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PHYSICAL REVIEW B VOLUME 3, NUMBER 3 1 FEBRUARY 1971

Canted Spin Phase in Gadolinium Iron Garnet

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The behavior of ferrimagnetic garnets with three magnetic sublattices in an external field has been investigated. In the field-temperature plane we determined the stability limits of the collinear phases with respect to angle formation between the various sublattice moments. The application of our molecular-field analysis to gadolinium iron garnet (GdIG) shows that the angle formation between the two strongly coupled iron sublattices cannot be neglected. By measuring the Faraday effect in fields up to 10 kOe, we were able to observe the angled spin phase in GdIG in the vicinity of the compensation point.

I. INTRODUCTION

A magnetic field applied to a ferrimagnet in which the sublattice moments are collinear tends to align all the moments parallel to itself, in opposition to the exchange interactions which try to maintain the ferrimagnetic antiparallel configuration. Under certain conditions of field and temperature, this competition can result in spin configurations in which the individual sublattice moments form angles with each other and with the field even in isotropic crystals. This phenomenon has been studied in a number of investigations both experimentally and theoretically.[1-4]

In the case of three-sublattice iron garnets, one expects that with increasing field, angles are first formed predominantly between the rare-earth sublattice moment and the net iron moment. Only at much higher fields is the antiparallel alignment of the iron moments broken up. This assumption is based on the fact that the exchange coupling of the rare-earth sublattice to the iron sublattices is weak compared with the dominant coupling between the two iron sublattices. Thus, in the past, molecular-field theory has been applied to calculate the magnetic response of garnets to an external field,[1-3] but angle formation between the two iron moments has been neglected. This simplifies the theoretical treatment, and the instability criterion for the collinear phases is a two-sublattice relationship.[1] Even in small fields, however, the simplified treatment can lead to wrong results, especially if the compensation point does not occur at very low temperatures.

In the present work (Sec. II) we derive the correct instability criterion for a three-sublattice system. This criterion, together with the molecular-field equations, determines the phase boundaries in the field-temperature plane. As an application, we calculate the magnetic-phase diagram for gadolinium iron garnet (GdIG), the magnetic properties of which can be reasonably well described by a simple three-sublattice model. As all magnetic ions in this material have an S ground state, crystal-field and anisotropy effects are small[5] and can be neglected.[6] The values of the molecular-field coefficients are available from various sources; in our calculations for GdIG we use Anderson's set[7] which was obtained from a fit of magnetization measurements. The results agree well with our Faraday rotation measurements which are reported in Sec. III. The Faraday rotation is very sensitive to changes in the spin configuration and therefore provides an excellent tool for detection of the boundaries of the

angled phase.

II. INSTABILITY CRITERION FOR COLLINEAR PHASES AND ITS APPLICATION TO GdIG

For *two-sublattice* systems it has been shown quite generally that there exists a critical temperature below which, in a closed region of the field-temperature plane, the sublattice moments form angles with each other and with the field. Outside this region the moments and the field are collinear, and all phase transitions are second order if anisotropy can be neglected.[1,4] In the following we derive the instability criterion for the collinear phases of a *three-sublattice* system.

We start from the molecular-field equations for a magnetically isotropic three-sublattice system in an external field $\vec{H}_0$:

$$\vec{M}_i = M_i^0 \frac{\vec{H}_i}{H_i} B_{S_i}\left(\frac{g\mu_B S_i H_i}{k_B T}\right), \quad i = 1, 2, 3 \tag{1}$$

where B_{S_i} is the Brillouin function for spin S_i, g is the Landé factor, μ_B is Bohr's magneton, k_B is Boltzmann's constant, and T is the temperature. $\vec{M}_i$ denotes the magnetic moment of the ith sublattice with magnitude M_i^0 at zero temperature, and the $\vec{H}_i$'s are the effective fields acting on the sublattices:

$$\vec{H}_i = \vec{H}_0 + \sum_{j=1}^{3} \lambda_{ij}\vec{M}_j, \quad \lambda_{ij} = \lambda_{ji} \tag{2}$$

where the λ_{ij}'s denote the molecular-field coefficients.

Since each sublattice moment is parallel to its molecular field [see Eq. (1)] we obtain from Eq. (2)

$$\vec{M}_i \times \vec{H}_0 = -\sum_{j=1;\ j\neq i}^{3} \lambda_{ij}\vec{M}_i \times \vec{M}_j \,. \tag{3}$$

Introducing spherical coordinates (see Fig. 1), we can write

$$\begin{aligned} &\vec{H}_0 = H_0(0, 0, 1), \\ &\vec{M}_i = M_i(\sin\theta_i \cos\phi_i, \sin\theta_i \sin\phi_i, \cos\theta_i). \end{aligned} \tag{4}$$

Note that a rearrangement of Eq. (3) gives the conditions

$$\begin{aligned} &\sum_{i=1}^{3} M_i \sin\theta_i \cos\phi_i = 0, \\ &\sum_{i=1}^{3} M_i \sin\theta_i \sin\phi_i = 0, \end{aligned} \tag{5}$$

i.e., the total magnetization is parallel to the applied field.

