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Insights from scanning tunneling microscopy experiments into correlated electron systems

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Chapter 1

Introduction

1.1 Motivation

When people ask me what I do for a living, I often tell them I work with extremely sensitive electron microscopes [1] studying superconductors [2]. This is not entirely true, as only one of the chapters in this thesis focuses on a material that is actually superconducting at low temperatures, but somehow superconductivity has always been a fascinating phenomenon to me, and I enjoy explaining the concept to other people who do not know about it. As an undergraduate student, I first heard about copper-oxide materials that can superconduct up to relatively high temperatures ($\sim$ 100 K), which fascinated me. While learning about these, eventually I also learned more about the broader group of strongly correlated materials. In conventional metals, electron-electron interactions are substantially screened, and as a result their properties can be well understood as a liquid of non-interacting charged particles. In correlated materials, however, electronic interactions are no longer screened and consequentially a whole range of exotic electronic states of matter can be encountered in them.

One of these exotic states of matter which is not encountered in normal metals is the so called Mott insulating state. Mott insulators are one of the many surprises nature throws at us, and are arguably the poster child for correlated systems. Many transition metal oxides show Mott insulating behavior. The reasons for this will be explained later on, but for now one could say their crystal structure is sort of "optimized" for the existence of Mott physics.

From normal band structure calculations, which typically ignore interactions between electrons, Mott insulators are actually expected to conduct like normal metals [3]. However, when these materials are cooled down, their electrical conductivity is reduced and they eventually become insulating. Nevill Mott first predicted that electron-electron correlations are the driving factor behind the metal-to-insulator transition [4–6], and hence this state of electronic matter was named after him.

One of the simplest models that explains Mott insulators is the Hubbard model [7]. I will try to explain this with a cartoon picture. Let us consider a material with one orbital per atom, and one electron per atom (*i.e.* half filling, figure 1.1). In

solids, atoms are arranged into a crystallographic lattice. The atomic orbitals overlap (partially), which allows electrons to hop from one atomic site to the next. This hopping is quantified by the hopping parameter, t, which is proportional to the overlap of neighboring wavefunctions: the more the wavefunctions overlap, the easier it is for an electron to hop between sites. An analogy can be made here with traffic on the highway [8, 9]. The hopping parameter quantifies the size of the highway: the larger the hopping term, the bigger the highway which means that cars (electrons) have an easier time traveling around. When hopping is reduced, however, the road becomes too narrow for all the cars to go through smoothly, and a traffic jam forms: transport (electrical conductivity) is reduced. This electronic traffic jam is exactly what occurs in a Mott insulator. This is not all there is to it though. An additional parameter in the Hubbard model is the on-site coulomb repulsion, usually indicated with U. In our analogy, this is sort of equivalent to the size of each car on the highway. After all, the size of our highway is only a meaningful quantity if we consider the size (and amount) of the cars that need to drive on it. The larger the cars are, the larger the roads also need to be to sustain smooth travel of the cars on it. It then becomes clear that it is meaningful to discuss the quantity U/t, which casually said quantifies the "Mottness" of a material. When $U/t \ll 1$, electrons are still free to hop, and the material stays conducting. In the other case, when $U/t \gg 1$, there is not enough bandwidth and since electrons strongly repel each other, hopping between neighboring sites is prevented (figure 1.1). The lowest energy for such a system is reached when each site has exactly one electron, preventing double occupancy (reducing U). At this point, I think the earlier remark about transition metal oxides being "optimized" for Mott physics can be further elucidated: these materials have a relatively small hopping parameter and large Coulomb interaction energy, enabling the Mott behavior observed in them. Interestingly, the picture sketched up to this point also suggests some degree of experimental control. If we think for example about the hopping parameter, reducing or enhancing the distance between lattice sites should enhance or decrease hopping, thereby tuning the "Mottness" of the material. Indeed, it has been experimentally demonstrated that metal-to-insulator transitions can occur when applying strain to a crystal [10, 11].

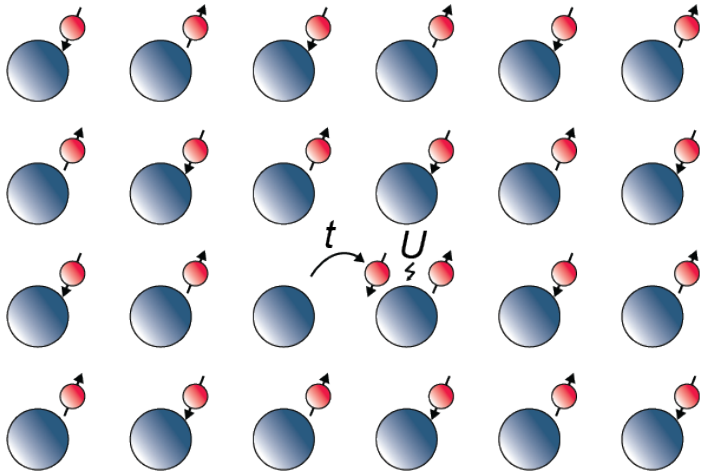

Figure 1.1: Cartoon picture of a Mott insulator. The ground state of the system (at half filling) is realized when all electrons (depicted in red) are evenly distributed over the lattice (depicted in blue) and their spins are antiferromagnetically aligned (indicated by the arrows). When electrons doubly occupy a site, the a Coulomb energy penalty needs to be paid, quantified by U.

