Spin-orbit induced non-collinear spin structure
in deposited transition metal clusters

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Abstract

The influence of the spin-orbit coupling on the magnetic structure of deposited transition metal nanostructure systems has been studied by fully relativistic electronic structure calculations. The interplay of exchange coupling and magnetic anisotropy was monitored by studying the corresponding magnetic torque calculated within ab-initio and model approaches. It is found that a spin-orbit induced Dzyaloshinski-Moriya interaction can stabilise a non-collinear spin structure even if there is a pronounced isotropic ferromagnetic exchange interaction between the magnetic atoms.

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As the complexity and technological applicability of nanostructured magnetic materials grows it has become important to develop a reliable quantitative theoretical framework in which to understand them. In principle this is available from relativistic density functional theory (RDFT) \[1\]. The magnetic behaviour of complex magnetic systems is described effectively in terms of 'local moments', even in metallic systems, which fluctuate as the temperature is increased from 0K. Within the effective single electron picture of DFT, the moments arise from local magnetic fields associated with the different atomic sites which affect the electronic motions and are self-consistently maintained by them. The spin-orbit coupling (SOC) effects on the electronic structure determine the magnetic anisotropy linking the magnetic and spatial structure of a material. As demonstrated by Staunton et al. \[2\] and others, magnetic torque calculations enable magnetic anisotropy to be studied reliably and magnetic structures to be determined. We have recently developed a method to study ab-initio the magnetic structures of complex nanostructures using this approach \[3\]. It is based on the framework of relativistic density functional theory using the local spin density approximation (LSDA) for exchange and correlation effects \[4\]. The electronic structure was determined in a fully relativistic way from the basis of the Dirac equation for spin-polarized potentials which is solved using the Korringa-Kohn-Rostoker (KKR) multiple scattering formalism \[5\].

In this letter we set out to understand the relativistic effects on the magnetic structure of nanoclusters in simpler terms of a model ‘moment’ Hamiltonian. In this context the mapping of energetic properties obtained from first principles calculations of complex magnetic systems onto a Heisenberg Hamiltonian has proved over recent years to be a very robust and successful scheme \[6, 7\]. Moreover for many systems the results concur with those from an ab-initio ‘disordered local moment’ theory in which no prior mapping to a Heisenberg system is assumed \[8, 9\]. An extensively used approach to calculate the isotropic exchange interaction parameter $J_{ij}$ for two magnetic moments on sites $i$ and $j$ for use in the classical Heisenberg Hamiltonian was worked out by Lichtenstein et al. \[6\] using perturbation theory and the so-called Lloyd formula. A corresponding fully relativistic approach was later introduced by Udvardi et al. \[7\] that produces an exchange interaction tensor $J_{ij}$ for use in a prescribed extended Heisenberg Hamiltonian. This scheme generates in particular a Dzyaloshinski-Moriya (DM)-type interaction \[10, 11\], which may explain many interesting phenomena including the magnetic ground state configuration of nanostructures \[12, 13\] as
well as magnetic thin films [14, 15]. Here we explore what form the effective moment Hamiltonian should have by a detailed study of its building blocks. With an investigation of Fe$_2$, Co$_2$ and Ni$_2$ dimers deposited on Pt(111) as most simple cluster examples we identify strong DM-type interactions as well as an additional substrate-generated effect in Ni$_2$ (for calculational details see [3]). The conclusions of these results can then be applied straightforwardly to larger clusters and nanostructures.

We start by considering the magnetic torque vector $\vec{T}_i(\hat{e}_i)$ acting on an atomic magnetic moment on a site $i$ ($i = 1$ or 2) and aligned along direction $\hat{e}_i$. The torque vector is defined in terms of the change in energy $E(\{\hat{e}_k\}) = E(\hat{e}_1, \hat{e}_2)$ of the system when changing the orientation of the magnetic moment, $\hat{e}_i$ on site $i$, $\vec{T}_i(\hat{e}_i) = -\partial E(\{\hat{e}_k\})/\partial \hat{e}_i$. The component $T_{i,\hat{u}}(\hat{e}_i) = -(\partial E(\{\hat{e}_k\})/\partial \hat{e}_i) \cdot (\hat{u} \times \hat{e}_i)$ of $\vec{T}_i(\hat{e}_i)$ with respect to the axis $\hat{u}$ can be determined from first-principles using an expression derived by Staunton et al. [2]. Following on from this the derivative $\partial^2 E/\partial \hat{e}_i \partial \hat{e}_j$ describing the change in energy upon changing the orientation of two magnetic moments on sites $i$ and $j$ can also be obtained [6, 7]. By making use of the rigid spin approximation (RSA) [16] this approach leads to a fitting of the magnetic energy landscape $E(\{\hat{e}_k\})$ at low temperatures obtained from first principles calculations onto a Heisenberg model Hamiltonian. With SOC included an anisotropy in the exchange interaction may occur. We then use these quantities in an extended classical Heisenberg Hamiltonian for the ‘spins’, $\{\hat{e}_k\}$ of the following form, (e.g.,[7, 17]):

