

# Low-temperature magnetization anisotropy in ferromagnetic materials with quenched orbital angular momentum

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A phenomenological theory of magnetic anisotropy and low-temperature magnetization anisotropy, which arise because of anisotropic quenching of the orbital angular momentum, is constructed. A relation is found between the parameters of the theory and the electronic spectrum of the magnetic material. It is shown that for magnetic materials whose magnetic anisotropy is of this origin the spin-orbit interaction energy and the spin and orbital contributions to the magnetization and susceptibility of the paraprocess can be determined from the magnetization curves in the "easy" and "difficult" directions. The application of the theory to pure Co and to some intermetallic compounds of the type  $\text{RCo}_5$ , where  $R = \text{Y, La, Ce, and Th}$ , is examined. The values obtained for the spin-orbit coupling constant per cobalt atom are close to the values known for  $3d$ -metal ions and the values obtained for the spin and orbital magnetic moments agree with the values obtained by neutron diffractometry for Co and  $\text{YCo}_5$ . © 1995 American Institute of Physics.

## 1. INTRODUCTION

Magnetization anisotropy, i.e., the change in the magnitude of the magnetic moment  $M$  of a single crystal in the region of the paraprocess as the orientation of the magnetic moment changes from the easy into the difficult direction, was first predicted at the end of the 1950s by E. R. Callen and H. B. Callen.<sup>1</sup> They predicted that when the magnetic crystalline anisotropy energy  $E_a$  is taken into account, the occupancy of the excited states at finite temperatures  $T$  depends on the orientation of the magnetization with respect to the axes of the crystal, and this results in anisotropy of the magnitude of the moment. Since at low temperatures only the ground state is occupied, this effect should vanish in the limit  $T \rightarrow 0$ .

Magnetization anisotropy was first observed experimentally at the beginning of the 1970s by G. Aubert and P. Escudier<sup>2</sup> in Ni single crystals. The magnitude of the effect, i.e., the quantity  $\Delta M/M_{\parallel}$ , where  $\Delta M$  is the difference of the values of the moment  $M$  measured along the easy ( $M_{\parallel}$ ) and difficult directions, was equal to only  $\approx 10^{-4}$ .

In the mid-1970s, proceeding from arguments similar to Ref. 1, we studied the magnetization anisotropy in the compounds  $\text{RCo}_5$ , where  $R = \text{Tb, Dy, and Nd}$ , accompanying spin-flipping transitions.<sup>3</sup> A giant effect  $\Delta M/M_{\parallel} \approx 10^{-1}$  was first observed in these compounds. We viewed the observed effect as entirely only the anisotropy of the average value of the magnetic moment of the R ions, and the calculations performed in Ref. 3 also showed that the effect should vanish in the limit  $T \rightarrow 0$ . According to our later investigations,<sup>4</sup> however, the effect remains substantial even in the limit  $T \rightarrow 0$  and the quantity  $\Delta M/M_{\parallel}$  is determined completely by the change in the magnetization of the cobalt sublattice. Additional investigations, performed in Ref. 4 on  $\text{Y}(\text{Co, Ni})_5$  single crystals, where the Y ions are nonmagnetic, made it possible to obtain a quantitative estimate of the observed low-temperature magnetization anisotropy in the cobalt sublattice. According to the data presented in Ref. 4 this effect

amounts to 4% or  $0.3\mu_B$  per formula unit, where  $\mu_B$  is the Bohr magneton, and is virtually temperature-independent.

The results obtained in Ref. 4 were later completely confirmed by experiments on  $\text{YCo}_5$  (Ref. 5) and  $\text{LaCo}_5$  (Ref. 6) in strong magnetic fields. It has now been determined experimentally that the low-temperature magnetization anisotropy is observed in Co (Ref. 7) and in an entire series of highly anisotropic intermetallic compounds containing Fe, Co, or Ni.<sup>8,9</sup> We note that in  $\text{CeCo}_5$  the effect reaches  $\approx 12\%$ .<sup>10</sup> The magnetization curves obtained in these experiments have the characteristic form displayed in Fig. 1. In the region of the paraprocess they are practically linear, the magnetization and susceptibility along the easy axis being greater than along the difficult axis.

Besides the temperature dependence of the effect, there is one other very important factor, first noted in Ref. 4. The quantity  $\Delta M/M_{\parallel}$  is found to be of the same order of magnitude as the ratio of the anisotropy energy  $E_a$  to the exchange energy  $E_{ex}$ :  $\Delta M/M_{\parallel} \sim E_a/E_{ex}$ . It is clear that for magnetic ions, in which the intra-atomic Coulomb interaction is strong compared to the crystal field, so that the orbital angular momentum is a good quantum number, the ratio  $\Delta M/M_{\parallel}$  at low temperatures will be a second-order quantity relative to  $E_a/E_{ex}$  or  $E_a/E_{sl}$ , where  $E_{sl}$  is the spin-orbit interaction energy. For this reason, it seems to be entirely natural that the sublattice of  $3d$  atoms with strongly collectivized magnetic electrons plays the main role in the formation of the magnetic moment of these systems with strong low-temperature magnetization anisotropy.

Most theoretical investigations of the magnetization anisotropy in collectivized magnetic materials are performed numerically in parallel with calculations of the magnetic anisotropy constants. As an example, we cite one of the latest treatments, Ref. 11, which also contains a detailed bibliography concerning this question. The magnitude of the magnetization anisotropy obtained for pure  $3d$  metals agrees with the existing experimental data. The numerical character of the results obtained, however, makes it hard to analyze the

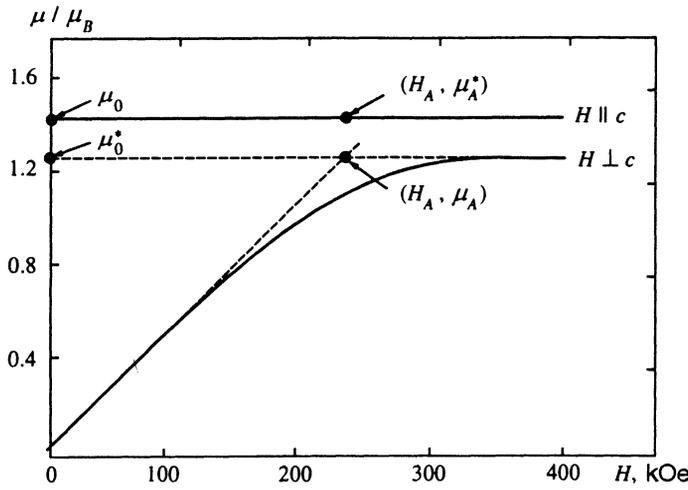


FIG. 1. Typical form of the magnetization curves of magnetically uniaxial single crystals of the intermetallic compounds  $\text{RCO}_5$  ( $\text{R}=\text{Y, La, Ce, Th}$ ). The curves were measured by applying a magnetic field parallel ( $\mathbf{H} \parallel \mathbf{c}$ ) and perpendicular ( $\mathbf{H} \perp \mathbf{c}$ ) to the easy magnetization axis. Solid lines—experimental data of Ref. 10 for  $\text{CeCo}_5$  at 4.2 K; dashed lines—continuation of the linear sections of the function  $\mu(H)$  for  $\mathbf{H} \perp \mathbf{c}$ ; points—characteristic parameters of the theory (see text);  $\mu$ —average value of the magnetic moment per cobalt atom.

experimental data, even for the rare systems where the corresponding calculations have been performed, to say nothing about most ferromagnetic materials, for which such calculations have not yet been performed. For this reason, our objective in the present work is to construct a simple phenomenological model that describes the magnetic anisotropy and magnetization anisotropy of systems with strongly quenched orbital angular momentum.

