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## Is the magnetic anisotropy proportional to the orbital moment?

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The relation between orbital moment and magnetic anisotropy is investigated by model calculations, which show that only a part of the spin-orbit coupling contributes to the anisotropy. A large part of the anisotropy energy, about 50% for iron series elements and nearly 100% for rare-earths, is stored in the nonrelativistic part of the Hamiltonian. A feature important for x-ray magnetic circular dichroism is that the orbital moment of heavy atoms rotates with the spin moment, whereas in light atoms, the orbital moment is recreated in each different direction. In the discussion, we consider three examples of current interest in different areas of magnetism, namely, spin-orbit coupling in  $Gd^{3+}$  and  $Eu^{2+}$ , surface anisotropy of  $Nd_2Fe_{14}B$ , and multiferroic magnetization switching using rare-earths. © 2011 American Institute of Physics. [doi:10.1063/1.3562445]

### I. INTRODUCTION

It is well established that the spin-orbit coupling (SOC) is the origin of both orbital moment and anisotropy and that magnetocrystalline anisotropy requires an orbital moment.<sup>1</sup> However, the relation between orbital moment and magnetic anisotropy has recently attracted renewed attention. Based on a widely quoted paper,<sup>2</sup> it has become popular to equate the spin-orbit energy with the magnetocrystalline anisotropy energy (MCA). It is correct that the SOC modifies the nonrelativistic Hamiltonian by a spin-orbit term, but the anisotropy energy is generally very different from the spin-orbit energy. For example, the  $4f$  shells in rare-earth magnets exhibit a rigid spin-orbit coupling, and the anisotropy energy is essentially equal to the crystal-field (CF) energy.<sup>3,4</sup> This effect is exploited in high-performance permanent magnets such as  $SmCo_5$  and  $Nd_2Fe_{14}B$ .<sup>3,4</sup>

The ratio of spin-orbit energy SOC energy to crystal-field or band energy has far-reaching implications, from the physical understanding of orbital-moment quenching to the interpretation of experimental methods such as x-ray magnetic circular dichroism (XMCD). For example, the orbital-moment contribution to an XMCD signal may involve two different mechanisms, namely, the rotation of the orbital moment and its collapse. A transparent real-space interpretation is in terms of orbital currents,<sup>5</sup> which are particularly strong in magnets with rotational symmetry, such as monatomic transition-metal wires. Intriguingly, the rigid rare-earth spin-orbit coupling (large SOC) has recently been questioned for  $4f^7$  ions ( $Gd^{3+}$  and especially  $Eu^{2+}$ ).<sup>6</sup>

### II. CALCULATIONS AND RESULTS

To investigate the spin-orbit and crystal-field contribution to the magnetic anisotropy, we use the quasiclassical model Hamiltonian

$$E = -\lambda \mathbf{S} \cdot \mathbf{L} + A_o \left( L_x^2 + L_y^2 + L_z^2 \right) - A \left( L_z^2 - \frac{1}{2} \left( L_x^2 + L_y^2 \right) \right) \quad (1)$$

where  $\lambda$  is the spin-orbit coupling constant. The effective crystal- or ligand-field parameters  $A > 0$  and  $A_o > A$  describe the nonrelativistic electronic structure of the magnet. Note that hybridization (band structure) and electrostatic crystal fields have very similar effects on orbital moment and anisotropy, in molecules known as ligand-field interaction.<sup>5,7</sup> In the present model,  $0 \leq L^2 \leq 1$ , corresponding to fully quenched ( $L^2 = 0$ ), partially quenched ( $0 < L^2 < 1$ ), and unquenched ( $L^2 = 1$ ) orbital moments. For simplicity, we also assume that  $S^2 = 1$ . Other choices of  $L$  and  $S$  do not yield additional physics.

The parameter  $A_o$  describes the quenching of the orbital moment due to the crystal field. Physically, the  $d$  and  $f$  electrons prefer eigenstates whose overlap with neighboring electron clouds are small. The creation of an orbital moment corresponds to a local electron current which partially enters otherwise avoided regions and therefore costs CF energy.<sup>4,5</sup> Both  $A$  and  $A_o$  may exceed 1 eV, especially in transition-metal magnets, and the condition  $A_o > A$  ensures that there are no orbital moments in the absence of SOC. The case  $A_o = A$  is approximately realized in free-standing monatomic wires, where a small SOC gives rise to a huge orbital moment.<sup>4,8</sup> Bringing such a wire into contact with a substrate destroys the orbital moment and effectively reinstates  $A_o > A$ .