$$H = -\frac{1}{2} \sum_{i,j(i\neq j)} J_{ij} \hat{e}_i \cdot \hat{e}_j - \frac{1}{2} \sum_{i,j(i\neq j)} \hat{e}_i J^S_{ij} \hat{e}_j - \frac{1}{2} \sum_{i,j(i\neq j)} \vec{D}_{ij} \cdot [\hat{e}_i \times \hat{e}_j] + \sum_i K_i(\hat{e}_i).$$

(1)

Here the exchange interaction tensor $J_{ij}$ has been split into its conventional isotropic part $J_{ij}$, its traceless symmetric part $J^S_{ij}$ and its anti-symmetric part $J^A_{ij}$. We assume that the latter one is represented in terms of the DM vector $\vec{D}_{ij}$ with $D^\gamma_{ij} = e^{\alpha\beta\gamma} J^\alpha_{ij} J^\beta_{ij}$ (with $\epsilon^{\alpha\beta\gamma}$ the Levi-Civita symbol). Finally, the anisotropy constants $K_i(\hat{e}_i)$ account for the so-called on-site anisotropy energy associated with each individual moment oriented along $\hat{e}_i$. From our ab-initio calculation of the torques $T_{i,\hat{u}}(\hat{e}_i)$ we can test whether such a model Hamiltonian is justified and also find the values of the $J_{ij}, K_i, J^S_{ij}$, and $\vec{D}_{ij}$ parameters.

For the model Eq. (1) the rate of change in energy when a moment on site $i$ is rotated about an axis $\hat{u}$ can be partitioned into contributions from different terms of Heisenberg
Hamiltonian $T_{i,\hat{u}}^{(\hat{e}_i)} = T_{i,\hat{u}}^{\text{iso}} + T_{i,\hat{u}}^{S} + T_{i,\hat{u}}^{\text{DM}} + T_{i,\hat{u}}^{K}$. The contribution to the torque from the DM coupling is given by:

$$T_{i,\hat{u}}^{\text{DM}} = \sum_{j \neq i} (\vec{D}_{ij} \cdot \hat{u})(\hat{e}_i \cdot \hat{e}_j) - \sum_{j \neq i} (\vec{D}_{ij} \cdot \hat{e}_i)(\hat{u} \cdot \hat{e}_j) \quad (2)$$

The last term derives from the single site anisotropy term, i.e. $T_{i,\hat{u}}^{K} = \frac{\partial K(\hat{e}_i)}{\partial \hat{e}_i} \cdot (\hat{u} \times \hat{e}_i)$. By focussing on nanoclusters of collinear ‘spin’ arrangements, the magnetically anisotropic terms can be determined. Note that the last term in Eq. (2) does not contribute to the torque in the case of a collinear magnetic structure. Also the sum of the DM contributions $T_{i,\hat{u}}^{\text{DM}}$ from all sites in a nanocluster to the total torque vanishes in this case. On the other hand, the anisotropy of the exchange interaction, represented by the symmetric tensor $J_{ij}$, gives rise to a finite torque hence contributing to the total magnetic anisotropy energy of the nanocluster.

We now see how well our ab-initio calculations of deposited Fe, Co and Ni dimers, using our first-principles multiple scattering formalism, fit such model torques. Firstly we find that the dimers, when constrained to be magnetically collinear exhibit a pronounced out-of-plane magnetic anisotropy \[18\]. The collinear magnetic moments $\hat{e}_1$ and $\hat{e}_2$ have been assumed to be orientated at an angle to the surface normal ($z$-axis) and are expressed in terms of polar angles $\theta$ and $\phi$, as $\hat{e}_1 = \hat{e}_2 = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$. Fig. 1 shows the atomic configuration together the projection of the moments onto the surface ($xy$-plane). In parallel to the fixed frame of reference ($x, y, z$) we use a second one ($x', y', z'$) rotated by $\phi$ with respect to the fixed one with $\hat{z} = \hat{z'}$ (see Fig. 1). In all calculations the torque $T_{i,\hat{u}}^{\hat{e}_i}$ is taken around the $y'$ axis, i.e. $\hat{u} = \hat{y'} = (-\sin \phi, \cos \phi, 0)$, for $\theta$ fixed to $\pi/4$ as a function of the azimuthal angle $\phi$. From the model Eq. (1) an expansion of the anisotropy energy term $K(\hat{e}_i)$ in terms of $l = 2$ spherical harmonics, gives the single site contribution to the torque in present configuration to be $T_{i,\hat{u}}^{K} = -2[K_{2,1} + K_{2,2} \cos(2\phi) + K_{2,2}' \sin(2\phi)]$ whereas $T_{i,\hat{u}}^{\text{DM}}$ deduced from the model has a $\cos(\phi)$ ($\sin(\phi)$) variation when $\vec{D}_{1,2}$ lies in the $yz$ ($xz$) plane.