## 2. ENERGY

In this section we shall derive an expression for the energy  $E_a$  of a magnetic material with strongly quenched orbital angular momentum  $\mathbf{L}$ . To make the exposition as clear as possible, however, we start with the opposite limiting case of a magnetic material with a localized magnetic moment and well-defined orbital and spin angular momenta. An excellent example of such magnetic materials is systems based on  $4f$  elements. The magnetic anisotropy of such systems is related to the dependence of the energy of the “charge cloud” of  $4f$  electrons on the orientation of the cloud in the crystal field. We emphasize that the shape of the cloud does not change as the cloud rotates (the crystal field is weak and the orbital angular momentum  $L$  is a good quantum number), and the only source of the anisotropy energy is the Coulomb interaction energy of the cloud with its environment. For this reason, in the present case it is entirely natural to express the magnetic anisotropy energy as a function of the angles determining the direction of the orbital angular momentum  $\mathbf{L}$  or, in the case of a strong spin-orbit interaction, the total angular momentum  $\mathbf{J}$ . In the case of a magnetic material with uniaxial symmetry this expression has the standard form

$$E_a = K_1 \sin^2 \vartheta + K_2 \sin^4 \vartheta + \dots, \quad (1)$$

where  $\vartheta$  is the angle between the angular momentum (usually the total angular momentum) and the anisotropy axis and  $K_i$  are the anisotropy constants.

An entirely different situation obtains when the orbital angular momentum in the ground state is strongly quenched. Irrespective of whether the strong crystal field or the collectivized character of the magnetic electrons is responsible for the quenching,<sup>12</sup> as the spin moment rotates the charge cloud

of the magnetic electrons at a site does not rotate together with it, only the shape of the cloud changes slightly. The change in shape is due to the mixing of orbital states with the ground state as a result of the spin-orbit interaction and the interaction with the external magnetic field. On the one hand, this mixing results in partial unquenching of the orbital angular momentum and therefore a decrease of the spin-orbit and Zeeman interaction energies. On the other hand, the change in the shape of the “charge cloud” results in an increase of the interaction energy of the cloud with the crystal field. The latter contribution to the energy is anisotropic, since different excited states are mixed with the ground state, depending on the direction of the external field and the spin moment. Here the situation is identical to that in van Vleck paramagnets. In first order perturbation theory the ground state has the form

$$|gr'\rangle = |gr\rangle + \sum_{exc} \frac{\langle exc|\hat{V}|gr\rangle}{E_{gr} - E_{exc}} |exc\rangle, \quad (2)$$

where  $|gr\rangle$  and  $|exc\rangle$  are the unperturbed ground and excited states,  $E_{gr}$  and  $E_{exc}$  are their energies, and

$$\hat{V} = -\lambda(\hat{\mathbf{L}} \cdot \hat{\mathbf{S}}) - \mu_B \mathbf{H} \cdot (\hat{\mathbf{L}} + 2\hat{\mathbf{S}}). \quad (3)$$

In Eq. (3)  $\hat{\mathbf{L}}$  and  $\hat{\mathbf{S}}$  are the orbital and spin angular momentum operators at a lattice site,  $\lambda$  is the spin-orbit interaction parameter, and  $\mathbf{H}$  is the external magnetic field. From Eq. (2) we obtain the following expression for the anisotropy energy and the average value of the orbital angular momentum:

$$E_a = \sum_{exc} \frac{|\langle gr|\hat{V}|exc\rangle|^2}{E_{gr} - E_{exc}}, \quad (4)$$

$$\langle \mathbf{L} \rangle = \sum_{exc} \langle gr|\hat{\mathbf{L}}|exc\rangle \frac{\langle exc|\hat{V}|gr\rangle}{E_{gr} - E_{exc}} + \text{H.c.}$$

Hence one can see that, assuming the spin angular momentum operator to be a  $c$  number,<sup>1</sup> we obtain in the absence of an external field

$$E_a = -\frac{1}{2}\lambda S \langle \mathbf{L} |_{\mathbf{S}} \rangle, \quad (5)$$

where  $\langle \mathbf{L} |_{\mathbf{S}} \rangle$  is the average value of the projection of the orbital angular momentum on the direction of the spin angu-

lar momentum, i.e., in this approximation the magnetic anisotropy is related to the anisotropy of the orbital angular momentum. The easy and difficult directions are those in which the orbital angular momentum is least and most strongly quenched, respectively.

The simplest phenomenological expression for the energy  $E_a$  in the case of uniaxial anisotropy, taking into account explicitly both contributions to it (the anisotropic quenching of the orbital angular momentum and the spin-orbit interaction) evidently has the form

$$E_a = -\lambda(\mathbf{L}\mathbf{S}) + r_{\parallel}(L_{\parallel})^2 + r_{\perp}(L_{\perp})^2, \quad (6)$$

where  $\mathbf{L}$  and  $\mathbf{S}$  are the average values of the orbital and spin angular momenta at a lattice site,  $r$  is a quenching hardness coefficient, and  $\parallel$  and  $\perp$  denote projections parallel and perpendicular to the symmetry axis. Minimizing the energy (6) with respect to  $\mathbf{L}$  gives

$$L_{\tau} = \frac{\lambda S_{\tau}}{2r_{\tau}}, \quad \tau = \parallel, \perp, \quad (7)$$

and at the minimum the expression (5) is obtained for the value of  $E_a$ .

This concept of the magnetic anisotropy in magnetic materials with quenched orbital angular momentum is well known (see, for example, the review by Irkhin in Ref. 13). In application to magnetic ions with a localized moment, it has been used many times for performing calculations both in the theory of EPR (Ref. 14) and in other problems (Ref. 15). In the methods employed, specifically, in the effective spin-Hamiltonian method,<sup>15</sup> the contributions of the orbital and spin angular momenta to the magnetization are not separated and an anisotropic  $g$ -factor is used to describe the magnetization anisotropy. In the present work, however, an explicit separation of the contributions of the orbital and spin systems to the magnetization at the outset appears to us to be preferable for the following two reasons.