For small and moderate spin-orbit coupling ( $L^2 < 1$ ), minimization of Eq. (1) with respect to  $L_z$  and  $L_x$  yields

$$\lambda S_z = 2(A_o - A)L_z \quad (2a)$$

$$\lambda S_x = 2(A_o + A/2)L_x. \quad (2b)$$

Due to the tensor character of the orbital term in Eq. (1),  $\mathbf{S}$  and  $\mathbf{L}$  are generally noncollinear, but there is no mixing if  $\mathbf{S}$  is parallel to any of the magnet's principal axes. For spin orientations in the  $z$  and  $x$  directions, the orbital moments are

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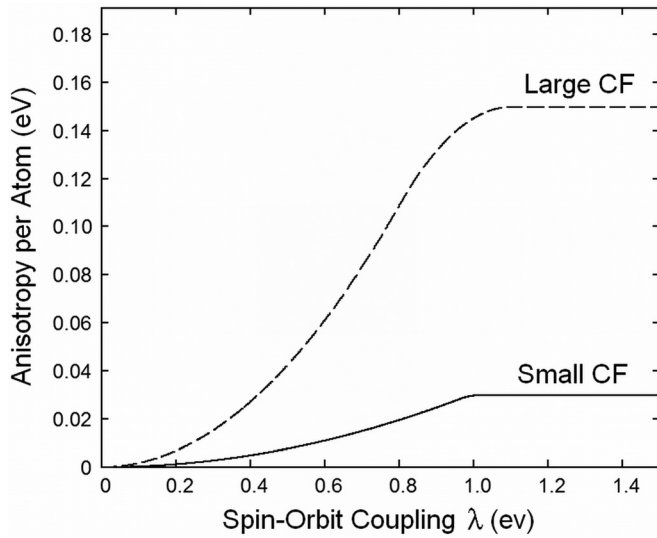


FIG. 1. Anisotropy per atom as a function of the spin-orbit coupling (SOC) strength for two different crystal fields (CF). Iron-series atoms have large CF but small SOC, whereas rare-earths have small CF due to screening but large SOC. For the palladium and platinum series, the ratio of the two parameters is intermediate.

$[1/2]\lambda/(A_0 - A)$  and  $[1/2]\lambda/(A_0 + A/2)$ , respectively. The anisotropy energy is obtained by substituting the orbital moments into Eq. (1) and comparing the total energies for the two directions:

$$E_a = \frac{\lambda^2}{4} \left( \frac{1}{A_0 - A} - \frac{1}{A_0 - A/2} \right). \quad (3)$$

The corresponding first uniaxial anisotropy constant  $K_1 = E_a/V_{\text{at}}$ , where  $V_{\text{at}}$  is the volume per atom. Equation (3) is of the familiar type  $K_n \sim \lambda^{2n}/\Delta A^{2n-1}$ , where  $n$  is the order of the anisotropy. This result was first obtained by Bloch and Gentile<sup>1</sup> for cubic anisotropy ( $n = 4$ ).

By substituting  $L_z$  and  $L_x$  into Eq. (1) we can separately evaluate the SOC energy,  $E_{\text{SOC}} = 2E_a$ . In other words, only 50% of the spin-orbit energy translates into anisotropy energy. The other 50% are used for the increase in crystal-field energy, which accompanies the formation of the orbital moment.

Minimization of Eq. (1) and taking into account that  $L^2 \leq 1$  yields a number of limiting and intermediate scenarios. Figure 1 shows two anisotropy energies as a function of the spin-orbit constant  $\lambda$ . For small  $\lambda$ , the behavior is quadratic, as in Eq. (3). For large  $\lambda$ , the anisotropy reaches a plateau at  $E_a = 3A/2$ . The two limits corresponds to the well-known cases of 3d (iron-series) and 4f (lanthanide) anisotropies, respectively. Figure 2 shows the anisotropy energy relative to several spin-orbit energy terms and illustrates that there is no simple relation between spin-orbit coupling and anisotropy energy.