In Fig.2 the ab-initio results for the torque $T_{i}^{\text{dir}}$ are compared with those deduced from the model Eq. (1). As one notes, the torques for the two atoms are different but are related with respect to their $\phi$-dependence according to the $C_s$ symmetry of the system. On comparing the torque of the two Co atoms of Co$_2$/Pt(111) obtained directly from the electronic structure calculations with those from the model (see Fig. 2a-c) we find the symmetric part of the
The exchange interaction tensor $T_i^S$ to be negligible. The contribution $T_i^{\text{DM}}$ due to the DM interaction Eq. (2) is shown in Fig. 2b. Clearly, $T_1^{\text{DM}}$ and $T_2^{\text{DM}}$ vary with $\cos \phi$ and are opposite in sign. In accordance with the $C_s$ symmetry, the DM vector $\vec{D}_{12}$ lies in the $yz$ plane (Fig. 1). For the contribution $T_i^K$ due to the on-site anisotropy one finds $K_{2,2}^i$ to be very small and the dominating terms $K_{2,1}$ and $K_{2,2}$ to be practically the same for both atomic sites leading to $T_1^K \approx T_2^K$.

The contribution to $T_i^K$ connected with $K_{2,1}$ does not depend on $\phi$, while that connected with $K_{2,2}$ varies with $\cos(2\phi)$ (Fig. 2c). As one can see, the torque $T_i^{\text{mod}}$ derived from the model Hamiltonian reproduces the results $T_i^{\text{dir}}$ calculated directly rather well. The remaining deviation is primarily due to the limitations of the model Hamiltonian with respect to the dependency of magnetic energy on the magnetic moment orientations $E(\{\hat{\mathbf{e}}_k\})$. From the decomposition of the $T_i$’s via the model Hamiltonian it becomes clear that its $\phi$-dependence is dominated by the DM-contribution while the $K_{2,2}$-contribution gives rise to a minor additional modulation. Owing to the large negative values of $T_1(K_{2,1})$ and $T_2(K_{2,1})$ an out-of-plane anisotropy results for the total system. This can be seen also from Fig. 2d, where the total torque $T_i^{\text{dir}} = T_1^{\text{dir}} + T_2^{\text{dir}}$ for Co$_2$ on Pt(111) is shown together with the individual contributions $T_i^{\text{dir}}$ and the corresponding DM terms $T_i^{\text{DM}}$. Once more one sees that the $\phi$-dependence of the individual torques is determined by $T_i^{\text{DM}}$ while that of the total torque $T_i^{\text{tot}}$ is set by the $K_{2,2}$ on-site anisotropy terms. This also holds for Fe$_2$ (Fig. 2d) for which the DM terms are even more dominant, i.e. the $\phi$-dependence of the individual torques are nearly exclusively due to $T_i^{\text{DM}}$. 

FIG. 1: Magnetic configuration of the transition metal dimers deposited on a Pt(111) substrate. The large (small) spheres present Pt surface (subsurface) atoms. The medium size spheres represent the dimer atoms with the projection of their magnetic moments onto the surface ($xy$-plane) represented by arrows. The projection of the DM-vector $\vec{D}_{12}$ onto the surface is represented by a short arrow.
FIG. 2: Magnetic torque $T_i$ for dimers on Pt(111) for $\theta = \pi/4$ as a function of the azimuthal angle $\phi$. Left: Torque for Co$_2$/Pt(111): a) results $T_i^{\text{dir}}$ of direct calculations (up and down triangles) compared with results $T_i^{\text{mod}}$ obtained on the basis of the Heisenberg-type Hamiltonian (Eq. (1)); b) $T_i^{\text{DM}}$ contribution to $T_i^{\text{mod}}$ due to the DM interaction (Eq. (2)); c) contribution $T_i^K$ to $T_i^{\text{mod}}$ due to the on-site anisotropy. Right: Magnetic torque for the two atoms of Fe$_2$ (d), Co$_2$ (e) and Ni$_2$ (f). The results calculated directly are presented by up ($T_i^{\text{dir}}$) and down ($T_i^{\text{dir}}$) triangles and their sum by thick solid line ($T_{\text{tot}}^{\text{dir}} = T_1^{\text{dir}} + T_2^{\text{dir}}$). The thin solid lines give the DM contributions according to Eq. (2).