First, such a separation corresponds exactly to the physics of the situation and thus allows us to introduce the microscopic characteristics of the material into the theory as phenomenological parameters (the spin-orbit interaction parameter  $\lambda$  and the parameters  $r_{\parallel}$  and  $r_{\perp}$  which characterize the hardness of the quenching of the orbital angular momentum and are directly related to the electronic spectrum). This also simplifies dramatically the analysis of the experimental results, making it possible, specifically, to relate the parameter  $\lambda$  directly to the anisotropy field  $H_A$  and at least to estimate the upper and lower limits of the contributions of the orbital and spin systems separately to both the magnetization and the susceptibility of the paraprocess, using only magnetization curves in the easy and difficult directions (see Sec. 4).

Second, the quenching of the orbital angular momentum is most often associated with the collectivized character of the magnetic electrons, and the computational methods developed for magnetic ions are inapplicable in this case. Moreover, a systematic calculation of the magnetic anisotropy energy and the dependence of the magnitude of the magnetization on the magnetization direction in these sys-

tems is necessarily associated with the separation of the contributions of the orbital and spin angular momenta to the magnetization.

Such a calculation is most easily performed by the tight-binding method. In the spirit of the works by Brooks<sup>16</sup> and Bruno,<sup>17</sup> in the absence of spin-orbit interaction the Bloch functions of an electron in a crystal can be represented as linear combinations of atomic orbitals

$$|\nu\mathbf{k}\sigma\rangle = \sum_m a_{\nu m\sigma}(\mathbf{k})|m\mathbf{k}\sigma\rangle, \quad (8)$$

$$|m\mathbf{k}\sigma\rangle = N^{-1/2} \sum_{\mathbf{R}} e^{i\mathbf{k}\mathbf{R}}|m\mathbf{R}\sigma\rangle.$$

Here  $\nu$  is the band number,  $\mathbf{k}$  is the wave vector,  $\sigma$  is the projection of the spin on the quantization axis  $\xi$ , which we assume to be parallel to the direction of the average spin of the crystal  $\bar{S} = N\mathbf{s}$  ( $N$  is the number of lattice sites and  $\mathbf{s}$  is the average spin at a site). The states  $|m\mathbf{R}\sigma\rangle$  appearing in the expression (8) are a combination of atomic functions at the  $\mathbf{R}$ th site that possess the symmetry corresponding to the point symmetry of the lattice. Specifically, for the  $d$ -transition metals  $m$  runs over the standard values  $xy$ ,  $yz$ ,  $zx$ ,  $x^2 - y^2$ , and  $3z^2 - r^2$ .

The functions  $|\nu\mathbf{k}\alpha\rangle$  are the eigenfunctions of the Hamiltonian  $\hat{\mathcal{H}}_0$ , which includes the translational energy of the electron, the energy of interaction of the electron with the periodic potential and the interaction with the external and exchange fields

$$\hat{\mathcal{H}}_0|\nu\mathbf{k}\alpha\rangle = \mathcal{E}_{\nu\sigma}(\mathbf{k})|\nu\mathbf{k}\sigma\rangle. \quad (9)$$

The orbital angular momentum is the sum of the orbital angular momenta at each site

$$\hat{\mathcal{L}} = \sum_{m_1, m_2} \langle m_1 | \hat{\mathbf{l}} | m_2 \rangle \sum_{\mathbf{R}\sigma} |m_2\mathbf{R}\sigma\rangle \langle m_1\mathbf{R}\sigma|, \quad (10)$$

and the matrix elements do not depend on the coordinates of a site and the projection of the spin

$$\langle m_2 | \hat{\mathbf{l}} | m_1 \rangle \equiv \langle m_2\mathbf{R}\sigma | [(\mathbf{r} - \mathbf{R}) \times \hat{\mathbf{p}}] | m_1\mathbf{R}\sigma \rangle, \quad (11)$$

and vanish for  $m_1 = m_2$ . The average value  $\langle \hat{\mathcal{L}} \rangle$  is different from zero (in the absence of an external magnetic field) only when the spin-orbit interaction

$$\hat{V}_{sl} = \zeta \sum_{\substack{m_1, m_2 \\ \sigma_1, \sigma_2}} \langle m_2\sigma_2 | (\hat{\mathbf{l}}\hat{\mathbf{s}}) | m_1\sigma_1 \rangle \sum_{\mathbf{R}} |m_2\mathbf{R}\sigma_2\rangle \langle m_1\mathbf{R}\sigma_1|, \quad (12)$$

is taken into account as a perturbation. Using the creation and annihilation operators  $c_{\nu\mathbf{k}\sigma}^+$  and  $c_{\nu\mathbf{k}\sigma}$  for an electron in the states  $|\nu\mathbf{k}\sigma\rangle$  [Eq. (8)] these expressions can be put into the form

$$\hat{\mathcal{H}}_0 = \sum_{\nu\mathbf{k}\sigma} \mathcal{E}_{\nu\sigma}(\mathbf{k}) \hat{n}_{\nu\mathbf{k}\sigma},$$

$$\hat{V}_{sl} = \zeta \sum_{\substack{\nu_1 \nu_2 \mathbf{k} \\ m_1 m_2 \sigma_1 \sigma_2}} a_{\nu_2 m_2 \sigma_2}^*(\mathbf{k}) a_{\nu_1 m_1 \sigma_1}(\mathbf{k}) \\ \times \langle m_2 \sigma_2 | (\hat{\mathbf{l}} \hat{\mathbf{s}}) | m_1 \sigma_1 \rangle c_{\nu_2 \mathbf{k} \sigma}^+ c_{\nu_1 \mathbf{k} \sigma}, \quad (13)$$

$$\hat{\mathcal{L}} = \sum_{\substack{\nu_1 \nu_2 \mathbf{k} \\ m_1 m_2 \sigma}} a_{\nu_2 m_2 \sigma}^*(\mathbf{k}) a_{\nu_1 m_1 \sigma}(\mathbf{k}) \langle m_1 | \hat{\mathbf{l}} | m_1 \rangle c_{\nu_2 \mathbf{k} \sigma}^+ c_{\nu_1 \mathbf{k} \sigma}.$$

The energy  $E_a \equiv \langle V_{sl} \rangle$  and the average values of the operators of the projections of the orbital angular momenta  $\langle \mathcal{L}_\tau \rangle$ ,  $\tau = x, y$ , and  $z$ , can now be calculated by the standard methods of perturbation theory:

$$E_a = -(\zeta^2/2) \sum_{\nu \mathbf{k} \sigma \tau} \{ w_{\nu \tau}^{\sigma-\sigma}(\mathbf{k}) + [w_{\nu \tau}^{\sigma\sigma}(\mathbf{k}) - w_{\nu \tau}^{\sigma-\sigma}(\mathbf{k})] \alpha_\tau^2 \} n_{\nu \mathbf{k} \sigma}, \\ \langle \mathcal{L}_\tau \rangle = -\zeta^2 \sum_{\nu \mathbf{k} \sigma} w_{\nu \tau}^{\sigma\sigma}(\mathbf{k}) \sigma n_{\nu \mathbf{k} \sigma} \alpha_\tau. \quad (14)$$

