Journal of Alloys and Compounds, 191 (1993) 67–70
JALCOM 463

Temperature dependence of single-ion anisotropy and magnetostriction coefficients of rare earth ferromagnets in terms of quantum theory

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(Received July 29, 1992)

Abstract

The quantum methods for calculation of the temperature dependences of single-ion magnetocrystalline anisotropy and magnetostriction coefficients in a wide temperature range are under discussion in this paper. The experimental data on anisotropy of Tb single crystal and intermetallic compounds of the R_2Co_{17} type, where R ≡ Pr, Nd, Sm, are analysed on the basis of quantum theory. A good agreement of the theory with experiment is achieved.

1. Introduction

For a long period of time, in theoretical papers on the calculation of the temperature dependences of ferromagnet single-ion magnetocrystalline anisotropy (MCA) and magnetostriction coefficients ($k_i(T)$ and $\lambda_i(T)$), the spontaneous magnetization M_S was used as the theory parameter [1–3]. Therefore, the interpretation of the MCA and magnetostriction data of different ferromagnets could be made only after the experimental determination of $M_S(T)$.

Naturally, the next step in the improvement of MCA theory was the inclusion of the $M_S(T)$ dependence in theoretical calculations and the explicit determination of $k_i(T)$ and λ_i(T) functions. As was shown by the first author of this paper, this progress can be made only on the basis of the quantum microscopical approach [4]. In this paper, we discuss the main concepts of such an approach and give the formulae for $M_S(T)$, $k_i(T)$ and $\lambda_i(T)$, which are used to interpret experimental data on Tb and R_2Co_{17} (R ≡ Pr, Nd, Sm) single crystals.

2. Main concepts of quantum theory

A simple quantum theory for $k_i(T)$ and $\lambda_i(T)$ temperature dependences over a wide temperature range can be developed using Bogolubov's theorem for the model system's free energy [5], where the main Hamiltonian H_0 characterizes the exchange energy and the perturbation Hamiltonian H' gives the single-ion anisotropy energy. In the rare earth metals, the ratio between the energies of anisotropy E_A and exchange E_{ex} is not small and adopts the values $E_A/E_{ex}=0.1$–0.4. However, the theorem is valid when there is no restriction of the small value of the perturbation Hamiltonian. In this instance, it estimates also the free energy, though sometimes rather approximately.

$$\langle H'\rangle_{H_0+H'} \leqslant F(H_0+H')-F(H_0) \leqslant \langle H'\rangle_{H_0} \quad (1)$$

The calculation of relative values of ferromagnet MCA and magnetostriction coefficients of different orders in the quantum theory is reduced to the calculation of the thermodynamical average values for Stevens' operators O_l^0 (J_z):

$$k_l^m(T)/k_l^m(0)=\lambda_l^m(T)/\lambda_l^m(0)=\langle O_l^0\rangle_T/\langle O_l^0\rangle_0=L_l^J(T) \quad (2)$$

where J is the quantum number of total angular momentum [1–4]. The relative magnetization is calculated also using these operators, such that

$$\sigma_J(T)=\langle O_1^0\rangle_T/\langle O_1^0\rangle_0=L_1^J(T) \quad (3)$$

A guarantee of the validity of the theory can be given when averaging eqns. (1)–(3) involves the total Hamiltonian. Such calculations usually are made with the help of the Green's function method [4]. The search for the thermodynamic functions is made using the following equations:

$$\langle O_l^0\rangle_T=\sum_{p=-J}^{J} O_l^0(p)A_{2p}$$

$$\sigma_J=\frac{1}{J}\sum_{p=-J}^{J} pA_{2p} \quad (4)$$

0925-8388/93/$6.00

The coefficients A_{2p} are formed by magnon filling functions $\phi_p(T)$. They depend on the angular coordinates of the magnetization vector M and all the interactions, which determine the multimode magnon spectrum of rare earth ferromagnets. Here, we will analyse only two simple cases based on the consideration of one spin-wave branch and $\phi(T)$ function. When the energy of MCA is large, the single-mode approach is valid only in the low temperature range ($T<0.3T_c$). At $J>l/2$, we have [6]

$$\sigma_J = 1-\phi/J \tag{5}$$

$$L_2^J = 1-3\phi/J+3\phi^2/J(J-\tfrac{1}{2}) \tag{6}$$

$$L_4^J = 1-10\phi/J+45\phi^2/J(J-\tfrac{1}{2})-105\phi^3/J(J-\tfrac{1}{2})(J-1) +105\phi^4/J(J-\tfrac{1}{2})(J-1)(J-\tfrac{3}{2}) \tag{7}$$

$$L_6^J = 1-21\phi/J+210\phi^2/J(J-\tfrac{1}{2})-1260\phi^3/J(J-\tfrac{1}{2})(J-1) +4725\phi^4/J(J-\tfrac{1}{2})(J-1)(J-\tfrac{3}{2})-10\,395\phi^5/ J(J-\tfrac{1}{2})\ldots(J-2)+10\,395\phi^6/J(J-\tfrac{1}{2})\ldots(J-\tfrac{5}{2}) \tag{8}$$

