The values of Y obtained at $T = 120$ K are listed in Table 2.1 and all are consistent with the assumption $Y \ll 1$.

From the definition of Y the longitudinal length scale $t\sqrt{A}$ and the anisotropy A are deduced. The anisotropy at $T = 120$ K of o-TaS₃ and NbSe₃ is on the order of 10^3 and 10^2 , respectively. The measured anisotropy of the thin samples are smaller than those of the thicker samples. Most likely, this is because a more reliable fit can be made to the data of thicker samples and samples that have a higher anisotropy.

We have also determined Y at other temperatures and Y is approximately temperature independent. To get a better estimate of $A(T)$, thicker crystals and a more accurate determination of the crystal's thickness are needed. In Chapter 3, another technique is used to measure the anisotropy of NbSe₃ in both the crystallographic a*-axis as the c-axis.

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3. Charge-Density wave devices fabricated with a Focused-Ion Beam

Abstract

We have fabricated a variety of Charge-Density Wave (CDW) devices using a focused-ion-beam (FIB) process. The FIB is used to etch any desired geometry in crystals, like constrictions, channels, tears, trenches, zigzag patterns etcetera. We have studied the electrical transport of such devices. This study includes: finite size effects, conduction perpendicular to the chains, geometrical effects and CDW junctions. This Chapter provides a characterization of the devices fabricated with a FIB. We have found complete mode-locking on CDW constrictions, indicating that the high-quality crystal properties are preserved after FIB processing. This makes the process a useful technique to study CDW dynamics on the length scale of the CDW phase coherence length.

3.1 Introduction

A new approach in the fabrication of mesoscopic CDW devices consists of using a Focused-Ion Beam (FIB) to make patterns in CDW crystals [1]. The FIB setup has three modes of operation: etching, imaging and deposition. In the etching mode, a 30 kV gallium ion beam is scanned across the crystal to etch any desired pattern. An image of the patterned structure can directly be obtained by using the same ion beam at low current (1-4 pA). In the deposition mode, the FIB is used to crack a precursor gas (e.g. Pt-complex molecule) resulting in a local deposition of material. This opens a way to make CDW heterostructures. We have made a CDW heterostructure by first etching a gap all the way through a NbSe₃ crystal. Subsequently, the gap is filled in situ with platinum by FIB deposition creating a CDW-Pt-CDW junction. The transport properties of the CDW-Pt-CDW junctions are reported elsewhere [2]. A major advantage of a FIB is that patterning of CDW crystals does not involve numerous lithographic steps and alignment of markers. The result of patterning with a FIB can be observed directly without breaking vacuum. This makes this technique much less time-consuming than Reactive Ion Etching (RIE). In this Chapter, we present transport measurements of CDW devices fabricated using a FIB. We have used NbSe₃ and o-TaS₃ crystals, which are grown with high purity. These two materials are the most widely studied CDW conductors. Processing with a FIB is expected to cause implantation of gallium ions in the crystal lattice. These ions might act as CDW pinning centers. We will show that unintentional damage caused by a FIB is minimal and that the high-quality properties of these materials are preserved after FIB processing.

3.2 Sample quality after FIB processing

Figure 3.1a is an image of a 2.5 μm wide NbSe₃ crystal on top of gold voltage probes. The crystals' thickness is estimated to be 0.5 μm , using the room temperature resistivity $\rho = 1.86 \Omega\mu\text{m}$. A constriction is made by etching one side of the crystal leaving a 1 μm wide wire. We have measured the residual resistance ratio $r_R = R(295 \text{ K})/R(5 \text{ K})$ of the wire before and after etching. Both r_R of the constriction and the unetched wire are 43. For large crystals, r_R is a measure for the impurity concentration. For small crystals, r_R depends on the dimensions of the crystals. Measurements of r_R on undoped NbSe₃ crystals as a function of the thickness has been performed by McCarten *et al.* [3]. Our measured values of r_R agree with their earlier measurement of r_R . Impurity implantation of Ga ions does not appear to affect single-electron transport in NbSe₃ over the temperature range of interest.

Another measurement to test the quality of the NbSe₃ crystals after FIB

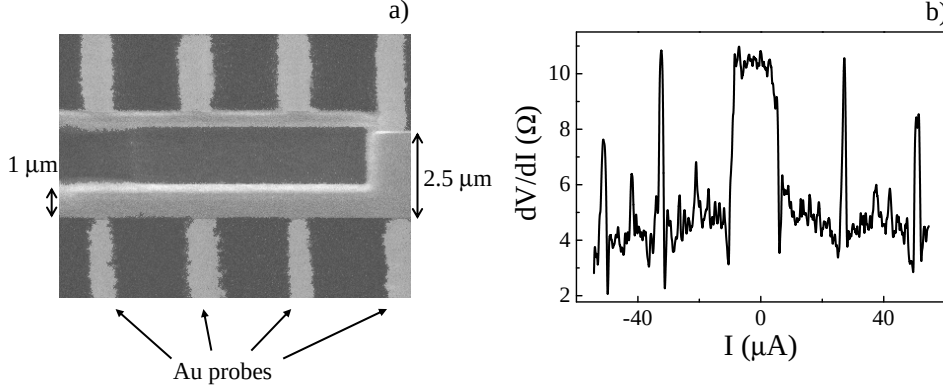


Fig. 3.1: a) Focused-Ion Beam image of a $2.5 \mu\text{m}$ wide NbSe_3 crystal on top of gold voltage probes. The upper part is etched with a FIB, leaving a $1 \mu\text{m}$ wide channel. b) Shapiro step measurement of the constriction at $T = 50 \text{ K}$. The CDW mode-locks completely to the external 130 MHz signal.

processing involves mode-locking of the CDW to an external radio-frequency signal. In such measurements, a dc-bias is combined with an ac-signal and mode-locking of the CDW is seen as Shapiro steps in the current-voltage $I(V)$ curve and peaks in the differential resistance dV/dI . Figure 3.1b shows the dV/dI at $T = 50 \text{ K}$ of the etched part using the two voltage probes indicated by the arrows in Fig. 3.1a. The first peaks beyond the threshold field reach the same level as at low bias; the CDW is completely mode-locked to the external frequency. From the position of the peaks, we can estimate the cross section using the appropriate values at $T = 50 \text{ K}$ taken from Bardeen *et al.* [4]. The value of the cross section $S = 0.34 \mu\text{m}^2$, agrees with the one obtained from room temperature resistivity within 10%.

Complete mode-locking of the CDW to an ac-signal is generally considered an indication of the high quality of the crystals used. The transport measurements on the etched wire demonstrate that FIB processed samples still show coherent CDW dynamics throughout the whole cross section. Any unintentional damage has not degraded the CDW dynamics. Measurements of r_R have often been used to determine the impurity concentration in NbSe_3 . The measurements on the etched wire show no noticeable increase of impurity concentration by FIB processing. After FIB processing, the crystals retain their high-quality properties.

3.3 Conductance anisotropy measurements

When etching structures in crystals, geometrical effects have to be taken into consideration. The electric field is non-uniform in the vicinity of con-

strictions or other etched structures. The non-uniformity of the electric field is more pronounced in anisotropic materials. Anisotropic materials can be mapped onto the isotropic case by scaling the transverse dimensions with $\sqrt{A}$, where $A = \rho_{\perp}/\rho_{\parallel}$ is the resistivity anisotropy. For NbSe₃, $\rho_{\parallel}$ is the b-axis resistivity and $\rho_{\perp}$ is either the c-axis or the a*-axis resistivity. In transport measurements, the non-uniform electric field will show up as an additional resistance, the so-called ‘spreading resistance’, also discussed in Chapter 2.

We have determined the spreading resistance of a tear as a function of the tear length L_{tear} by resistance measurements on stainless steel and copper strips. The spreading resistance ΔR is determined by measuring the resistance of the strip with the tear R_{tear} minus the resistance of the strip without the tear R . The distance between the voltage probes is kept constant and is much larger than the width of the strip (1.5 to 3 cm wide strips were used). The resistance of the strip without the tear is equal to the sheet resistance $R_{sheet} = Rw/l$ times the number of squares between the voltage probes l/w , where l is the distance between voltage probes and w is the width of the strip. In the anisotropic case, the number of squares is $\sqrt{A}$ times smaller compared to the isotropic case, because the width has to be rescaled by $\sqrt{A}$. Therefore, the spreading resistance divided by $R_{sheet}\sqrt{A}$ provides a universal curve of the spreading resistance independent of the material and the anisotropy of the material, see Fig. 3.2a. This is confirmed by numerical calculations of the Laplace equation with the proper boundary conditions depicted by the solid line in Fig. 3.2a. Measurements of the spreading resistance of a tear in NbSe₃ can be collapsed onto this universal curve resulting in the anisotropy:

$$A = \left(C \frac{R_{sheet}}{\Delta R} \right)^2 \quad (3.1)$$

where the constant C can be obtained from Fig. 3.2a. For a 50% tear, C is about 0.5. This way, the anisotropy of NbSe₃ as a function of temperature is found. We have used a FIB to etch a tear in a NbSe₃ crystal, which was placed on an array of gold probes. The tear length was 50% of the width ($w = 4 \mu\text{m}$ and $l = 100 \mu\text{m}$), see inset of Fig. 3.2b. We have measured the resistance of the segment with the tear as a function of temperature. The resistance of another segment, not containing a tear, was measured simultaneously. ΔR was calculated by subtracting the resistance of the segment with the tear from the segment without the tear. The anisotropy is calculated by using Eq. 3.1. Fig. 3.2b shows the resulting c-axis anisotropy $A_c = \rho_c/\rho_b$ as a function of temperature, with ρ_c and ρ_b the resistivity along the crystallographic c-axis and b-axis respectively. This temperature dependence agrees with measurement by Ong and Brill [5].

To measure the a*-axis anisotropy $A_{a*} = \rho_{a*}/\rho_b$, with ρ_{a*} the resistivity in the a*-axis, we have etched a narrow trench across the width of a NbSe₃

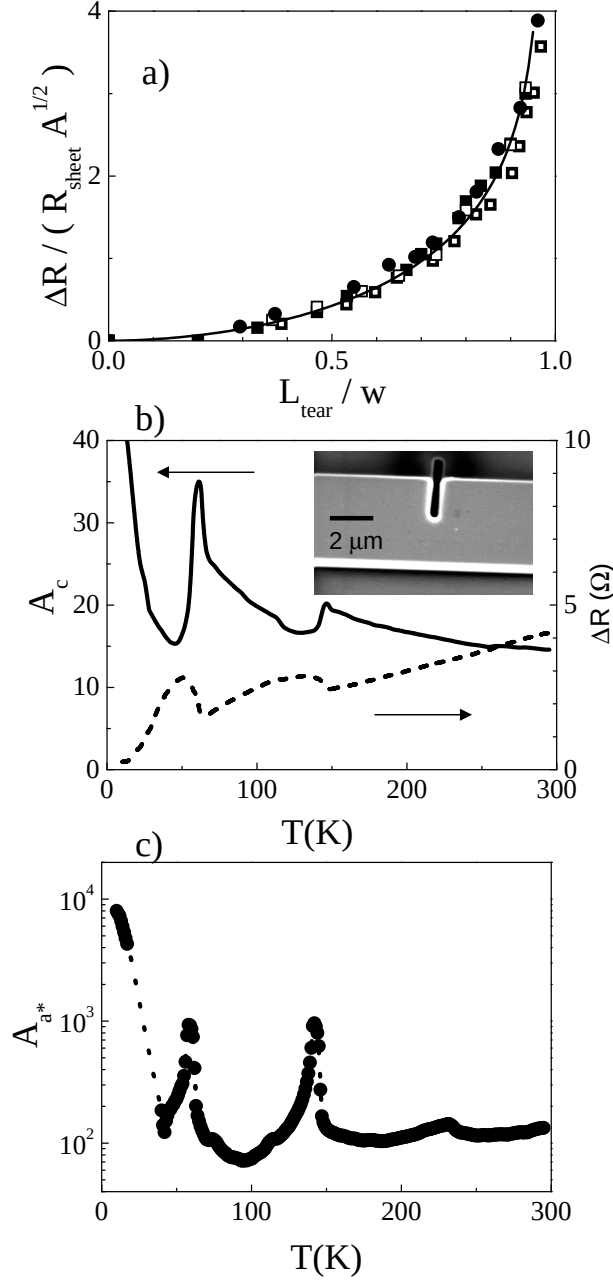


Fig. 3.2: a) Normalized spreading resistance versus the fractional tear length L_{tear}/w (w is the width of the strips). The circles (squares) are measurements on copper (stainless steel). The line is a numerical solution of the Laplace equation with the appropriate boundary conditions. b) c-axis anisotropy of NbSe_3 as a function of temperature. The spreading resistance ΔR (dashed line) is added for clarity. c) a^* -axis anisotropy of NbSe_3 as a function of temperature.

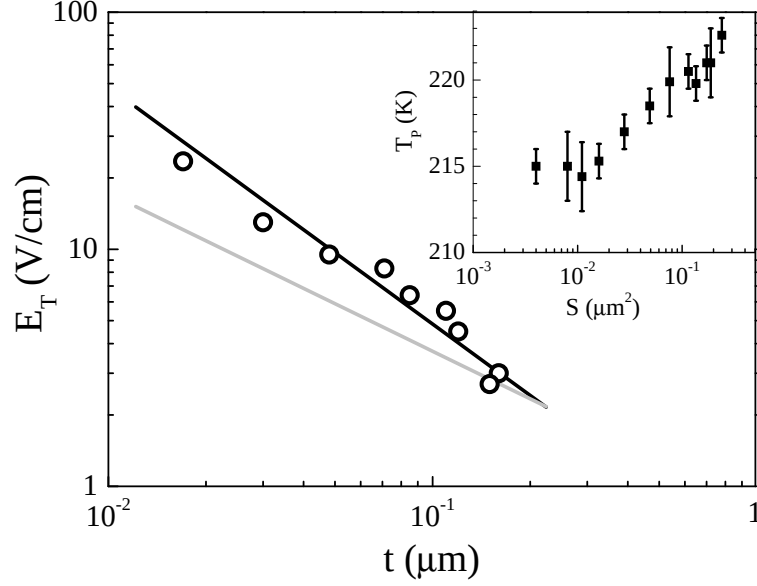


Fig. 3.3: Threshold field E_T at $T = 120$ K of one o-TaS₃ wire as a function of the thickness t , while the width is maintained the same. The black solid line shows a $1/t$ dependence and the grey line a $1/t^{2/3}$ dependence. The inset shows the Peierls transition temperature T_P as a function of the cross sectional area S of two o-TaS₃ samples.

crystal. The tear's time of exposure to the FIB is taken half of the time needed to etch all the way through the crystal. Therefore, the depth of the trench is estimated to be 50% of the thickness. ΔR was determined by subtracting the resistance of the segment with the trench from the resistance of an unetched segment. Again, ΔR is mapped unto the universal curve, resulting in a temperature dependence of A_{a*} , shown in Fig. 3.2c. A_{a*} varies from about 100 at room temperature to 10^4 at low temperature.

3.4 Thickness dependent threshold-field of o-TaS₃

We have determined the threshold field E_T of an o-TaS₃ wire as a function of the thickness t , while maintaining the same width w . The thickness is reduced by exposing the crystal to a FIB. The sample was made by putting an o-TaS₃ crystal on a sapphire substrate with another wide o-TaS₃ crystal perpendicular on top of it serving as an evaporation mask. Gold was evaporated and the crystal mask was removed. We have determined E_T at $T = 120$ K and measured the resistance as a function of temperature to determine the Peierls transition temperature. We estimated t from the room temperature resistance and $w = 1.6 \mu\text{m}$ from FIB images. Subsequently, a

thin layer was removed over the entire length of the crystal (100 μm) with a FIB. These measurements and milling steps were repeated several times. Figure 3.3 shows E_T at $T = 120$ K as a function of t . The solid line indicates a $1/t$ dependence of E_T . The same dependence of E_T on the thickness was found by Borodin *et al.* who did not use any etching procedure [6].

The $1/t$ dependence of E_T is expected for 2D weak pinning. The dotted line in Fig. 3.3 shows a $1/t^{2/3}$ dependence expected for 1D weak pinning, see Chapter 5. The data follows the $1/t$ dependence above $t = 60$ nm. For smaller thicknesses (below about 60 nm), the $1/t^{2/3}$ dependence fits the data better.

For small crystals, surface pinning is likely to be important and E_T should be proportional to the surface-to-volume ratio. For flat crystals ($w \gg t$), this leads to $E_T \propto 1/t$ [3]. Since for two-dimensional weak pinning, E_T is also proportional to $1/t$, a distinction between surface pinning and bulk pinning is hard to make. E_T for two-dimensional weak pinning is proportional to the impurity concentration n_i and it is expected that E_T would increase more rapidly than $1/t$ with decreasing thickness as more implantation of Ga is expected for the thinnest wire. It is remarkable that E_T follows the $1/t$ -dependence and no clear effect of implanted Ga impurities is found.

The inset of Fig. 3.3 shows the Peierls transition temperature T_P as a function of the cross section S of two o-TaS₃ samples. S has been determined from the room temperature resistance and T_P has been determined by the position of the dip in the $\text{dln}R/\text{d}T(T)$ curve. T_P decreases when S is reduced. This behavior has also been observed by Borodin *et al.* [6].

3.5 Non-metallic behavior in NbSe₃ channels

We have measured the resistance-temperature $R(T)$ curves of channels of several NbSe₃ samples. When $w > 500$ nm, we always observe the normal (semi-)metallic behavior, see bottom curve of Fig. 3.4a. When the width is reduced below 500 nm, we observe a decrease of the residual resistance ratio and the $R(T)$ curve becomes more flat, as shown by the middle curve of Fig. 3.4a. In this case, the width is comparable to the thickness. When $R(295 \text{ K})/L$ is larger than 1-10 $\text{k}\Omega/\mu\text{m}$, the Peierls transitions vanish and the transport changes from metallic to insulating behavior. The typical width for this change in behavior is 100-200 nm. For very thin and narrow channels ($t < 200$ nm and $w < 200$ nm), the resistance follows a power-law as a function of temperature below $T = 100$ K, see top curve of Fig. 3.4a. The exponent of the power-law relation is about 3. The $I(V)$ curve at $T = 10$ K of this insulating channel is shown in Fig. 3.4b. For low bias, the $I(V)$ curve is linear. Above 10^{-2} V, the $I(V)$ follows a power-law with an exponent of about 3 as well. Zaitsev-Zotov *et al.* have also

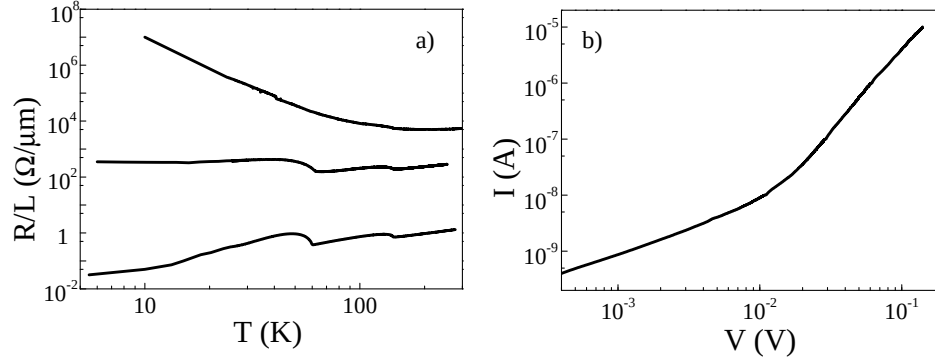


Fig. 3.4: a) Unit length resistance R/L of three NbSe₃ channels as a function of temperature on a log-log scale. The bottom curve resembles the expected semi-metallic behavior of bulk NbSe₃. The middle curve is data taken from a 250 nm wide constriction. The top curve shows non-metallic behavior of a channel with cross section $100 \times 150 \text{ nm}^2$. The resistance follows a power-law with exponent 3 at low temperature. b) The $I(V)$ curve at $T = 10 \text{ K}$ on a log-log scale of the non-metallic channel. At high bias the $I(V)$ also follows a power-law with exponent 3.

observed a change from metallic to insulating behavior in NbSe₃ crystals [7]. In their case, they reduced the thickness of the wire with an SF₆ plasma, while we reduce the width of the wires. Also, similar non-metallic behavior with power-law dependence is observed in cleaved and ultrasonically cleaved NbSe₃ nanowires, which have not undergone any etching procedure [7] and Chapter 6. Since this behavior is observed in those crystals, it is likely that the insulating properties are caused by the finite size of the transverse dimensions rather than implanted impurities or unintentional damage to the crystal by the FIB.

