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Low energy electron transmission through layered materials and chiral organic films

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SUMMARY

Molecules, crystal structures, and viruses are too small to see by the naked eye. Nonetheless, they undeniably impact our daily lives. Microscopy was developed to explore this microcosmos on smaller and smaller scales. Typically, we mean optical microscopy when we think about microscopy. However, electron microscopy allows us to look at different properties and smaller length scales than light microscopy. Throughout this thesis, we draw parallels between electron and light waves, because the electron behaves as a quantum mechanical wave.

Instead of shining light at a sample and collecting what is reflected or transmitted, in electron microscopy we send electrons to the sample and detect how they are reflected or transmitted. We conduct this experiment with a low energy electron microscope (LEEM). Instead of detecting color, i.e., the wavelength of light, we can detect (and adjust) the electron energy, related to its wavelength. In both light- and electron microscopy we call the wavelength-dependent intensity a spectrum.

The spectra of thin materials presented in this thesis can largely be understood from interference, again a concept we know from optics. Optical interference answers questions like: Why do soap bubbles appear in all colors of the rainbow, while they are made from colorless soap solution? The answer is that the thickness of a bubble's wall is a few 100 nanometers, comparable to the wavelength of the light. When the light bounces back and forth between the front and back surfaces of the soap bubble wall, the transmission of some wavelengths that fit (twice) the optical path length is enhanced, and other wavelengths are reflected. The same is true for our thin samples and the electron wavelength, although on a scale of less than 1 nanometer. The electron energy needed for this is between 0 and 25 eV, which is considered very low for electron microscopy. Thus, we use a LEEM.

For example, graphene is a crystalline layer of carbon only one atom thick. When we have two layers of graphene, the electron wave bouncing back and forth between these layers interferes just like the light wave does in a soap bubble wall. By changing the electron energy, thus the electron wavelength, we can measure the spectra and find a characteristic reflection minimum and transmission maximum at 2.5 eV above the vacuum level. Upon adding more layers of graphene, these spectral features split, making for a spectral fingerprint of the layer count. The intensity and broadness of these spectral features are related to the probability of inelastically scattering the electron, characterized by the inelastic mean free path determined in Chapter 3. The inelastic mean free path tells us, how far an electron will travel on average before it hits

something and loses energy. The more often the electron bounces back and forth between the layers without being disturbed, the sharper the features. In optical terms, the finesse of the cavity (that is light bouncing between two mirrors in optics), increases with the lifetime of the resonance state.

We apply this interference model to different two-dimensional materials, reaching from the conducting graphene, via the insulating hexagonal boron nitride (hBN), to the semiconducting molybdenum disulfide (MoS_2). MoS_2 is different from graphene and hBN, as the molybdenum atoms are located in a 2D plane sandwiched between a plane of sulfur atoms above and below. Thus, the samples of bi- and tri-layer MoS_2 we measure in Chapter 5 are relatively thick compared to tri-layer graphene. Comparison to calculated reflection and transmission spectra shows that we can measure and model the electron interactions even in this rather complicated compound.

Another central concept in physics is symmetry: We ask ourselves, whether an electron can know if it hits the front or the back side of the sample. For a sample consisting of only graphene, the front side and the rear side of the sample are indistinguishable. However, if we have a sample with graphene on one side and hexagonal boron nitride on the other, there may be a difference in electron reflection. In Chapter 4 we consider such a sample with broken symmetry and show that inelastic scattering is vital to change reflection depending on the sample orientation.

In the final chapter, we are concerned with a different type of symmetry breaking. A chiral object is one that cannot be rotated and/or moved to look like its mirrored version. The typical example of chirality (from Greek *cheir*=hand) are our hands, which are mirror versions of each other but clearly distinct. On the length scale of molecules, handed molecules have the same chemical composition but a different arrangement in space. Remarkably, the arrangement plays a role in biological processes.

In Chapter 6 we use ultraviolet (UV) light to liberate ('photoemit') electrons out of a surface covered with chiral molecules. We investigate the interplay between the handedness of light polarization with the handedness of chiral organic molecules. The chirality of the molecules has been reported to couple to the spin of transmitted electrons and spin-filter them: a phenomenon known as the chirality-induced spin selectivity (CISS) effect. We demonstrate control of the light polarization on 400 nm long gold rods. As the size of the gold rods is comparable to the UV wavelength, the electric field (of the light wave) can excite a resonant wave in the gold electrons when it is aligned with the long axis. This enhancement of the electric field leads to a larger electron emission.

Furthermore, we show photoemission from chiral BINAP molecules. However, we could not see a significant difference between excitations with light of opposite chirality. This may be due to insufficient order of the molecular film. We characterize our setup and estimate that it is sensitive enough to detect a 1% emission difference between excitation with right-handed and left-handed circularly polarized light. Imaging the CISS effect with nanometer resolution, as opposed to measuring averages over a whole sample, will allow for a better understanding and for building better devices utilizing the effect.