Deducing model parameters of ferrimagnets from high-field magnetization curves

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The maximum magnetic field available for acquisition of quality magnetization data has risen significantly in recent times, the current limit being just short of a megaoersted. This opens possibilities for determining model parameters of various magnets by way of expressions containing directly observable quantities. Here such expressions are derived for anisotropic two-sublattice ferrimagnets.

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I. INTRODUCTION

Phenomenological description of ferrimagnets involves several model parameters that are to be determined from experiment. As a data source one normally uses low-temperature magnetization curves measured in a strong pulsed field capable of breaking the initial antiparallel orientation of the sublattice moments. The low temperature is important in two ways. First, it ensures the saturated state of the sublattices, effectively eliminating the intrasublattice exchange from the problem. Second, it renders harmless the inevitable magnetocaloric effect since sublattice magnetizations and anisotropy constants are independent of temperature at $T \lesssim 10$ K. (Ferrimagnets with very low Curie points are not considered herein, the more typical objects of our interest being iron-rich or cobalt-rich intermetallic compounds with heavy rare earths.)

In the absence of magnetic anisotropy the two-sublattice ferrimagnet at $T=0$ is readily tractable analytically. This was first done by Tyablikov,[1] soon followed by Schlömann's clearer exposition using the notation that was to become standard.[2] Schlömann himself admitted that the applicability of his simple approach to real ferrimagnets was impaired by their non-negligible anisotropy. Allowing for the anisotropy is in principle straightforward but results in a loss of the analytical tractability, whereas the number of adjustable parameters becomes unmanageably large. The latter applies particularly to rare-earth materials, which generally require more than a single anisotropy constant at low temperatures.[3]

During the half-century that has elapsed since Schlömann's work a few ingenious experimental techniques have been developed,[4] allowing in many cases to circumvent the unknown rare-earth anisotropy, which is still awaiting a proper theoretical treatment. In its absence the situation with quantitative interpretation of high-field magnetization data remains unsatisfactory because blind numerical fitting produces insufficiently reliable results. A well-documented example is Er_2Co_{17}, where several authors obtained for the first anisotropy constant of the Er sublattice values differing by as much as a factor of 2 (for a recent discussion see Ref. 9).

Meanwhile, in the wake of the steady progress in generating high magnetic fields, ever more experimental data are obtained in ever stronger fields. Trying to bring the lagging theory abreast with todays demands, we decided to reexamine the old model with a view to finding explicit expressions for the model parameters in terms of quantities directly deducible from experimental magnetization curves.

II. MODEL

We consider a two-sublattice ferrimagnet at a temperature sufficiently low to ensure that both sublattices are saturated. We exclude systems that do not saturate as $T \to 0$, e.g., rare earths with an even number of $4f$ electrons in a strong crystal field and a relatively weak exchange field. It is convenient to label the sublattices with subscripts T and R, where T refers to the sublattice with a larger magnetization, $M_T > M_R$. T may, but does not have to mean transition metal, nor does R necessarily stand for "rare earth." Let the magnetic field be applied along a high-symmetry crystallographic direction and denote $\theta_T = \widehat{\boldsymbol{HM}}_T$, $\theta_R = \widehat{\boldsymbol{HM}}_R$ [see Fig. 1(b)]. The energy of the system is presented as follows:

$$E = \lambda M_T M_R \cos(\theta_T + \theta_R) - M_T H \cos\,\theta_T - M_R H \cos\,\theta_R + K_T \sin^2\,\theta_T + E_{aR}(\sin\,\theta_R). \quad (1)$$

Terms in this expression describe the intersublattice exchange interaction (the exchange constant λ being positive), the Zeeman interactions of the two sublattices as well as their respective anisotropy energies. It has been assumed that the anisotropy of the sublattice T can be with sufficient accuracy described by a single anisotropy constant, which is justified if T stands for Fe or Co. As against that, no specific assumption has been made about the form of the function $E_{aR}(\sin\,\theta_R)$. From general principles it should be expansible in even powers of $\sin\,\theta_R$,

