SIC: Exchange interactions in magnetic oxides

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In the last years there has been a general strong interest in finding materials with specific or even parametrisable magnetic properties. A lot of the promising candidates are strongly correlated electronic systems which in many ways are still a challenge to be properly described theoretically regarding their electronic ground-state properties. One important example of such materials are transition metal monoxides (TMO), specifically MnO, FeO, CoO, and NiO. They are charge-transfer insulators, well known for strong correlation effects associated with the TM 3d electrons. Originating from the Anderson-type superexchange, their equlibrium magnetic structures are of the antiferromagnetic II (AFII) order, characterized by planes of opposite magnetization which are stacked in (111)-direction (see Fig. 1).
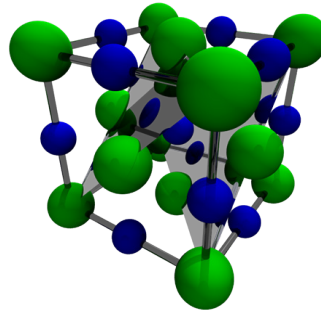

Fig. 1.

A first-principles description of these systems is rather difficult since the conventional local spin density approximation (LSDA) fails to reproduce correctly their insulating ground state or predicts much too small band gaps and magnetic moments. This is associated with an unphysical self-interaction of an electron with itself, occurring in the Hartree term of the LSDA energy functional on account of the local approximation applied to the exchange–correlation energy functional. This self-interaction becomes important for localized electrons like d electrons of TM elements in their monoxides. In the latter, the self-interactions push the localized electron orbitals into the valence band, usually resulting in too strong a hybridization with the other valence electrons (see Fig. 2, upper

panel). This problem was recognized many years ago and a remedy was proposed by Perdew and Zunger [1] to simply subtract the spurious self-interactions from the LSDA functional, orbital by orbital, for all the localized states. The resulting SIC–LSDA approach treats both localized and itinerant electrons on equal footing, leading to split d- and f-manifolds and describing the dual character of an electron (see Fig. 2, lower panel). We implemented the self-interaction corrections method within the the multiple scattering theory and applied it to study electronic magnetic properties of TMO [2,3,4].
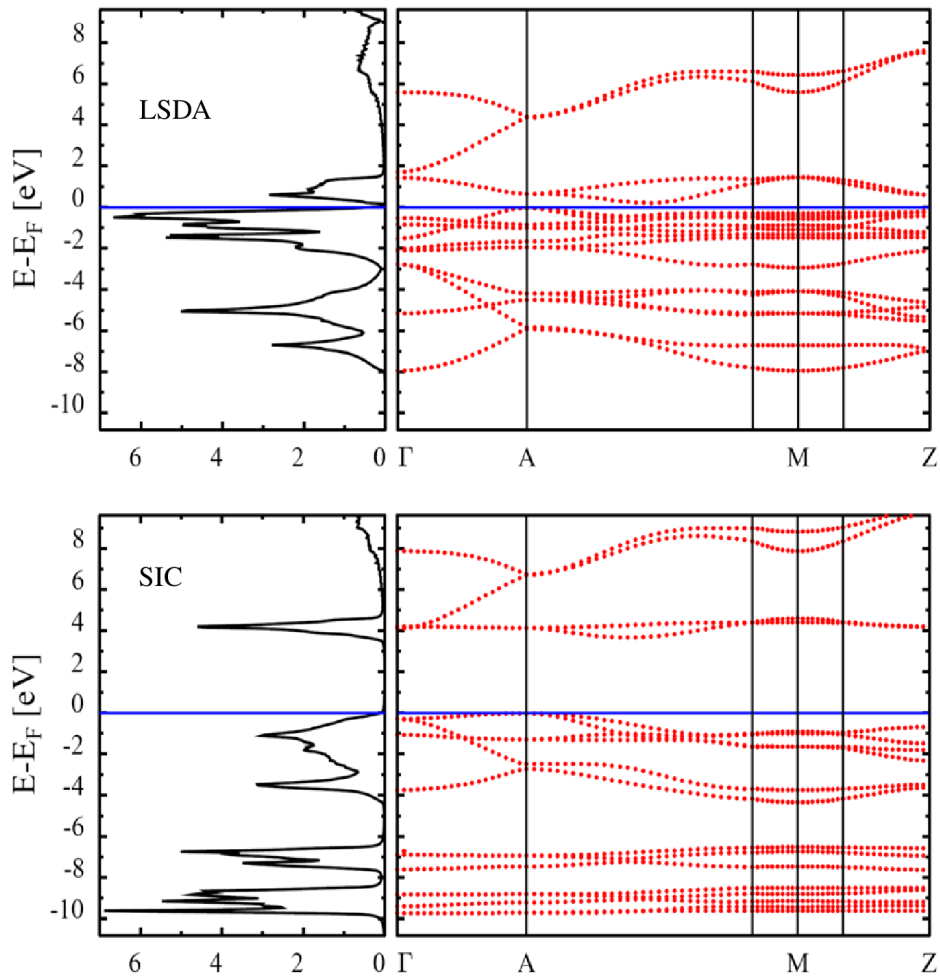