3.6 Conclusions and future work

We have shown that a FIB is a practical tool to fabricate mesoscopic CDW structures. We have completed the necessary characterization of possible unintentional damage to the crystals. We conclude that the damage is minimal and that the crystals retain their high-quality properties.

For future work, more-complicated structures can be made. For instance, to investigate phase slip on a local scale, a FIB can be used to make artificial pinning sites in a predetermined pattern. It is also possible to make structures to investigate CDW shear interactions [8, 9]. A few more-complicated structures are shown in Fig. 3.5. Compared to lithographic techniques, the fabrication process of these structures takes very little time.

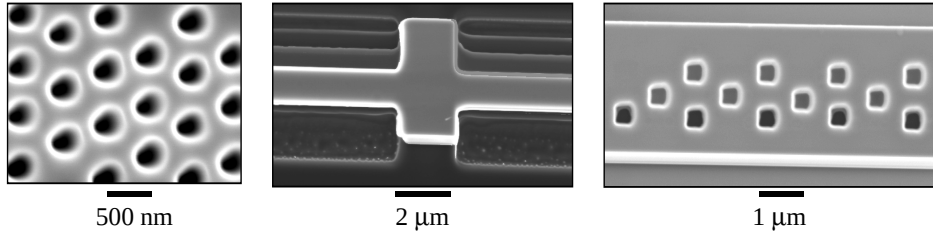


Fig. 3.5: Scanning Electron Microscope pictures of a 700 nm thick NbSe_3 crystal with FIB patterned structures. Left: A pattern of 200 nm wide and 700 nm deep holes. The holes are 500 nm apart from each other. Middle: Exemplary structure for measurements on CDW shear or phase slip processes. Right: A pattern of 300 nm wide and 700 nm deep square holes.

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4. Negative Resistance and Local Charge-Density Wave dynamics

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Abstract

Charge-density wave dynamics is studied on a submicron length scale in NbSe₃ and o-TaS₃. Regions of *negative absolute* resistance are observed in the CDW sliding regime at sufficiently low temperatures. The origin of the negative resistance is attributed to the different forces that the deformed CDW and quasi-particles feel: the force on the CDW is merely caused by a difference of the electric potentials, while the quasi-particle current is governed by an electrochemical potential difference.

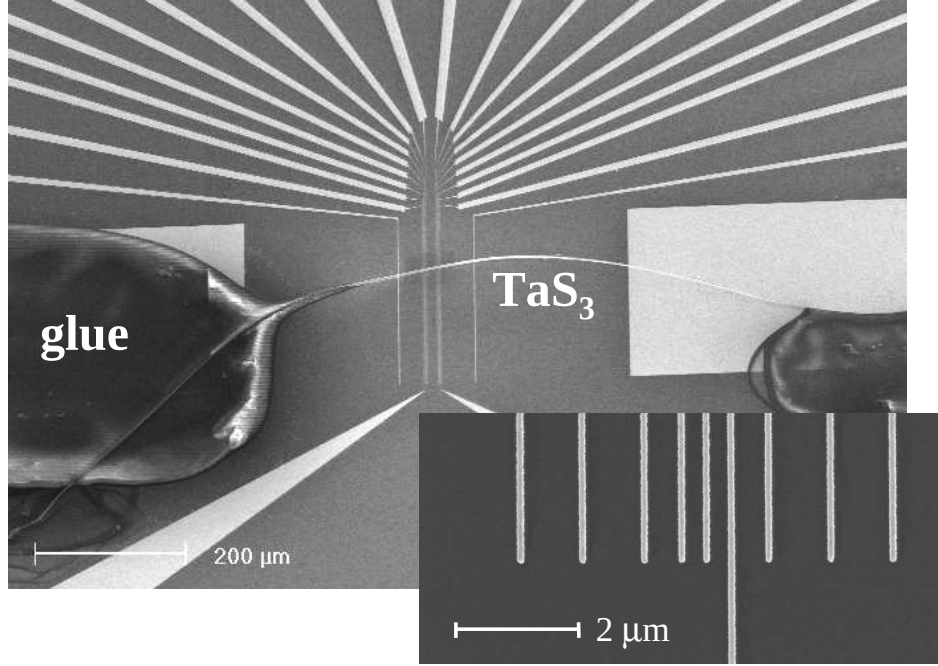


Fig. 4.1: A thin TaS_3 crystal on top of an array of voltage probes to study CDW dynamics on submicron length scales. The spacing between the big (current) pads on either side of the picture is 0.5 mm. The inset shows an enlargement of the main figure with 9 voltage probes that are 100 nm wide; the smallest distance between adjacent probes is 300 nm. Each sample has two of these probe-sets that are separated 12 μm from each other.

4.1 Introduction

In metallic and superconducting devices, reduction of sizes has revealed a variety of new mesoscopic phenomena. For CDW conductors, the mesoscopic regime has only been studied for small transverse dimensions [1, 2, 3] because samples of (sub)micron sizes in the chain direction could not be fabricated in a controlled way. Consequently, many aspects of microscopic CDW dynamics are still unknown. Nevertheless, some early studies on a micron-scale revealed interesting mesoscopic features related to the CDW phase distribution [4, 5, 6]. More recently, artificial submicron CDW devices have been fabricated [7, 8].

4.2 Experiments

Current-voltage characteristics (IV s) are recorded on high-quality NbSe_3 and TaS_3 crystals with probe spacings in the submicron range (see the inset of Fig. 4.1). On these short length scales IV curves vary strongly from segment to segment. For some segments the absolute resistance becomes negative, indicating that the moving CDW forces single-particle carriers in a direction opposite to that of the rest of the sample. Our results show that $1\text{ }\mu\text{m}$ is the typical length scale for this new phenomenon in CDW dynamics.

Experiments were carried out on single NbSe_3 and o-TaS_3 crystals with cross sections of 0.2 to $1\text{ }\mu\text{m}^2$. Both materials have a very anisotropic, chain-like structure [9]. NbSe_3 exhibits CDW transitions at $T_{\text{P}1} = 145\text{ K}$ and $T_{\text{P}2} = 59\text{ K}$. At low temperatures a small portion of the conduction electrons remains uncondensed, providing a metallic single-particle channel. In contrast, in o-TaS_3 all electrons condense into the CDW state. As a result, the linear resistance shows semiconducting behavior below the transition temperature of $T = 220\text{ K}$.

A common technique to contact small CDW whiskers consists of putting the crystals on top of metal probes that are evaporated on an insulating substrate. Then a droplet of glue (ethyl cellulose dissolved in ethyl acetate) is put on the crystals to keep them fixed on the metal probes. In previous studies the smallest probe widths were on the order of $2\text{ }\mu\text{m}$ and their smallest separations were typically $10\text{ }\mu\text{m}$. By using standard e-beam lithographic techniques, we have fabricated gold wires that are 50 nm high and 100 nm wide. The smallest probe separation is 300 nm as illustrated in the inset of Fig. 4.1. It is important to note that to study microscopic CDW dynamics, our results show that both the probe width and separation must be sufficiently small.

Electrical contact between o-TaS_3 and the gold wires has only been obtained after heating the crystals to $120\text{--}130\text{ }^\circ\text{C}$ for several minutes to one hour [10]. During this annealing step, sulfur that has accumulated at the surface oxidizes leaving behind a clean interface. For NbSe_3 , we heat the samples so that the thin crystals do not start floating in the glue solvent. When the substrate is heated to $80\text{ }^\circ\text{C}$ the solvent evaporates quickly, giving the crystals no opportunity to float.

A series of measurements has been performed to characterize the crystals. Cross sections (S) have been determined from measuring the resistance R for segments with different separation L at room temperature. Here, L is defined as the distance between the middle of two voltage probes. We find that R scales with L . Cross sections are then calculated using the literature values of the room-temperature resistivity: $\rho = 2\text{ }\Omega\mu\text{m}$ [3] for NbSe_3 and $\rho = 3\text{ }\Omega\mu\text{m}$ for o-TaS_3 [11]. Another test involves the measurement of Shapiro steps when biasing the samples with both a dc and ac drive. At $T = 120\text{ K}$, we obtain complete mode locking for both types of crystals

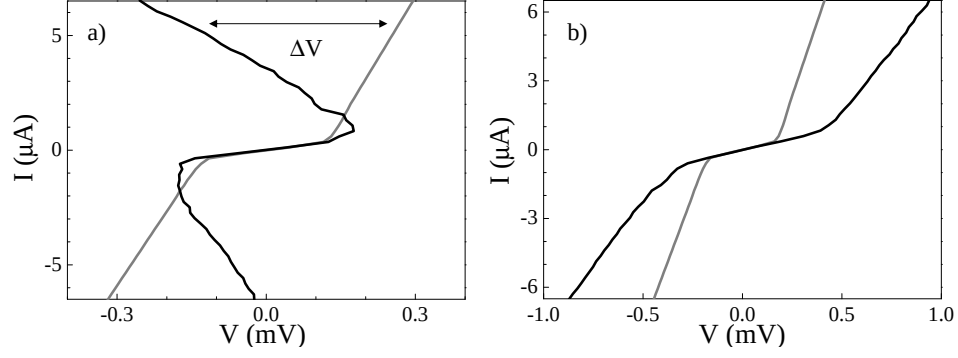


Fig. 4.2: Two current-voltage characteristics of a TaS_3 crystal with a cross section of $0.5 \mu\text{m}^2$. The curves are taken at $T = 120 \text{ K}$ on adjacent $1 \mu\text{m}$ -long segments. Grey lines correspond to a measurement on a $31.6 \mu\text{m}$ long segment for which the voltage has been scaled by a factor of $1/31.6$. They represent the expected, averaged nonlinear CDW behavior. a) the absolute value of the resistance becomes negative for high positive bias. The deviation ΔV from the expected behavior is linear in I_{CDW} . b) the curve shows less CDW current at a given field, i.e., it is less nonlinear. When adding up the two curves one approximately recovers the expected, average CDW behavior.

indicating their high quality. The step-width scales with frequency yielding sample cross sections that compare well with the values obtained from the resistance measurements (within 7 %). Threshold fields (E_T 's) also point at a good sample quality. At $T = 120 \text{ K}$, $E_T = 0.58 \text{ V/cm}$ for a NbSe_3 crystal of $S = 0.2 \mu\text{m}^2$ and $E_T = 1.23 \text{ V/cm}$ for a TaS_3 crystal of $S = 0.5 \mu\text{m}^2$.

We have systematically studied IV characteristics of various segments. IV curves have been obtained in the current-biased, normal four-probe configuration: Current is injected at the large gold pads (see Fig. 4.1) and probe pairs measure the voltage across the various segments. We first discuss the results on o- TaS_3 crystals that have been obtained in the temperature range of 94 K - 220 K .

When L is larger than about $10 \mu\text{m}$, we always observe the expected behavior for CDWs as indicated by the grey lines in Fig. 4.2. On a micron scale, however, the shape of the IV varies from segment to segment. Most small segments show the same nonlinear behavior as observed in the large segments. Some segments on the other hand exhibit an IV that shows a negative absolute resistance (NAR). NAR is observed at high currents (see the positive current branch in Fig. 4.2a for $I > 3.5 \mu\text{A}$). Note that at smaller currents the effect of negative resistance shows up as a negative differential resistance (NDR). A small segment adjacent to the NAR region has an IV

curve that is less nonlinear with a higher threshold (Fig. 4.2b). As stated above, these differences average out when measuring on larger length scales and deviations between various $I(E = V/L)$ -curves are small if $L > 10 \mu\text{m}$.

Taking a closer look at the NAR curve in Fig. 4.2a, one sees that the deviation (ΔV) from the expected IV curve (grey line) is proportional to the CDW current I_{CDW} . We can express this deviation with a parameter α defined as $\alpha = \Delta V/(R_0 I_{CDW})$, where R_0 is the linear resistance around $V = 0$. From Fig. 4.2a, we find that $\alpha \approx 0.36$ for the positive current branch and $\alpha \approx 0.2$ for the negative current branch.

We have also determined α at other temperatures. At $T = 200 \text{ K}$, we find no unusual behavior: all IV characteristics can approximately be collapsed on one $I(E)$ -curve so that $\alpha \approx 0$. For the segment of which the 120 K data are shown in Fig. 4.2a, NDR sets in at $T = 160 \text{ K}$ with $\alpha = 0.17$ and 0.11 for the positive and negative current branch respectively. As temperature decreases, α increases and NAR develops. At $T = 94 \text{ K}$, $\alpha \approx 0.96$ for the positive current branch and $\alpha \approx 0.6$ for the negative current branch. Situated $20 \mu\text{m}$ from this segment, another segment with $L = 0.8 \mu\text{m}$ also starts to develop NAR at temperatures below 100 K .

In total, we have studied four different o-TaS₃ samples. All four samples showed NDR in one or two segments, each time involving different probe pairs. In the thinnest samples, these NDR regions yield NAR at high currents. From measuring $R_0(L)$ and $E_T(L)$, we have found no indications of serious damage of the crystals (cracks) in the NDR and NAR regions. We have also measured the two-probe resistances between all probe pairs at several temperatures. Again no unusual features have been found. The two-probe resistance is generally two orders of magnitude larger than the four-probe resistance. They only vary by a factor five from each other and no trend was observed for the segments that show NDR or NAR (e.g. no systematically lower contact resistance).

We have also studied two NbSe₃ samples on a micron scale. In one sample, the absolute resistance became negative. Another sample exhibits only NDR. The conditions to find NDR and NAR in NbSe₃, however, appear to be more stringent. We only observe it below the second Peierls transition (T_{P2}) in the close vicinity ($\sim 1 \mu\text{m}$) of a current contact. As in the case with TaS₃, IV 's are asymmetric with respect to interchanging the positive and negative current branches.

Next, we present the IV characteristics of one NbSe₃ sample. We have systematically studied the four-probe IV curves of each segment in the temperature range 25 K - 120 K . When the current contacts are far from the voltage probes ($> 10 \mu\text{m}$), we observe the expected behaviour for all segments at all temperatures, even if the probe spacing is smaller than $1 \mu\text{m}$. When the current is injected closer to the voltage probes, IV curves become more asymmetric for temperatures below the second Peierls transition ($T_{P2}=59 \text{ K}$). Fig. 4.3 shows typical IV curves for a segment with $0.4 \mu\text{m}$

probe spacing. The quantity d is the distance between the current contact and the middle between voltage probes. As d decreases the asymmetry becomes more pronounced and for $d=3.2\text{ }\mu\text{m}$ NDR sets in at the negative current branch. When current is injected only $1.2\text{ }\mu\text{m}$ from the voltage probes (Fig. 4.3), the negative differential resistance is largest and at high negative current the absolute resistance becomes negative.

To summarize our data, the central result is the observation of a negative absolute resistance (NAR) in the IV curves of both o-TaS₃ and NbSe₃. In all cases it is a local effect occurring on a micron scale. Although early reports on macroscopic CDW dynamics have shown NDR in NbSe₃ [12, 13] and in TaS₃ [4, 10, 14] crystals, we are not aware of any measurement nor prediction of NAR in CDW samples. In the remainder, we provide a qualitative explanation for this new effect.

4.3 Qualitative explanation for Negative Absolute Resistance

This section describes a model for Negative Absolute Resistance (NAR) applicable to o-TaS₃. Its applicability to NbSe₃ is not clear, since NbSe₃ is a metallic conductor below the Peierls transitions where screening of charges could be much stronger. The model considers an inhomogeneous region in between two homogeneous regions. The two homogenous regions are large so that the average value of relevant quantities can be taken. The inhomogeneous region is small so that relevant quantities are local, emphasizing that the observation of NAR is a finite-size effect.

The basic ingredient of our model describing local dynamics is that CDWs and quasi-particles are driven by different forces. At low temperatures, when the quasi-particle density is much smaller than the density of the condensed electrons, the force exerted on the CDW is mostly related to a difference of the *electric* potential Φ [15]. In contrast, the quasi-particle current is governed by a difference of the *electrochemical* potential, i.e., by the voltage drop $V = U(x_1) - U(x_2)$.

Starting from the experimental data, we can draw the potential and current distributions along the sample defining the conditions for which NAR can be observed. First of all, the slope, dU/dx , in the NAR region is opposite to the mean slope of $U(x)$ along the sample (see Fig. 4.4a). The quasi-particle current I_q is equal to $-\sigma_q dU/dx$ where σ_q is the quasi-particle conductance per unit length. It therefore also has an opposite sign compared to the rest of the sample. Since the total current, $I = I_q + I_{CDW}$, is the same along the sample, I_{CDW} in the NAR region must be larger than in the rest of the sample (Fig. 4.4b). At the same time as illustrated in Fig. 4.4a, one sees that the force on the CDW is larger in the NAR region because the gradient of the *electric* potential $E = -d\Phi/dx$ is larger here. Finally, one

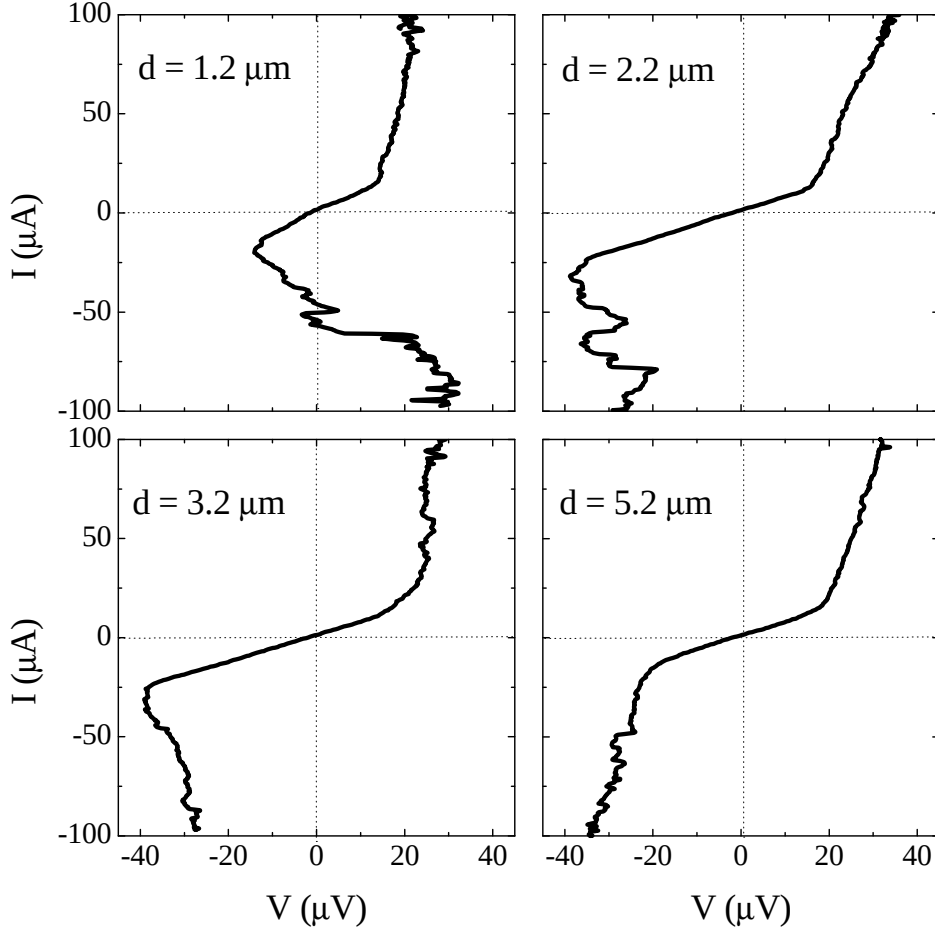


Fig. 4.3: Four current-voltage characteristics of a NbSe_3 sample with a cross section of $0.2 \mu\text{m}^2$. All four curves are taken at $T = 40 \text{ K}$ on the same $0.4 \mu\text{m}$ -long segment, while the distance to the current contact is varied. Here, d is the distance from the current contact to the middle between voltage probes. The IV curve becomes more asymmetric when d is decreased and starts to develop negative differential resistance. The curve closest to the current contact exhibits negative absolute resistance for larger currents than $-50 \mu\text{A}$.

