A study of electron scattering through noise spectroscopy

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SUMMARY

Unlike bulk metals, for which the electrical resistance is due to scattering of the traversing electrons on lattice defects, lattice vibrations, grain boundaries, and impurities, electrons can travel across a quantum conductor without any scattering. For this reason transport in quantum conductors is said to be ballistic. The resistance in a quantum conductor is due to scattering of electrons at the contacts between the quantum conductor and the leads connecting it to the world outside. Atomic contacts are among the most widely studied systems of this kind, holding a special place in the field of nanoscience and nanotechnology. Unlike two-dimensional electron gas systems, the traveling electrons have a high density of states at the Fermi energy. The electrons are transmitted through discrete quantum channels and are susceptible to the local potential of the atoms in the contact. Hence, the above systems serve as a test bed to study the local interaction of electrons probing the properties of the atoms or molecules.

One of the techniques that has been widely used for probing the different physical properties of atoms or molecules is the mechanically controllable break junction technique, widely known as MCBJ. A bulk wire is thinned down to an atomic size contact by stretching. By stretching of the contact the resistance increases in discrete steps. The origin of these discrete steps is completely different from what we know from experiments in 2-dimensional electron gas systems. In our case the discrete steps are more associated with abrupt changes in the local arrangement of atoms in the contact. We are able to control the process of thinning down of the contact in the sub-Ångstrom regime. This gives us freedom to vary the coupling of the central atoms in the contact with the bulk leads. The electronic traveling states at the Fermi energy are quite sensitive to the co-ordination of the atoms; thus we have a tool to manipulate the effective electron transmission through the atomic contact just by stretching. This process creates atomically sharpened leads that are used not only for the study of the local properties of the atoms but also for the properties of foreign molecules inserted in the junction. The interaction of the electrons with these molecules shows up as distinct features in current-voltage spectroscopy. Electron-electron or electron-vibron interactions show characteristic signatures of the atomic or molecular species in the contact. While these interaction signals have now been studied in current-voltage spectroscopy by many groups, here we have studied the effect of these interac-

tions on the current noise. The current fluctuates around its the mean value due to scattering of the electron at the contact, when the transmission probability is smaller than 1. This fluctuation is white in nature and known as shot noise. The shot noise in a conductor is proportional to the electronic charge and to mean current value. The full shot noise in a conductor in absence of any interactions is given as $S_I(A^2/Hz) = 2eI\delta(f)$, which is attained in the limit of low transmission probability, $\tau \ll 1$, for all channels. For perfect transmission ($\tau = 1$) the shot noise is fully suppressed, but in general the noise per channel is suppressed by a factor $F = (1-\tau)$. The measure of this deviation is known as the Fano factor F. Due to electron-electron and electron-vibron scattering the shot noise value deviates from the non-interacting value, which can be seen in the case of electron-vibron scattering as a kink in the noise as a function of bias voltage. In our research we have looked into these deviations in order to study the interactions taking place in atomic or molecular conductors. This thesis can be divided into two parts: (a) Inelastic signatures in noise due to electron-vibron interactions. (b) Study of itinerant magnetism in the atomic contact: electron-electron interactions.

In the first part of this thesis we have looked into inelastic scattering effects in the noise. Previously, a cross-over in the sign of the signal in the differential conductance due to interaction of the conduction electrons with vibrons had already been seen. This interaction leads to a downward step in the differential conductance at the bias corresponding to the vibron energy for a quantum conductor having perfectly transmitting eigen channels. On the other hand, a upward step in differential conductance is found for quantum conductors with low-transmission eigen channels. The cross over was found around $\tau = 0.5\mathrm{G}_0$ for a single channel Landauer conductor. This can be understood in terms of a vibron mediated single-particle scattering effect. Since shot noise is the second cumulant of the current (the conductance is the first cumulant) it is more sensitive to subtle interactions of the conduction electrons. Our noise measurements on Au atomic chains show linear deviations at the vibronic energy of the Au atomic system. The effective change in noise is proportional to the effective change in the conductance due to the electron-vibron interaction. The cross over in the sign of the inelastic correction in noise is observed at a conductance value of $0.95\mathrm{G}_0$. This cross-over is higher than the predicted value of $0.85\mathrm{G}_0$. The positive inelastic correction in noise observed for high transmission is quite intuitive because of decrease in transmission probability due to inelastic scattering. However, the negative correction cannot be understood in a simple single-electron picture, but can be explained by processes involving two-electrons and a vibron: Two electrons, with energies E and $E-\hbar\omega_0$, are emitted from the left lead, with the higher energy electron (at energy E) emitting a vibron. The two electrons will then compete for the same outgoing states. This Pauli exclusion interaction makes the energy distribution of occupied outgoing states more uniform, and hence this lowers the noise. The observed positive inelastic correction in noise close to $\tau = 1$ is very close to the theoretical value while the negative correction deviates from the theoretical value, although this is very much model dependent. While adding conductance fluctuations in electron transport to the picture phenomenologically explains a shift in the cross-over point, analytically it is not entirely satisfactory.

Similar measurements on Au-O-Au atomic chains show similar linear deviations in noise due to inelastic scattering of electrons. For Au atomic chains as well as for Au-O-Au atomic chains we have seen a second kink in the noise at higher energies. This could be a higher order vibronic processes taking place in the atomic chain.