The situation is noticeably different for Ni$_2$ on Pt(111) (Fig. 2f) for which the DM terms give only minor contributions to the individual torques $T_1$ and $T_2$. In contrast to Fe$_2$ and Co$_2$ the difference between the torques of different atoms cannot be attributed to DM coupling. The period of oscillations of the torques as functions of $\phi$ are different compared to those of the torques created by DM coupling varying as $\sin(2\phi)$. Clearly an effect which is not contained in the model Hamiltonian Eq. (1) is evident and must derive from interactions of the Ni moments with those induced in the Pt substrate. As with a DM-type interaction it produces torques such that $T_1 = -T_2$ but its $\sin(2\phi)$ variation would arise if a term of the form $(\vec{A} \cdot \hat{e}_1)(\vec{B} \cdot \hat{e}_1) - (\vec{A} \cdot \hat{e}_2) \cdot (\vec{B} \cdot \hat{e}_2)$ were added to Eq. (1), where $\vec{A}$ lies along $x$ and $\vec{B}$ along $y$. These Ni$_2$ dimer results indicate that an effective Heisenberg model must be used with caution for systems where the magnetic structure of a nanocluster is strongly influenced by the spin polarisability of the substrate.

For the Co$_2$ and Fe$_2$ dimers, however, the model works very well. For our chosen geometry $\theta = \pi/4$ and $\phi = 0$ and using the symmetry properties of the elements of the exchange tensor,
TABLE I: Components $D_{ij}^\alpha$ of the DM vector $\vec{D}_{ij}$, the isotropic exchange constant $J_{ij}$ (in meV) and the tilt angle $\alpha$ (in degrees) (see text) for the dimers Fe$_2$ and Co$_2$ on Pt(111). The data labelled *direct* have been obtained from the direct first principles calculations of the torque. The data below have been obtained by mapping first principles results onto the model Hamiltonian, Eq. (1).

|       | direct | model |
|-------|--------|-------|
| Fe$_2$ | 6.04   | 0.00  |
| Co$_2$ | 3.69   | 0.00  |
| Ni$_2$ | -0.02  | 0.00  |

one finds for Co$_2$/Pt(111) with $T_1^K \approx T_2^K$ the total torque $T = T_1 + T_2 = -(J_{ij}^{Sxx} - J_{ij}^{Szz}) + 2T_1^K$. Finally, the magnetic anisotropy energy (MAE) of the dimer being the difference in energy when the magnetic moments are both oriented along $\hat{e}_b$ and $\hat{e}_a$, is given by the integral $-\int_{\hat{e}_a}^{\hat{e}_b} (\vec{T}_1(\hat{e}) + \vec{T}_2(\hat{e}))d\hat{e}$. Obviously this has no contribution from the DM interaction. For Co$_2$/Pt(111) we find the exchange parameters $J_{ij}^{xx}$, $J_{ij}^{yy}$ and $J_{ij}^{zz}$ to be nearly the same implying that the total MAE of the dimer is nearly exclusively due to the on-site anisotropy. The values $K_{2,1} = 1.5$ meV and $K_{2,2} = 0.39$ meV for Co$_2$/Pt(111) lead as mentioned above to a pronounced out-of-plane anisotropy, i.e. in the ground state the total magnetisation points along the surface normal.

Taking the difference between the individual torques one arrives at the relation $D_{ij}^y = \frac{T_1 - T_2}{2}$ allowing $D_{ij}^y$ to be deduced directly from the ab-initio torque $T_i^{\text{dir}}$ calculations. Table [I] shows the corresponding results for dimers Fe$_2$ and Co$_2$ on Pt(111) in comparison with data derived from a mapping to our model Hamiltonian, Eq. (1).