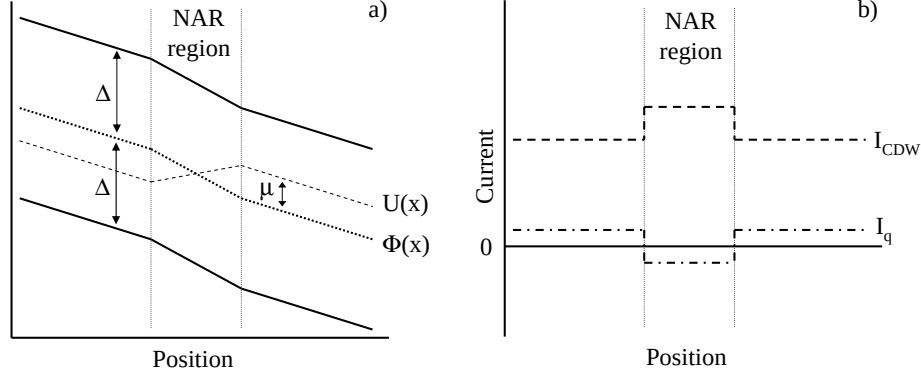


Fig. 4.4: (a) Band bending and (b) current distributions around the NAR region. Φ coincides with the middle of the Peierls gap under the chosen calibration of the electrostatic potential and U is the electrochemical potential. Edges of the Peierls gap are represented by the solid lines.

should keep in mind that the difference $(U - \Phi)$ defines shifts of the chemical potential (μ) with respect to the midgap position. These shifts are related to CDW phase deformations, i.e., $\mu = (U - \Phi) \propto d\varphi/dx$ [1, 16, 17], where φ is the CDW phase.

We now concentrate on the physical conditions that lead to the potential and current distributions shown in Fig. 4.4. If we assume that K in the relation $I_{CDW} = K\dot{\varphi}$ is the same along the sample, then the jump in I_{CDW} (see Fig. 4.4b) corresponds to a larger CDW phase velocity $\dot{\varphi}$ in the NAR region. Such a variation of $\dot{\varphi}$ must be provided by phase slip, but the corresponding CDW deformations in Fig. 4.4a have the wrong sign [18]. This contradiction means that NAR results from a variation of $K \equiv I_{CDW}/\dot{\varphi}$ rather than from a variation of the CDW velocity only.

Taking $\dot{\varphi}$ to be the same along the sample, we assume that $I_{CDW} = (1 + \alpha)K\dot{\varphi}$ in the NAR region (the origin of α will be discussed later), and that $I_{CDW} = K\dot{\varphi}$ for the rest of the sample. Since the total current along the sample is constant, one has $K\dot{\varphi} + I_q = (1 + \alpha)K\dot{\varphi} + V/R_0$. The voltage drop in the NAR region is then given by:

$$V = R_0 (I_q - \alpha I_{CDW}), \quad (4.1)$$

where $R_0 = l/\sigma_q$ with l the length of the NAR region. Eq. 4.1 implies that for $I_{CDW} > I_q/\alpha$ the sign of V is negative. Note that $\Delta V = \alpha R_0 I_{CDW}$ is the deviation from the regular IV curve introduced earlier.

There is, however, a limitation for the CDW to keep the same phase velocity $\dot{\varphi}$ through an NAR region. Phase slippage is expected to set in as soon as the CDW deformation reaches its limiting value: $\mu_{\max} = V_{ps}/2e$, where V_{ps} is the phase-slip voltage [15]. Therefore, ΔV , is limited by the phase-

slip voltage and $\Delta V_{\max} = (I_{CDW}/I)V_{ps}$. Thus, NAR cannot be observed in segments having $l > l_{\max} \sim \alpha V_{ps}/E_T$, and for total currents $I > V_{ps}\sigma_q/l\alpha$. For typical values ($V_{ps} \sim 1$ mV, $\alpha \sim 0.1$ and $E_T \sim 1$ V/cm) one gets $l_{\max} \sim 1$ μm , in good agreement with the experimental data.

A variation of $K = (2e/\pi)N_c(1 - b)$ (see Ref. [19]) and thus a nonzero α can be related to a change in the number of the conducting chains in the sample's cross section (N_c), or to a change in the parameter b . Here, b describes the reduction of “condensed” electrons with increasing temperature and contains the so-called “back-flow current”¹ which depends on the scattering times. A plausible explanation might be that increased scattering in a NAR region -due to for example a macroscopic defect, e.g. a line dislocation- leads to a quasi-particle current that flows in a direction opposite to that in the rest of the sample. Hall measurements on $\text{K}_{0.30}\text{MoO}_3$ have shown [20] that back-flow currents produce typical α -values of $0.1 - 0.2$, in good agreement with our high-temperature data.

Theoretical estimates based on a microscopic approach [19, 21] indicate that the contribution of b is too small to account for the observed low-temperature α -values of ~ 1 . However, at low temperatures the experimentally obtained values are likely to be overestimated. The measured values of α are very sensitive to a correct determination of R_0 . Furthermore, in TaS_3 at $T < 100$ K, R_0 greatly depends on the CDW deformations which undoubtedly occur near defects. In that case R_0 is smaller than expected from the simple Arrhenius law, $R_0 \sim \exp(\Delta/T)$, where Δ is the Peierls gap [22, 23].

More detailed calculations on the local CDW dynamics are needed to explain all experimental details. These should also address the asymmetry of the IV curves in the NAR region. It is most likely caused by the absence of symmetry between electron-like and hole-like excitations and the built-in CDW deformations that shift the chemical potential.

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¹ Here, the term back-flow current refers to the *non-equilibrium* part of the CDW current (see e.g. Ref. [19]) that for example gives rise to a contribution to the Hall effect. This current should be distinguished from the 'balancing' or 'compensating' current measured in other experiments (see e.g. W.P. Beyermann et al., Phys. Rev. Lett. **56**, 1489 (1986)).

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5. One-dimensional collective pinning in NbSe_3

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Abstract

NbSe_3 structures were fabricated with widths comparable to the Fukuyama-Lee-Rice phase-coherence length of bulk samples in the three-dimensional collective pinning limit. We observe a crossover from two-dimensional to one-dimensional collective pinning, for samples already in the two-dimensional limit, when the crystal width (crystallographic c -axis) is less than $1.6\ \mu\text{m}$, corresponding to the phase-coherence length in this direction. Our results show that surface pinning is negligible in our samples, and provide a means to probe the dynamics of single domains giving access to a new regime in charge-density wave physics.

Incommensurate Charge-Density waves (CDWs), such as in NbSe₃, interact with impurities and defects that pin the CDW. When a force larger than a certain threshold value is exceeded the CDW begins to slide. The interaction of the CDW with impurities can be described using the mean-field model of Fukuyama, Lee, and Rice (FLR) [1, 2]. They consider the CDW as an elastic medium, that can deform in the presence of impurities and under the influence of a force. The FLR model is valid when the CDW's deformation is elastic. It does not consider inelastic properties of CDWs like phase slip processes.

5.1 Fukuyama-Lee-Rice model

The charge density of the CDW is periodically modulated in space [3], and may be written as

$$\rho(\vec{r}, t) = \rho_0 + \rho_1 \cos[\vec{Q} \cdot \vec{r} + \phi(\vec{r}, t)], \quad (5.1)$$

where ρ_1 is the CDW amplitude and $\vec{Q} = 2\vec{k}_F$ is the CDW wave vector and $\vec{k}_F$ is the Fermi wave vector. The CDW phase ϕ describes the local position of the CDW with respect to the underlying lattice. The elastic strain in the CDW is proportional to the gradient of the CDW phase $\vec{\nabla}\phi$. Under the influence of an electric field the CDW can slide with respect to the lattice [4] and the charge density becomes a function of space and time $\rho(\vec{r}, t)$. The velocity of the CDW (or the CDW current) is proportional to the time evolution of the phase $\delta\phi/\delta t$. However since impurities and other lattice defects pin the CDW, depinning occurs only above a threshold electric field E_T . Near E_T , the CDW is strained by the electric field.

The time-independent FLR model starts with the Hamiltonian

$$\mathcal{H} = \frac{1}{2}K \int d\vec{r} (\vec{\nabla}\phi)^2 + V_0 \rho_1 \sum_i \cos[\vec{Q} \cdot \vec{R}_i + \phi(\vec{R}_i)] + \int d\vec{r} \frac{\rho_{eff} \vec{\mathcal{E}} \cdot \vec{Q} \phi}{Q^2} \quad (5.2)$$

The first term describes the elastic energy, where K is the elastic force constant of the CDW; the second term describes the interaction with point pinning sites located at $\vec{R}_i$, where V_0 is the impurity potential; and the third term describes the coupling of the CDW phase to an electric field $\vec{\mathcal{E}}$, where ρ_{eff} is an effective condensed charge density.

FLR considered two cases of pinning depending on the impurity spacing $l_i = \sqrt[3]{1/n_i}$, where n_i is the impurity concentration.

- In the strong pinning limit ($V_0 \rho_1 / K l_i \gg 1$), pinning sites locally pin the phase of the CDW and amplitude collapse is needed for local phase motion [5].

- In the weak pinning limit ($V_0\rho_1/Kl_i \ll 1$), the elastic energy dominates and the CDW is collectively pinned by elastic deformations of its phase on lengths l_ϕ larger than the impurity spacing l_i .

The length scale l_ϕ is known as the Fukuyama-Lee-Rice length, which is analogous to the Ovchinnikov-Larkin length in pinned vortex lattices. These phase deformations allow the CDW to gain pinning energy by taking advantage of fluctuations in the interaction with randomly distributed impurities on that length scale. The elastic-energy cost of these deformations ($\vec{\nabla}\phi \sim \pi/l_\phi$) is balanced by the impurity-energy gain within a phase coherent volume. For simplicity, we consider the case where l_ϕ is isotropic; the appropriate scaling for the anisotropic case is described in the appendix of this chapter, following Refs. [2] and [6].

Fluctuations in the interaction with randomly distributed impurities, within a phase-coherent volume l_ϕ^3 , are on the order of $\sqrt{N}$, where the number of impurities $N = n_i l_\phi^3$. Without any confinement (3D), FLR showed, using a random-walk argument, that the pinning-energy gain of a phase coherent volume is

$$E_{pin} = -V_0\rho_1\sqrt{n_i l_\phi^3}. \quad (5.3)$$

Within a phase coherent domain the phase varies less than π . The elastic energy term in Eq. 5.2 becomes

$$E_{el} = \frac{1}{2}Kl_\phi^3\left(\frac{\pi}{l_\phi}\right)^2. \quad (5.4)$$

Without an applied electric field $\mathcal{E}$, the total energy density, within a phase coherent volume of size l_ϕ^3 , is

$$\frac{E_{el} + E_{pin}}{l_\phi^3} = \frac{1}{2}K\left(\frac{\pi}{l_\phi}\right)^2 - V_0\rho_1\sqrt{\frac{n_i}{l_\phi^3}} \quad (5.5)$$

Minimizing the total energy density (Eq. 5.5) with respect to l_ϕ yields the phase coherence length

$$l_\phi = \left(\frac{2\pi^2 K}{3V_0\rho_1\sqrt{n_i}}\right)^2. \quad (5.6)$$

The threshold electric field E_T is determined from the electrical energy needed to overcome the total energy per unit volume:

$$\frac{\rho_{eff}E_T\phi_T}{Q} = V_0\rho_1\sqrt{\frac{n_i}{l_\phi^3}} - \frac{K}{2}\left(\frac{\pi}{l_\phi}\right)^2, \quad (5.7)$$

where ϕ_T is the angle through which the CDW has to evolve before it depins.

Tab. 5.1: The phase coherence length l_ϕ and the threshold field E_T for weak pinning in case of no confinement (3D), confinement in the thickness t (2D) and for confinement in both width w and thickness (1D). l_ϕ is given for the isotropic case. To compare l_ϕ to measurements, the anisotropy of the material should be taken into account. The appropriate scaling is described in the appendix.

confinement	l_ϕ	E_T
none	$\left(\frac{2\pi^2 K}{3V_0\rho_1\sqrt{n_i}}\right)^2$	$\frac{Q(V_0\rho_1\sqrt{n_i})^4}{4\rho_{eff}\phi_T\left(\frac{2}{3}\pi^2 K\right)^3}$
thickness	$\left(\frac{\pi^2 K}{V_0\rho_1\sqrt{n_i}}\right)\sqrt{t}$	$\frac{Q(V_0\rho_1\sqrt{n_i})^2}{2\rho_{eff}\phi_T(\pi^2 K)}\frac{1}{t}$
width-thickness	$\left(\frac{2\pi^2 K}{V_0\rho_1\sqrt{n_i}}\right)^{2/3}\sqrt[3]{wt}$	$\frac{Q(V_0\rho_1\sqrt{n_i})^{4/3}}{2\rho_{eff}\phi_T(2\pi^2 K)^{1/3}}\left(\frac{1}{wt}\right)^{2/3}$

When the CDW is confined to a thickness t less than the 3D phase-coherence length in that direction, the CDW does not deform in that direction and the optimization of the pinning energy occurs in 2D defined by the width (along the crystallographic c-axis in NbSe₃) and length (along b-axis). The phase coherent volume l_ϕ^3 is now cut-off by the thickness and becomes $l_\phi^2 t$. In this case, the pinning energy in Eq. 5.3 changes to

$$-V_0\rho_1\sqrt{n_i l_\phi^2 t}, \quad (5.8)$$

and again minimizing the total energy results in a depinning field proportional to $1/t$. When the CDW is only confined in the width direction, E_T has the same form with t replaced by the width w .

When the CDW is confined in both thickness and width, phase deformations occur along the chains (1D), and the pinning energy becomes

$$-V_0\rho_1\sqrt{n_i l_\phi w t} \quad (5.9)$$

resulting in a threshold field proportional to $(1/wt)^{2/3}$.

These results assume that there is no surface pinning so that the elastic energy per unit volume is unchanged from the 3D case. Table 5.1 summarizes results for the phase-coherence length l_ϕ and the depinning field E_T in the 3D, confined 2D and confined 1D cases.

5.2 Previous pinning studies in NbSe₃

The FLR length in high quality CDW materials, like NbSe₃, can be micrometers, allowing study of finite-size effects [7, 8]. Previous studies by McCarten *et al.* [6] showed a crossover from 3-dimensional (3D) to 2-dimensional (2D) collective pinning in NbSe₃ when the crystal thickness (along the crystallographic a^* -axis) was smaller than the phase-coherence length l_ϕ in that direction. Finite-size effects play a key role in understanding CDW physics, since nearly all properties of pinned and moving CDWs depend on the dimensionality imposed by the crystal geometry.

The width (along the crystallographic c -axis) of ribbon-shaped NbSe₃ crystals is almost always much larger than l_ϕ in this direction, so previous work has focused on the 2D limit. Here, we have prepared structures of varying widths both larger and smaller than l_ϕ . We observe a crossover from two-dimensional to one-dimensional (1D) pinning for the first time in CDW systems, and use this crossover to deduce the value of the CDW's phase-phase correlation length in the width direction. The 1D limit provides new opportunities for studying CDW physics, including the dynamics of single phase-coherent CDW domains [9].

5.3 Fabrication techniques

We have used two techniques to fabricate small NbSe₃ structures to probe width-dependent pinning and the 2D to 1D crossover. In the first technique, a crystal is placed on top of a gold contact pattern. The crystal width is then reduced by using either a focused-ion beam (FIB) or by reactive-ion (SF₆) etching. The crystal width can be controllably reduced to ~ 200 nm, provided that the thickness is comparable to or smaller than the final width. Measurements are then performed in a 4-point configuration. A full description of the fabrication process can be found in [10, 11]. In all cases, the initial crystals were thinner than the corresponding phase-coherence length so that the pinning was initially in the 2D limit.

The second technique does not use etching to reduce the crystal size. Instead, bulk NbSe₃ crystals are mixed into pyridine and then shaken in an ultrasonic bath. The ultrasound cleaves the crystals, producing a suspension of NbSe₃ nanowires with typical cross sections ($t \times w$) of $50 \text{ nm} \times 100 \text{ nm}$ and lengths of $20 \text{ } \mu\text{m}$. A drop of this suspension is put on a Si substrate coated with insulating SiO₂ and left to evaporate. The position of the nanowires is determined with respect to predefined markers on the substrate, and e-beam lithography followed by Ti and Au depositions are used to define a contact pattern on top of the wires. Both 2-point and 4-point structures are made with typical contact separations of 2 to $5 \text{ } \mu\text{m}$. The two fabrication techniques complement each other, producing samples with cross sections

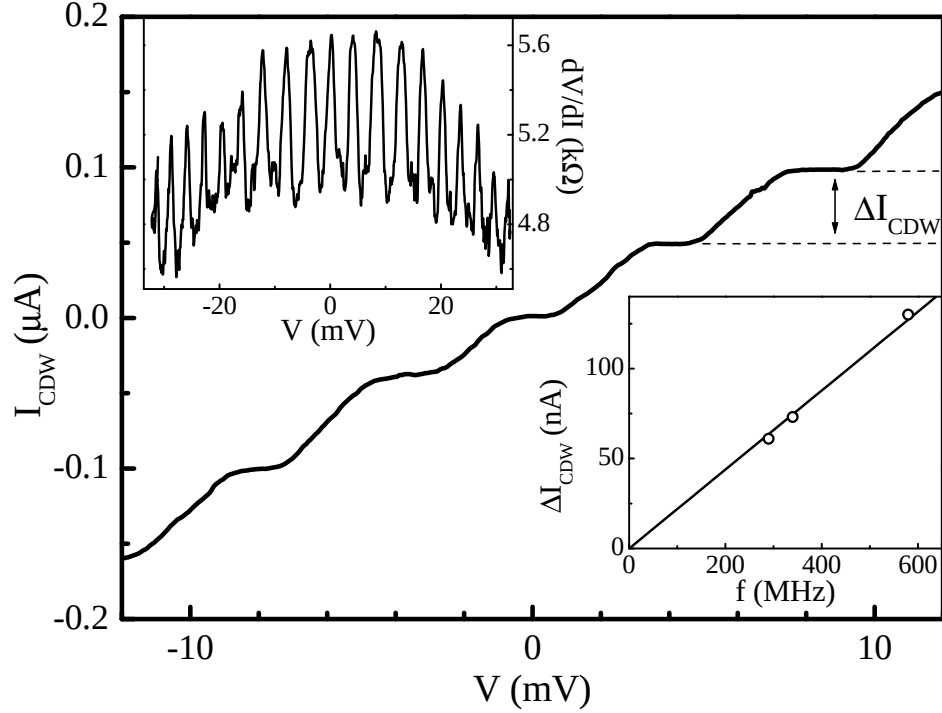


Fig. 5.1: CDW current I_{CDW} as a function of voltage at $T = 50$ K shows flat plateaus, when a radio-frequency signal is applied, indicating complete mode-locking. The applied frequency is 340 MHz. The top-left inset show the differential resistance as a function of voltage. Peaks reaching the same level as at $V = 0$ indicate complete mode-locking. The bottom-right inset shows the distance between steps ΔI_{CDW} as a function of the applied radio-frequency. A linear fit provides an estimate for the number of participating chains in the cross section.

varying by nearly 5 orders of magnitude from 400 nm^2 to $20 \text{ } \mu\text{m}^2$.

