Atomic-scale contrast mechanism in atomic force microscopy

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Despite the early success of atomic force microscopy in imaging atomic structures on layered materials, much of the hope and expectation for the high resolution capability of this method have been subverted by puzzling experimental results and a poor understanding of the basic contrast mechanism. The fact that very high repulsive forces can be applied between the tip and the sample without losing atomic resolution has led to several different explanations of the imaging process. These include multiple tip-sample contact imaging, large elastic deformations of the sample, and friction-dominated image formation. Until very recently it was only possible to atomically resolve materials having layered structures, such as HOPG, BN, MnPS$_3$, mica, TaS$_2$ and TaSe$_2$. Here, we compare those images with the ones collected from surfaces of the predominantly ionic crystals LiF and PbS. Based on the comparison of these two classes of imaged materials, contrast mechanisms are discussed.

I. Introduction

With the atomic force microscope (AFM) [1] a new method has been introduced to look at the structure of solid surfaces as well as to study minute forces on a sub-nanometer scale. Additionally, AFM on a nanometer scale has given valuable information about technologically interesting insulating samples (for a recent review on force microscopy see [2]). Imaging with the AFM has been performed with attractive as well as repulsive forces, but highest spatial resolution is only possible in the regime of strong repulsive (contact) forces. In this regime atomic resolution on layered materials has ben readily obtained [3].

Despite this early success AFM still suffers from a lack of understanding of the basic contrast mechanism. In particular, studies of layered materials such as highly oriented pyrolytic graphite (HOPG) at atomic resolution have yielded a variety of puzzling observations. It is possible to image these surfaces with loading forces (i.e., repulsive forces between tip and sample) of up to $10^{-6}$ N without losing atomic resolution. These conditions are too extreme for a single atom tip in contact with the surface to be stable, and it is likely that a single atom would puncture the topmost graphite layer. Together with the fact that single atom defects have not been observed so far by AFM, this leads to the conclusion that the contact area is likely to be much larger than a typical single atomic dimension.

Researchers have invoked several explanations to account for these observations. These include tips interacting with the sample at numerous contacts ('multiple tips'), microflakes of sample material being dragged across the surface, the shearing of layers of sample material and a large elastic response of the sample and/or a mediating surface layer.

The demonstration of atomic resolution on harder, non-layered crystals represents an important step since it overcomes the problems of elastic response of layered materials. Individual atoms of ionic crystal surfaces, such as LiF [4], NaCl [5] and PbS have now been resolved with AFM. In formulating an imaging mechanism for these ionic materials, previous explanations valid only for layered samples can be excluded.

In this paper we summarize some of the high resolution AFM work performed in our laboratory. From atomic scale images of the layered compounds HOPG, MnPS$_3$, MoS$_2$, and TaSe$_2$, the effects of friction and multiple contact points are reviewed, and the contribution of a lateral, frictional force to the observed signal is identified as a major source of image contrast on layered materials. We then present atomically resolved images of the nonlayered ionic crystals LiF and PbS. These data are compared to Helium scattering data, and are discussed in the light of recent theoretical efforts. A monatomic tip - model is in reasonable agreement with the experimental data, pointing to a different contrast mechanism for the two classes of materials discussed here.

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II. Experimental procedure

We use electron tunneling to measure the deflection of the probing cantilever. It is our experience that this method takes advantage of the superior sensitivity of the tunneling current and is very reliable as long as there is a good tunneling signal. Before each experiment, we freshly evaporate a gold layer on the lever backside. PtIr tunneling tips work more reliable than tungsten tips since they are more inert. We find these experimental details to be critical for the operation of the AFM. These conditions can be maintained over several days without needing to change either cantilever or tunneling tip.

As force probes we use SiO$_2$ cantilevers which are kindly provided by the Institut de Microtechnique at the Université de Neuchâtel. Force constants are between 0.1 and 0.7 N/m, standard values for this type of AFM experiments. Often no additional tip is attached to the cantilever; the micro-roughness of the SiO$_2$ is sufficient for atomic-scale imaging of flat surfaces, and nearly every cantilever gives atomic resolution.

The instrument is described in detail in previous publications [2, 6]. The key feature is an eccentric mechanism which allows a pure mechanical approach of sample and probe in steps as small as 50 Å. The microscope is operated on an antivibration table at scan rates between 20 and 80 Hz. Pictures are taken in the variable deflection mode, with both constant tunneling current and variable tunneling current conditions. We operate the instrument in the regime of repulsive contact forces in a nearly constant force mode. By performing force vs. distance curves adhesive forces and repulsive forces in the contact zone are determined. For the repulsive forces typical values of $10^{-8}$ N are found.

The samples with layered structures were mounted on sample holders and freshly cleaved with adhesive tape before being transferred to the microscope. As it is the case for HOPG and mica, also MnPS$_3$, MoS$_2$, and TaSe$_2$ all provide large flat areas upon cleavage, ideally suited for fast scan imaging. The non-layered samples are cleaved with a knife edge to expose a (001) surface. All the experiments described here are conducted at ambient conditions.

III. Friction-dominated imaging of layered materials

Soon after the first demonstration of atomic resolution on HOPG by AFM [3], several other layered compounds were imaged, such as highly oriented pyrolytic boron nitride [7] and TaSe$_2$ [8]. Since then atomic-scale resolution of layered materials by AFM has become standard. The interpretation of the data, however, is difficult, and many unresolved questions remain. In this chapter we will demonstrate that frictional forces are a very important contrast source for AFM of layered materials.

Figure 1 shows AFM images of HOPG. This surface consists of a hexagonal net of two inequivalent carbon atoms in a sense that every other carbon atom sits directly above a carbon atom in the layer beneath (A sites); the alternating atoms sit above the centers of the rings in the layer below (B sites). Figure 1a shows a hexagonal structure, every second corner appearing brighter than its neighbor. The observed periodicity corresponds to the known lattice spacing of 2.46 Å, within the experimental uncertainty of ±0.1 Å. This pattern seems to reflect the atomic arrangement and the inequivalency of the atomic sites. A measurable difference in the force experienced by a tip over an A site and over a B site is, however, not predicted by calculations [9]. The experimental corrugation height is 0.1 to 0.3 Å, a value which is also found by other researchers [10] and compares well to Helium scattering data [11].

More often however an AFM image of HOPG only shows every second atom, as in Fig. 1b. The protrusions are spaced 2.5 ± 0.1 Å. These changes in the appearance of images of HOPG can be explained by changes in the relative amplitudes and phases of three dominant Fourier components [12], and can thus be caused by changes in the tip-sample contact.