Up to this far, I have not discussed the spin degree of freedom of the electrons. One can imagine electrons temporarily being able to visit neighboring lattice sites if their spins are anti-aligned: When all spins point in the same direction, the Pauli exclusions principle would forbid the electrons from occupying the same lattice site. Therefore, the lowest possible energy configuration for this system is achieved when all spins align antiferromagnetically, allowing for virtual hopping to occur [6]. This antiferromagnetic picture can, and has been confirmed many times, for example by magnetic measurements [12, 13] and neutron scattering experiments [14, 15].

1.2 Thesis outline

In this thesis, an assortment of correlated electron systems is discussed. To some degree, all of them seem to have a connection to Mott physics. Earlier, it was already mentioned that Mott insulators are frequently encountered among the 3d transition metal oxides. Perhaps most famously among them are the copper oxides, or cuprates, due to their prominent reputation as high-temperature superconductors. Cuprates consist of metallic copper-oxide layers, sandwiched between inert buffer layers that act as charge reservoirs. They can be doped by tuning the amount of excess oxygen introduced in the crystals during growth. Both electron- and hole doping reveals complex electronic phases in the cuprates, among which is the unconventional superconducting state that can last up to 133 K [16]. The cuprates are discussed in more detail in reference [9].
In conventional superconductors, electron-electron pairing is driven by electron-phonon

interaction. This is well described with Bardeen-Cooper-Schrieffer theory [17, 18]. However, the pairing mechanism behind the superconductivity observed in the cuprates remained a mystery for a long time. Only last year, substantial evidence was found for charge-transfer superexchange interaction driving pairing in the cuprates [19]. At sufficiently high doping, however, it was believed for a long time that superconductivity here is actually of the normal BCS kind [9]. **In chapter 2, we present our STM results measured on an overdoped cuprate compound, and actually find that this is not the case at all.**

But cuprates are not the only compounds where Mott physics is encountered. I have sketched a picture of how competition between orbital overlap (bandwidth) and Coulomb repulsion could lead to such a state of matter, and mentioned Mott states often occur in the 3d transition metal oxides for this reason. However, Mott states are not limited to this class of materials. Perhaps most surprisingly, even in graphene, Mott physics is believed to be responsible for an insulating state that occurs at low temperature, below 4 K [20, 21]. I need to explain this a little bit further though, as this state does not occur in a single, pristine sheet of graphene. Therefore, it came as a big surprise when in 2018, it was experimentally shown that correlated electronic states can occur in twisted bilayer graphene [21]. When two sheets of graphene are stacked on top of one another, their electronic structure starts to hybridize. By changing the stacking angle between the two lattices, the degree of hybridization between the two sheets is effectively varied, thereby influencing the overall band structure of the bilayer. It was first predicted theoretically that at certain so called "magic" angles, an extremely flat band is formed close to the Fermi level [22]. By now, this picture has indeed also been confirmed experimentally, for example by STM [23] and ARPES [24]. When the chemical potential is tuned into this flat band, at sufficiently low temperatures, correlated insulator behavior and superconductivity can be observed [20, 21], demonstrating beautifully how exotic states of matter emerge when bandwidth is reduced and electron-electron correlations start to take over. Twisted bilayer graphene and experimental progress on the topic is discussed in more detail in reference [25].
In the case of this magic-angle twisted bilayer graphene, the bandwidth is heavily dependent on the exact stacking angle between the two sheets. When the angle deviates from the magic angle, already by few tenths of a degree, the correlated states disappear as Coulomb interactions become more and more screened. Therefore, uniformity of the graphene sheets and exact control of the twist angle are important when twisted bilayer graphene device is fabricated. Because of the nature of the conventional fabrication process, however, strain is often incorporated in bilayer devices. **In chapter 3, we study twisted bilayer graphene devices, and quantify their local twist angle and strain on the nanoscale.**

Another material that exhibits Mott physics is Sr_2IrO_4. Like in the twisted graphene bilayer, the presence of Mott physics in this compound is quite surprising. The highest occupied orbital in Sr_2IrO_4, or iridates, is the 5d orbital which is filled with 5 electrons. Because the 5d orbital extends spatially more away from the nucleus than the 3d orbital, the hopping term should be larger, and Mott physics is not necessarily

expected because Coulomb interactions get screened. Nevertheless, iridates are insulating, contrary to what is expected based on band structure calculations. It took a while before it was understood why Mott physics occurs here. The key element turns out to be the strong spin-orbit coupling present in this compound [26]. The 5d states are divided into a t_{2g} band (lower energy) and an e_g band (higher energy). This happens because of crystal field splitting: certain d-orbitals become energetically favored over others based on interaction with the surrounding crystal. Considering the crystal field alone, however, the bandwidth of the t_{2g} manifold is too large and correlations still have a negligible effect. The spin-orbit interaction changes this. When it is included, calculations show that 2 narrow bands split off from the other bands forming a filled $J_{eff} = 3/2$ band and a half-filled $J_{eff} = 1/2$ band [26]. These bands are so narrow that even small correlations become relevant, leading to a $J_{eff} = 1/2$ Mott insulating ground state. In this way, iridates are yet another example where a reduction of bandwidth leads to fascinating electronic behavior. More information about the iridates can be found in reference [27].
We decided to measure iridates with scanning tunneling microscopy and noise spectroscopy measurements. **In chapter 4, we describe how to build and characterize the hardware needed to do noise spectroscopy measurements in a conventional, low temperature scanning tunneling microscope setup.** Our noise spectroscopy measurements on the iridate sample show substantial noise enhancement from the normal poissonian value expected for uncorrelated tunneling events. **In chapter 5, we present our noise spectroscopy measurements on iridates, and explain how random telegraph noise could lead to the observed noise enhancements.**