5.4 Measurement results

The threshold for CDW sliding E_T was determined from differential resistance (dV/dI) measurements in a helium flow cryostat using a standard lock-in amplifier technique. To characterize current homogeneity within the samples, we have applied large amplitude ac signals to mode lock the internal CDW “washboard” frequency to multiples of the applied frequency at $T = 50$ K and $T = 120$ K. We have observed complete mode locking on samples prepared by both fabrication techniques, indicating homogeneous current flow and that the fabrication techniques do not reduce the high

quality of the crystals, see Fig. 5.1. A linear fit of the distance between mode-locked steps as a function of applied frequency has an error of less than 2%. From this fit, we have determined the sample cross sectional areas and these agree to within 10% with cross sections determined from the room-temperature resistance. Similar results are obtained by comparing the 2- and 4-point resistances, indicating that contact resistances are less than 10% of the nanowire resistance.

Figure 5.2 shows the threshold field E_T as a function of R/L for the FIB and SF₆ etched samples as well as for the much smaller sonicated samples. R/L is the room-temperature resistance between voltage probes, and with the good assumption that the room-temperature resistivity is independent of sample size, R/L is inversely proportional to the cross section $A = wt$. When R/L is less than $\approx 1 \text{ } \Omega/\mu\text{m}$ corresponding to a cross-sectional area larger than $2 \text{ } \mu\text{m}^2$, E_T does not depend on the cross section. For $R/L > 1 \text{ } \Omega/\mu\text{m}$, E_T increases strongly with decreasing cross section. On the log-log plot of Fig. 5.2, E_T follows the solid line with a slope of 2/3 over more than two decades in the abscissa. Data presented here were obtained at $T = 120 \text{ K}$, where contributions to the measured voltage due to the current conversion process at current contacts are minimal.

Figure 5.3 shows the dV/dI curves for four nanowires. The R/L values are shown next to each curve. For low R/L values, the threshold for sliding is easy to determine visually by a sharp decrease of the differential resistance. As the cross section decreases the threshold for sliding is more gradual and harder to determine due to thermal rounding [6, 13, 14, 15]. This contributes to the increased scatter in the data of Fig. 5.2 for very small crystals ($R/L > 1 \text{ k}\Omega/\mu\text{m}$). For nanowires with $R/L > 10 \text{ k}\Omega/\mu\text{m}$, the threshold for sliding could not be determined at $T = 120 \text{ K}$.

We have also investigated E_T 's temperature dependence between $T = 100 \text{ K}$ to $T = 130 \text{ K}$. We have obtained plots where E_T increases with R/L following a straight line on a log-log scale similar to Fig. 5.2. The exponents at $T = 130 \text{ K}$ and $T = 120 \text{ K}$ are 0.67, and increase when decreasing temperature further to a value of 0.76 at $T = 100 \text{ K}$. This increase may be due to increasing contributions to E_T from phase slip processes with decreasing temperature. Typical phase slip voltages are $V_{ps} = 1 \text{ mV}$ at $T = 120 \text{ K}$, which for a $10 \text{ } \mu\text{m}$ long sample is a contribution to E_T of 1 V/cm , while at $T = 100 \text{ K}$ V_{ps} can be 10 mV and its contribution to E_T 10 V/cm .

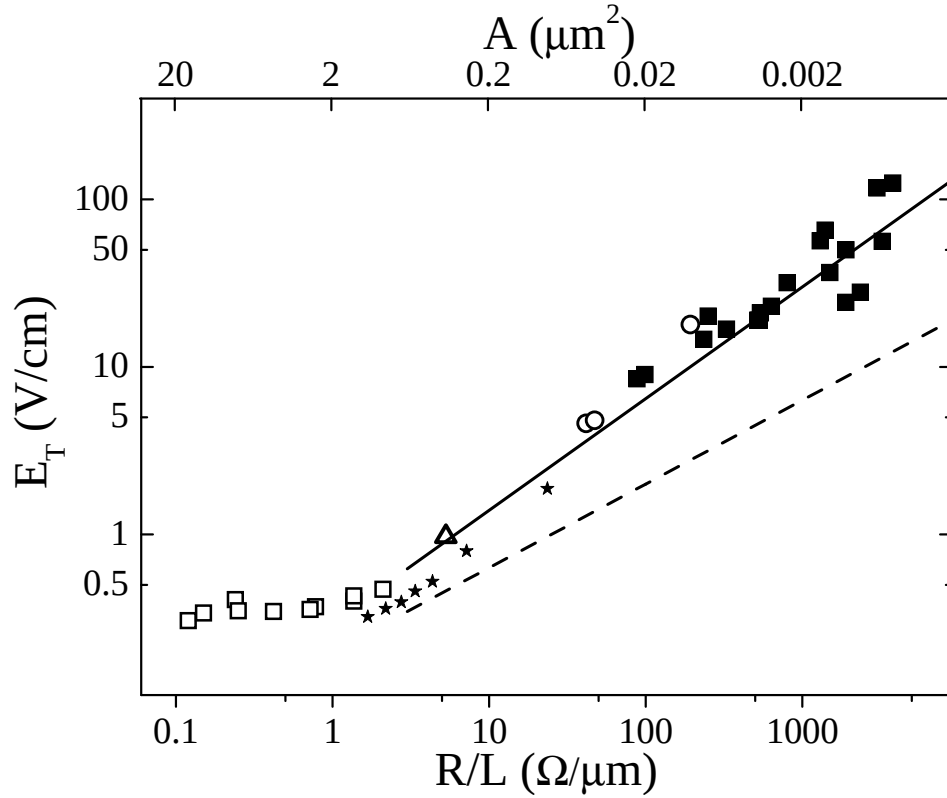


Fig. 5.2: The threshold field E_T at $T = 120$ K as a function of the room-temperature resistance R over the voltage probe separation L . The cross section $A = wt$ is inversely proportional to R/L . The open symbols are data for the three FIB processed crystals, with thicknesses of (squares) $0.56 \mu\text{m}$, (triangle) $0.58 \mu\text{m}$, and (circles) $0.20 \mu\text{m}$. The stars are data for an SF_6 etched crystal with a thickness of $0.7 \mu\text{m}$. These are all smaller than the lower bound of the bulk (3D) CDW phase-phase correlation length in the thickness direction (a^* -axis) of $0.80 \mu\text{m}$ measured by X-ray diffraction [12]. For reference, E_T for an unprocessed crystal with $t = 0.5 \mu\text{m}$ is 0.2 V/cm (Ref. [6]). Each closed (solid) square represents the data of a different nanowire. The solid line displays the $A^{-2/3} = (R/L)^{2/3}$ dependence expected for 1D pinning. The dashed line displays the $A^{-1/2} = (R/L)^{1/2}$ dependence expected for surface pinning.

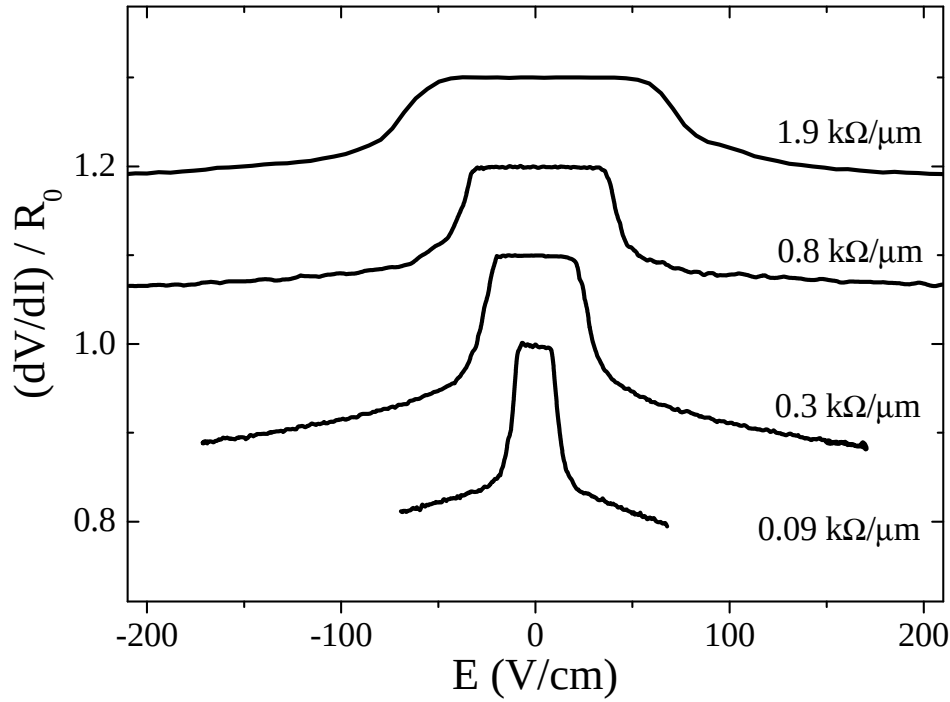


Fig. 5.3: The differential resistance dV/dI normalized by the zero-bias resistance R_0 for four nanowires as a function of electric field at $T = 120$ K. The curves have offsets of 0, 0.1, 0.2, and 0.3. The room-temperature resistance per unit length R/L is shown next to each curve. The onset of CDW conduction becomes more rounded for higher R/L values.

5.5 Conclusions

The CDW in the crystal larger than $2 \mu\text{m}^2$ in Fig. 5.2 begins in the 2D collective pinning regime where $E_T \propto 1/t$, and where pinning related CDW deformations are cut off by the crystal thickness. The crossover to a width-dependent E_T when crystals are etched to small widths is due to a crossover from 2D to 1D collective pinning, in which transverse CDW deformations are cut off by both the thickness and width. This is strongly supported by the measured exponent $E_T \propto A^{-2/3}$ for crystals ranging over two orders of magnitude in cross section, which corresponds with the prediction for 1D collective pinning in Table 5.1. The crossover from 2D pinning to 1D pinning is more easily seen in thick samples, because the phase-coherence length in the width direction is larger for thicker samples. As l_ϕ is confined in the thickness direction, l_ϕ in the width direction will be proportionally smaller as well. The crossover width depends on the thickness of the crystal as $\sqrt{t}$ and is thus smaller for thinner crystals. The crossover width of the sample represented by the squares in Fig. 5.2 is $1.6 \pm 0.2 \mu\text{m}$. This value is a factor of two larger than the bulk (3D) CDW correlation length in the c^* -axis¹ of $0.75 \mu\text{m}$ measured by X-ray diffraction [12]. This provides strong additional support for a 2D to 1D pinning origin.

The value of the phase-coherence length l_ϕ for thickness confinement is determined using literature values and the thickness of our largest sample $t = 560 \text{ nm}$. The value for l_ϕ in the width direction is about $3.2 \mu\text{m}$. This is remarkably close to the observed crossover width of $1.6 \mu\text{m}$, since the expression for l_ϕ is the result of an order-of-magnitude calculation. A more accurate determination of the phase-coherence length and the crossover width involves numerically solving the FLR Hamiltonian with the proper boundary conditions.

An increase of E_T with decreasing crystal cross section could also arise due to pinning by crystal surfaces [7, 8]. In this case E_T is determined by the crystal surface-to-volume ratio. For thin but wide ($w \gg 1 \mu\text{m}$) crystals, $E_T \propto 1/t$ for both surface pinning and 2D collective pinning, making these mechanisms difficult to distinguish [6]. But in crystals where the CDW is confined in both width and thickness, E_T due to surface pinning is proportional to $(w + t)/wt$. Since the width-to-thickness ratio w/t shows only a small sample-to-sample variation compared with the nearly three orders-of-magnitude variation of cross-sectional area in Fig. 5.2, this simplifies to $E_T \propto A^{-1/2}$. This differs from the 1D collective pinning prediction $E_T \propto A^{-2/3}$, and as indicated by the dashed line in Fig. 5.2 is clearly inconsistent with the experimental data. This indicates, somewhat surprisingly, that surface pinning remains unimportant even in the high purity crystals

¹ The crystallographic c^* -axis is at a 19.5 degree angle with the c -axis, both are perpendicular to the chain direction (b -axis)

with cross sections as small as 400 nm^2 .

In the 1D limit, the total energy of a phase coherent domain is proportional to $(wt)^{2/3}$. Accounting for NbSe_3 's anisotropy and using $K = 3.5 \text{ meV/\AA}$, $V_{0\rho_1} = 4 \text{ meV}$ and $n_i = 2.5 \times 10^{16} \text{ cm}^{-3}$ taken from Refs. [6] and [16], we estimate the total energy of a 1D domain in a crystal with $R/L = 2 \text{ k}\Omega/\mu\text{m}$ to be only $\approx 4.5 \text{ kT}$ at $T = 120 \text{ K}$, compared with bulk 3D values of 10^5 K . This small energy explains the pronounced thermal rounding in the depinning transition visible in the dV/dI for these small crystals evident in Fig. 5.3.

The number of pinning centers per unit length decreases with decreasing cross-sectional area. Eventually the separation between pinning centers is comparable to the phase-coherence length along the length. This changes pinning for samples with small cross-sectional areas. The separation of pinning centers for our smallest sample is estimated to be 100 nm (using $n_i = 2.5 \times 10^{16} \text{ cm}^{-3}$ and $wt = 400 \text{ nm}^2$), while the predicted 1D phase-coherence length, taken from Table 5.1, is 350 nm for the smallest sample. Assuming these numbers, this means that for all samples shown in Fig. 5.2 the phase-coherence length is larger than the separation between pinning centers and the weak pinning limit applies to all our samples.

The actual type of pinning centers within the samples are not known. We have used undoped NbSe_3 samples with bulk residual resistance ratios (RRR) which are known to be 300-400. From the RRR, we can estimate the impurity density. For Ti impurities, an RRR of 400 gives $n_i = 2.5 \times 10^{16} \text{ cm}^{-3}$, while for Ta impurities $n_i = 7.5 \times 10^{17} \text{ cm}^{-3}$ [6]. Using the Ta impurity density, the 1D phase coherence length becomes 110 nm instead of 350 nm for the smallest samples, but the impurity separation will now be only 3 nm (using $wt = 400 \text{ nm}^2$). Assuming only Ti and Ta impurities, this puts our samples comfortably in the weak pinning limit.

The samples will most likely contain a combination of charged impurities (like Ti) and isoelectronic impurities (like Ta), both of which are substitutional impurities. The relation between RRR and the impurity concentration for NbSe_3 is only known for Ti and Ta. It is therefore hard to estimate the actual impurity density when the impurity type is unknown. The actual impurity density is therefore probably somewhere between the values calculated for Ti and Ta.

For samples that have been processed by a FIB, Ga impurities may be implanted as interstitial impurities. This results in a higher impurity density and an increase of E_T compared to cleaved unetched samples of the same sizes. Evidence for Ga implantation can be seen in Fig. 5.2 by the somewhat larger E_T of a FIB processed sample (open squares) compared to an unprocessed sample of similar thickness (indicated by the 0.2 V/cm line in the graph).

All these considerations have no effect on the validity of the observation of the 2D to 1D crossover. However, depending on the actual impurity

density, these have consequences for the amount of observed rounding in the dV/dI . For a higher n_i , the pinning energy per phase coherent domain is too large to explain the amount of observed rounding in the dV/dI .

5.6 Summary

In summary, we have shown a dimensionality crossover from two-dimensional to one-dimensional weak pinning in NbSe₃. 1D pinning is observed when both the width and thickness are smaller than the CDW's bulk phase coherence length in these directions. The observation of 1D pinning behavior is not an artefact of the fabrication technique, since it is observed for samples prepared using three different methods. From the width dependence of E_T we estimate the CDW's phase-coherence length in that direction to be 1.6 μm , comparable to the value obtained by X-ray diffraction [12].

The ability to reach the 1D limit should provide new opportunities for studying CDW physics. For example, in ordinary size crystals the cross section contains a large number of phase correlated domains in the pinned state and of dynamically correlated domains in the depinned state. In 1D crystals, the cross section will contain a single domain in both the pinned state and in the depinned state at modest fields, and the length of the domain will be $\approx 1\text{-}10\ \mu\text{m}$. This will allow the detailed dynamics on the scale of a single domain to be explored with nanofabricated probes spaced along the crystal [9, 17]. Large-scale numerical simulations by Matsukawa [18, 19, 20] suggest that the CDW's dynamical correlation length should be larger or comparable to the pinned correlation length out to at least a few times E_T . Theoretically, the size of the critical regime near the depinning transition should be larger in 1D [21, 22, 23, 24, 25], and will therefore make it experimentally accessible, although equally interesting finite-size effects in the dynamics [23, 24, 25] should also become more prominent in 1D.

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Appendix: Appropriate scaling

This appendix describes the appropriate scaling for the anisotropy in the phase-coherence length, following Lee and Rice [2]. By rescaling the length scale in the transverse direction, an anisotropic system can be regarded as an isotropic system. The Ginzburg-Landau expansion contains the amplitude coherence lengths ξ_x , ξ_y and ξ_z , where ξ_z is along the chains. These are used to rescale the anisotropic system to the isotropic system. The following Table shows how the the rescaling is performed.

Anisotropic system	Isotropic system
x (a*-axis)	$(\xi_z/\xi_x)x$
y (c-axis)	$(\xi_z/\xi_y)y$
K	$(\xi_x\xi_y/\xi_z^2)K$
ρ_{eff}	$(\xi_x\xi_y/\xi_z^2)\rho_{eff}$

Estimates for the ratio $\xi_{b^*} : \xi_{a^*} : \xi_{c^*} = 1 : \frac{1}{9} : \frac{1}{25}$ as found in Ref. [6] come from X-ray measurements. The ratio $\xi_z : \xi_x : \xi_y$ should be nearly the same, because the c-axis and the c*-axis only differ by 19.5 degrees, whose cosine is 0.943.

6. One-dimensional conductance in Charge-Density Wave nanowires

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Abstract

We report a systematic study of the transport properties of coupled one-dimensional metallic chains as a function of the number of parallel chains. When the number of parallel chains is less than 2000, the transport properties show power-law behavior on temperature and voltage, characteristic for one-dimensional systems.

6.1 One-dimensional metals

Electron-electron interactions in one-dimensional (1D) metals give dramatically different behavior as opposed to those of a three-dimensional Fermi liquid of electrons. Depending on the details of the electron-electron interaction, several phases are possible, such as a Luttinger Liquid (LL) or a 1D Wigner Crystal (WC) for low electron density. The tunneling density-of-states (TDOS) of these 1D phases exhibit power-law behavior on the larger one of either eV or $k_B T$ [1, 2, 3], with V the voltage and T the temperature.

Power-law behavior has been observed in seemingly different 1D systems, such as ballistic single-wall [4] and diffusive multi-wall carbon nanotubes [5], degenerately doped semiconductor nanowires [6], and fractional quantum Hall edge states [7]. Each of these systems revealed new behavior, but also raised questions. In multiwall carbon nanotubes (MWCN), the different nanotube shells interact with each other and the question arises how this interaction affects the 1D properties. It is still under debate whether the observed power-law behavior in MWCNs should be described by LL or by Environmental Coulomb Blockade Theory (ECBT) [5].

Theoretically, it has been shown that the LL state survives for a few 1D chains coupled by Coulomb interactions [8, 9]. However, when *interchain* hopping $t_\perp$ is taken into account, the LL state is destroyed for low temperatures. For example, for low $t_\perp$ (weak coupling), electrons may be confined to the chains by the Mott insulator gap. When $t_\perp$ is large, Fermi liquid is recovered [10]. The LL state exists at finite $t_\perp$ for not too low temperatures.

It is less clear what the situation is when many (> 10) chains are coupled together in the presence of disorder. A single impurity in a 1D chain forms a tunnel barrier [11, 12], but with increasing number of impurities the situation may be different. For instance, many impurities can form a potential landscape different from a series of tunnel barriers.

Recent theoretical investigations show that a LL state can also be stabilized in the presence of impurities for system with more than two coupled chains [13]. Furthermore, formation of a WC is expected in the limit of strong Coulomb interactions, or low electron density [2, 14]. All these considerations lead to a large parameter space, and at present a full theory of 1D transport in disordered, multichannel systems is still lacking.