Here  $\alpha_\tau$  are the direction cosines of the vector  $\vec{\mathcal{J}}$ ,  $n_{\nu \mathbf{k} \sigma} \equiv f(\mathcal{E}_{\nu \sigma}(\mathbf{k}))$  is the Fermi function,

$$w_{\nu \tau}^{\sigma\sigma}(\mathbf{k}) = \sum_{\nu'} \frac{Q_{\nu \sigma, \nu' \sigma'}^{\tau}(\mathbf{k})}{\mathcal{E}_{\nu' \sigma'}(\mathbf{k}) - \mathcal{E}_{\nu \sigma}(\mathbf{k})},$$

$$Q_{\nu \sigma, \nu' \sigma'}^{\tau}(\mathbf{k}) = \sum_{\substack{m_1 m_2 \\ m_1' m_2'}} a_{\nu m_2 \sigma}^*(\mathbf{k}) a_{\nu m_2' \sigma'}(\mathbf{k}) a_{\nu' m_1' \sigma'}^*(\mathbf{k}) a_{\nu' m_1 \sigma}(\mathbf{k}) \\ \times \langle m_2 | \hat{\mathbf{l}}_\tau | m_1 \rangle \langle m_1' | \hat{\mathbf{l}}_\tau | m_2' \rangle. \quad (15)$$

It is simple to show that

$$\sum_{\nu} w_{\nu \tau}^{\sigma\sigma}(\mathbf{k}) n_{\nu \mathbf{k} \sigma} = - \sum_{\nu} w_{\nu \tau}^{\sigma\sigma}(\mathbf{k}) (1 - n_{\nu \mathbf{k} \sigma}), \\ \sum_{\nu \sigma} w_{\nu \tau}^{\sigma-\sigma}(\mathbf{k}) n_{\nu \mathbf{k} \sigma} = - \sum_{\nu \sigma} w_{\nu \tau}^{\sigma-\sigma}(\mathbf{k}) (1 - n_{\nu \mathbf{k} \sigma}). \quad (16)$$

For this reason,  $\langle \hat{\mathcal{L}} \rangle$  and  $\vec{\mathcal{J}}$  are antiparallel for a less than half-filled shell and parallel for a more than half-filled shell.

In the simplest variant of Stoner's model, setting  $\mathcal{E}_{\nu \sigma}(\mathbf{k}) = \mathcal{E}_\nu(\mathbf{k}) - 0.5\sigma\Delta$ , we obtain from Eq. (14)

$$\langle \mathcal{L}_\tau \rangle = -\zeta \alpha_\tau \sum_{\nu \nu' \mathbf{k}} \frac{Q_{\nu \nu'}^{\tau}(\mathbf{k})}{\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})} (n_{\nu \mathbf{k} \uparrow} - n_{\nu \mathbf{k} \downarrow}), \quad (17)$$

so that the summation extends only over the layer of singly occupied states near the Fermi surface. Since

$$\sum_{\nu \mathbf{k}} (n_{\nu \mathbf{k} \uparrow} - n_{\nu \mathbf{k} \downarrow}) = 2\mathcal{S} \equiv 2NS, \quad (18)$$

the average value of the projection of the orbital angular momentum at a site on the  $\tau$ th axis can be written in the form

$$L_\tau \equiv N^{-1} \langle \mathcal{L}_\tau \rangle = 2\zeta S \alpha_\tau \bar{w}_\tau, \quad (19)$$

$$\bar{w}_\tau = -(2NS)^{-1} \sum_{\nu \nu' \mathbf{k}} \frac{Q_{\nu \nu'}^{\tau}(\mathbf{k})}{\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})} (n_{\nu \mathbf{k} \uparrow} - n_{\nu \mathbf{k} \downarrow}), \quad (20)$$

where  $\bar{w}_\tau$  is virtually independent of  $S$ .

The magnetic anisotropy energy (14) in this approximation separates into three terms:  $E_a = E_a^{(0)} + E_a^{(1)} + E_a^{(2)}$ . The first term

$$E_a^{(0)} = -\zeta^2 \sum_{\nu \nu' \mathbf{k} \tau} \frac{Q_{\nu \nu'}^{\tau}(\mathbf{k})}{\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})} n_{\nu \mathbf{k}}^p \quad (21)$$

remains finite and, when the ferromagnetic splitting vanishes ( $\Delta \rightarrow 0$ ,  $n_{\nu \mathbf{k} \sigma} \rightarrow n_{\nu \mathbf{k}}^p$ ), it is isotropic and does not depend on  $S$ . Therefore it is an additive constant and can be dropped. The second term

$$E_a^{(1)} = -\frac{\zeta^2}{2} \sum_{\nu' \mathbf{k}} \frac{Q_{\nu \sigma, \nu' \sigma'}^{\tau}(\mathbf{k})}{[\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})]^2 - \Delta^2} \left\{ [\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})] (n_{\nu \mathbf{k} \uparrow} + n_{\nu \mathbf{k} \downarrow} - 2n_{\nu \mathbf{k}}^p) + 2 \frac{\Delta^2}{\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})} n_{\nu \mathbf{k}}^p - \Delta (n_{\nu \mathbf{k} \uparrow} - n_{\nu \mathbf{k} \downarrow}) \right\} \quad (22)$$

is also isotropic, but its expansion in powers of the small quantity  $\Delta$  starts with terms proportional to  $\Delta^2$ . For this reason, it is proportional to  $S^2$  and can be written in the form

$$E_a^{(1)} = -2N\zeta^2 S^2 \bar{W}_0, \quad (23)$$

where  $\bar{W}_0$  is determined in an obvious way from Eq. (22), by analogy to Eq. (20), and also depends weakly on  $S$ . Finally,

$$E_a^{(2)} = -2N\zeta^2 S^2 \sum_{\tau} \bar{W}_\tau \alpha_\tau^2, \quad (24)$$

where

$$\bar{W}_\tau = \frac{\Delta}{N(2S)^2} \sum_{\nu \nu' \mathbf{k}} \frac{Q_{\nu \nu'}^{\tau}(\mathbf{k})}{[\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})]^2 - \Delta^2} \left\{ n_{\nu \mathbf{k} \uparrow} - n_{\nu \mathbf{k} \downarrow} - \frac{\Delta (n_{\nu \mathbf{k} \uparrow} + n_{\nu \mathbf{k} \downarrow})}{\mathcal{E}_{\nu'}(\mathbf{k}) - \mathcal{E}_\nu(\mathbf{k})} \right\}. \quad (25)$$

Therefore, dropping the  $S$ -independent constant (21), we can write the magnetic anisotropy energy per site in the form

$$E_a = -2\zeta^2 S^2 \left( \bar{W}_0 + \sum_{\tau=x,y,z} \bar{W}_\tau \alpha_\tau^2 \right). \quad (26)$$