### III. DISCUSSION

Let us discuss three examples of importance in different areas of magnetism, namely, the spin-orbit coupling in half-filled 4f shells, the surface anisotropy of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , and the exploitation of the SOC in multiferroic switching without strain.

A recent paper has questioned the applicability of the rigid-coupling approach to 4f<sup>7</sup> ions, by arguing that  $L = 0$

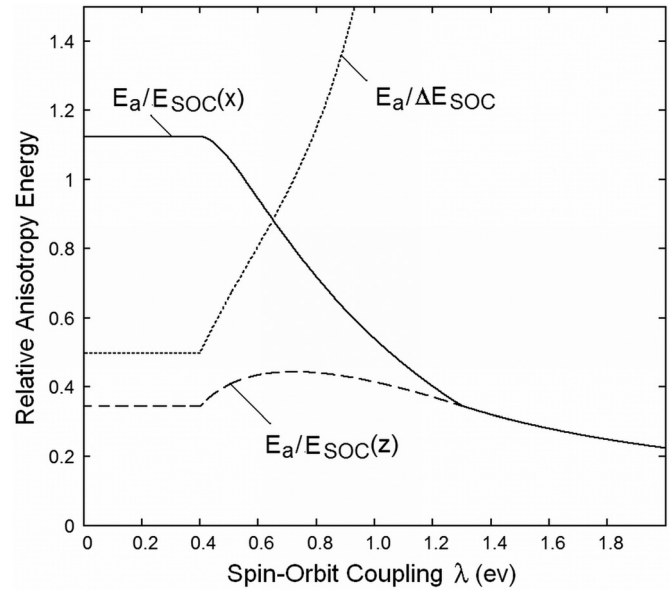


FIG. 2. Ratios of anisotropy energy  $E_a$  and spin-orbit coupling SOC for different directions. Reference 2 corresponds to a horizontal line with a ratio of one.

for ions such as  $\text{Gd}^{3+}$  and  $\text{Eu}^{2+}$ , so that  $L$  and  $S$  decouple.<sup>6</sup> However, the total  $L$  is only one consideration. Both quenched wave functions,  $\cos(m\phi)$  and  $\sin(m\phi)$ , and quenched wave functions

$$|\Psi_{\pm}(m)\rangle = |\Psi_{\cos}(m)\rangle \pm i|\Psi_{\sin}(m)\rangle, \quad (4)$$

yield  $L = 0$  for Gd (Fig. 3), but this does not mean that the orbital moments decouple from the spin moment. The Hund's-rules ground state of  $\text{Gd}^{3+}$  has seven  $\uparrow$  electrons, so that all available orbital states ( $m = -3, -2, -1, 0, 1, 2, 3$ ) are occupied and individual electrons cannot interact with the crystal field. This argument also applies to the possible partial quenching of one-electron 4f states caused by intra-atomic Coulomb interactions. If the orbital degrees of freedom ( $\pm m$ ) were decoupled from the spin, they would yield, for example, a contribution to the heat capacity. However, Eu spectroscopy does not support such low-lying excitations, and the lowest-lying excited term ( $S = 5/2$ ) has an excitation energy of about 4.0 eV,<sup>9</sup> as compared to typical CF energies of order 20 meV. Reference 6 contains the interesting argument that  $\text{Eu}^{2+}$  crystal fields tend to be larger than  $\text{Gd}^{3+}$  crystal fields. However, this does not invalidate the involvement of terms with  $S = 5/2$ , that is, terms with nonzero  $L$  and with net spin-orbit coupling.

A practically important feature in rare-earth permanent magnetism is the behavior of neodymium in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , whose anisotropy is reduced at the (001) surface. The

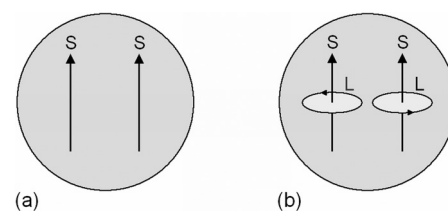


FIG. 3. One-electron character of a Gd 4f shell: (a) quenched and (b) unquenched. In both cases, the total  $L$  equals zero.

