Are Lattice Relaxations in Thin-Film Nanodevices Driven by Quantum-Size or Interface Proximity Effects?

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The out-of-plane layer relaxations in thin films have been investigated by means of self-consistent density functional theory (DFT). Interface proximity relaxation effects, due to the presence of independent surfaces (or interfaces), have been compared to the relaxation effects driven by quantum-size induced interference between the two interacting surfaces in metallic thin films. We find that the most dominating contribution is due to the interface proximity relaxations and that quantum-size induced effects in fact are small. Our findings give a simple transparent picture of the driving mechanism behind thin film layer relaxations, which can be understood in terms of Fermi surface nesting. Furthermore, the ability to neglect quantum interference makes it possible to estimate thin film relaxations from independent surface or interface relaxation profiles through superposition. Our result provides a limit in the ability to tailor properties due to structural relaxations through size modification of thin film nanodevices.

In the famous gedanken experiment by Democritus a piece of matter is divided into successively smaller and smaller pieces until we finally end up with the individual atoms. Today we may perform a simple extension of this thought experiment. Since individual atoms are very different from the original bulk material, we realize that cutting the material into smaller pieces can be used to alter its properties. This straightforward extension of the Democritus gedanken experiment can be seen as a basis of modern nanoscience, where materials are tailored by a reduction in dimension to have new unique properties not present in the bulk material. This ability is of great scientific interest with numerous technological applications.

From a mathematical point of view the Democritus cutting procedure means that we change the boundary conditions from the periodic Born-von-Karman condition of the crystal, to the boundary condition of the nonperiodic gas phase for the individual atoms. From a physical point of view we have two major effects from reducing the dimension of a material: the presence of a surface or interface, where the relative number of surface atoms to bulk atoms increases when we make a material smaller, and if the material is small enough, the interfaces or the surfaces are close and may interact. These two effects, which we refer to as surface proximity effects and quantum-size (or -interference) effects, respectively, are important for understanding unique properties of materials on the nanoscale.

Recently we have performed a theoretical study of lattice relaxation in thin film nanodevices. The problem is to understand the dominating mechanism behind structural relaxations. Are relaxations due to quantum interference or interface proximity, or do these two effects compete? By analyzing the separate effects on the out-of-plane lattice relaxations of free standing thin metallic films, calculated from *ab initio* theory based on DFT, we find that the influence of quantum-size effects on the out-of-plane lattice relaxation is neglible. This is true even for very thin films of materials where we normally could expect a large electronic effect due to quantum interference. Instead the structural modification is dominated by interface proximity relaxation. This relaxation is driven by the independent interface/surface perturbation, which can be understood in terms of electronic screening. It is therefore possible to establish a qualitative relationship between structural relaxation in thin film nanodevices and the Fermi surface of the unperturbed bulk material, from which the screening can be estimated. Figure 1 shows the out-of-plane relaxation of a thin film of Rh in comparison with the Fermi surface nesting.

Thus, the ability to neglect quantum-size effects gives us a simple transparent picture of the dominant mechanism behind thin film relaxation, which can be analyzed in terms of nesting properties of the Fermi surface of the unperturbed bulk material. Furthermore, the neglect of interface interference effects makes it possible to superimpose relaxation profiles of independent interface/surface relaxations to estimate the relaxation profile in complex thin film nanodevices. This construction provides a structural limit of the ability to tailor electronic properties of thin fil
modifications.

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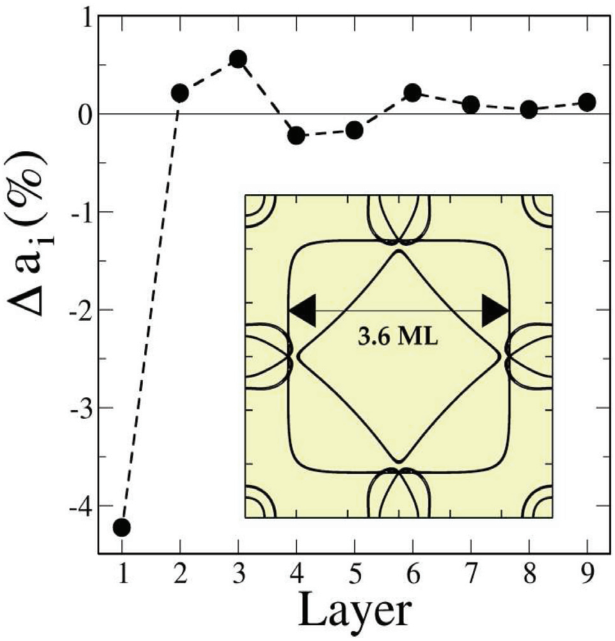

Fig. 1.
The out-of-plane relaxation in the lattice constant a_i for free-standing films of Rh. The periodicity of the relaxation can be estimated from the nesting features of the bulk Fermi surface shown in the inset.