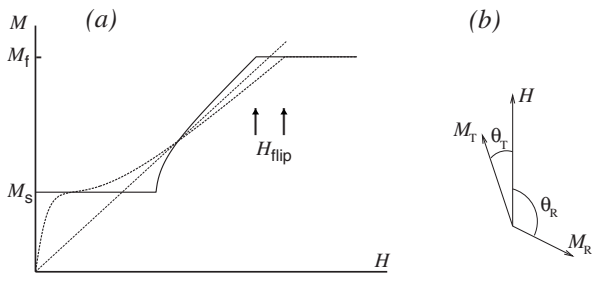

FIG. 1. (a) Schematic magnetization curves of an anisotropic ferrimagnet ($K_T=0$). The field is applied either in the direction of easy magnetization (solid curve) or in the hard direction (dashed curve). The thin sloping line corresponds to $M=\lambda^{-1}H$. (b) Mutual orientation of the sublattice magnetizations and applied magnetic field.

$$E_{aR} = K_R \sin^2 \theta_R + \cdots, \tag{2}$$

but we place no restriction whatsoever on how rapidly, if at all, this expansion might converge. Such precautions are not out of place if R is to mean rare earth.[3]

The parameters entering in Eq. (1) can be divided in two groups. The first group includes the parameters that we shall regard as known: M_T, M_R, and K_T. These are obtainable, at least in the case of rare-earth transition-metal intermetallics, from low-field magnetization data and some general considerations (e.g., taking a free-ion value for M_R, taking for K_T the anisotropy constant of an isomorphous auxiliary compound with R=Y, La, or Lu, etc.). The parameters of the second group, λ, K_R, and any higher-order coefficients in Eq. (2) are essentially unknown. The goal of this work is to find λ and K_R.

Regarding the symmetry of the crystal we make only a broad assumption that it is higher than triclinic so that at least one high-symmetry direction can be found. This is then taken to be the direction of the applied field. Caution should be exercized when relating K_T and K_R to any conventional notation. E.g., for hexagonal crystals K_T is equivalent to the standard K_1 if $\boldsymbol{H} \| [001]$, however, K_T equals $-K_1$ if $\boldsymbol{H} \| [100]$ and the sublattice moments rotate in the $\boldsymbol{ac}$ plane, finally, K_T is nil if the moments prefer to rotate in the basal plane.

The remaining steps include setting the necessary conditions of equilibrium, $\partial E/\partial \theta_T = \partial E/\partial \theta_R = 0$ or

$$\lambda M_T M_R \sin(\theta_T + \theta_R) = M_T H \sin \theta_T + K_T \sin 2\theta_T, \tag{3}$$

$$\lambda M_T M_R \sin(\theta_T + \theta_R) = M_R H \sin \theta_R + E'_{aR}(\sin \theta_R)\cos \theta_R, \tag{4}$$

and solving these simultaneous equations for θ_T and θ_R. The obtained values are then used to compute the magnetization in the direction of the applied field,

$$M = M_T \cos \theta_T + M_R \cos \theta_R. \tag{5}$$

III. DETERMINATION OF λ

It appears generally impossible to obtain from Eqs. (3) and (4) closed expressions for θ_T and θ_R. We shall therefore examine special cases. A particularly useful one is that of $\theta_R = \pi/2$, i.e., when the smaller sublattice magnetization makes a right angle with the applied field. At this point the anisotropy of the sublattice R effectively cancels out, as the last term in Eq. (4) vanishes due to the factor $\cos \theta_R$. So we find from Eq. (4)

$$\sin(\theta_T + \theta_R) = \cos \theta_T = \frac{H}{\lambda M_T}. \tag{6}$$

Setting this expression in Eq. (3), we obtain thence

$$\sin \theta_T = \frac{M_R}{M_T + \dfrac{2K_T}{\lambda M_T}}. \tag{7}$$

By Eqs. (5) and (7) with $\theta_R = \pi/2$, the magnetization at the orthogonality point is given by

$$M_\perp = M_T \cos \theta_T = \sqrt{M_T^2 - \left(\frac{M_R}{1 + 2K_T/\lambda M_T^2}\right)^2}. \tag{8}$$

Denote $H_\perp$ the corresponding (orthogonality) field. By Eq. (6) one has

$$H_\perp = \lambda M_T \cos \theta_T = \lambda M_\perp, \tag{9}$$

whence

$$\lambda = \frac{H_\perp}{M_\perp}. \tag{10}$$

Thus, finding the intersublattice exchange constant of an anisotropic ferrimagnet is as simple as locating the orthogonality point in the magnetization curve. At that point the system is momentarily released from the clutches of the unknown but presumably strong anisotropy of the sublattice R. The anisotropy of the other sublattice, T, which we regard as known, is readily allowed for. The only precondition is that K_T should be much less in magnitude than the exchange energy, λM_T^2. This seems to be fulfilled in most cases, at least for T=Fe or Co. Then Eq. (8) can be recast in an approximate form, corrected for K_T to first order,

$$M_\perp \approx \sqrt{M_T^2 - M_R^2} + 2\left(\frac{M_R}{M_T}\right)^2 \frac{K_T}{\lambda \sqrt{M_T^2 - M_R^2}}. \tag{11}$$