6.2 Tunneling density-of-states

The tunneling density-of-states (TDOS) can be determined by looking at transport measurements across a tunnel barrier. The differential conductance $G = dI/dV$ in such transport measurements is proportional to the TDOS. When power-law behavior is expected for the TDOS of 1D phases, this should be observable in transport measurements as a power-law in the

differential conductance of systems that exhibit such 1D phases. Equivalently, the current-voltage characteristic will also exhibit power-law behavior, with the exponent of the TDOS plus one.

The tunneling density-of-states (TDOS) of a single 1D wire is a power-law on energy:

$$\nu(E) \propto E^\alpha, \quad (6.1)$$

where E is the larger one of either eV or $k_B T$ [1, 2, 3], with V the voltage and T the temperature. The exponent α depends on the number of channels within the wire. For tunneling into the bulk of a Luttinger liquid (with two spin channels), the exponent α is:

$$\alpha_{bulk} = \frac{g + g^{-1} - 2}{4}, \quad (6.2)$$

with g the spin-independent electron-electron interaction parameter. For tunneling into a Luttinger liquid near the end of the wire, the exponent α is:

$$\alpha_{end} = \frac{g^{-1} - 1}{2}. \quad (6.3)$$

These results are derived for a single wire with two channels without disorder. The exponents for bulk- and end-tunneling have been measured in Single Wall Carbon Nanotubes (SWCN) by using different contact geometries. SWCN have four channels and values have been found of $\alpha_{bulk} \approx 0.3$ and $\alpha_{end} \approx 0.6$. Taking into account that SWCN have four channels instead of two, the interaction parameter $g \sim 0.2 - 0.3$ [4].

6.3 Multi-channel wires

It is well-known that Coulomb blockade is smeared due to quantum fluctuations in the leads. The smearing is modelled by tunneling of an electron into a so-called ‘environment’, where the electron interacts with the electromagnetic modes of the environment. This model, Environmental Coulomb Blockade Theory (ECBT), describes tunneling into a wide metal wire with many modes, where the width of the wire is much larger than the Fermi wavelength. For ECBT, the tunneling density of states also depends on energy as a power-law:

$$\nu \propto E^\alpha. \quad (6.4)$$

Here, α depends on the impedance Z of the environment:

$$\alpha_{ECBT} = \frac{e^2}{\pi \hbar} Z(0) = 2 \frac{Z(0)}{R_Q}, \quad (6.5)$$

with e the electron charge, $\hbar$ Planck’s constant, and $Z(0)$ the zero-frequency impedance. When $Z(0)$ is smaller than the quantum resistance $R_Q =$

$2\pi\hbar/e^2$, coulomb blockade is smeared by fluctuation of charge in the environment. For $Z(0) \gg R_Q$ ($\alpha \gg 1$), ECBT is no longer applicable and full coulomb blockade is recovered.

It follow that in both opposite confinement limits (wide wires and narrow wires), the TDOS is a power-law, however with different exponents. To make a connection with both these limits, tunneling into a multi-channel wire with an arbitrary number of transverse modes was considered by Matveev and Glazman [3]. They found that between these limits, the exponent α depends on the number of channels N . When all channels contribute equally the exponent for $N \gg 1$ is approximately

$$\alpha(N) \sim \sqrt{\frac{V_0}{\pi\hbar v_F N}}, \quad (6.6)$$

with V_0 a zero-momentum interaction potential and v_F the Fermi velocity. The exponent decreases with increasing cross sectional area A of the wire as $\alpha \propto 1/\sqrt{A}$. In the limit $N \rightarrow \infty$, $\alpha \rightarrow 0$ and the transport is ohmic, as expected.

6.4 Disorder in wires

Impurities or disorder in a wire change the properties of 1D electron systems dramatically. A few impurities in a wire effectively divide the wire into several segments, which can be viewed as a series of clean 1D electron systems connected by tunnel barriers. For non-interacting electrons in random disorder, which means many impurities, electrons are repeatedly backscattered and are thus localized. Localization for non-interacting electrons is by now well-understood, with models such as Anderson localization and Variable range hopping. When the localization length is smaller than the inter-particle distance, the electrons can be treated classically and the TDOS exhibits a Coulomb gap. In the opposite limit, where the localization length is larger than the inter-particle distance, electron systems with long-range Coulomb interaction can be considered as Wigner crystals.

6.5 Wigner crystallization

When the Coulomb interaction between electrons is larger than the kinetic energy of the electrons, the electrons will be localized and may form a Wigner crystal. The Wigner crystal may interact with impurities in much the same way as CDWs do. When the localization length is larger than the impurity spacing, the weak pinning regime, the TDOS follows a power-law [2]:

$$\nu \propto E^\alpha, \quad (6.7)$$

Tab. 6.1: The value of the power-law exponent α for three different models, with indicative values found either experimentally or numerically.

Model	exponent	found values
LL	$\frac{g+g^{-1}-2}{4}$	0.3-0.6 [4]
ECBT	$\frac{e^2}{\pi\hbar}Z(0)$	$\alpha < 2$ [3]
WC	$\frac{\sqrt{1+\eta}}{2}$	2-5 [2]

where α is $\alpha_{WC} = \frac{\sqrt{1+\eta}}{2}$, with

$$\eta = \frac{e^2}{2\pi\hbar v_F} \frac{1}{\epsilon_0\epsilon} \ln \frac{L_0}{a}, \quad (6.8)$$

where ϵ is the wire's dielectric permittivity (ϵ_0 for vacuum), L_0 is a localization length, and a is a lattice electron constant. Using $v_F = 2.5 \times 10^7$ cm/s, $\epsilon \sim 1$ -5, and $\ln L_0/a \sim 4$ -6, gives values for α_{WC} of 2-5.

6.6 Charge-Density Wave nanowires

We show that charge-density wave (CDW) nanowires exhibit the characteristic behavior for 1D transport. We use undoped NbSe₃, which consist of metallic chains weakly coupled by Van der Waals forces. When the nanowires consist of thousands of chains, the transport measurements clearly show the bulk CDW characteristics, see Ref. [15] and Chapter 5. In this limit, CDWs can be viewed as the classical analogue of a LL-state. However, we find that at low temperatures our nanowires with less than 2000 chains show power-law dependencies on both voltage and temperature characteristic for 1D transport.

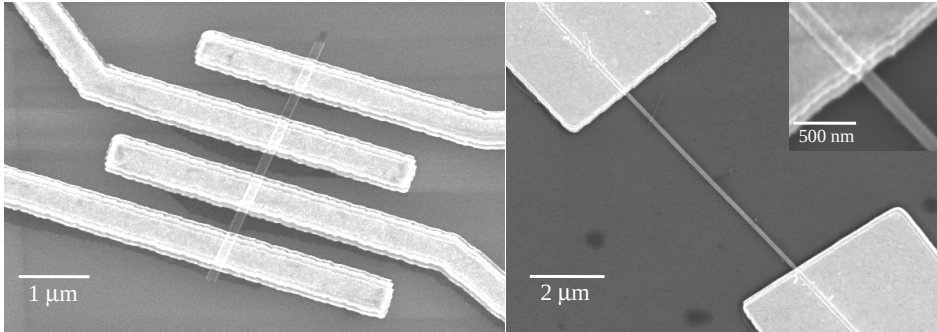


Fig. 6.1: Scanning Electron Microscope images of NbSe₃ nanowires. Left: A four-probe device. Right: A two-probe device. Right inset: A zoom-in on the contact of the two-probe device.

The monoclinic unit cell of NbSe₃ contains six metallic chains along the b direction. Its lattice parameters are $a = 10.0$ Å, $b = 3.5$ Å, $c = 15.6$ Å, and $\theta = 109.5^\circ$ [16]. NbSe₃ is a metallic CDW material with a partially gapped Fermi surface. Two Peierls transitions occur at $T_{P1} = 145$ K and $T_{P2} = 59$ K. Below the second Peierls transition nearly all of the electrons are condensed into both CDWs. The few remaining uncondensed electrons, with a very low electron density of $n = 1.1 \times 10^{18}$ cm⁻³ [17], provide metallic conduction down to the lowest temperatures.

NbSe₃ nanowires were made from bulk NbSe₃ crystals by ultra-sonically cleaving the crystals in a pyridine solution. After several hours of cleaving, a suspension of NbSe₃ nanowires with widths ranging from 30 nm to 300 nm and lengths from 2 to 20 μ m, emerges. A drop of the suspension is deposited onto a degenerately doped Si substrate with predefined markers. The Si can serve as a backgate, but is connected to ground in the measurements shown here. Nanowires are selected and located with an optical microscope with respect to the predefined markers. Subsequently a contact pattern is defined with e-beam lithography. Gold and a Ti sticking layer are deposited within minutes after a 3 second dip in ammonium buffered hydrofluoric acid to optimize contact resistances. Both two-probe and four-probe samples were made, see Fig. 6.1. In total, 21 nanowires have been measured. One wire (4P sample in Fig. 6.2) has also been measured after prolonged exposure to air. We found that the room-temperature resistance of this nanowire monotonically increased with exposure time (a factor 2 of resistance increase after 1100 hours). Most likely, the surface slowly oxidizes thereby reducing the number of chains that participate in conduction.

6.7 Measurements of NbSe₃ nanowires

Figure 6.2 shows the zero-bias resistance R per unit length L as a function of T for several nanowires plotted on a log-log scale. All samples, both large and small, show metallic behavior at room temperature. The samples with the largest cross sectional areas are at the bottom of the graph and show the two Peierls transitions clearly. These samples remain metallic down to $T = 4.2$ K, similar to bulk crystals. The samples with the smallest cross sectional areas are at the top of the graph. When the room-temperature unit-length resistance R_{300K}/L is larger than 10 k Ω/μ m, the Peierls transitions become increasingly less visible while the transition temperatures show a small monotonic decrease of up to 20% for the smallest sample, see inset of Fig. 6.2.

The most remarkable behavior is the change from metallic to non-metallic behavior at low temperatures when the number of parallel chains decreases. This transition occurs around $R_{300K}/L = \rho/A = 4$ k Ω/μ m corresponding to a cross section $A = 500$ nm² and a total number of 2000 chains (bulk NbSe₃

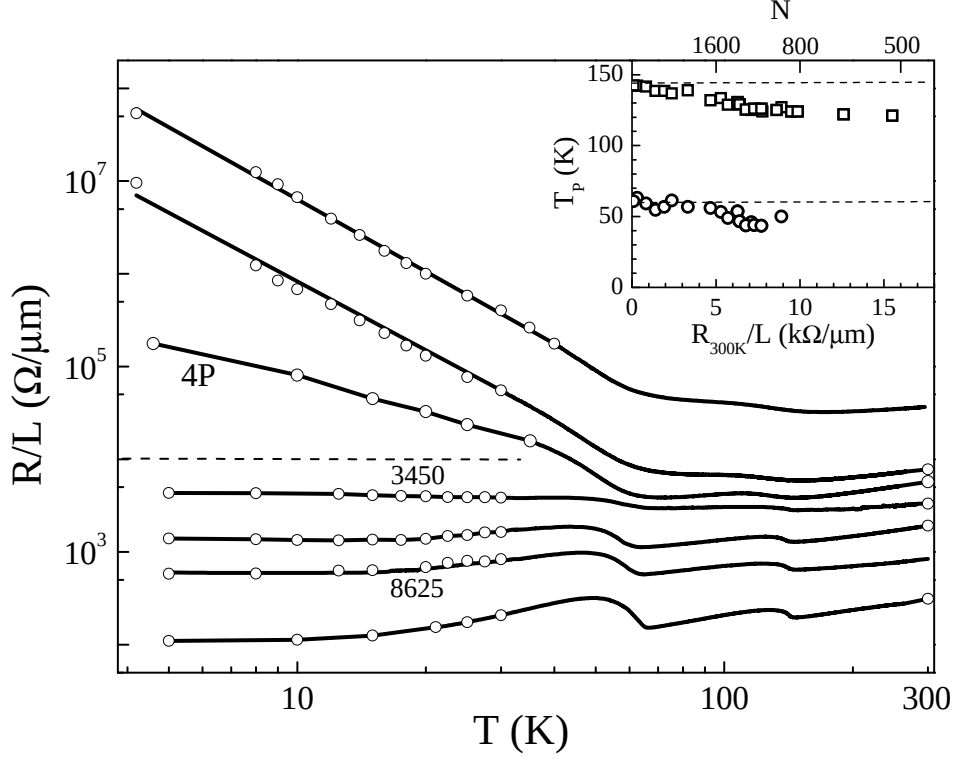


Fig. 6.2: Resistance per unit length R/L as a function of T on a log-log scale. Small nanowires (high R_{300K}/L value) show metallic behavior at room temperature, but show non-metallic behavior at low temperature. All curves are two-probe measurements except for the one indicated by 4P. The open circles are data points taken manually from IV curves. The number of chains, deduced from Shapiro-step measurements, is indicated in the figure for two samples. The number of chains deduced from room-temperature resistance measurements (not indicated in the graph) are from bottom to top: 25640, 9490, 4170, 2430, 1400, 1030, 220. Inset: Peierls transition temperature as a function of R_{300K}/L and the corresponding number of parallel chains.

resistivity $\rho = 2 \text{ } \Omega\mu\text{m}$).

To characterize the sample quality, we have performed Shapiro-steps measurements at $T = 120 \text{ K}$ and $T = 50 \text{ K}$ on two samples that show the Peierls transitions. A Figure of the mode-locking experiment at $T = 50 \text{ K}$ can be found in Chapter 5. We were able to mode-lock the CDW completely to an external radio-frequency signal on both samples. From the distance between mode-locking steps we deduced the number of chains in the cross section, indicated by the numbers in Fig. 6.2. The cross sectional area deduced from the number of chains agrees to within 10% of the value obtained from room-temperature resistance. This observation indicates that the wires are homogeneous and that the contact resistance at $T = 120 \text{ K}$ is small compared to the nanowire resistance.

Of the 7 curves presented in Fig. 6.2, one has been obtained from a four-probe measurement, indicated by 4P. For this sample, the 4P resistance is equal to the two-probe resistance to within 10% over the entire temperature range 4.2 K to 300 K. The interface of the contact and the nanowire is transparent and therefore, the non-metallic behavior at low temperature seen for this sample is not an effect of the interface.

At low temperature for all samples with $R_{300\text{K}}/L > 5 \text{ k}\Omega/\mu\text{m}$, power-law behavior $R \propto T^{-\alpha}$ is observed, as illustrated in Fig. 6.2 and in Fig. 6.3. In the inset of Fig. 6.3 we have plotted all the α -values as a function of $R_{300\text{K}}/L \propto 1/A$. The exponents seem to increase as the number of chains becomes smaller. The exponents can also be divided into two groups around $\alpha = 1$ and $\alpha = 2$. The $R(T)$ curves have also been fitted to the thermal activation and Mott hopping law $R \propto \exp(T_0/T)^\delta$, where $\delta = 1$ for thermal activation and δ depends on dimensionality for Mott hopping [18]. Neither model provides a satisfactory fit to the data.

Figure 6.4 shows the differential resistance per unit length $(dV/dI)/L$ as a function of the voltage per unit length E at $T = 4.2 \text{ K}$ for three samples. The nanowire with the largest cross section is at the bottom of the graph. When the cross section gets smaller, the threshold field for sliding E_T increases as expected, see Ref. [15] and Chapter 5. A peak in the differential resistance develops for $E < E_T$, which grows as the cross section gets smaller. The top $(dV/dI)/L$ curve follows a power-law $I \propto V^\beta$ at high bias, with $\beta = 1.7$ and on temperature with $\alpha = 2.2$. Therefore, power-law behavior is observed simultaneously with CDW features. In this respect our results are different from earlier attempts to observe 1D transport in CDW conductors [19, 20], where complete disappearance of both transitions was reported.

Figure 6.5 shows the current-voltage (IV) curves for a single sample at several temperatures. At $T = 50 \text{ K}$, the IV is almost linear, but at $T = 4.2 \text{ K}$ the IV is highly non-linear. All IV curves collapse unto a single master curve, by plotting $I/T^{1+\alpha}$ versus $eV/k_B T$, where $\alpha = 2.15$ is the power-law exponent from the $R(T)$ curve. The scaled IV curves are non-linear above $eV \approx 80 k_B T$, and for $eV > 80 k_B T$, the IV follows a power-

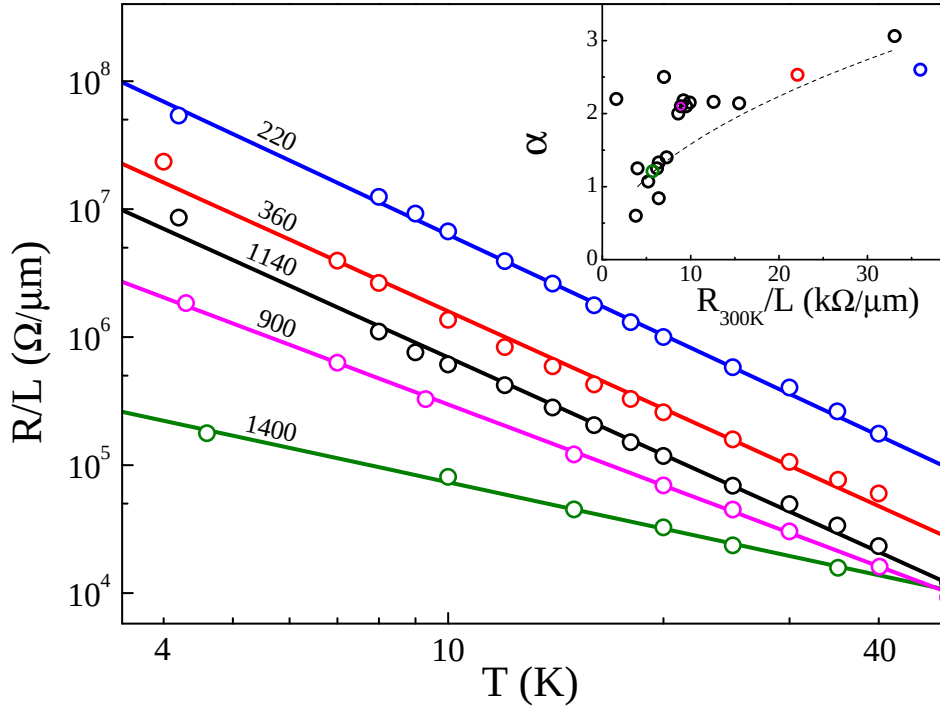


Fig. 6.3: Unit-length resistance R/L as a function of temperature T on a log-log scale. The lines are linear fits to the data from which the power-law exponent α is deduced. The number of chains, deduced from two-probe room-temperature resistance measurements, is indicated for each nanowire. The inset shows the exponents for all samples measured as a function of the room-temperature resistance per unit length.