It is obvious that even in approximations which are more complicated than Stoner's model, specifically, in spin-polarized calculations taking into account the dependence of the exchange splitting on the band number and the wave vector, the same expressions (19) and (26) will be obtained from Eq. (14). The  $S$ -dependence of the parameters  $\bar{w}_\tau$ ,  $\bar{W}_0$ , and  $\bar{W}_\tau$  appearing in them will remain weak even in this case, and to a first approximation they can be regarded as constants.<sup>2</sup>

For a magnetic material with uniaxial symmetry we obtain from Eqs. (19) and (26)

$$L_\tau = 2\zeta \bar{w}_\tau S \tau, \quad \tau = \parallel, \perp, \\ E_a = -2\zeta^2 [(\bar{W}_0 + \bar{W}_\parallel) S_\parallel^2 + (\bar{W}_0 + \bar{W}_\perp) S_\perp^2]. \quad (27)$$

Therefore

$$E_a = -\zeta \frac{\bar{W}_\perp - \bar{W}_\parallel}{\bar{w}_\perp - \bar{w}_\parallel} (\text{LS}) - 2\zeta^2 S^2 \left( \bar{W}_0 - \frac{\bar{W}_\perp \bar{w}_\parallel - \bar{W}_\parallel \bar{w}_\perp}{\bar{w}_\perp - \bar{w}_\parallel} \right). \quad (28)$$

The second term here, neglecting the anisotropy of the magnitude of the spin moment, is isotropic and can be regarded simply as a correction to the exchange-interaction energy in a spin system as a result of its interaction with the system of orbital angular momenta. The anisotropic first term in Eq. (28) and the expression for  $L_\tau$  in Eq. (27) are obtained immediately by minimizing Eq. (6), if we set

$$\lambda = 2\zeta \frac{\bar{W}_\perp - \bar{W}_\parallel}{\bar{w}_\perp - \bar{w}_\parallel}, \quad r_\tau = \frac{\bar{W}_\perp - \bar{W}_\parallel}{2\bar{w}_\tau(\bar{w}_\perp - \bar{w}_\parallel)}, \quad \tau = \parallel, \perp. \quad (29)$$

The last step in deriving a phenomenological expression for the energy of a magnetic metal with a uniaxial symmetry is the addition to Eq. (6) of an expansion of the Ginzburg-Landau energy of the spin system in powers of  $\mathbf{S}^3$  and the Zeeman interaction energy with the external field. Introducing the notation  $\boldsymbol{\mu}_s = 2\mathbf{S}$  and  $\boldsymbol{\mu}_l = \mathbf{L}$  for the spin and orbital magnetic moments of a site, which are expressed in Bohr magnetons, we obtain the final expression for the energy in the form

$$E = a\boldsymbol{\mu}_s^2 + b\boldsymbol{\mu}_s^4 - 0.5\lambda(\boldsymbol{\mu}_s\boldsymbol{\mu}_l) + r_\parallel(\boldsymbol{\mu}_l^\parallel)^2 + r_\perp(\boldsymbol{\mu}_l^\perp)^2 - \boldsymbol{\mu}_B\mathbf{H}(\boldsymbol{\mu}_s + \boldsymbol{\mu}_l). \quad (30)$$

### 3. MAGNETIZATION CURVES

We assume for definiteness that  $r_\parallel < r_\perp$ , so that the  $\hat{Z}$  axis is the easy-magnetization axis, and the external field  $\mathbf{h} = \mu_B\mathbf{H}$  makes an angle  $\theta$  with it. Since the anisotropy energy in the basal plane is neglected, the vectors  $\boldsymbol{\mu}_s$ ,  $\boldsymbol{\mu}_l$ , and  $\mathbf{H}$  and the  $\hat{Z}$  axis lie in the same plane, and minimizing the energy (30) with respect to the variables  $\mu_l^\parallel, \mu_l^\perp, \mu_s$ , and  $\vartheta$  (where  $\vartheta$  is the angle between  $\boldsymbol{\mu}_s$  and the  $\hat{Z}$  axis) leads to the following system of equations:

$$\mu_l^\parallel = \gamma_\parallel \mu_s \cos(\vartheta) + h \cos \theta / (2r_\parallel), \quad (31)$$

$$\mu_l^\perp = \gamma_\perp \mu_s \sin(\vartheta) + h \sin \theta / (2r_\perp),$$

$$\sin(\vartheta)\cos(\vartheta) = 2h \frac{(1+r_\perp)\sin(\theta)\cos(\vartheta) - (1+r_\parallel)\sin(\vartheta)\cos(\theta)}{\lambda\mu_s(r_\parallel - r_\perp)}.$$

It is convenient to write the last equation of this system in the form

$$8b\mu_s^3 - (4a + \lambda\gamma_\parallel)\mu_s = 2h(1 + \gamma_\parallel) \frac{\cos \theta}{\cos \vartheta} \quad (32a)$$

for  $\cos \vartheta \neq 0$  and in the form

$$8b\mu_s^3 - (4a + \lambda\gamma_\perp)\mu_s = 2h(1 + \gamma_\perp) \quad (32b)$$

for  $\cos \vartheta = 0$ . In Eqs. (31)–(32)

$$\gamma_\tau = \lambda / (4r_\tau), \quad \tau = \parallel, \perp. \quad (33)$$

We shall study the solutions of these equations in two simple special cases: when the external field is parallel and perpendicular to the easy axis.

#### a) $\theta=0$ , $\mathbf{H}$ parallel to the easy axis

It is obvious that in this case both the spin and orbital angular momenta are oriented parallel to the  $\hat{Z}$  axis irrespective of the magnitude of the field, i.e.,  $\mu_l^\perp = 0$  and  $\vartheta = 0$ . For  $H=0$  we obtain

$$\mu_s(H=0) \equiv \mu_s^0 = 0.5\sqrt{(4a + \lambda\gamma_\parallel)/(2b)},$$

$$\mu_l^\parallel(H=0) \equiv \mu_l^0 = \gamma_\parallel \mu_s^0. \quad (34)$$

Assuming the external field is weak compared to the exchange field, we can represent the spin moment in the form

$$\mu_s = \mu_s^0 + \delta\mu_s \quad (35)$$

and solve Eq. (32a) in an approximation linear in  $\delta\mu_s$ . As a result, the field dependence of the total moment  $\mu = \mu_s + \mu_l$  is linear

$$\mu(H) = \mu_0 + \chi_\parallel H / \mu_B, \quad \chi_\parallel = \mu_B^2 \left\{ \frac{1}{2r_\parallel} + \frac{(1 + \gamma_\parallel)^2}{8b(\mu_s^0)^2} \right\}. \quad (36)$$

#### b) $\theta=\pi/2$ , $\mathbf{H}$ parallel to the difficult axis

In this case we obtain immediately from the last equation of Eqs. (32)

$$\sin \vartheta = \frac{H}{H_A}, \quad H_A = \frac{\lambda\mu_s^0(\gamma_\parallel - \gamma_\perp)}{2\mu_B(1 + \gamma_\perp)}. \quad (37)$$