In the same approximation the orthogonality field is given by

$$H_\perp \approx \lambda \sqrt{M_T^2 - M_R^2} + 2\left(\frac{M_R}{M_T}\right)^2 \frac{K_T}{\sqrt{M_T^2 - M_R^2}}. \tag{12}$$

We are now ready to give a prescription for finding λ. Suppose we have available an experimental magnetization curve $M(H)$ of a certain ferrimagnet, measured to a sufficiently high field, and we know the parameters M_T, M_R, and K_T. Then these are the steps to follow:

(1) Compute a starting ordinate of the orthogonality point,

$$M_\perp^{(0)} = \sqrt{M_T^2 - M_R^2}. \tag{13}$$

(2) Find the corresponding abscissa in the magnetization curve, $H_\perp^{(0)}$.

(3) Correct the ordinate for K_T,

$$M_\perp^{(1)} = M_\perp^{(0)} + 2\left(\frac{M_R}{M_T}\right)^2 \frac{K_T}{H_\perp^{(0)}}. \tag{14}$$

At this stage it should become clear if the correction is worthwhile.

(4) Find a new abscissa $H_\perp^{(1)}$ that corresponds to the corrected ordinate $M_\perp^{(1)}$.

(5) Evaluate $\lambda = H_\perp^{(1)}/M_\perp^{(1)}$.

As the fields available in experiment grow ever stronger, it becomes increasingly likely that the spin-flip transition point might be reached, beyond which the magnetization is fully aligned with the field, $\boldsymbol{M}_T \uparrow \uparrow \boldsymbol{M}_R \uparrow \uparrow \boldsymbol{H}$, and therefore saturated. In this case one can directly measure the full (final or flip) magnetization,

$$M_f = M_T + M_R \tag{15}$$

along with the routine spontaneous magnetization,

$$M_s = M_T - M_R. \tag{16}$$

For such an eventuality it appears advantageous to rewrite Eqs. (13) and (14) directly in terms of M_s and M_f,

$$M_\perp^{(0)} = \sqrt{M_s M_f}, \tag{13'}$$

$$M_\perp^{(1)} = M_\perp^{(0)} + 2\left(\frac{M_f - M_s}{M_f + M_s}\right)^2 \frac{K_T}{H_\perp^{(0)}}. \tag{14'}$$

Theoretically, it is indifferent for the success of the above algorithm whether the field is applied along an easy or a hard magnetization direction. The former has, however, two practical advantages: (i) the spontaneous magnetization M_s can be determined from the same curve and (ii) the spin-flip transition is reached in a lower field [see Fig. 1(a)]. Any discontinuities that may be present in the magnetization curve (which are associated with field-induced first-order phase transitions) do not hinder the determination of λ by the described method.

It follows from Eqs. (8)–(11) that in a special case of $K_T=0$ the orthogonality point is a crossing point of magnetization curves taken along different high-symmetry crystal directions.

IV. DETERMINATION OF K_R

Let the system be close to the spin-flip transition point, $H \approx H_{\text{flip}}$. Then both orientation angles in Fig. 1(b), θ_T and θ_R, are small and the equilibrium conditions Eqs. (3) and (4) can be linearized by replacing the sines therein with their arguments

$$(\lambda M_T M_R - M_T H - 2K_T)\theta_T + (\lambda M_T M_R)\theta_R = 0, \tag{17}$$

$$(\lambda M_T M_R)\theta_T + (\lambda M_T M_R - M_R H - 2K_R)\theta_R = 0. \tag{18}$$

Note that the expansion Eq. (2) has been truncated after the first term; this is now justified by the smallness of θ_R. At $H=H_{\text{flip}}$ the determinant of the system of homogeneous linear Eqs. (17) and (18) must vanish,

$$(\lambda M_T M_R - M_T H_{\text{flip}} - 2K_T)(\lambda M_T M_R - M_R H_{\text{flip}} - 2K_R) - (\lambda M_T M_R)^2 = 0. \tag{19}$$