The physical sense of ϕ follows from eqn. (5) and its temperature dependence has to be calculated. In particular, in the case of 'easy plane' ferromagnets, $\phi(T)$ is given by the expression [4]

$$\phi(T)=\left(\frac{2}{\pi}\right)^{1/2}\frac{E_g^{\,2}}{(4\pi AJ)^{3/2}[D_{2J-1}-JA(0)]^{1/2}}\sum_{n=1}^{\infty}\frac{\kappa T}{nE_g}K_1\frac{nE_g}{\kappa T} \tag{9}$$

where E_g is the energy gap of the low cooperative mode and $K_1(x)$ is the Bessel function. All the other indexes are the same as in ref. 4. Formulae similar to eqns. (5)–(8) can be derived for a wider temperature region. The key approach for this is a small value of the MCA energy with respect to the exchange energy. Again, all the ferromagnetic specifications are reflected by the unique function ϕ, which is different for each particular case [7].

$$\sigma_J = 1-\frac{\phi}{J}\left\{1-\frac{(2J+1)\phi^{2J}}{(1+\phi)^{2J+1}-\phi^{2J+1}}\right\} \tag{10}$$

$$L_2^J = \{2J(J+1)-3J(1+2\phi)\sigma_J\}/P_J(1) \tag{11}$$

$$L_4^J = \{8J^2(J+1)^2-16J(J+1)-5J[8J(J+1)-9] \times(1+2\phi)\sigma_J+70J(J+1)(1+2\phi)^2 -105J(1+2\phi)^3\sigma_J\}/P_J(3) \tag{12}$$

$$L_6^J = \{16J^3(J+1)^3-128J^2(J+1)^2+192J(J+1) -J[168J^2(J+1)^2-966J(J+1)+1575/2] \times(1+2\phi)\sigma_J+[756J^2(J+1)^2-2457J(J+1)] \times(1+2\phi)^2-5J[504J(J+1)-945](1+2\phi)^3\sigma_J +3465J(J+1)(1+2\phi)^4-\frac{10\,395}{2} \times J(1+2\phi)^5\sigma_J\}/P_J(5) \tag{13}$$

$$P_J(1)=J(2J-1)$$

$$P_J(3)=J(2J-1)(2J-2)(2J-3) \tag{14}$$

$$P_J(5)=\frac{1}{2}J(2J-1)\ldots(2J-5)$$

For the low temperature region, eqns. (5)–(8) immediately follow from eqns. (10)–(14) ($\phi^{2J}\ll 1$). At the same time, for the high temperatures they transform to the relatively simple relation [6]

$$L_l^J(T)=\frac{(3\sigma_J)^l(2J+l+1)!}{(2l+1)!!(2J+2)^l(2J+1)!} \tag{15}$$

If ϕ is considered as a theory parameter, the $L_l^J(T)$ functions can be easily expressed from the relative magnetization in the 0 K – T_c temperature region. The quantum functions $L_l^J(\sigma_J)$ differ from the classical functions only for small J numbers.

It should be noted, that the new theory parameter X can be introduced in the more convenient form

$$(1+\phi)/\phi=\exp(X/J)\quad(1+2\phi)=cth\frac{X}{2J}$$

$$\sigma_J=B_J(X) \tag{16}$$

where $B_J(X)$ is the well known Brillouin function. This parameter has a very distinct physical interpretation. Thus, supposing that the Fourier image of the exchange integral, which depends on the wave vector, is equal to zero, we obtain the Weiss anisotropic molecular field approach, where X is the ratio between the Zeeman energy and the thermal energy ($X=gJ\mu H/\kappa T$).

3. Comparison of theory with experiment

Figure 1 shows calculations based on eqns. (5), (6) and (9) of the temperature dependences of the relative magnetization $\sigma_J(T)$ [4] and the uniaxial MCA coefficient $k_2^0(T)$ for Tb metal as a function of $\tau=T/T_c$, where $T_c=219$ K [8]. Experimental data, as well as the theoretical data, calculated from the previous classical theories for the temperature range $0<T<0.4T_c$ [9, 10] are also given in this figure. It is seen that, when σ_J acts as a calculated physical constant, neither the Weiss molecular field approach (curve 1) nor Akulov's classical formula (curve 2) can give a good description of the experimental data. Only quantum theory is able to provide a good description (curve 3). The high efficiency of quantum theory is also illustrated by very good fitting of the experimental data on $k_2^0(T)$, $k_4^0(T)$ and $\lambda_2^{\alpha,\,2}(T)$

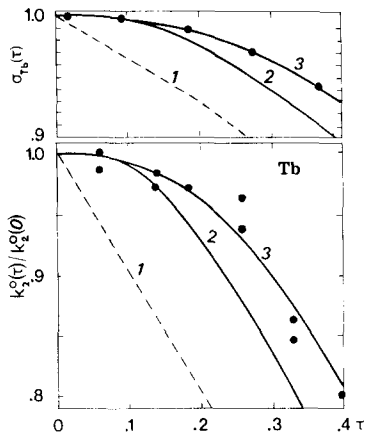

Fig. 1. Relative temperature dependences of the spontaneous magnetization and second-order magnetic anisotropy coefficient k_2^0 for Tb single crystal. The points are the experimental data [9, 10] and the curves are the theoretical approximation on the basis of (1) Akulov classical approach [1], (2) Weiss molecular field method [2] and (3) Green's function method [4].