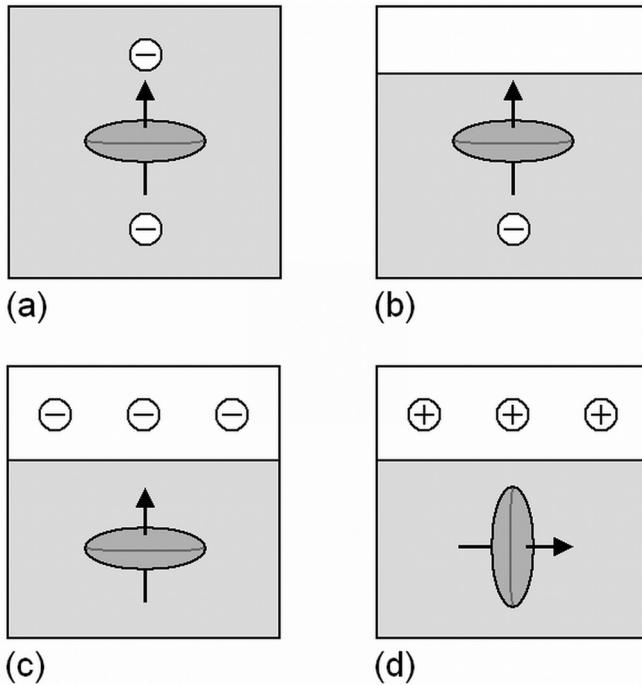


FIG. 4. Crystal-field of an oblate  $4f$  shell ( $\text{Nd}^{3+}$ ): (a) schematic neighborhood in bulk  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , (b) (001) surface in  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , and (c)-(d) multiferroic switching (schematic).

corresponding loss in coercivity is typically compensated by the use of dysprosium, but element-strategic consideration discourage the use of Dy, and current experimental and theoretical work is tackling this problem.<sup>11,12</sup> Figures 4(a)–4(b) shows how the reduced surface anisotropy can be explained by the CF interaction of the Nd atoms at the surface.  $\text{Nd}^{3+}$  has a negative Stevens coefficient (oblate  $4f$  shell), so that the orientation of the neighboring crystal-field charges in bulk  $\text{Nd}_2\text{Fe}_{14}\text{B}$  (positive crystal-field coefficient  $A_2^0$ ) yield easy-axis anisotropy. The effect is weakened at the (001) surface, because a negative crystal-field charge is removed from the top, Fig. 4(b). The resulting surface anisotropy then modifies the coercivity.<sup>13</sup>

Multiferroic materials have recently attracted much attention, for example, in spin electronics. Traditional systems exploit magnetoelastic strain, but the switching of the preferential magnetization direction can also be realized by crystal-field interactions.<sup>5</sup> Precursors of this mechanism are easy-axis changes in  $\text{Sm}_2\text{Fe}_{17}\text{N}_x$  due to interstitial nitrogen<sup>14,15</sup> and changes of the magnetization of Ni/Cu(001) thin films due to adsorbed hydrogen.<sup>16</sup> Figures 4(c)–4(d) illustrate the switching mechanism, which is triggered by the change in the electric dipole moment of the ferroelectric. In

lowest order, the magnetization direction is given by the electrostatic quadrupole interaction of the rare-earth  $4f$  shells with the crystal-field gradient.<sup>3,4</sup> Since the crystal-field source is the ferroelectric layer, the present interaction is proportional to electrostatic dipole-field gradient and therefore decreases as  $1/D^4$  with increasing distance  $D$  between  $4f$  ion and electrostatic dipole. By comparison, uniaxial anisotropies exhibit a  $1/D^3$  dependence.

#### IV. CONCLUSIONS

In summary, we have investigated the intriguing interplay between spin-orbit and crystal-field interactions in ferromagnets. Magnetization rotations in a crystal change both the spin-orbit and crystal-field energies, and only a part of the spin-orbit energy translates into anisotropy, namely about 50% for itinerant magnets and about 1% for rare-earth magnets. The latter mechanism allows an intuitive description of effects such as surface anisotropies in rare-earth permanent magnets and multiferroic switching of rare-earth spins due to electric fields. The case of  $4f^7$  ions is less transparent, although we suggest that the vanishing orbital moment is unrelated to the one-electron quenching of the iron-series orbital moments.

#### ACKNOWLEDGMENTS

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