Hence it follows that

$$K_R = \frac{\frac{\lambda}{2}(M_T + M_R) - \frac{H_{\text{flip}}}{2} - K_T\left(\frac{1}{M_T} - \frac{\lambda}{H_{\text{flip}}}\right)}{\frac{1}{M_R} - \frac{\lambda}{H_{\text{flip}}} + \frac{2K_T}{M_T M_R H_{\text{flip}}}}. \tag{20}$$

One thus determines K_R from a measured critical field H_{flip} and the intersublattice exchange constant λ found in the previous section. Equation (20) is particularly useful for uniaxial (i.e., hexagonal, tetragonal, or trigonal) crystals, provided the field is applied along the symmetry axis [001] (which need not be direction of easy magnetization). In this case K_R is the conventional first anisotropy constant. Other field directions are less convenient because then K_R is equivalent to a combination of anisotropy constants and to tell which combination it is, one requires prior knowledge of the preferred azimuthal orientation of the sublattice moments in Fig. 1(b). Still such measurements may provide useful additional information on higher-order anisotropy constants of the sublattice R.

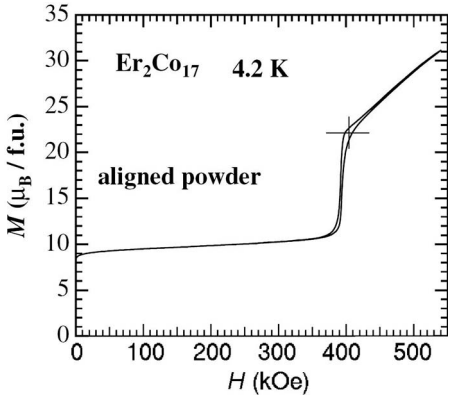

FIG. 2. Experimental easy-direction magnetization curve of Er_2Co_{17} taken from Ref. 9. The cross marks the position of the orthogonality point at $H_\perp=405$ kOe, $M_\perp=22.1\mu_B$/f.u.

V. DISCUSSION

As an application of the formulas obtained in the previous sections, let us consider a well-researched binary intermetallic compound Er_2Co_{17}. A number of thourough studies were carried out on single crystals and the intersublattice exchange constant λ was variously reported to be either 16.7 kOe f.u./μ_B (Ref. 10) or 19.1 kOe f.u./μ_B (Ref. 11). A more recent paper[9] adopts an intermediate value, $\lambda=18.3$ kOe f.u./μ_B obtained by judicious averaging of several independent results. We shall now demonstrate how the same value can be deduced directly from the experimental magnetization curve of Ref. 9 reproduced in Fig. 2. Taking the sublattice moments of Ref. 9, $M_T \equiv M_{\text{Co}}=28.5\mu_B$/f.u. and $M_R \equiv M_{\text{Er}}=18\mu_B$/f.u., we find from Eq. (13) $M_\perp^{(0)}=22.1\mu_B$/f.u. The corresponding abscissa in the magnetization curve is $H_\perp^{(0)}=405$ kOe as marked with a cross in Fig. 2. The presence of a slight hysteresis in the experimental curve does not interfere with our analysis—we simply take the average of the two branches. We further borrow from Ref. 9 a value of $K_T \equiv K_1^{\text{Co}}=-9$ K/f.u., inferred from the literature on Y_2Co_{17}, and attempt to correct $M_\perp^{(0)}$ by means of Eq. (14). The correction turns out to be $-0.26\mu_B$/f.u., which we deem insignificant and choose to neglect. Our final result thus is $\lambda=405/22.1=18.3$ kOe f.u./μ_B. This agrees in all three digits with the educated guess of Ref. 9.

We are now in a position to demonstrate *a posteriori* the applicability of the two-sublattice model of Sec. II to Er_2Co_{17}. One of the concerns might be that the Er atoms occupy two nonequivalent sites in the hexagonal Th_2Ni_{17}-type structure of Er_2Co_{17}. Experimental evidence suggests, however, that the molecular fields on the two rare-earth sites in this structure are close to each other: Thus, the ^{161}Dy Mössbauer spectra of the isomorphous Dy_2Fe_{17} are