The reason that the factor  $\mu_s^0$  appears instead of  $\mu_s$  in the expression for  $H_A$  is clear from Eq. (32a): In a field oriented perpendicular (and only perpendicular!) to the easy axis the spin angular momentum does not change as it rotates:

$$\mu_s \equiv \mu_s^0 \text{ for } H \leq H_A. \quad (38)$$

As a result, the projections of the total angular momentum on the axis have the form

$$\mu_\parallel = \mu_s^0(1 + \gamma_\parallel)\cos \vartheta = \mu_s^0(1 + \gamma_\parallel)\sqrt{1 - (H/H_A)^2},$$

$$\mu_\perp = \mu_A H / H_A, \quad \mu_A = \mu_s^0(1 + \gamma_\perp)(1 + \gamma_\parallel), \quad (39)$$

where  $\mu_A$  is the value of the magnetic moment in a field equal to the anisotropy field  $H_A$ . In strong fields ( $H > H_A$ ) the paraprocess starts once again, and as with Eq. (36) we obtain

$$\mu(H > H_A) = \mu_A + \chi_\perp(H - H_A) / \mu_B,$$

$$\chi_\perp = \mu_B^2 \left\{ \frac{1}{2r_\perp} + 2 \frac{(1 + \gamma_\perp)^2}{16b(\mu_s^0)^2 + \lambda(\gamma_\parallel - \gamma_\perp)} \right\}. \quad (40)$$

It is simple to show that for parallel ordering of the spin and orbital angular momenta ( $\lambda > 0$ ) the inequalities

$$\chi_\parallel > \chi_\perp > 0, \quad \mu_0 > \mu_A \quad (41)$$

always hold.

### 4. DETERMINATION OF THE PHENOMENOLOGICAL PARAMETERS FROM THE EXPERIMENTAL MAGNETIZATION CURVES

The magnetization curves obtained in the preceding section are broken lines (see Fig. 1) with five main parameters

$\mu_0$ ,  $\mu_A$ ,  $H_A$ ,  $\chi_{\parallel}$ , and  $\chi_{\perp}$ . Instead of the last two parameters it is more convenient to use the parameters  $\mu_A^*$  (the magnetic moment in a field equal to  $H_A$  but parallel to the easy axis) and  $\mu_0^*$  (the value of the magnetic moment, directed parallel to the difficult axis, would have in a zero field)

$$\mu_A^* = \mu_0 + \chi_{\parallel} H_A / \mu_B, \quad \mu_0^* = \mu_A - \chi_{\perp} H_A / \mu_B. \quad (42)$$

It seems natural to find all five phenomenological parameters appearing in the energy (30) from these five values which can be quite easily determined from experiment. Indeed, using Eqs. (34), (37), and (39) we obtain

$$H_A = 0.5 \lambda \mu_B^{-1} (\mu_0 - \mu_A) \quad \text{or} \quad \lambda = 2 \frac{\mu_B H_A}{\mu_0 - \mu_A}. \quad (43)$$

We wrote out this important relation in two different forms for the following reasons. First, it is of interest to compare the expression for the anisotropy field (43) with the classical expression

$$H_{A1} = 2K_1 / I_S \quad (44)$$

( $I_S$  is the saturation magnetization), which follows immediately from Eq. (1) including the Zeeman interaction ( $I_S H$ ) and in the linear approximation ( $K_2 = 0$ ). In magnetic materials with a strong magnetization anisotropy, for example in  $\text{CeCo}_5$ ,<sup>10</sup> the parameter  $I_S$  becomes hard to determine and the expression (43) for  $H_A$  is physically more natural than the expression (44).

Second, it is obvious from Eq. (43) that the spin-orbit interaction parameter  $\lambda$  can be determined very simply in terms of the three quantities  $\mu_0$ ,  $\mu_A$ , and  $H_A$ , which can be found from simple magnetometric experiments. We emphasize that for magnetic metals it is very hard to determine  $\lambda$  by other methods.

The expressions for the quenching hardnesses  $r_{\parallel}$  and  $r_{\perp}$  for known  $\lambda$  can be easily obtained from Eqs. (34) and (39):

$$\begin{aligned} r_{\parallel} &= \lambda \mu_s^0 / (4\mu_l^0) \equiv \lambda \mu_s^0 / [4(\mu_0 - \mu_s^0)], \\ r_{\perp} &= \lambda (\mu_s^0 + \mu_l^0 - \mu_l(H_A)) / [4\mu_l(H_A)] \\ &= \lambda (\mu_0 - \mu_A + \mu_s^0) / [4(\mu_A - \mu_s^0)]. \end{aligned} \quad (45)$$

The only unknown parameter appearing in Eq. (45) is  $\mu_s^0$ , the magnitude of the spin moment in the absence of an external field. In accordance with Eq. (34), it is determined in terms of the Ginzburg-Landau parameters  $a$  and  $b$ , which we can try to determine using the two experimental values  $\chi_{\parallel}$  and  $\chi_{\perp}$  (or, which is the same thing,  $\mu_0$  and  $\mu_A^*$ ) which have not yet been used. It is found, however, that in this model we have the identity

$$\frac{1}{\mu_0 - \mu_0^*} - \frac{1}{\mu_A^* - \mu_A} = \frac{1}{\mu_0}. \quad (46)$$

On the other hand, this identity makes it possible to find the smaller, and therefore harder to determine experimentally, susceptibility  $\chi_{\perp}$  in terms of  $\chi_{\parallel}$ . On the other hand, it decreases the number of independent parameters obtained from experiment from 5 to 4, which eliminates the possibility of determining  $\mu_s^0$  uniquely on the basis of data on the magne-

tization curves only (Fig. 1).<sup>4</sup> Nonetheless, on the basis of these data it is simple to determine the maximum and minimum values of  $\mu_s^0$ ; the true value lies between them.

Indeed, it is obvious that  $\mu_s^0 \leq \mu_A = \mu_s^0 + \mu_l^{\perp}(H_A)$ . The equality in this expression holds only if the orbital moment in the difficult direction is completely quenched, i.e.,  $r_{\perp} \rightarrow \infty$ . For this reason, taking into account Eq. (34),

$$\begin{aligned} (\mu_s^0)_{\max} &= \mu_A, \quad (\mu_l^0)_{\min} = \mu_0 - \mu_A, \\ (r_{\parallel})_{\max} &= \lambda \mu_A / [4(\mu_0 - \mu_A)]. \end{aligned} \quad (47)$$

