Ab initio calculation of magnetic structure of small iron nanoclusters

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Abstract

Local spin and orbital magnetic moments at atoms in free iron clusters with bulk geometry are presented for a range of cluster sizes. The site- and size-dependence of these properties is quite complex and distinctly differs from a simple average between the properties of bulk and surface systems. The local spin magnetic moments depend on the effective coordination number in a linear way.

Keywords: Clusters; Magnetic moments; Iron

We investigate free spherical BCC iron clusters, with the interatomic distances taken from the bulk. The electronic and magnetic structure was calculated within the local density approximation, using a fully relativistic spin-polarized real-space multiple-scattering (MS) method, as implemented in the sprkkr code [1]. The scattering potential was obtained by XαMS calculations, as described in Ref. [2]. The clusters were surrounded with empty spheres located on sites corresponding to further coordination spheres.

The dependence of the local spin and orbital magnetic moments from the distance from the cluster surface is displayed in Fig. 1 for three cluster sizes. We also present analogous information for the planar crystal surfaces, obtained from relativistic tight-binding Korringa–Kohn–Rostoker (KKR) calculations. Spin magnetic moments \( \mu_{\text{spin}} \) of iron clusters of bulk geometry were calculated in the past using an ab initio XαMS method [3] and a parameterized tight-binding model Hamiltonian [4–6]; we include those results in Fig. 1 as well.

The depth dependence of magnetic moments at and below planar and spherical-like surfaces follow quite similar patterns, however, the differences in details persist even at relatively large cluster sizes. The oscillatory behaviour of magnetic profiles is more pronounced at clusters than at planar surfaces. Atoms belonging to the same coordination sphere may have different orbital magnetic moments \( \mu_{\text{orb}} \) because magnetization lowers the symmetry of clusters. Small clusters have a wider spread in \( \mu_{\text{spin}} \)'s than large clusters.

There are distinct differences between \( \mu_{\text{spin}} \)'s calculated by different authors. These differences are larger for the inner atoms than for the outer ones and seem to be too significant to be attributable to various “technical factors”. It appears that even a relatively simple task of computing the magnetic structure of fixed-geometry clusters is quite a complex one in fact.

Fig. 2 displays the dependence of \( \mu_{\text{spin}} \) on the effective coordination number \( N_{\text{eff}} \), defined as \( N_{\text{eff}} = N_1 + 0.25 \times N_2 \), where \( N_1 \) is the number of the nearest neighbours and \( N_2 \) is the number of the second-nearest neighbours of respective atoms [7]. This dependence can be described by a linear function with quite a high accuracy for all the closed-shell cluster sizes between 15 and 89 atoms if \( N_{\text{eff}} \leq 8.5 \); only the smallest cluster of all, comprising 9 atoms, steps out of the line. On the other
hand, there is quite a large spread of $\mu_{\text{spin}}$ for bulk-like atoms (with $N_{\text{eff}} = 9.5$).

Apparently the magnetic properties of iron clusters are not a mere average of properties of its bulk and surface constituents. The site- and size-dependence of the local $\mu_{\text{spin}}$s and $\mu_{\text{orb}}$s exhibit quite a complex structure and it is quite difficult to determine whether the limits of convergence have been reached already or not.

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