Fig. 2

To characterize the magnetic properties of TMO we investigated thermally induced magnetic fluctuations, which are treated using a mean-field disordered local moment (DLM) picture of magnetism [2]. This involves the assignment of a local spin-polarization axis to all lattice sites. The orientations vary slowly on the time-scale of electronic motion. To determine the magnetic properties of TMO we investigated the spin fluctuations that characterize the paramagnetic state.
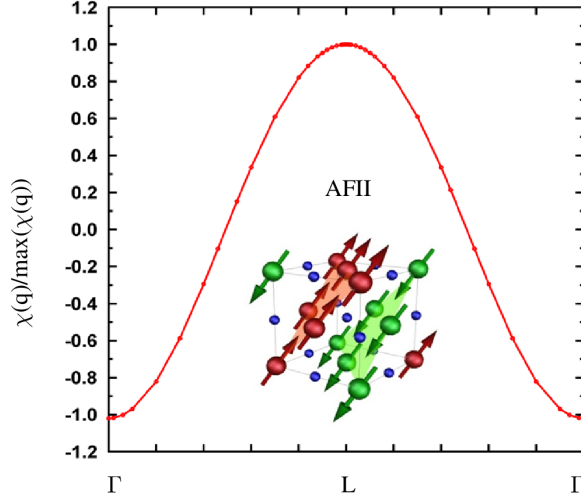
Fig. 3

In Fig. 3, we present the results of our paramagnetic spin susceptibility calculations for NiO. These show the paramagnetic state to be dominated by spin fluctuations with wave vector q_{max} = (0.5; 0.5; 0.5) (in units of $2\pi/a$), which corresponds to the symmetry point L in the Brillouin zone. This indicates that the system will order into the AF II structure, that concurs with the experimentally observed ground state of this system and also calculations at T = 0 K, where the most stable structure was determined by comparing the total energies of different magnetic configurations. We examined the temperature dependence of the static spin susceptibility in particular looking for a divergence which would signify that paramagnetic states become unstable with respect to the formation of a spin density wave, characterized by the wave vector q_{max}. Our paramagnetic susceptibility calculations indicate, that like NiO, the other members of the TMO series have a tendency to order into the AF II structure. The temperatures at which we predict this ordering to occur are shown in Fig. 4. We find a good agreement with the experiment, with the exception of NiO where we underestimated the temperature by about of a third. This suggests that some additional physics, not at work in the other TMOs, may be of relevance to the determination of the ordering temperature of NiO.
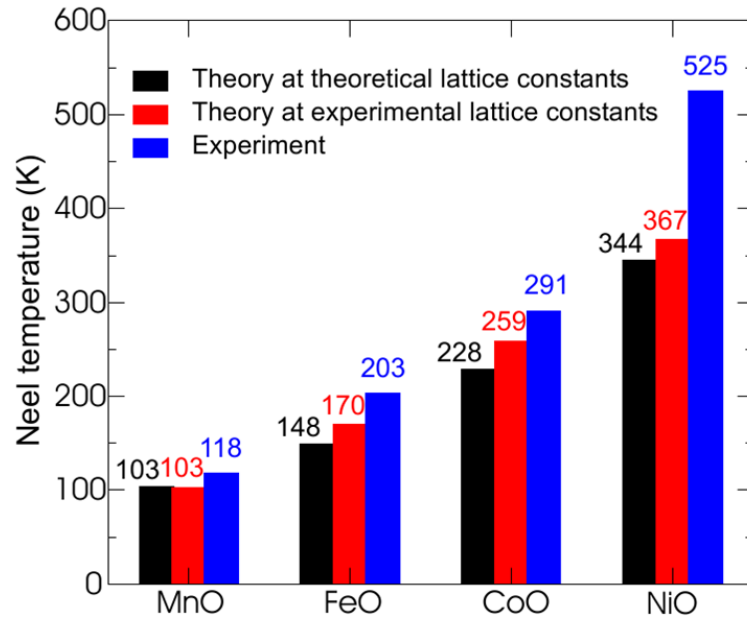
Fig. 4

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