The limit of the possible values of these parameters on the other side can be easily determined from the experimental value of the susceptibility  $\chi_{\parallel}^{\text{exp}}$  in the easy direction. As is clear from Eq. (36), there are two positive contributions of  $\chi_{\parallel}$ : The direct response of the orbital system to the external field and the response of the spin system (first and second terms, respectively), and they are both positive for  $\lambda > 0$ . For this reason, the minimum possible value of  $r_{\parallel}$  is determined from the condition  $(r_{\parallel})_{\min} = \mu_B^2 / 2\chi_{\parallel}^{\text{exp}}$ . Hence

$$\begin{aligned} (r_{\parallel})_{\min} &= \lambda (\mu_0 - \mu_A) / [4(\mu_A^* - \mu_0)], \\ (\mu_s^0)_{\min} &= \mu_0 (\mu_0 - \mu_A) / (\mu_A^* - \mu_0), \\ (\mu_l^0)_{\max} &= \mu_0 (\mu_A^* - \mu_0) / (\mu_A^* - \mu_A). \end{aligned} \quad (48)$$

The equations (47) and (48) make it possible to determine also the limits of the true value of the magnetomechanical factor  $g'$  in a zero field

$$g' = (\mu_l^0 + \mu_s^0) / (\mu_l^0 + 0.5\mu_s^0) \approx 2(1 - \varepsilon), \quad \varepsilon = \mu_l^0 / \mu_s^0. \quad (49)$$

For  $\varepsilon$  we have the inequality

$$(\mu_0 - \mu_A) / \mu_A \leq \varepsilon \leq (\mu_A^* - \mu_0) / (\mu_0 - \mu_A). \quad (50)$$

Finally, the upper limit of the possible values of the contribution of the spin system to the susceptibility in the easy direction is easily determined from Eqs. (47) and (36):

$$\begin{aligned} (\chi_{\parallel}^{(s)} / \chi_{\parallel})_{\max} &= (\chi_{\parallel}^{\text{exp}} - \mu_B^2 / [2(r_{\parallel})_{\max}]) / \chi_{\parallel}^{\text{exp}} \\ &= 1 - (\mu_0 - \mu_A)^2 / [\mu_A (\mu_A^* - \mu_0)]. \end{aligned} \quad (51)$$

Naturally, the lower limit of this ratio is zero, and it corresponds to the case (48).

## 5. ANALYSIS OF THE EXPERIMENTAL DATA

Quite accurate measurements of the magnetization curves for magnetically-uniaxial single crystals at low temperatures are available for pure Co (Ref. 7) and the intermetallic compounds with a crystal structure of the  $\text{CaCu}_5$  type:  $\text{YCo}_5$ ,<sup>5</sup>  $\text{LaCo}_5$ ,<sup>6</sup>  $\text{CeCo}_5$ ,<sup>10</sup> and  $\text{ThCo}_5$ ,<sup>18</sup> as well as for a number of other highly anisotropic intermetallic compounds:<sup>9</sup>  $\text{Y}_2\text{Fe}_{17}$ ,  $\text{YFe}_3$ ,  $\text{YNi}_3$ ,  $\text{YCo}_3$ ,  $\text{Pr}_2\text{Co}_{17}$ , and  $\text{Nd}_2\text{Co}_{17}$ .

The typical form of the magnetization curves of these single crystals is shown in Fig. 1. In the rotation and paraprocess regions they can be approximated well by straight lines. This makes it possible to determine the values of  $\mu_0$ ,  $\mu_A$ ,  $H_A$ , and  $\chi_{\parallel}$ , and in some cases also  $\chi_{\perp}$ . The experimental data for compounds of the type  $\text{RCO}_5$  and Co are

TABLE I. Experimental values of the magnetic moments  $\mu_0$ ,  $\mu_A$ , and the difference  $\Delta\mu = \mu_0 - \mu_A$ , the susceptibility  $\chi_{\parallel}$  of the paraprocess ("longitudinal" susceptibility),  $\chi_{\perp}$  ("transverse" susceptibility) per cobalt atom, and the anisotropy field  $h_A = \mu_B H_A$  according to measurements of the magnetization curves at  $T = 4.2$  K for Co,<sup>7</sup> YCo<sub>5</sub>,<sup>5</sup> LaCo<sub>5</sub>,<sup>6</sup> CeCo<sub>5</sub>,<sup>10</sup> and ThCo<sub>5</sub> (Ref. 18) single crystals.

	Co [7]	YCo <sub>5</sub> [5]	LaCo <sub>5</sub> [6]	CeCo <sub>5</sub> [10]	ThCo <sub>5</sub> * [18]	ThCo <sub>5</sub> ** [18]
$\mu_0/\mu_B$	1.7291	1.666	1.693	1.424	0.920	1.446
$\mu_A/\mu_B$	1.7211	1.604	1.633	1.253	0.800	1.368
$\Delta\mu/\mu_B$	0.0080	0.062	0.060	0.171	0.120	0.078
$\chi_{\parallel}/\mu_B^2, 10^{10} \text{ erg}^{-1}$	406	550	1040	1450	6400	910
$\chi_{\perp}/\mu_B^2, 10^{10} \text{ erg}^{-1}$	313	—	—	1200	2200	—
$h_A, 10^{-16} \text{ erg}$	0.98	14.3	15.6	22.2	8.7	14.6

\*The exact composition corresponds to the formula Th<sub>0.965</sub>Co<sub>5.07</sub>. \*\*The exact composition corresponds to the formula Th<sub>0.95</sub>Co<sub>5.10</sub>. The quantity  $h_A$  for cobalt is indicated as an average over the data for the constants  $K_1$  (Refs. 24 and 25) and the magnetization (Ref. 7); the relative error in the values of  $\mu_0$ ,  $\mu_A$ ,  $\Delta\mu$ ,  $\chi_{\parallel}$ ,  $\chi_{\perp}$ , and  $h_A$  is (in percent) approximately in the case of cobalt 0.05, 0.05, 20, 5, 5, and 10; for the series RCo<sub>5</sub> the errors are 0.5, 0.5, 2, 10–30, 10–20, 2–5, respectively.

presented in Table I. All characteristics besides  $H_A$  are presented per Co atom. In this connection, two remarks are in order.

First, we assume that in the compound studied RCo<sub>5</sub> (R=Y, La, Ce, Th) the R atoms are completely nonmagnetic. This assumption is completely reasonable and generally accepted in the case R=Y, La, and Th.<sup>4,5,8,18–21</sup> At the same time the cerium atoms may contribute to both the anisotropy and magnetization of the compound CeCo<sub>5</sub>.<sup>10</sup> The question of the magnitude of each of these contributions remains open, however, and we shall assume here that they are negligibly small compared to the corresponding contributions from the cobalt sublattice.

Second, it is well known that the Co atoms in the RCo<sub>5</sub> crystal lattice occupy two nonequivalent crystallographic positions 2c and 3g. The energy  $E_a$  as well as the values of the magnetic moments of the atoms and their anisotropy in these positions can be different. If this is indeed so, the spin-orbit interaction constant  $\lambda$  must be found and the contributions of the spin and orbital subsystems to the magnetization must be separated separately for the 2c and 3g sublattices. This, however, requires that the contributions of these sublattices to the magnetization curves first be separated experimentally. This problem can be solved by using local methods for measuring the magnetic moments of atoms together with the standard methods of magnetization measurements. This problem has not been solved completely for any compound. At the same time, for  $H=0$  the magnetic moment of the Co atom in the 2c and 3g positions for YCo<sub>5</sub> is the same, according to Refs. 19 and 20, and according to Ref. 18 it does not differ by more than 3% and the relative contribution of the orbital component of the magnetic moment in these positions is 0.26 (5) (2c) and 0.16 (4) (3g). These data suggest that the values  $\lambda$ ,  $\mu_S$ , and  $\mu_I$  presented below (Table II), which must be regarded as averages, will not differ too much from the corresponding characteristics for cobalt atoms occupying different crystallographic positions.