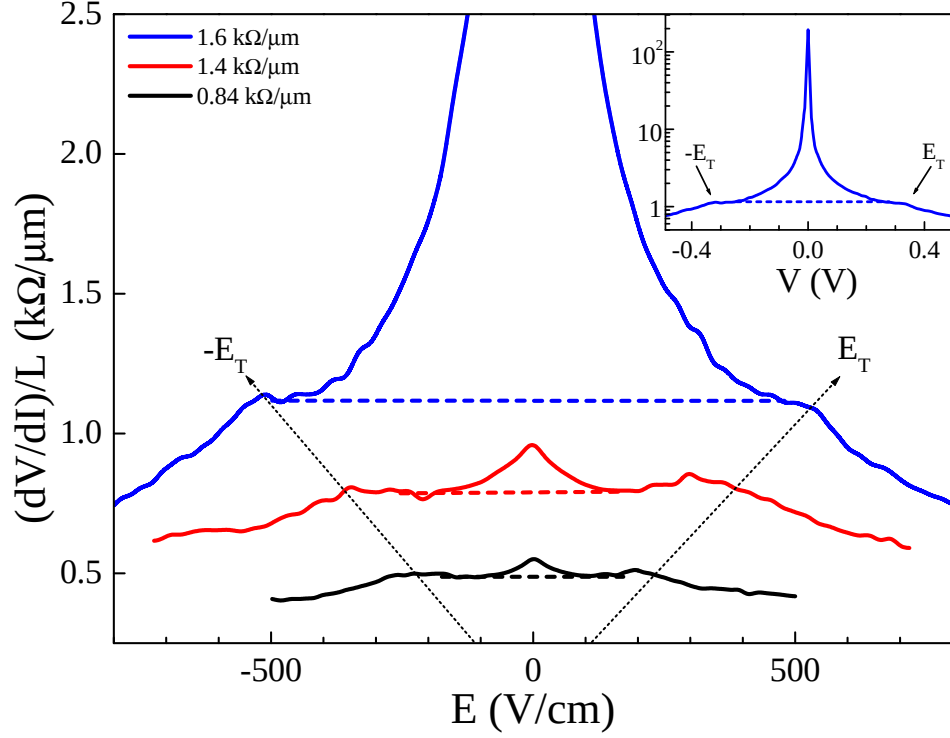


Fig. 6.4: Differential resistance per unit length $(dV/dI)/L$ as a function of the voltage per unit length E of three nanowires at $T = 4.2 \text{ K}$. The threshold field for sliding E_T increases when the cross sectional areas gets smaller, indicated by the dashed arrows. Below E_T where the CDW is pinned, a peak in dV/dI is observed. The dashed lines indicates the shape of the dV/dI expected for bulk samples. The room-temperature unit length resistance is displayed for each curve. The inset shows the $(dV/dI)/L$ of the top curve as a function of the voltage V ; the maximum resistance is $190 \text{ k}\Omega/\mu\text{m}$ and $L = 6.2 \mu\text{m}$.

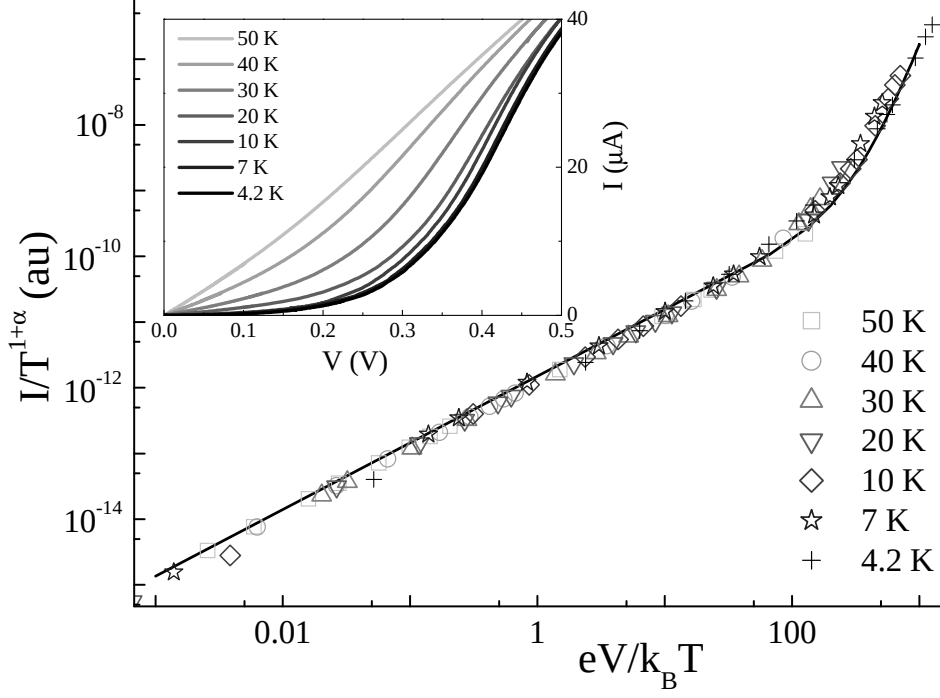


Fig. 6.5: Scaled current-voltage IV curves on a log-log scale. The IV s collapse onto a universal curve from $T = 4.2$ K to $T = 50$ K for $\alpha = 2.15$. The inset shows the unscaled IV s ($L = 1.4 \mu\text{m}$).

law with $\beta = 4.2$ for this sample. It is important to note that sliding of the CDW occurs at higher voltages [15] as is evident from the measurement at $T = 50$ K. At this temperature, the threshold field for sliding is the lowest and we still observe linear behavior in the inset of Fig. 6.5.

The scaled master curve can be fitted to a general equation to describe IV s for bosonic excitations in 1D [21, 22]:

$$\frac{I}{T^{1+\alpha}} = C \sinh\left(\gamma \frac{eV}{k_B T}\right) \left| \Gamma\left(1 + \frac{\beta}{2} + i\gamma \frac{eV}{\pi k_B T}\right) \right|^2, \quad (6.9)$$

where Γ is the complex gamma function, C is a proportionality constant (in arbitrary units) and γ determines the position of the “knee” in the IV curve. The parameters α and β are the two experimentally determined exponents for the temperature and voltage dependence respectively. The fit to the data is depicted by the solid line in Fig. 6.5, with $C = 2.3 \times 10^{-11}$ and $\gamma = 77^{-1}$. IV s have also been measured for another sample with $\alpha = 2.16$ and $\beta = 4.7$ and again a similar fit could be made. The fit parameters for this sample are $C = 1.1 \times 10^{-11}$ and $\gamma = 100^{-1}$.

6.8 Discussion and possible models

The power-law behavior in NbSe₃ nanowires results from a reduction of cross section below about 2000 parallel chains. The data clearly show that CDW sliding does not play a role. Power-law dependence is observed below 50 K suggesting that the low electron density below the second Peierls transition is important. The measurements show that these carriers dominate transport and govern the metallic to nonmetallic transition. The CDW state still exists, because power-law behavior is observed simultaneously with the threshold field for CDW sliding (Fig. 6.4). Furthermore, $T_{P1,2}$ decreases only slightly with the number of chains (inset of Fig. 6.2). This is in agreement with the expectation that already a small number of chains (≈ 10) stabilizes the CDW state [23]. In the remainder of this paper, we discuss the 1D transport mechanism and compare our results with different models.

When comparing NbSe₃ nanowires to other systems that show power-law behavior, we have to take into account that NbSe₃ is a diffusive conductor. Therefore comparison to a single channel LL without disorder, such as SWCNs, is inappropriate. We also have to take interaction between chains into account. A suitable system to compare NbSe₃ nanowires to is MWCNs, which are diffusive conductors with interaction between the nanotube shells. MWCN have been modelled using ECBT [5, 24, 25]. The IV follows a power-law on energy, with an exponent $\beta \equiv \alpha + 1 = 2Z/R_Q + 1$ that depends on the impedance Z of the environment [3], see Eq. 6.5. Here, R_Q is the quantum resistance.

Following Ref. [5], we model the NbSe₃ nanowires as lossless transmission lines with $Z = \sqrt{l/c}$. The kinetic inductance per unit length $l = m^*/(e^2 n A)$, where the mass of the charge carriers $m^* = 0.24m_e$ [26], with m_e the electron mass, e the elementary charge. The capacitance per unit length $c = 4\epsilon_0\epsilon_r/(\pi \ln(4d/w))$, for a rectangular wire above a ground plane at distance d , with ϵ the permittivity ($\epsilon_r = 3.9$ for Si-oxide), and w the wire's width. Taking $A = 500 \text{ nm}^2$, the kinetic inductance $l = 16 \text{ nH}/\mu\text{m}$ is much larger than the wire's geometrical inductance ($\text{pH}/\mu\text{m}$). For $d = 1 \mu\text{m}$ and $w = 50 \text{ nm}$, $c \approx 10 \text{ aF}/\mu\text{m}$, so that $Z = \sqrt{l/c} \approx 1.6 R_Q$ and $\beta = 4.2$, in agreement with the exponent measured in the IV at high bias.

The assumption of a lossless transmission line is valid when the inductive part ωl is larger than the resistive part R/L of the impedance, i.e. for high bias: $\hbar\omega = eV > \hbar(R/L)/l \approx 0.4 \text{ meV}$, for $R/L = 10 \text{ k}\Omega/\mu\text{m}$. We have observed the power-law in the IV well above 0.4 mV and therefore the assumption of a lossless transmission line is valid.

ECBT theory may also explain an increase of α for smaller wires, because the impedance depends on the cross section through the kinetic inductance. Since the capacitance to a ground plane depends only weakly on the width of the wires, $\alpha \propto \sqrt{1/A}$ (dashed line in the inset of Fig. 6.3). This dependency is in agreement with the derived exponent as a function of the number of

modes in a quasi-1D wire [3]. However, ECBT is derived for a single tunnel barrier connected to the impedance of the environment, which may not be the case for NbSe₃ nanowires. Also, ECBT does not explain the observation $\beta \neq \alpha + 1$ and the knee at $eV \approx 80 k_B T$. Note that the observation $\beta \neq \alpha + 1$ has also been found in InSb nanowires [6].

An alternative model to describe NbSe₃ nanowires is the case of a 1D disordered conductor with low electron density, where Wigner crystallization may occur. Wigner crystallization occurs when the Coulomb energy E_C is larger than the kinetic energy E_F of the electrons or, or in other words, when the coupling parameter $\Gamma_C = E_C/E_F$ is large. In NbSe₃, Γ_C is large below T_{P2} , because the electron density and hence E_F is exceptionally small. A WC can adjust its phase in the presence of disorder to optimize the pinning energy gain, much like CDW systems or vortex lattices. When the localization length is larger than the distance between impurities, the TDOS follows a power-law [2]. The power-law exponent is determined by the localization length and high values of 3-6 are predicted, similar to values we found.

6.9 Conclusions

In conclusion, we have studied the transport properties of weakly coupled metallic wires in the presence of disorder. We find power-law behavior characteristic for 1D systems when the number of chains is smaller than about 2000. A model to describe NbSe₃ nanowires is not available. Ingredients for such a model should include interaction between metallic chains, disorder, confinement and the low electron concentration.

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7. Charge-Density Wave Point-contacts

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Abstract

This chapter describes the measurements of insulating point contacts in NbSe₃. The point contacts were fabricated with a Focused Ion-Beam (FIB) or created in a Mechanically Controlled Break Junction (MCBJ). The point contacts are typically smaller than 100 nm, connecting two large parts of a single Charge-Density wave conductor. Differential conductance measurements of a FIB point contact at low temperatures show peaks in the differential conductance at ± 105 mV and ± 190 mV, corresponding to the two gaps of NbSe₃. Low-temperature measurements of a MCBJ of NbSe₃ shows peaks in the differential conductance at ± 81 mV and ± 196 mV.

7.1 Introduction

The CDW ground state can be described by an order parameter: $\Delta = |\Delta|e^{i\phi}$. $|\Delta|$ is related to the energy gap for single particle excitations, similar to the energy gap in the ground state of superconductors. The derivative of the phase ϕ is related to the movement of the CDW condensate. In superconductors, $d\phi/dx$ is proportional to velocity of the condensate (i.e. the supercurrent); in CDWs, $d\phi/dt$ is proportional to the velocity of the condensate. The similarities of the ground state properties are apparent in experiments involving tunneling spectroscopy. The field of superconductivity is in this respect far more explored than CDW systems. This is mainly due to the available fabrication techniques for superconductor junctions, weak links and heterostructures of superconductors.

The density-of-states (DOS) of conventional superconductors have peaks at $\pm\Delta$ around the Fermi level. Using the semi-conductor model for electron tunneling in superconductors, these DOS peaks give rise to peaks in the differential conductance (dI/dV) of superconducting tunnel junctions. In an NIS junction the peaks appear at voltages $\pm\frac{\Delta}{e}$ while in an SIS junction the peaks appear at voltages $\pm\frac{2\Delta}{e}$. Applying the same model to CDW junctions, peaks in the dI/dV should appear at $\pm\frac{2\Delta}{e}$ for CDW-I-CDW junctions and at $\pm\frac{\Delta}{e}$ for N-I-CDW junctions, where the tunnel barrier is denoted with I.

Tunneling properties of CDW materials have previously been investigated using several techniques. Most experiments concern tunneling from a metal to the CDW material perpendicular to the chains. For instance, Scanning Tunneling Microscopy (STM) studies of Dai *et al.* [1] on NbSe₃ reveal conductance peaks at $\Delta_1 = 101.2$ mV and $\Delta_2 = 35$ mV. Fournel *et al.* [2] have made tunneling devices by placing Pb on the top face of a NbSe₃ crystal creating a Pb-I-NbSe₃ junction. Gap features of the lower transition at $\Delta_2 = 35$ mV were observed when the Pb was superconducting. Sorbier *et al.* [3] have used the same Pb-I-NbSe₃ geometry, but have suppressed the superconducting state of Pb with a small magnetic field. Their measurements did not resolve gap features. The temperature dependence of NbSe₃'s gap features (above $T = 77$ K) was experimentally obtained by He Haifeng and Zhang Dianlin [4] using a Sn-I-NbSe₃ junction. The gap features of both Peierls transitions in NbSe₃ were observed simultaneously in transport measurements by Ekino and Akimitsu [5]. They have used Au-NbSe₃ junctions in the a-b plane to measure electron tunneling at $T = 4.2$ K and found gap features at $V = 90$ mV and $V = 37$ mV. These experiments all concern tunneling from a metal or superconductor perpendicularly to the chains of the CDW material.

Ekino and Akimitsu [5] have observed electron tunneling between two NbSe₃ crystals, instead of between a metal and a NbSe₃ crystal. In their experiment, the a-c plane of one crystal was pushed against the b-c plane of another crystal and therefore tunneling occurs again perpendicular to

Tab. 7.1: Experimental gap values of NbSe₃ of both the upper (Δ_1) and lower (Δ_2) transition. The direction for electron tunneling is indicated in the fourth column. The last row of the Table show the measurements of this Chapter. ARPES is Angular Resolved Photo Emission Spectroscopy. The last two gap values are from samples described in this Chapter. Sample A is made using a Focused-Ion Beam (FIB) and sample C is a Mechanically Controllable Break junction (MCBJ).

Type of measurement	Δ_1 (mV)	Δ_2 (mV)	axis	Ref.
ARPES	110	45	-	[8]
Pb-I-NbSe ₃	-	35	a*	[2]
Sn-I-NbSe ₃	80-85	-	a*	[4]
STM	101.2	35	a*	[1]
Au-NbSe ₃	100	-	b	[7]
Au-NbSe ₃	90	37	c	[5]
NbSe ₃ -NbSe ₃	143	60	a/c	[5]
NbSe ₃ -NbSe ₃	-	26	a*	[6]
NbSe ₃ -NbSe ₃ (FIB)	95	53	b	Sample A
NbSe ₃ -NbSe ₃ (MCBJ)	98	41	b	Sample C

the chains. Latyshev *et al.* [6] have used a Focused-Ion Beam (FIB) to create overlap junctions. Overlap junctions are made by etching a single crystal such that a conduction path is created in the a*-axis. Tunneling occurs between layers in the b-c plane. The differential conductance shows a clear peak at $V = 25 - 27$ mV. This peak vanishes at the lower Peierls transition. Their data goes up to $V \sim 50$ mV and does not reveal a peak due to the upper transition. Additionally, they made NbSe₃ point contacts by pushing two crystals against each other. The differential conductance of these point contacts show merely a shoulder which vanishes at the lower Peierls transition. Their data does not show gap features of the upper transition.

The first experiments where tunneling occurred along the chains, reported by Sinchenko *et al.* [7], made use of a Au-NbSe₃ point contact. They found gap features, above $T = 77$ K, at $\Delta_1 = 100$ mV for several point contacts with different resistances. The gap features vanish near the Peierls transition temperature $T_{P1} = 142$ K.

In Table 7.1, gap values of tunneling experiments on NbSe₃ are summarized including the experiments described in this Chapter. These experiments are the first where the temperature dependence of the gap is measured over the entire temperature range $T = 4.2$ K to above the first Peierls tran-

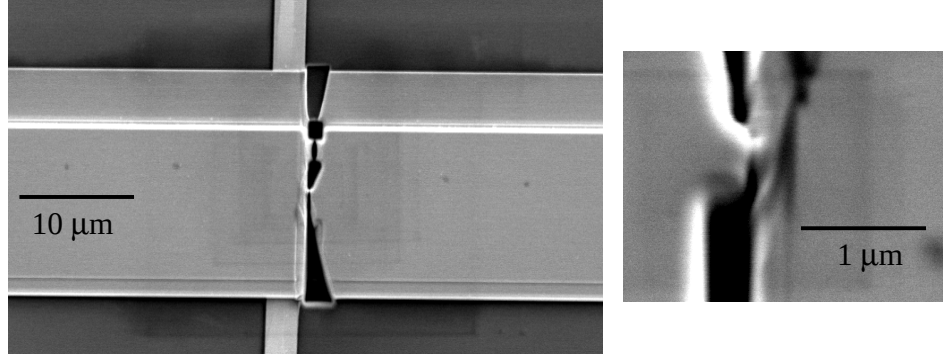


Fig. 7.1: Scanning Electron Microscope images of a NbSe_3 point-contact (sample A), fabricated with a Focused Ion Beam. The constriction is close to a gold voltage probe. The right image is a high magnification image of the constriction. The constriction is about 100 nm long.

sition $T_{P1} = 145$ K. The samples are different from previous experiments in that they are single crystal point contacts and they concern tunneling along the chains in NbSe_3 . All previous experiments mentioned above found features in the dI/dV at $eV = \Delta$, while the samples described in this Chapter show features at $eV = 2\Delta$.

Point contacts in NbSe_3 are made with two techniques: a Focused-Ion Beam (FIB) and a Mechanically controllable Break junction (MCBJ). The measurements of three samples are discussed in this Chapter; sample A and B are made using a FIB and sample C is made in an MCBJ. The point contact made with an MCBJ is in a single NbSe_3 crystal. The point contact was broken and contact was regained controllably several times. This point contact is different from measurements of Ekino and Akimitsu [5] where two different faces of NbSe_3 crystals are pushed together. The point contacts made with a FIB are single crystal NbSe_3 wires and transport is along the chains in the crystallographic b-axis. These point contacts are different from previous studies, where transport occurred perpendicular to the chains [1], from different NbSe_3 crystals [5], or from a metal to the CDW crystals [7].

7.2 Fabrication of point-contacts

7.2.1 NbSe_3 Focused-Ion Beam point-contacts

Samples are made by placing NbSe_3 crystals on top of a silicon oxide substrate with lithographically created Au contacts. Typical thicknesses of the crystals is a few hundred nanometer. Point-contacts were made by etching two trenches in a NbSe_3 crystal using a FEI/Philips FIB-200 focused ion

beam at low magnification. Finer cuts at the point-contact were made at high magnification. Images were taken by exposing the point-contact with a low current ion-beam. The detailed geometry of the point-contact can not be inferred from the FIB images taken in between etching. Instead the resistance of the point-contact, measured in a four-point configuration, is used to monitor the size. Fine cuts were made until the resistance of the point-contact was higher than $150\ \Omega$ at room temperature.

Figure 7.1 shows a Scanning Electron Microscope (SEM) image of sample A taken after the transport measurements were performed. The room-temperature resistance of this point-contact is $165\ \Omega$. In the diffusive limit, the Maxwell resistance [9] provides an estimate for the constriction's size

$$R_M = \frac{\rho}{2D}, \quad (7.1)$$

with ρ the resistivity. Using the room-temperature resistivity of NbSe₃ $\rho = 1.86\ \Omega\mu\text{m}$, the size of the constriction is $D \sim 6\ \text{nm}$. The estimate neglects the resistivity anisotropy of NbSe₃ and the series resistance of the bulk crystal, providing a lower limit of the constriction's size.