Now let ϵ_i, $i = 1, 2, 3$ denote small deviations from a collinear configuration:

$$\theta_i = n_i\pi + \epsilon_i, \quad n_i = 0 \text{ or } 1. \tag{6}$$

Keeping only terms linear in the ϵ_i, we obtain

$$\vec{M}_i \times \vec{H}_0 \approx \sigma_i H_0(\xi_i, -\eta_i, 0),$$

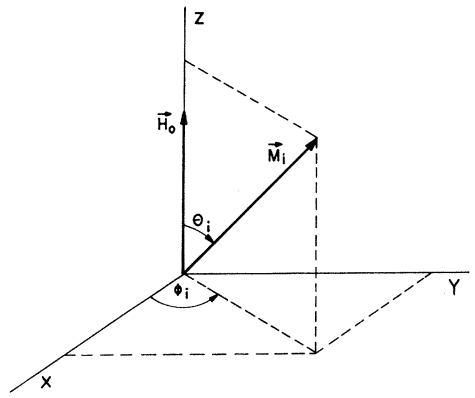

FIG. 1. Spherical coordinates for sublattice magnetization $\vec{M}_i$ (external field $\vec{H}_0$ in z direction).

$$\vec{M}_i \times \vec{M}_j \approx \sigma_i\sigma_j(\xi_i - \xi_j, -(\eta_i - \eta_j), 0), \tag{7}$$

where

$$\sigma_i = \nu_i M_i, \qquad \nu_i = \cos(n_i\pi) = \pm 1$$

and

$$\xi_i = \epsilon_i \sin\phi_i, \quad \eta_i = \epsilon_i \cos\phi_i.$$

Equation (3) then reduces to

$$\begin{aligned} H_0\xi_i &= -\sum_{j=1}^{3} \lambda_{ij}\sigma_j(\xi_i - \xi_j), \\ H_0\eta_i &= -\sum_{j=1}^{3} \lambda_{ij}\sigma_j(\eta_i - \eta_j), \end{aligned} \qquad i = 1, 2, 3 \tag{8}$$

where we have omitted the restriction $j \neq i$ in the summations, since the diagonal terms give no contribution. Rewriting Eqs. (8) in the form

$$\sum_{j=1}^{3} \left[\delta_{ij}\left(H_0 + \sum_{k=1}^{3} \lambda_{ik}\sigma_k\right) - \lambda_{ij}\sigma_j\right] \begin{matrix}\xi_i \\ \eta_i\end{matrix} = 0, \tag{9}$$

we see that the condition for nontrivial solutions ξ_i or η_i, respectively, is

$$\det\left[\delta_{ij}\left(H_0 + \sum_{k=1}^{3} \lambda_{ik}\sigma_k\right) - \lambda_{ij}\sigma_j\right] = 0. \tag{10}$$

Evaluating the determinant we obtain the instability criterion for the collinear phases

$$\begin{aligned} &H_0^2 + H_0[\lambda_{12}(\sigma_1 + \sigma_2) + \lambda_{13}(\sigma_1 + \sigma_3) + \lambda_{23}(\sigma_2 + \sigma_3)] \\ &\quad + (\sigma_1 + \sigma_2 + \sigma_3)(\lambda_{12}\lambda_{13}\sigma_1 + \lambda_{12}\lambda_{23}\sigma_2 + \lambda_{13}\lambda_{23}\sigma_3) = 0. \end{aligned} \tag{11}$$

For a given collinear configuration, defined by the signs of the σ_i's, we now find the stability limits by simultaneously solving (1), (2), and (11).

For the application to GdIG we use Anderson's[7] molecular-field coefficients[8] which (in Oe g/emu) are[9]

$$\lambda_{11} = -61\,000, \quad \lambda_{12} = -91\,060, \quad \lambda_{13} = -900,$$

$$\lambda_{22} = -29\,000, \quad \lambda_{23} = -4075,$$
$$\lambda_{33} = -200.$$