well represented by a single set of hyperfine parameters even at room temperature.[12] Furthermore, the molecular field on Er is so strong that it prevails over the crystal field on either of the Er sites. (The total exchange splitting of the ground multiplet of Er, $18\mu_B(\lambda M_{\mathrm{Co}}) \approx 630$ K, is much greater than the respective crystal-field splitting, ≈ 10 K, as estimated using the leading crystal field parameter of Tm_2Fe_{17} from Ref. 13.) Consequently, the low-temperature moment of Er is close to the free-ion saturation moment, $9\mu_B$. This is corroborated by the comparison of the saturation magnetizations of Er_2Co_{17} and Y_2Co_{17} in Ref. 9. The energy gap to the first excited level of Er is $1.2\mu_B(\lambda M_{\mathrm{Co}}) \approx 42$ K. At $T \lesssim 10$ K it is justified to regard all Er atoms in Er_2Co_{17} as one saturated magnetic sublattice. Likewise, the Co atoms, distributed over four different sites, can be viewed as a single Co sublattice because the parallel orientation of the Co moments is ensured by the very strong Co-Co exchange (as manifest in the exceptionally high Curie temperatures of R_2Co_{17}, ~ 1200 K). Finally, we confirm that the exchange energy, $\lambda M_{\mathrm{Co}}^2 \approx 1000$ K, is indeed much greater than $|K_T| \approx 9$ K.

Proceeding now to $K_R \equiv K_1^{\mathrm{Er}}$, we note that the discrepancy between Refs. 9 and 10 is no less than a factor of 2, that is $K_1^{\mathrm{Er}} = 110$ K/f.u. versus 225 K/f.u., respectively. Our contribution to the dispute will be to check if the calculations of Ref. 9 are consistent. We note that those calculations yielded $H_{\mathrm{flip}} = 755$ kOe for the critical field of spin-flip (cf. Fig. 5 of Ref. 9). Setting this value as well as $\lambda = 18.3$ kOe f.u./μ_B into Eq. (20), we get $K_1^{\mathrm{Er}} = 108$ K/f.u. Within the limits of error this coincides with the input value $K_1^{\mathrm{Er}} = 110$ K/f.u. used by Yoshii *et al.*[9] and thus confirms the correctness of their numerical calculations. Of course, $H_{\mathrm{flip}} = 755$ kOe is at this stage a mere extrapolated value since the magnetization in Ref. 9 was measured only up to 550 kOe. Technically, conditions for an experimental observation of the spin flip in Er_2Co_{17} are ripe, as long-pulsed fields in excess of 850 kOe are now available both at Los Alamos[14] and at Dresden.[15]

Returning to the calculations of Yoshii *et al.*,[9] we note that their allowance for K_1^{Co} was unnecessary. To demonstrate this point, Eq. (19) needs to be solved for H_{flip}. In principle, this can be done exactly (the larger of the two solutions of the quadratic equation should be taken), but it is more convenient to use an approximate expression corrected for anisotropy to first order,

$$H_{\mathrm{flip}} = \lambda(M_T + M_R) + 2\left(\frac{K_T + K_R}{M_T + M_R} - \frac{K_T}{M_T} - \frac{K_R}{M_R}\right).$$

The first term in this expression is the well-known isotropic result of Schlömann.[2] Now in the case of Er_2Co_{17} the contribution to H_{flip} due to $K_T \equiv K_1^{\mathrm{Co}} = -9$ K/f.u. equals

$$2K_1^{\mathrm{Co}}\left(\frac{1}{M_{\mathrm{Co}} + M_{\mathrm{Er}}} - \frac{1}{M_{\mathrm{Co}}}\right) = 3.6 \text{ kOe}$$

that is about 0.2 mm on the scale of Fig. 5 of Ref. 9.

The value of the last remaining parameter in the calculation of Yoshii *et al.*[9] $K_2^{\mathrm{Er}} = -30$ K/f.u. does not appear physically meaningful on account of the rather arbitrary neglect of the sixth-order anisotropy constant K_3^{Er}. One could have equally well set $K_2^{\mathrm{Er}} = 0$ and adjusted K_3^{Er} to fit the data. According to the linear theory of magnetic anisotropy,[3,16] there is no reason to believe that the low-temperature value of K_3^{Er} should be any smaller in magnitude than that of K_2^{Er}. It would be interesting to probe these quantities by observing a spin flip with $\boldsymbol{H} \| [100]$.

In Summary, we have derived expressions for the key model parameters of anisotropic ferrimagnets, λ and K_R, in terms of quantities directly deducible from low-temperature magnetization curves. K_R need not be small and higher-order anisotropy constants of the sublattice R may be nonzero. The method also allows for K_T, regarded as known. Whether such an allowance is really necessary can be readily assessed within the same approach.

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