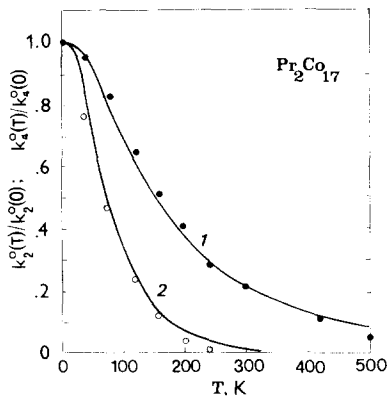

Fig. 2. Relative temperature dependences of second- (curve 1) and fourth- (curve 2) order anisotropy coefficients for Pr sublattice in Pr_2Co_{17} intermetallic compound. The points are the experimental data [11] and the lines are the calculations on the basis of eqns. (11), (12) and (17).

for R_2Co_{17} intermetallic compounds (R ≡ Pr, Nd, Sm), taken from our previous papers [11, 12] and plotted here in Figs. 2 and 3. As is known, these intermetallics are characterized by a ferromagnetic-type magnetic order between the R and Co sublattices. The main contribution to their magnetization originates from the Co sublattice, whereas the anisotropy type and the value of the anisotropy coefficients arise mainly from the R sublattice. Due to the small value of R sublattice magnetization, especially when R ≡ Sm, the experimental determination of the $M_S(T)$ dependence is hampered, so it creates some problems for the interpretation of these data in the ordinary way (as $\sigma_R(T)$ function). However, eqns. (10)–(16) allow us to calculate $k_2^0(T)$, $k_4^0(T)$ and $\lambda_2^{\alpha,2}(T)$ dependences for these compounds without the knowledge of the $M_R(T)$ data. Because of the small value of the R sublattice MCA

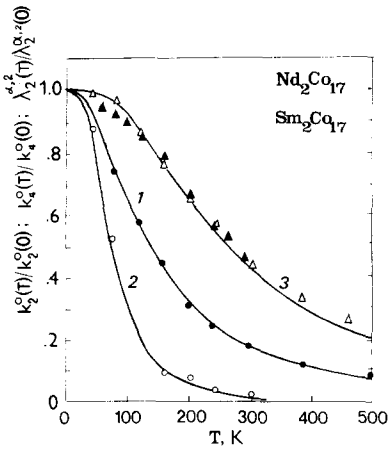

Fig. 3. Relative temperature dependences of second- (curves 1, 3) and fourth- (curve 2) order anisotropy and magnetostriction coefficients for Nd and Sm sublattices in Nd_2Co_{17}, Sm_2Co_{17} and $(Sm_{0.4}Y_{0.6})_2Co_{17}$ intermetallic compounds. The lines are the calculations on the basis of eqns. (11), (12) and (17). The points are the experimental data: △, $k^0_{2Sm}(T)/k^0_{2Sm}(0)$ for Sm_2Co_{17} [11]; ▲, $\lambda^{\alpha,2}_{2Sm}(T)/\lambda^{\alpha,2}_{2Sm}(0)$ for $(Sm_{0.4}Y_{0.6})_2Co_{17}$ [12]; ●, $k^0_{2Nd}(T)/k^0_{2Nd}(0)$, ○, $-k^0_{4Nd}(T)/k^0_{4Nd}(0)$ for Nd_2Co_{17} [11].

energy in comparison with the R–Co intersublattice exchange energy, the angular dependence of M_R and non-collinearity of M_R and M_{Co} in an external magnetic field applied in the hard direction can be neglected. Thus, the parameter X will be calculated from the very simple expression

$$X = \xi\sigma_{Co}(T)/\kappa T \tag{17}$$

where ξ is the parameter of isotropic intersublattice exchange interaction and σ_{Co} the relative magnetization of the Co sublattice which is determined from $M_S(T)$ for Y_2Co_{17}. As is seen from Figs. 2 and 3, the appropriate choice of the ξ parameter (respectively 460 K, 510 K and 750 K for compounds with Pr, Nd and Sm) gives a very good description of the experimental dependences and allows us to make a conclusion in favour of the single-ion MCA mechanism in R sublattices of such intermetallics.

4. Conclusions

The relatively simple quantum theory for the calculation of ferromagnet temperature dependences of anisotropy and magnetostriction coefficients, developed in refs. 4–6 and summarized here, can be successfully applied to interpret the experimental temperature dependences of these magnetic constants for magnetically ordered crystals with an arbitrary value of the total angular momentum of the magnetoactive ions. The obvious virtue of this theory is the possibility to derive the explicit theoretical formulae for $k_i(T)$ and $\lambda_i(T)$ functions and, at the same time, to calculate the tem-

perature dependence of ferromagnet magnetization over a wide temperature range.

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