The closeness of the results justifies once more the use of the model Hamiltonian for Fe$_2$ and Co$_2$. In addition one notes that $D_{ij}^y$ has an appreciable value compared to the isotropic exchange constant $J_{ij}$. Fixing the azimuthal angle $\phi$ to be $\pi/2$ and performing similar steps one finds $D_{ij}^x$ to be 0. This is also in line with the $C_s$ symmetry of the investigated dimer systems (see Fig. [II]). $D_{ij}^z$, on the other hand, may take a non-zero value and is found to be comparable to $D_{ij}^y$ (see Table [I]).
Thus, the above analysis shows that the torques $T_1$ and $T_2$ may differ even if the total torque is zero, i.e. if the moments are aligned collinearly along the easy axis (surface normal). The difference between these torques is caused exclusively by the $D_{12}^y$ terms leading to a rotation around the $y$-axis. Minimizing the magnetic energy $E(\{\hat{e}_i\})$ of the 2 atom clusters leads to an outward tilting of the moments by an angle $\alpha$ given by $\alpha = \tan \left( \frac{D_{12}^y}{J_{12}} \right)$ with $J_{12}$ being the isotropic exchange coupling constant. The corresponding results given in Table I show that the DM interaction causes the deposited Fe$_2$ and Co$_2$ dimers to have an appreciable deviation from collinear configurations in spite of the pronounced ferromagnetic exchange coupling given by $J_{12}$. This effect of SOC is completely in line with the findings of Sandratskii and Kübler [19] for bulk systems.

The substrate clearly plays a crucial role in the DM interaction. Firstly, hybridisation with the substrate breaks the inversion symmetry for the dimer leading to a non-zero DM vector. This symmetry effect is also confirmed by test calculations on free dimers and on dimers embedded in a bulk Pt matrix. Secondly, the hybridisation with the substrate also allows the SOC effects of the substrate to be transferred to the magnetic 3d transition metal dimer. We have confirmed this by further calculations in which the SOC of the substrate and the dimer atoms were manipulated separately. Enhancing the SOC for a Co dimer leads primarily to an increase of the on-site anisotropy $K_{2,2}$. However, enhancing the SOC for the Pt substrate leads to a strong increase in the anisotropy $K_{2,1}$ as well as to a larger difference in the individual torques on the two Co atoms, reflecting an increase of the DM interaction (Fig. 2). This behaviour is in line with Levi’s model of the indirect DM interaction between two spin moments [20], which is mediated by nearby atoms. As a consequence, the magnitude of the DM interaction is essentially determined by the SOC strength of the neighbouring atoms.

The effect of anisotropic exchange is even more spectacular in magnetic alloy nanoclusters, where magnetic atoms are separated by non-magnetic atoms with large SOC. This is demonstrated by our calculations for a FePt (2×1) 2D alloy cluster deposited on the Pt(111) surface pertinent to an experimental investigation by Honolka et al. [21]. Ab-initio torque calculations show the clear trend that the magnetic structure of this cluster is non-collinear. Here we present the results of Monte Carlo simulation based on an effective Heisenberg model with the isotropic and DM exchange coupling parameters calculated in accordance to Ref. [7]. Fig. 3 shows the non-collinear magnetic structure obtained for $T = 0$K. The
FIG. 3: Magnetic structure of a 91-atom FePt (2 × 1) 2D alloy cluster deposited on the Pt(111) surface determined via MC simulation for $T = 0$K. The arrows denote the orientation of the magnetic moments associated with the iron atoms.

non-collinearity between the Fe chains is essentially caused by the nearest neighbours Fe-Fe interchain DM interaction ($|\vec{D}| = 4.6$ meV) being of similar magnitude when compared to the isotropic exchange interaction ($J = 8.8$ meV). Within a Fe chain, however, the DM interaction is more than one order of magnitude smaller when compared to the isotropic exchange leading only to a slight screwing of the Fe magnetic moments along the chain. Thus, we see that the FePt cluster’s non-collinear structure is created by an enhanced DM coupling between Fe moments mediated by Pt atoms having large SOC, on the one hand side, while the isotropic exchange between these atoms separated by non-magnetic Pt is small enough to make both these couplings comparable.

In summary our investigation of deposited transition metal dimers as simple but realistic model systems has demonstrated that ab-initio magnetic torque calculations enable the impact of SOC on the magnetic interactions within nanostructures to be monitored in a very detailed way revealing subtle anisotropic effects. The analysis of directly calculated electronic structure quantities within a framework based on a Heisenberg model Hamiltonian gives further insight and identifies the role of the various contributions and also the limitations of such models. For the Fe and Co dimer systems studied here the DM interaction was found to be pronounced owing primarily to the SOC of the substrate. Moreover it leads to non-collinear magnetic configurations of the dimers in spite of the pronounced ferromagnetic coupling and out-of-plane anisotropy. These magnetically anisotropic interactions also have a profound effect in larger clusters and we have demonstrated this explicitly with a study of a deposited FePt cluster. In particular we infer that the magnetic structure around the edges of magnetic nanoparticles is likely to be significantly affected.
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