Since the values of  $\mu_0$  in Table I are scattered over a

quite wide range, it makes sense to point out that the R atoms in the compounds RCo<sub>5</sub> have valence 3 (Y, La), 4 (Th), and variable valence (Ce). For compounds in which the ions R have the same valence (Y, La) the values of both  $\mu_0$  and the other magnetic characteristics are virtually identical. Apparently, the change in the valence of the R ion produces a large change in the electronic states and, correspondingly, a change in the magnetic characteristics. Small changes in the cobalt concentration in the region of homogeneity of the compounds RCo<sub>5</sub> with trivalent R ions do not lead to any large changes in the magnetic moment of the Co atom.<sup>20,21</sup> On the other hand, small changes in the cobalt concentration in the region of homogeneity of the compound ThCo<sub>5</sub> can lead to marked changes in its magnetic properties and even to the appearance of metamagnetic transitions<sup>18</sup> in a sufficiently strong magnetic field. The data presented in Table I for ThCo<sub>5</sub> pertain to a low-field state, i.e., to a state before a metamagnetic transition occurs.

We estimated the spin-orbit interaction constant  $\lambda$  and the contributions of the spin and orbital systems to the magnetization and susceptibility of the paraprocess from the data given in Table II for Co and the compounds considered here. All calculations were performed in accordance with the preceding section. The results are summarized in Table II. It is evident that the values obtained for  $\lambda$  agree completely with the published data on the spin-orbit interaction constant of 3d elements.<sup>22</sup>

According to neutron-diffractometry data, which were generalized in Ref. 18, for Co and YCo<sub>5</sub> the ratio  $\mu_I/\mu_0$  is equal to 0.07 and 0.20, respectively. These values are close to the values that we obtained assuming the complete absence of a paraprocess in the spin subsystem. These results as well as the maximum values of the spin susceptibility, presented in Table II, and the proposed model itself force us to reconsider Wohlfarth's criterion for separating "site" and "itinerant" magnetic materials.<sup>23</sup>

TABLE II. The spin-orbit interaction constant  $\lambda$ , the spin magnetic moment  $\mu_s$ , and the orbital magnetic moment  $\mu_l$ , the relative orbital contribution  $\mu_l^\parallel/\mu_0$  to the magnetization, and the maximum relative contribution of the spin susceptibility  $\chi_\parallel^{(S)}$  to the susceptibility  $\chi_\parallel^{exp}$  of the paraprocess for pure cobalt and a series of intermetallics  $RCO_5$  per cobalt atom, calculated from experimental data, Table I, in accordance with the present theory in the approximation of a predominant orbital contribution to the paraprocess [data in the rows (a)] or in the approximation of zero orbital contribution to the magnetization and in the paraprocess with the magnetization in the "transverse" direction [data in rows (b)].

		Co	YCo <sub>5</sub>	LaCo <sub>5</sub>	CeCo <sub>5</sub>	ThCo <sub>5</sub> *	ThCo <sub>5</sub> **
$\lambda, 10^{-16}$ erg		240	460	520	260	140	380
$\frac{\mu_s}{\mu_B}, (H=0)$	(a)	1.647	1.48	1.33	1.20	0.63	1.24
	(b)	1.721	1.60	1.63	1.25	0.80	1.37
$\frac{\mu_l^\parallel}{\mu_B}, (H=0)$	(a)	0.082	0.19	0.36	0.23	0.29	0.21
	(b)	0.008	0.066	0.063	0.17	0.12	0.08
$\frac{\mu_l^\parallel}{\mu_0}, (H=0)$	(a)	0.05	0.11	0.21	0.16	0.32	0.15
	(b)	0.005	0.04	0.04	0.12	0.13	0.05
$\chi_\parallel^{(S)}/\chi_\parallel^{exp}$	(b)	0.90	0.69	0.86	0.28	0.67	0.67

\*The exact composition corresponds to the formula  $Th_{0.965}Co_{5.07}$ . \*\*The exact composition corresponds to the formula  $Th_{0.95}Co_{5.10}$ .

## 6. CONCLUSIONS

We have shown that the source of the magnetic anisotropy energy in systems with strongly quenched orbital angular momentum is the different degree of quenching of the orbital angular momentum in different crystallographic directions. The coefficients of the direct proportionality between the change in the spin-orbit interaction energy accompanying a reorientation of the magnetization and the magnitude of the unquenched orbital angular momentum were determined for a system of collectivized electrons in the tight-binding approximation. A simple phenomenological theory was constructed that describes both the magnetic crystallographic anisotropy and the anisotropy of the magnitude of the magnetization. A relation was derived between the magnitude of the change in the magnetization on reorientation of the magnetization in a single crystal with an anisotropy field. This relation makes it possible to determine directly the spin-orbit interaction constant  $\lambda$  from measurements of the magnetization curves for single crystals. Using such experiments and the present theory, it is also possible to separate the contributions of the orbital  $\mu_l$  and spin  $\mu_s$  moments to the magnetization and susceptibility of the paraprocess in a ferromagnetic material. The possibility of obtaining these important characteristics of a ferromagnetic material was demonstrated for the case of pure cobalt and intermetallic  $RCO_5$  compounds.

We emphasize once again that the magnetization anisotropy studied in this paper occurs in a wide temperature range, specifically, in the limit  $T \rightarrow 0$  and for  $T > T_c$ , where  $T_c$  is the Curie temperature. This effect should occur in all magnetically ordered materials, but it should be most pronounced in magnetic materials with strongly quenched orbital angular momentum and high magnetic anisotropy. Nonetheless, investigations of this effect in magnetic metals with a low magnetic anisotropy energy are also apparently of interest from the standpoint of determining the parameter

$\lambda$ , which is hard to measure by other methods. In this case quite accurate data on the change of the magnetization accompanying a change in its orientation in a crystal can be obtained by measuring the signal produced in a circuit by rotating the sample.

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<sup>1</sup>This will be a good approximation, for example, in the presence of a very strong exchange interaction.

<sup>2</sup>This dependence together with the anisotropy of  $S$  contribute to the higher-order magnetic-anisotropy constants.

<sup>3</sup>The second term in Eq. (28) will be absorbed by the first term of this expansion in  $\mu_s^2$ .

<sup>4</sup>It is readily shown that the required additional parameter can be obtained, for example, by measuring the field-dependence of the torque acting on the sample in a field.

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