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Spin polarization in half-metallic CrO_2

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In materials science, people are investigating materials with a high degree of spin polarization that could form the basis for a future device technology where the spin adds a degree of freedom with respect to the charge-based silicon technology [1]. One material that is being investigated is CrO_2, well known from its use in magnetic tapes. According to several LDA-based electronic structure calculations [2-4], CrO_2 is expected to have a spin polarization close to 100% several eV around the Fermi level.

Polarized and spin-resolved spectroscopic techniques are crucial in obtaining detailed information on the spin polarization. Below the Fermi level, spin-resolved photoemission [5] shows indeed a spin polarization close to 100%, see Fig. 1. However, above the Fermi level, recent spin-resolved circularly-polarized resonant photoemission experiments [6] show a behavior different from that expected from the LDA-based calculations. Within 0.2 eV above the Fermi level, the spin polarization is close to 100%. However, at higher energies above the Fermi level, the spin polarization rapidly decreases to around 50%. The value of 50% is close to what one would expect of electron addition to a local S=1 moment. CrO_2 therefore show a strong competition between quasiparticle and local moment character.

We have performed model calculations that take into account the dual character of the electronic structure of CrO_2. The results are given in Fig. 2. Qualitatively the results can be interpreted as follows. An added spin-up electron can form a quasiparticle with the positively polarized spin background. This leads to a quasiparticle peak at the Fermi level. However, an added spin-down electron is unable to form a quasiparticle with the Fermi sea of spin-up electrons. Therefore, close to the Fermi level only 100% spin-polarized quasiparticle weight is found. However, a higher energies above the Fermi level, the local moment character starts to dominate. To a local moment of S=1, one can add an spin-up electron forming a S=3/2, S_z=3/2 local moment. In the case of a spin down electron one can form S=3/2 and 1/2 local moments, both with S_z=1/2. The S=3/2 moment is close to the Fermi level, leading to a spin polarization of 50%. This is a many-body Slater determinant effect not properly taken into account in the single Slater-determinant based LDA approaches.

In conclusion, we have shown the importance of including the competition between local moment and quasiparticle character in describing the electronic structure of CrO_2. The contents of this paper were supported by the Laboratory for Nanoscience, Engineering, and Technology under a grant from the U.S. Department of Education. MvV is supported by the U.S. Department of Energy (DE-FG02-03ER460097).

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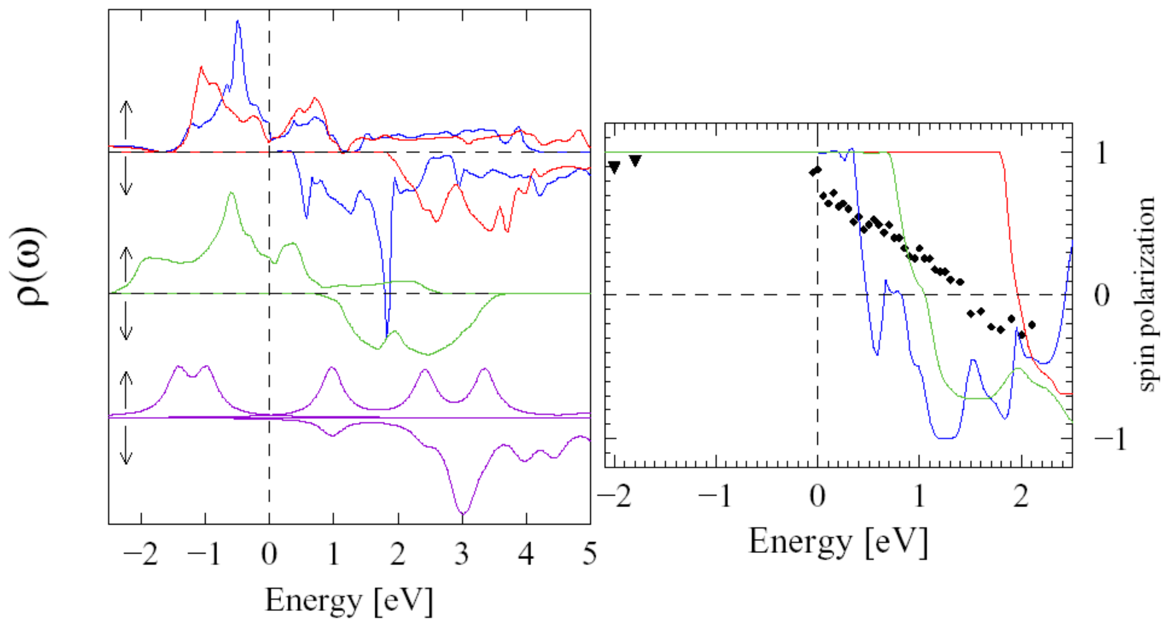

FIG. 1. The left panel shows the spin-polarized density of states for CrO_2 calculated with different methods: a conventional LDA calculation [2] (blue); an LDA+*U* calculation [3] (red); an LDA plus dynamical mean field calculation [4] (green).The violet lines are a configuration-interaction calculation for a CrO_6 cluster. The right panel shows the spin polarizations corresponding to the various LDA-based calculations. The experimental spin polarization is obtained from spin-resolved photoemission [5] (triangles) and spin-resolved polarized resonant O 1*s* Auger spectroscopy [6] (diamonds).

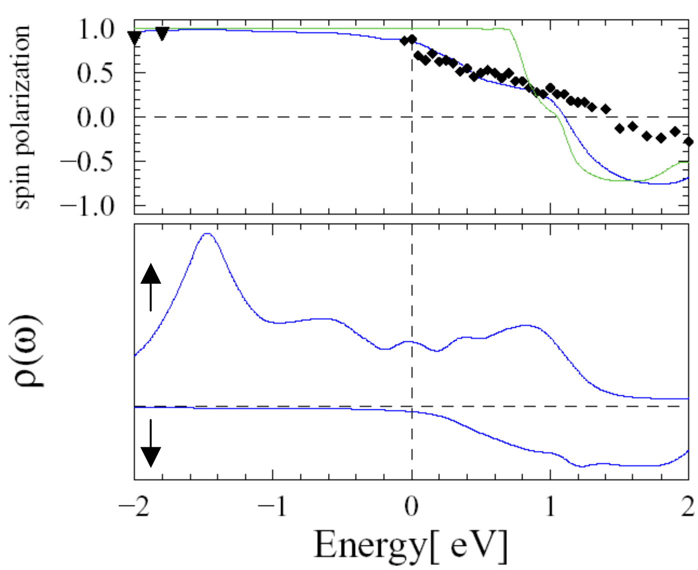

FIG. 2. The lower panel shows the spin-polarized density of states including the competition between quasiparticle and local-moment behavior as described in the text. The upper panel shows the spin polarization (blue). For comparison, the experimental spin polarization from spin-resolved photoemission [5] (triangles) and spin-resolved polarized resonant O 1*s* Auger spectroscopy [6] (diamonds), and the theoretical spin polarization obtained with and the LDA plus dynamical mean field [4] (green) are shown.