Another estimate of the size of the constriction can be made at low temperature using the Sharvin resistance for point contacts [10]

$$R_S = \frac{\hbar k_F}{e^2 D^2 n}, \quad (7.2)$$

with $\hbar$ Plank's constant, k_F the Fermi wave vector, e the electron charge, D the diameter of the constriction and n the electron density. The series resistance of the bulk crystal is two orders of magnitude smaller at $T = 4.2\ \text{K}$ than at room temperature and can therefore be neglected. Using the low-temperature charge carrier concentration $n = 1.1 \times 10^{18}\ \text{cm}^{-3}$ [11], $k_F = 0.22\ \text{\AA}^{-1}$ and the measured value of the constriction's resistance $R = 1.8\ \text{k}\Omega$ at $T = 4.2\ \text{K}$, the size of the constriction $D \sim 70\ \text{nm}$. From the SEM image, the size of the constriction is about $100\ \text{nm}$.

7.2.2 Mechanically controlled break junctions of NbSe₃

We have used a Mechanically Controllable Break-junction (MCBJ) configuration to make constrictions in NbSe₃. These are made by stretching or pulling the crystal until it partly breaks or shears. The crystals are fixed by an epoxy adhesive on an elastic substrate, which is clamped in a 3-point configuration consisting of two fixed counter supports and a moveable rod, as illustrated in Fig. 7.2. When the substrate is bent by the rod, the epoxy drops are pushed further apart and the crystal is stretched. Phosphor bronze sheets are cut into $18\ \text{mm} \times 4\ \text{mm}$ substrates. The thickness t of the substrates was either $0.7\ \text{mm}$ or $1.0\ \text{mm}$. Kapton tape (polyimide) is put on the substrate to form an insulating layer between the substrate and a sample.

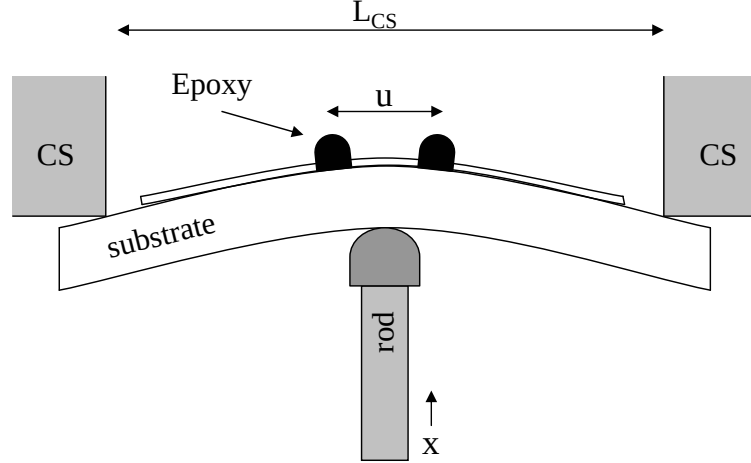


Fig. 7.2: Schematic drawing of a break-junction setup. A Phosphor bronze substrate is clamped in a 3-point bending configuration, consisting of two counter supports (CS) and a rod. Bending the substrate is controlled by moving the rod up or down.

A NbSe_3 crystal is put on top of the Kapton tape. Two stycast drops fix the crystal to the tape. The two drops are put as close together as possible to reduce the effect of external vibrations, typically the distance between the stycast drops is $u = 500 \mu\text{m}$. The substrate is mounted in the 3-point clamp configuration. A moveable rod pushes the substrate against two counter-supports a distance $L_{CS} = 16 \text{ mm}$ apart. When the rod is moved a distance δx , u changes by $\delta u = 6ut/L_{CS}^2 \delta x$ [12]. The displacement ratio $\delta u/\delta x$ is typically less than 1%.

Next, four Cu wires are used to create electrical contact between the sample and the measurement setup. Two different fabrication methods have been used:

- The Cu wires are glued directly to the crystal and the Kapton tape using silver paint.
- Four Au strips with a Cr sticking layer are deposited on the Kapton tape before putting the crystal down. The NbSe_3 crystal is put on the Au strips and fixed with stycast drops. Four Cu wires are connected to each Au strip with silver paint.

The silver paint is baked in an oven at 70-100 °C for 30 minutes. This generally improves the contact resistance to both the NbSe_3 crystal and the Au strips. Additionally, a drop of ethyl-cellulose on top of the NbSe_3 crystal also improves the contact to Au strips. The samples with the Au strips have the lowest contact resistances, as found by comparing the two-probe

resistance (typically 300-500 Ω) with the four-probe resistance (typically 250 Ω) at room temperature.

7.3 Point-contact transport measurements

Figure 7.3 shows the differential conductance of constriction A in the temperature range $T = 4.2 - 145$ K, below the upper transition temperature T_{P1} . The dI/dV at $T = 4.2$ K shows clear peaks at $V = \pm 105$ mV and $V = \pm 190$ mV, see Fig. 7.3c. The clear peaks at $V = \pm 105$ mV vanish at temperatures above about $2T_{P2}/3$, and become inclinations at higher temperatures. Above T_{P2} these features are completely gone. Similarly, the peaks at $V = \pm 190$ mV become inclinations above $T = 80$ K and above T_{P1} also these inclinations vanish.

The zero-bias resistance-temperature curve in Fig. 7.3b does not reflect the one of bulk NbSe₃ (see Chapter 1), which shows metallic behavior down to $T = 4.2$ K. Also, the two Peierls transition are not clearly visible as is the case for bulk NbSe₃. Instead, the resistance increases monotonically when the temperature is decreased. This non-metallic behavior implies that the constriction acts as an insulating part between two sides of the single NbSe₃ crystal. Figure 7.3b also shows the differential resistance at $V = 220$ mV as a function of temperature, indicated by the open stars. The high-bias differential resistance increases approximately linear with decreasing temperature over the entire temperature range 4.2 K-300 K.

One could argue that the appearance of the peaks in the dI/dV might be due to Joule heating, in which case the peaks in the dI/dV reflect the conductance peaks in the $R(T)$ curve of bulk NbSe₃ at the Peierls transitions. However, such a reason can immediately be ruled out, since the $R(T)$ curve of this insulating constriction (Fig. 7.3b) does not exhibit the conductance peaks at the Peierls transitions. Therefore, the appearance of peaks in the dI/dV is not due to Joule heating of the constriction.

The voltages at which features occur in the dI/dV are plotted as a function of temperature in Fig. 7.4 for sample A. At low temperature peaks in the dI/dV are used, but for higher temperatures the inclinations in the dI/dV curve are used. Our data closely matches X-ray data from Fleming *et al.* when scaled to our data. It is clear that the features disappear at the corresponding Peierls transitions, indicating that these features are related to the gaps of NbSe₃.

Sample B is a NbSe₃ constriction with a room-temperature resistance of 360 Ω , which is larger than the room-temperature resistance of the constriction of sample A. Sample B has been exposed to a FIB for a longer time and higher degree of Ga implantation in the constriction region is expected, which may influence the appearance of features in the dI/dV .

Figure 7.5 shows the differential conductance traces of sample B in the

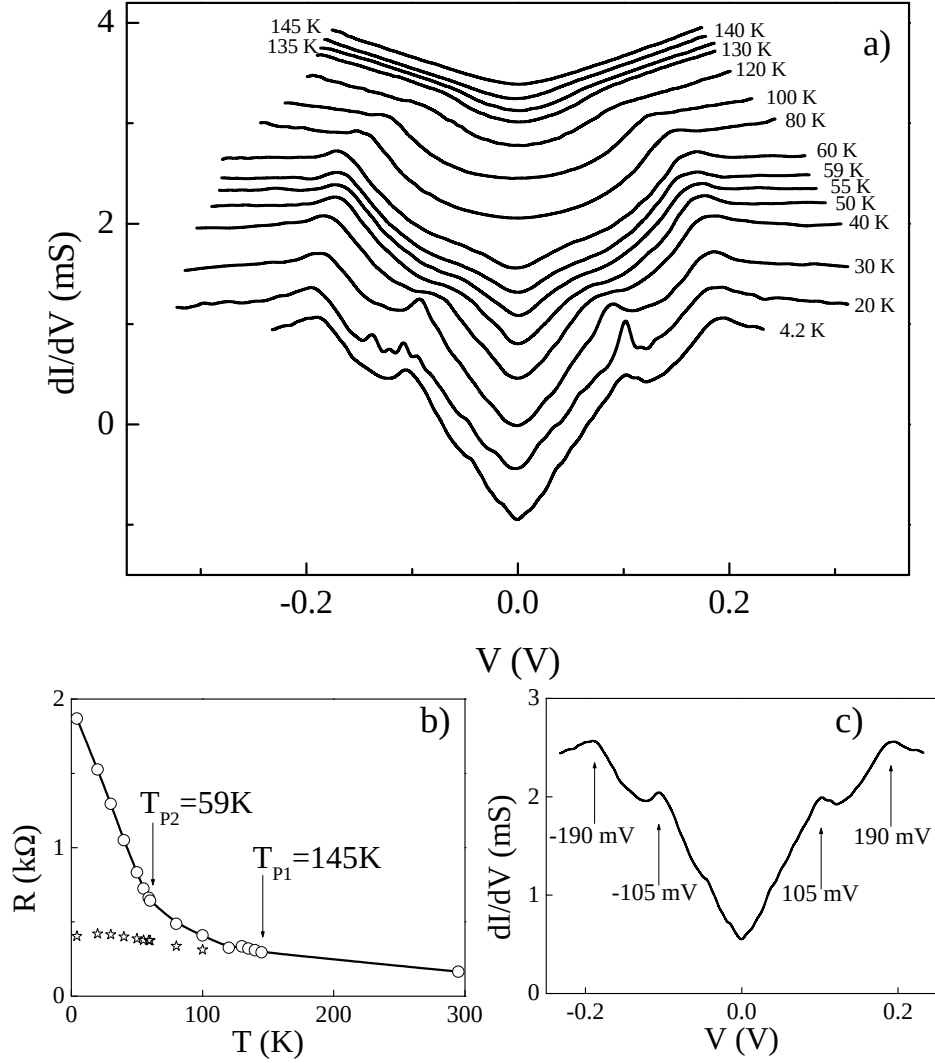


Fig. 7.3: Measurements of sample A. a) Differential conductance dI/dV as a function of voltage in the temperature range 4.2 K - 145 K. Offsets in mS below $T = T_{P2}$: -1.5 (4.2 K), -1.1 (20 K), -0.8 (30 K), -0.5 (40 K), -0.4 (50 K), -0.3 (55 K), -0.2 (59 K). b) Zero-bias resistance as a function of temperature (open circles). The open stars show the differential resistance dV/dI at $V = 220$ mV. c) dI/dV at $T = 4.2$ K shows two clear peaks at both positive and negative bias.

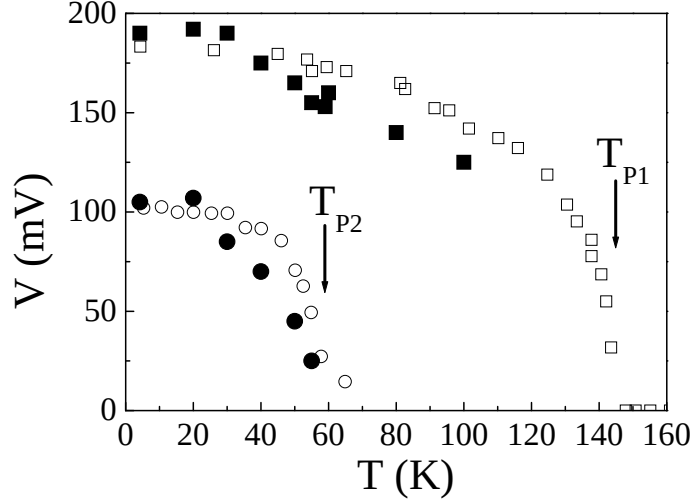


Fig. 7.4: The position of the dI/dV features as a function of temperature for sample A (filled symbols). The open symbols are X-ray data taken from Fleming *et al.* [13].

temperature range $T = 4.2 \text{ K} - T = 80 \text{ K}$. Sample B is a short non-metallic constriction. The differential conductance has a zero-bias dip and all traces show conductance peaks at about $V = \pm 170 \text{ mV}$ in this temperature range. These may correspond to the gap of T_{P1} . There are no conductance peaks in the traces that correspond to the gap of T_{P2} , but there are inclinations at around $\pm 120 \text{ mV}$ at $T = 4.2 \text{ K}$.

The zero-bias resistance versus temperature increases with decreasing temperature over the entire measured temperature range. The resistance increases strongly below $T = 59 \text{ K}$. The $\ln R/dT$ still reveals two dips of the Peierls transitions at $T = 143 \text{ K}$ and $T = 59 \text{ K}$, see inset of Fig. 7.5. Most likely, the dips are due to changes in the series resistance of the large part of the crystal as a function of temperature, and not due to the point contact itself. Since the $R(T)$ curve does not have a conductance peak, the peaks in the dI/dV are not due to Joule heating of the sample.

Sample C is a mechanically controlled break junction (MCBJ) of NbSe_3 . The sample has four silver paint contacts to the crystal. The initial contact resistances at room temperature were too high to perform a good 4P measurement. Therefore, the current and voltage probes were connected on each side of the crystal by silver paint. This created a 2-probe sample with four Cu wires connected to it, which eliminates the resistance of the leads. The two silver paint drops covered about 90% of the crystal length. The 2-probe resistance at room temperature is $4 \text{ k}\Omega$, measured with a multi-meter.

At $T = 4.2 \text{ K}$, the zero-bias resistance of the wire is $1.3 \text{ k}\Omega$. Several $I(V)$ traces were made, that showed switching behavior. The expected wire's

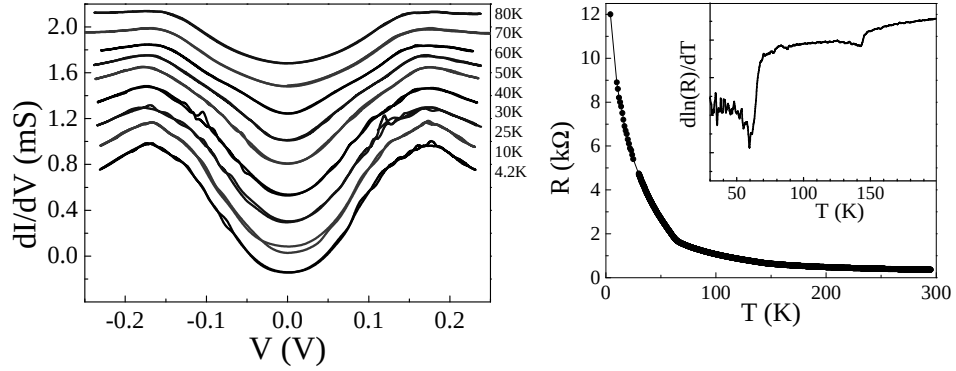


Fig. 7.5: Measurements of sample B. Differential conductance of non-metallic constriction. The traces have offsets, starting from $T = 4.2$ K to $T = 80$ K of: -0.3, -0.1, 0.1, 0.3, 0.5, 0.6, 0.7, 0.8, and 0.9 mS. The resistance versus temperature curve shows non-metallic behavior. The $d\ln R/dT$ still reveals two Peierls transitions at $T = 143$ K and $T = 59$ K.

resistance is on the order of $1\text{--}10\ \Omega$ and the switching indicates mechanical instability of the contacts. Bending the substrate decreased the 2P resistance to a few hundred Ohms and stopped switching events, after which stable $I(V)$ traces were made without any switching events.

Figure 7.6 shows the differential conductance dI/dV of the break-junction as a function of voltage V . The zero-bias resistance of the break-junction is $90\ \Omega$. Note that this value is much smaller than the resistance (about $2\ \text{k}\Omega$) at $T = 4.2$ K of non-metallic constrictions made with a FIB. The dI/dV shows a zero-bias dip, indicative for tunneling type behavior. Clear peaks in the dI/dV occur at ± 79 mV and ± 195 mV. The ratio of these voltages is 0.41, exactly the transition temperatures ratio $T_{P2}/T_{P1} = 59/145 = 0.41$. This is an indication that these peaks are related to the gaps of NbSe_3 . The temperature dependence of the conductance peaks and zero-bias resistance have not been measured.

7.4 Discussion

The appearance of clear peaks in the dI/dV for both MCBJ and FIB samples show that transport properties of the FIB samples are not qualitatively changed by any damage caused by Ga bombardment or implantation of Ga atoms. The position of the peaks in the dI/dV agrees with angular-resolved photoemission spectroscopy measurements, see Table 7.1. The peaks in the dI/dV can be explained when we model the samples as CDW-I-CDW junctions, i.e. two bulk CDW parts separated by a tunnel barrier. In analogy with the semi-conductor model for electron tunneling in superconductors,

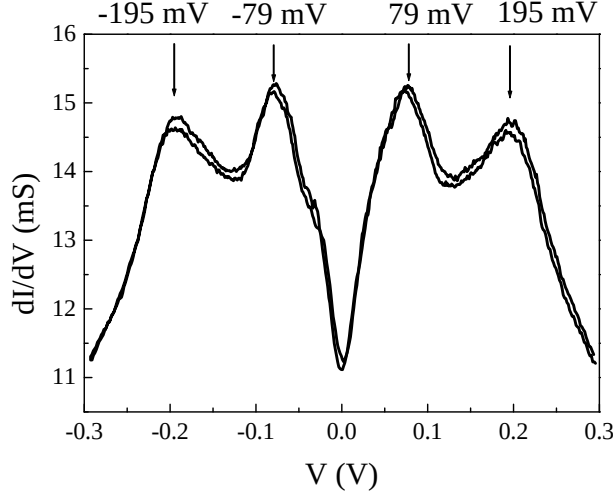


Fig. 7.6: Measurements of sample C. Differential conductance of a NbSe₃ break junction at $T = 4.2$ K. Clear peaks occur at voltages corresponding to the gaps. The peak positions ratio is identical to the transition temperatures ratio.

we expect peaks at $V = \pm \frac{2\Delta}{e}$ for each Peierls instability.

A direct way of determining the energy gap of a superconductor is through tunneling experiments. In these experiments, a thin insulating layer allows particles to tunnel from a normal metal to a superconductor (NIS junction) or from a superconductor to another superconductor (SIS junction). We will briefly discuss the shape of the dI/dV of an SIS junction and compare it to the measured dI/dV of the CDW samples A to C.

When two superconductors, in general with two different gaps Δ_{S1} and Δ_{S2} , are brought into contact with an insulating layer in between, the ground states of both superconductor align, as shown in Fig. 7.7. When a voltage is applied, the ground states of S_1 is shifted by eV . At zero temperature, a tunneling process from S_1 to S_2 is only possible when $eV \geq \Delta_{S1} + \Delta_{S2}$. Then an electron can tunnel from the lower band in S_1 to the quasi-particle band of S_2 . The dI/dV shows peaks at $eV = \pm(\Delta_{S1} + \Delta_{S2})$. At finite temperatures, quasi-particle bands are more occupied and tunneling between the quasi-particle bands is possible when the bands are aligned at $eV = |\Delta_{S1} - \Delta_{S2}|$. The dI/dV shows peaks near $eV = \pm(\Delta_{S1} - \Delta_{S2})$, see Fig 7.7.

