Comment on "Ferromagnetism of the Rh(001) Surface"

In a recent set of *ab initio* pseudopotential calculations, Morrison, Bylander, and Kleinman (MBK) [1] reported that the surface and subsurface layers of Rh(001) are ferromagnetic. This result is surprising in light of previous work on the magnetism of 4*d* overlayer and interlayers [2].

To calculate the local spin-density approximation (LSDA) exchange and correlation potentials, μ_{XC}, MBK use only the valence pseudocharge density; the effect of the core density is assumed to be included in the pseudopotential itself. This construction gives good results for paramagnetic properties and, as discussed by MBK, applying it to magnetic properties increases the tendency towards magnetism.

In order to understand the results of MBK, we calculated the magnetic properties of 9–13 layer films of Rh(001) and Fe(001) using the all-electron FLAPW method [3]. To mimic the μ_{XC} used by MBK, we construct a modified valence-only LSDA (VLSDA) such that the charge (paramagnetic) contributions are determined by the total density, whereas the magnetic contributions are determined by the valence density only.

A standard LSDA calculation for Rh(001) is definitely nonmagnetic; any initial configuration of moments quickly iterates to the nonmagnetic solution. The VLSDA, however, gives a self-consistent solution in good agreement with the results of MBK for the moments and work-function changes; the numerical differences that do exist can be explained by different parametrizations of μ_{XC} and differences between the all-electron and pseudopotential 4*d* orbitals. From these results alone, however, one cannot determine which solution—LSDA or VLSDA—is physically more reasonable.

We also have applied the VLSDA to Fe(001). The LSDA gives bulk (surface) moments of 2.25 (2.98) μ_B, while the VLSDA gives moments of 3.45 (3.77) μ_B. Even with its defects, the LSDA gives reasonable moments, whereas the VLSDA moments are unrealistically large.

The Stoner model of magnetism provides a framework to understand these results. In this model [4], there is a self-consistency condition relating the local exchange splitting to the magnetic moment and the Stoner parameter, *I*. In Fig. 1, the layer magnetic moments for Rh(001) are given as a function of *I*. The LSDA (using the value of *I* given in [5]) and VLSDA (placed on the curves) results are also given. [A similar plot for Fe(001) also correctly places the LSDA moments.] The results show interesting magnetic behavior and imply a Stoner parameter of ~1.5 eV for VLSDA, compared to the LSDA value of 0.65 eV [5]; the Fe(001) results for *I* differ by a similar factor.

This increase in *I* implies a large increase in the susceptibility enhancement. Including various other contributions [6] yields a total VLSDA susceptibility of at least $400\text{–}600\times10^{-6}$ emu mol^{-1}, compared to experimental [7] (or LSDA [6]) values of ~100 $(\sim90)\times10^{-6}$ emu mol^{-1}.

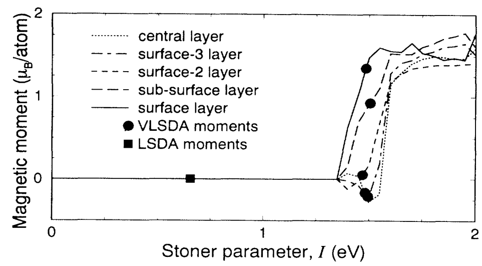

FIG. 1. Calculated moments for nine-layer Rh(001) film as a function of the Stoner parameter, *I*. The self-consistent LSDA and VLSDA moments are also given.

We have demonstrated that (1) in the LSDA, Rh(001) is nonmagnetic, (2) the VLSDA gives results in agreement with MBK, (3) the Fe VLSDA moments are significantly too large, and (4) the Stoner parameter *I* implied by the VLSDA is unphysically large. Thus, we believe that ferromagnetism of the Rh(001) surface is unlikely.

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