Most of the Au contacts show a linear deviation of the measured shot noise from Leosvik-Levitov noise above the vibronic energy. A linear dependence is expected for a strong coupling of vibrons with the phonons in the leads. However, in some cases when this decoupling could be reduced the noise measurements show a non-linear deviation above the vibronic energy of the contact. This non-linearity is due to the feedback of vibronic fluctuations on the charge statistics of the traversing electrons. The noise deviates quadratically above the vibronic energy in the experiment. However, its is too early to conclude on the relaxation rate of vibrons and make any definitive conclusion *w.r.t.* the existing theories.

A molecular system like Pt-D_2-Pt and Au-O-Au would be more appropriate to study the non-equilibrium vibron states feedback on traversing electron statistics. The disadvantage is that these systems are prone to $1/f$ noise. Typically, for Pt-D_2-Pt, the $1/f$ noise corner frequency at 34meV is 1MHz. For this reason we have developed a high-frequency low-noise cross-spectrum measurement setup, which can measure the noise in a molecular system in the MHz regime. This involves a custom-built low noise amplifier and a two-channel cross correlator. This setup is suitable to measure the inelastic noise signatures on single-mlecule systems.

For some of the Au atomic contacts we have seen an anomalously large suppression in noise at the vibronic energy. For the processes described above the suppression in noise is due to two-electron processes and is not larger than 20 – 30%. The anomalous contacts show a much higher suppression, even such that the total noise decreases at higher bias. We have investigated whether the effect can be explained by weak charge shuttling of the electrons, including the image charge in the model. We can only arrive at a sufficiently large effect for a very high amplitude periodic motion of the atom in the contact. Such anomalously large amplitudes have been suggested to arise due to non conservative current-induced forces acting on the atoms in the contact. This force may also explain earlier observations of anomalous current-induced breaking of atomic chains, hence it would be interesting to test the breaking voltages of the chains showing anomalous noise properties. Such a study is underway.

In the second part of the thesis we have looked into the role of magnetism in Pt atomic chains and ferromagnetic atomic contacts. It has been predicted that Pt atomic chains should develop a localized magnetic moment in the contact. An increased localized moment is expected due to the enhanced density of states which takes the suscepetibility beyond the Stoner limit, upon a stretching induced reduction of the atomic co-ordination. Being very close to the Stoner criterion and having high spin-orbit coupling, Pt is expected to show a transition from bulk paramagnetism to spontaneous magnetization in atomic chains. The presence of strong zero bias anomalies in differential conductance measurements for Pt atomic chains has been attributed to weakly localized magnetic moments. We have characterized the Pt atomic chains by differential conductance and noise measurements. The noise measurements shows the presence of at least 4–6 spin channels in the atomic contacts. The presence of more than one (two spin) channels is expected for transition metals, due to the participation of d orbitals in the conductance. We have observed a strong reduction in noise upon stretching of the contact and eventually a Fano factor falling on the minimum noise curve for two spin-degenerate channels (four spin channels) having one doubly occupied channel perfectly transmitting. The cloud of data points observed is clearly bound by this curve. This suggests absence of strongly spin polarized eigen channels in conductance. We propose that the conductance is dominated by weakly polarized channels, with magnetism possibly confined to channels that are poorly transmitted.

In contrast to Pt, Fe and Ni are ferromagnets in the bulk, and atomic contacts are considered to have localized moments due to the presence of partially filled d sub-orbitals. The interaction of the local magnetic moment with the continuum states gives rise to many body phenomena with the localized non-zero spin states being coupled to the sea of conduction electrons. For a spin-$\frac{1}{2}$ moment in a non-magnetic host metal this phenomenon is widely known as the Kondo effect. The signature of the Kondo effect in the differential conductance is seen as a zero bias anomaly. In experiments, the expected logarithmic dependence of the zero bias conductance*w.r.t.* bath temperature as shown by Calvo *et al.*, has led to a Kondo interpretation of the zero bias anomalies, despite the strong exchange splitting of the 'host'. We have followed the stretching dependence of the differential conductance and noise for such contacts. Applying the same analysis we find a Kondo temperature for Fe and Ni atomic contacts of 100K and 230K, respectively, in agreement with Calvo *et al.* The Kondo temperature of the contacts decreases with stretching of the contacts. Upon pushing the contacts towards the bulk limit a splitting of the Kondo resonance is observed. We tentatively attribute this to an interaction between two localized magnetic moments on two different atoms in the ferromagnetic atomic contact.

The noise measurements for the ferromagnetic contacts show sub-Poissonian noise with the presence of predominantly more than two channels (four spin channels) taking part in the conduction. We did not find any clear evidence for strongly spin polarized conductance channels below the limiting curve for spin degenerate systems, except for two contacts with ($G < 1G_0$), for which we have only a low accuracy. The Fano factor measured on the ferromagnetic atomic contact scales roughly linearly with the Kondo temperature and its resonance amplitude, with some scatter. The observed connection between the Fano factor and the weight of the zero bias anomaly supports the view that the zero bias anomaly originates from spin scattering by localized magnetic moments. However, whether this is true Kondo scattering as suggested by Calvo *et al.* can not be stated conclusively from our data.

The study of shot noise on atomic contacts gives insight into electron transport properties that are missing in simple conductance measurements. Future high-bias shot noise experiments on molecular systems could reveal details of the non-equilibrium vibron occupation and its interaction with the conduction electrons. The high frequency noise measurement set up is a sure way to go head to reveal more physical phenomena in nanoscale electron transport.