The dI/dV of the samples A and C show peaks at 2Δ for each Peierls instability. This is in agreement with what is expected for the dI/dV of an SIS junction when the two superconductors are the same ($\Delta_{S1} = \Delta_{S2}$). However, there are no peaks at $\Delta_1 + \Delta_2$ in any of the measured CDW samples. This indicates that there is no tunneling transport between the two different CDW states. Also the peaks for SIS junctions at $eV = \Delta_{S1} - \Delta_{S2}$

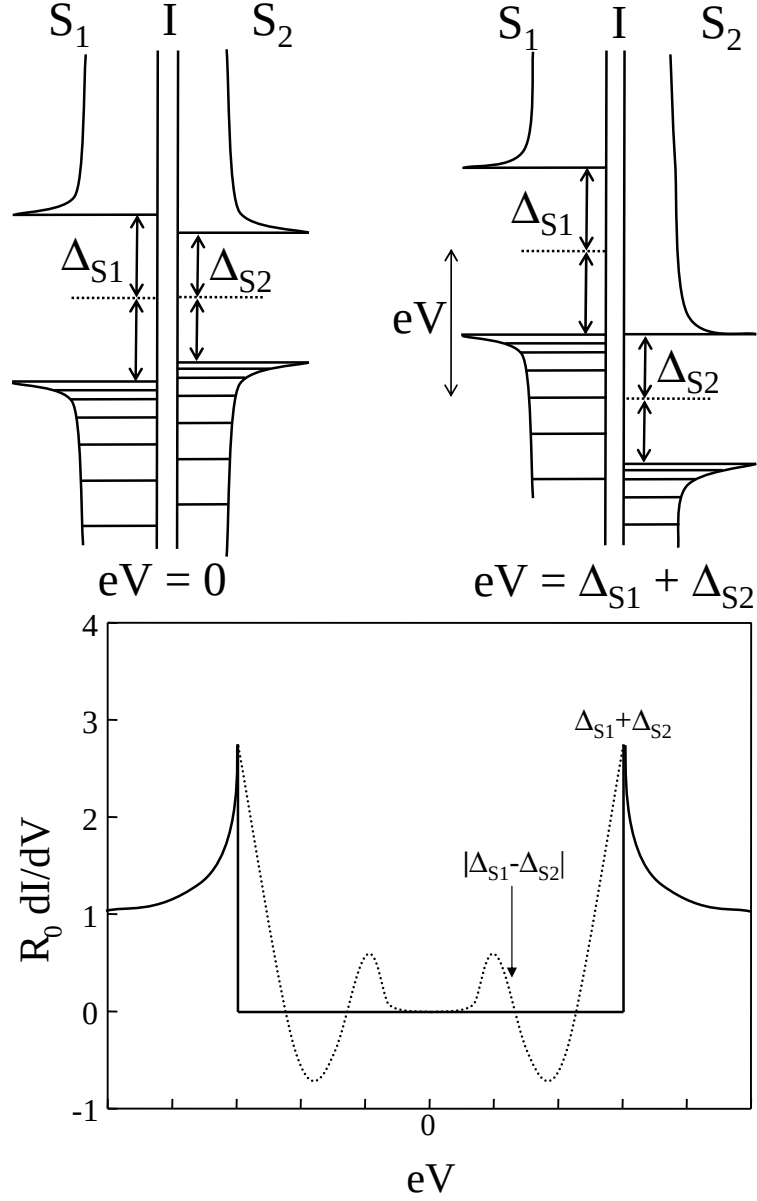


Fig. 7.7: Density-of-states diagrams of a superconductor-superconductor tunnel junction. Left: The ground states of the superconductors align, although Δ can be different. Right: At $T = 0$ K, an electron can tunnel from superconductor S_1 to the other superconductor S_2 , when $eV > \Delta_{S1} + \Delta_{S2}$. The resulting differential conductance trace, drawn schematically with a solid line, shows peaks at $\pm(\Delta_{S1} + \Delta_{S2})$. At finite temperatures, thermally excited states can tunnel and features appear near $\pm|\Delta_{S1} - \Delta_{S2}|$, drawn schematically with a dotted line.

which appear at finite temperatures are not observed in the CDW samples.

The measurements show that the width of the peaks do not vary significantly below $T_P/2$ for either Peierls transition, implying a temperature-independent broadening of the CDW DOS. In contrast to superconductor junctions, the dI/dV shows a substantial tail for voltages below the gap down to zero-bias, indicating that the DOS does not cut off sharply below the gap. This is consistent with optical absorption measurements [14] and calculations of the zero-bias CDW DOS with one-dimensional fluctuations [15, 16]. The suppression of the conductance peaks for temperatures higher than $2T_P/3$ are likely due to thermal fluctuations, which are strong in quasi-one-dimensional systems, in agreement with calculations of the DOS near the Peierls transition [17].

The results may be explained in terms of CDW-I-CDW tunneling in the presence of a transverse corrugation of the T_{P2} CDW energy gap. A transverse corrugation of the T_{P2} gap has been suggested based on low-bias behavior of metal-NbSe₃ tunnel junctions [3] and on comparison of thermal and electric conduction at low temperatures [18]. The dispersion of the quasi-particles states in momenta perpendicular to the chains is incorporated in the semi-conductor model for tunneling. The dispersion arises from the interchain coupling that results in imperfect Fermi surface nesting [19]. This dispersion is characterized by a single parameter $\epsilon = (t_\perp^2 \cos(bk_F))/(2t_b \sin(bk_F))$, with $t_\perp$ the dispersion bandwidth perpendicular to the chains, t_b the bandwidth parallel to the chains and b the interchain separation. The corrugation for the T_{P2} transition ϵ_2 is larger than the corrugation of the T_{P1} transition ϵ_1 . Also, Sorbier *et al.* [3] conclude that ϵ_2 is larger than the T_{P2} gap Δ_2 .

Figure 7.8 illustrates the dispersion, showing by brightness the DOS $N(E, k_\perp)$ as a function of energy and perpendicular wave vector within half a Brillouin zone and the DOS $N(E)$ at fixed wave vector $k_\perp$. Using this dispersion, the differential conductance is calculated by integrating the product $N(E, k_\perp)N(E - eV, k_\perp)$ over all energies E and wave vectors $k_\perp$ assuming a zero-temperature BCS DOS broadened by a Gaussian distribution with the experimentally measured width $\sigma = 24$ meV. The computed dI/dV curves for no corrugation ($\epsilon = 0$) and for $\epsilon_2 = 50$ meV are shown in Fig. 7.8, with ϵ_2 the magnitude of the corrugation of the T_{P2} Peierls transition. As expected for no corrugation, the dI/dV shows a strong peak at $eV = \Delta_1 + \Delta_2$. As ϵ_2 is increased, this peak splits in energy and shrinks in height. While all conductance peaks remain visible with a divergent DOS, with a broadened DOS the peaks at $\Delta_1 + \Delta_2 - \epsilon_2$ and $2\Delta_2$ merge at around 100 meV, resulting in a curve that closely resembles the measured data. The shift to higher voltage of the conductance peak of the T_{P2} gap also explains why our measured value of $\frac{\Delta_2}{\Delta_1} = 0.55$ is higher than the value $\frac{T_{P2}}{T_{P1}} = 0.41$. This same value $\frac{\Delta_2}{\Delta_1} = 0.41$ is confirmed by ARPES measurements [8].

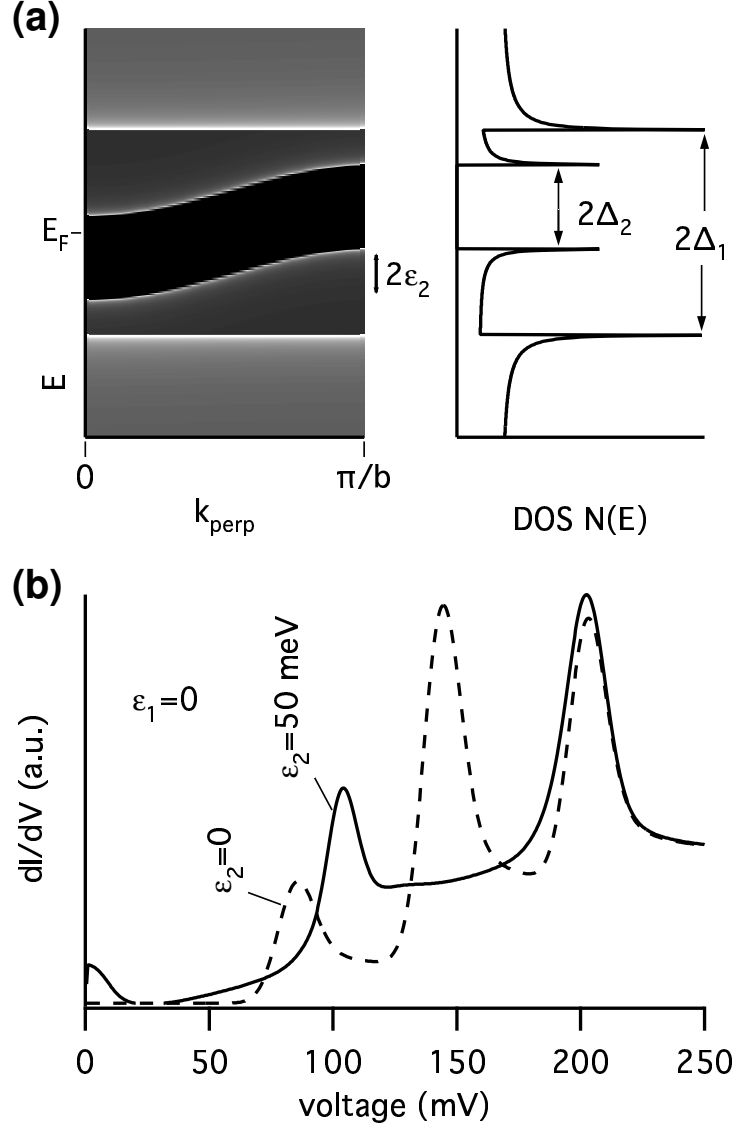


Fig. 7.8: Semiconductor model for transitions between quasi-particle excited states of the CDW, with corrugation ϵ_2 present in the transverse momentum direction of the $T_{P2} = 59$ K but not the $T_{P1} = 145$ K CDW states: (a) left : Density of states $N(E, k_{\perp})$ as a function of perpendicular wave vector and energy, showing how corrugation of the T_{P2} CDW changes the relative positions of the DOS maxima. Brighter regions denote higher DOS; right : Projection of $N(E, k_{\perp})$ at a fixed wave vector $k_{\perp} = \pi/b$; (b) Simulated dI/dV as a function of voltage V using BCS density of states broadened by 24 meV, and a corrugation $\epsilon_2 = 0$ (dashed line) and 50 meV (solid line). All calculations assume $\Delta_1 = 100$ meV and $\Delta_2 = 41$ meV.

In conclusion, an in-chain CDW point contact has been fabricated that behaves like a tunnel junction and demonstrates peaks in the low temperature differential conductance in agreement with values for the energy gaps $2\Delta_1$ and $2\Delta_2$ previously reported in literature. The device is reproducible and does not suffer from mechanical instabilities reported for other types of point contacts [7]. The peaks disappear at around two-thirds of the Peierls transition temperature, in agreement with calculations of fluctuation effects in the DOS of one-dimensional compounds.

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Abstract

This thesis described the work performed on crystals with a phase transition to a Charge-Density Wave (CDW). The electrical transport properties change when crystal sizes are smaller than characteristic length scales for CDWs, typically $1\text{ }\mu\text{m}$. In contrast to metals, semiconductors and superconductors, reduction of sizes is relatively unexplored in the case of CDWs. The development of methods to reduce sizes of CDW crystals are described in this thesis. Numerous finite-size transport effects are found as a result of the development of these new fabrication methods. These finite-size effects have led to the development of models to describe the microscopic aspects of CDWs.

Chapter 1 gives a short introduction to CDWs and an overview of the transport properties known for bulk crystals. A summary of the developed fabrication techniques is given.

The phase transition to a CDW occurs in crystals with an anisotropic crystal structure, as in parallel metallic chains. Electrical transport measurements are usually performed by making lateral contacts to the crystal and measuring the current-voltage characteristics. The current is injected perpendicular to the chains, in which direction the conductance is lower than along the chains, and the electric field near such a current injection contact is non-uniform. Typical length scales for this non-uniformity are on the same order as characteristic length scales of the CDW. The interpretation of microscopic effects are affected by the non-uniformity of the electric field.

One of the fabrication techniques to reduce sizes of CDW crystals makes use of a gallium Focused-Ion Beam (FIB). This technique is used in the work described in Chapters 5 and 7. Chapter 3 gives a characterization of this fabrication technique. Particular attention is given to the unwanted effects of gallium implantation on the CDW transport properties.

Measurements of the dynamics of the CDW on a submicron scale are described in Chapter 4. When the distance of electrical contacts is on a micron scale, the transport properties change dramatically. Regions of absolute negative resistance are observed. This means applying a current in one direction results in a voltage in the opposite direction. This may seem thermodynamically impossible, but it can be explained using a so-called 'two-fluid' model. The CDW is one fluid and quasi-particles are the other. The forces on the CDW are determined by the electrical field, while the forces on the quasi-particles are determined by the electro-chemical potential difference. The origin of Negative resistance is attributed to the different forces on the CDW and quasi-particles. A description of this model is given in Chapter 4.

The dynamic properties of CDWs depend strongly on the interaction of the CDW with impurities. The CDW adjusts itself by elastic deforma-

tions with typical length scales called the phase coherence length. Chapter 5 described measurements on crystals with sizes smaller than the phase coherence length. Good agreement is found using the model of Fukuyama, Lee and Rice.

Chapter 6 describes how transport properties of CDW crystals change when the number of parallel chains is reduced from millions to a few hundred. Below 2000 parallel chains, current-voltage and resistance-temperature characteristics show power-law behavior. Power-law behavior is known for one-dimensional metallic systems, but has not been observed in CDW systems up to now. A few models are proposed to describe the measurements. A new model has to be developed, which should include disorder, low electron density and interaction between metallic chains.

The last Chapter describes measurements on point-contacts in CDW crystals. Point-contacts are made using two fabrication techniques, namely with a FIB and in a mechanically controllable break-junction configuration. Break junctions are known for making single-atom contacts in gold. CDW point-contact of both fabrication techniques show transport properties, in which electrons or quasi-particles tunnel across a barrier in the chain direction. The differential conductance shows peaks at voltages expected from the value of the gap in the density-of-states. The measurements are modelled using the semiconductor model for electron tunneling in superconductors. To describe the observed features, the particular band structure of the used CDW crystals is incorporated in this model.

Samenvatting

Dit proefschrift beschrijft het werk verricht aan kristallen die een faseovergang naar een ladingsdichtheidsgolf, afgekort als CDW van het Engelse Charge-Density Wave, hebben. Wanneer afmetingen van deze kristallen kleiner zijn dan karakteristieke lengteschalen van de CDW, typisch $1\mu\text{m}$, veranderen de elektrische transporteigenschaften van de CDW. In tegenstelling tot metalen, halfgeleiders en supergeleiders is het verkleinen van afmeting voor CDWs nog een vrijwel onontgonnen gebied. Methoden die ontwikkeld zijn om afmetingen van deze CDW-kristallen kleiner te maken zijn beschreven en de uiteenlopende transporteffecten die als gevolg daarvan gevonden zijn. Deze nieuwe effecten hebben geleid tot de ontwikkeling van modellen om de microscopische aspecten van CDWs te kunnen beschrijven.

Hoofdstuk 1 geeft een beknopte introductie van CDWs en een overzicht van welke bijzondere transporteigenschaften al bekend zijn van grote CDW-kristallen; dit om te begrijpen hoe de transporteigenschaften veranderen wanneer afmetingen van kristallen klein zijn. Verscheidende methoden om de afmetingen klein te maken zijn samengevat.

De faseovergang naar een CDW komt voor in kristallen met een anisotrope kristalstructuur, zoals ketenachtige kristalstructuren. Elektrische transportmetingen door zulke kristallen wordt gewoonlijk gedaan door laterale elektrische contacten aan te brengen en stroom-spanningskarakteristieken te bepalen. De stroom wordt loodrecht op de ketens aangelegd; een richting waarin de elektrische geleiding lager is dan langs de ketens. Dit heeft tot gevolg dat er een niet-uniformiteit van het elektrisch veld in de buurt van het contact optreedt. De lengteschaal waarop dit gebeurt is van dezelfde orde van grootte als de karakteristieke lengteschalen van de CDW en heeft dus gevolgen voor de interpretatie van microscopische effecten. Hoofdstuk 2 beschrijft metingen aan en modellering van deze niet-uniformiteit van het elektrisch veld.

En van de fabricagetechnieken om kleine afmetingen van CDW-kristallen te maken is het gebruik van een gefocusseerde bundel van galliumionen, afgekort als FIB van het Engelse Focused Ion-Beam. Deze techniek is gebruikt voor het werk beschreven in hoofdstuk 5 en 7. Hoofdstuk 3 geeft een karakterisatie van deze fabricagetechniek, waar in het bijzonder gelet wordt op mogelijk ongewenste effecten van galliumimplantatie op de eigenschappen van de CDW.

Metingen aan de dynamische eigenschappen van de CDW op (sub)-micrometer schaal zijn beschreven in hoofdstuk 4. De transporteigenschaften veranderen opmerkelijk wanneer de afstand tussen elektrische contacten langs de ketenrichting zo klein zijn. Delen van het kristal laten negatieve weerstand zien, dat wil zeggen een aangelegde stroom in één richting geeft een spanningsverschil in de tegengestelde richting. Dit lijkt thermodynamisch onmogelijk, maar is te begrijpen met behulp van een zogenaamd twee-

vloeistoffenmodel. De ene 'vloeistof' is de CDW die beweegt ten gevolge van een elektrisch veld; de andere vloeistof bestaat uit quasi-deeltjes die bewegen ten gevolge van een elektrochemisch potentiaalverschil. Negatieve weerstand ontstaat door verschillen in de krachten die de twee vloeistoffen ondervinden. Dit model is beschreven in hoofdstuk 4.

De dynamische eigenschappen van de CDW is sterk afhankelijk van de interactie van de CDW met kristalroosteroneffenheden. De CDW past zich aan door te vervormen over een typische lengteschaal genaamd de fasecoherentielengte. Hoofdstuk 5 beschrijft metingen aan kristallen waarvan de afmetingen kleiner zijn dan de fasecoherentielengte. De metingen worden beschreven aan de hand van het Fukuyama-Lee-Rice model, waarmee een goede overeenkomst is gevonden.

Hoofdstuk 6 beschrijft hoe de transporteigenschappen van CDW-kristallen veranderen wanneer het aantal parallelle ketens verkleind is van een paar miljoen tot een paar honderd. Beneden de 2000 parallelle ketens laten stroom-spannings- en weerstand-temperatuurskarakteristieken een machtsverband zien. Deze machtsverbanden zijn bekend van één-dimensionale metallische systemen, maar waren niet eerder gezien in CDW-kristallen. Een aantal modellen om de metingen te beschrijven zijn voorgesteld. Het uiteindelijke model zal roosteroneffenheden, de lage elektronendichtheid en interactie tussen metallische ketens in rekening moeten brengen.

Het laatste hoofdstuk beschrijft metingen aan puntcontacten in CDW-kristallen. Twee verschillende fabricagetechnieken zijn gebruikt, namelijk met behulp van een FIB en in een breekjunctieconfiguratie. Breekjuncties zijn bekend om contacten van enkele atomen te maken. Deze twee technieken lieten beide transporteigenschappen zien waar elektronen of quasi-deeltjes 'tunnelen' over een barrière in de ketenrichting. De differentiële geleiding laat pieken zien bij spanningen die overeenkomen met de waarde verwacht voor het gebruikte CDW-kristal. Deze transporteigenschappen zijn gemodelleerd met het halfgeleidermodel voor elektronen in supergeleiders, met als toevoeging de bijzondere bandenstructuur van het gebruikte CDW-kristal.

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Curriculum Vitae

Erwin Slot

Born November 18, 1975 in Delft, The Netherlands.

In 1994 I became a student at the Department of Applied Physics at the Technical University of Delft. In 2000 I graduated for my Master Degree in the Quantum Transport group of Prof. Dr. ir. J. E. Mooij under the supervision of Dr. ir. H. S. J. van der Zant. At the end of 2000 I was accepted as PhD student by Van der Zant and Prof. Dr. P. H. Kes to work on a project to study the microscopic aspects of Charge-Density Waves. The project was split in two parts: 1) Studying the transport properties of CDWs, which was performed mainly at Delft University of Technology and 2) Imaging the motion of CDWs with a Scanning Tunneling Microscope at Leiden University. I did much of the research work in Delft including educational tasks as supervising undergraduate and Bachelor students and giving courses in classical mechanics and special relativity. In 2003 and 2004, I spent more time in Leiden at the Huygens and Kamerlingh Onnes laboratories, where the experiments on Charge-Density Wave break junctions were performed. Since November 2004 I work as an experimental physicist at Mapper Lithography, a company that is building a prototype for multi-beam electron lithography.

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