Task: Orbital moment and magnetic anisotropy

Evaluate the orbital magnetic moment and the magnetic anisotropy of a Co dimer in its $S = 2$ ground state. The magnetic anisotropy is the energy difference between magnetization directions along the dimer axis and perpendicular to it.

This task aims at providing an idea about the effects of spin-orbit coupling in metallic clusters.

Subtasks:

(i) At first, perform a calculation without spin-orbit coupling in order to obtain the desired $S = 2$ ground state.

Use a fixed geometry with a bond length of 1.946 Å. Use spacegroup 123 and take the dimer axis parallel to the z-axis.

Set the maximum number of iterations to 100. Use the default convergence criterion (Density). Do not forget to switch on the spin polarized mode and use the fixed spin moment method with a total spin moment of 4 μ_{B}. Enhance the Fermi smearing temperature for the sake of better convergence to 300 Kelvin. Use default settings else.

(ii) Copy all files from the scalar relativistic calculation to a separate directory and perform a full relativistic calculation (this means, spin-orbit coupling is included now). Take the quantization axis (this is the mean direction of magnetization) along (0 0 1). The latter value is the default setting. Switch off the fixed spin moment (the FSM method cannot be applied in full relativistic mode).

How large is the resulting orbital moment of the dimer?

Reduce the Fermi smearing temperature to 150 Kelvin and repeat the calculation (using the existing =.dens file). Check the orbital moment once more.

(iii) Repeat step (ii) for two further quantization axes, (1 0 0) and (0 1 0). More patience is required with respect to the reduction of the Fermi temperature now. Reduce from 300 Kelvin first to 200 Kelvin. Use the obtained =.dens file as input for a final calculation with 150 Kelvin. How large is the orbital moment now? How large is the energy difference between the three different quantization axes?

Solutions (fplo8.65):

(ii) For 300 Kelvin Fermi temperature, the orbital moment amounts to $1.77\mu_B$ per dimer and for 150 Kelvin to 1.97 μ_B per dimer. Such a sensitivity is caused by the small gap between HOMO and LUMO and by the different orbital character of both.

(iii) Both orientations of the magnetization perpendicular to the dimer axis are equivalent by symmetry and yield the same total energy and the same orbital moment of 0.34 μ_B per dimer (150 Kelvin).

The total energy is 1.6 mHartree lower (150 Kelvin), if the magnetization is directed parallel to the dimer axis. Like the orbital moment, this value is very sensitive to the Fermi temperature. Related published data amount to about 1 mHartree (Strandberg *et al.*, Nature Materials **6**, 648 (2007)) and to about 1.8 mHartree (Fritsche *et al.*, J. Comp. Chem. **29**, 2210 (2008)). The latter result was obtained with the FPLO code (100 Kelvin).