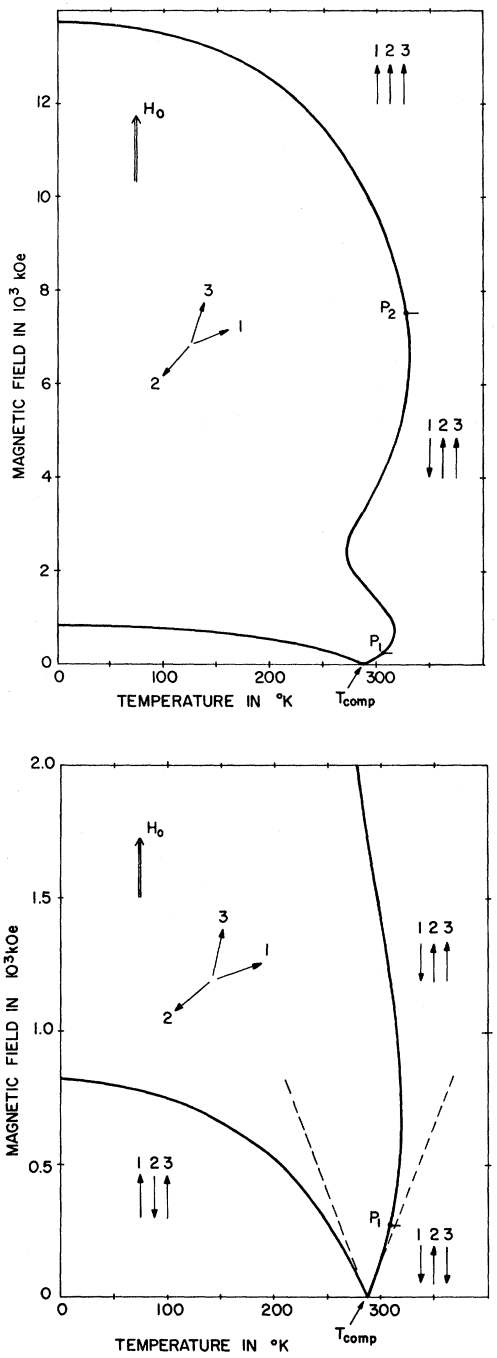

FIG. 2. Phase diagram for GdIG. Arrows 1, 2, 3 denote the directions of the *a*, *d*, *c* sublattice moments, respectively. The sublattices *c* and *a* are completely demagnetized at points P_1 and P_2, respectively. (b) is the lower portion of (a) on an enlarged-field scale. The broken lines give the slopes of the phase boundaries near the compensation point T_{comp}.

Furthermore, we have $S_1 = S_2 = \frac{5}{2}$, $S_3 = \frac{7}{2}$, and[7]

$$M^0 = M_1^0 - M_2^0 + M_3^0 = 94.8 \text{ emu/g},$$

so that

$$M_1^0 = 59.25, \quad M_2^0 = 88.875, \quad M_3^0 = 124.425 \text{ emu/g}.$$

Finally, we know the spin configurations in small fields. Below the compensation temperature, the moments of the rare-earth and the octahedrally coordinated iron sublattices are parallel to the field, while the moment of the tetrahedrally coordinated iron is antiparallel. Above the compensation temperature, all three moments are reversed.

The resulting phase diagram for GdIG is shown in Figs. 2(a) and 2(b). Due to the very strong exchange interactions, the angled phase only exists in extremely high fields, except at temperatures near the compensation point. There are two regions in the diagram where only collinear phases exist. These two are separated by the angled phase region. We see that by increasing the field isothermally it is possible to cross the upper phase boundary four times, in contrast to the two-sublattice case where this can happen at most twice.[1] The right-hand collinear region can be divided into three subregions. Let us illustrate this by considering the behavior of the sublattice moments along the phase boundary. Starting from T_{comp}, the two sublattice moments 1 and 3, which are antiparallel to the field H_0, decrease in magnitude. At P_1, the rare-earth moment vanishes and then increases parallel to H_0. At P_2 the same happens to the moment of the octahedrally coordinated iron.

If we neglect λ_{13}, λ_{23}, and H_0 with respect to λ_{12}, Eq. (11) takes the simplified form

$$\sigma_1 + \sigma_2 + \sigma_3 = -\frac{H_0(\sigma_1 + \sigma_2)}{\lambda_{13}\sigma_1 + \lambda_{23}\sigma_2} . \tag{12}$$

This is identical with the simplified criterion which is obtained from the approximation that in not too high fields the two iron moments remain antiparallel.[1] For GdIG, however, the simplified criterion gives wrong results even in very small fields. Calculations with criterion (12) yield phase boundaries, the slopes of which (near to the compensation point) have the wrong sign. So, in our case, the instability with respect to angle formation between the two iron sublattices cannot be neglected, although the angles are presumably very small in low fields.

III. FARADAY ROTATION MEASUREMENTS

According to the calculated phase diagram (Fig. 2), all three sublattice moments are reversed if the material is in a weak magnetic field and the temperature passes through the compensation point. In stronger fields, the sublattice reorientation takes place in a finite temperature interval. The width

of this interval is nearly proportional to the field for fields up to 100 kOe and is rather small in fields available in the laboratory. We have measured this width as a function of field by observing the temperature dependence of the optical Faraday rotation at constant field. The Faraday effect at optical wavelengths is related to the individual sublattice moments rather than to the total magnetization, so that the temperature dependences of the Faraday rotation and the magnetization in general are very different from each other.[10] In the compensating region the magnetization is small and shows comparatively little variation with temperature. In contrast, the magneto-optical rotation is quite large and the sublattice reorientation is clearly demonstrated by a corresponding reversal of the sense of the rotation.[11]

The Faraday effect was measured using monochromatic light of a wavelength $\lambda = 1.15$ μm and a magnetic field of up to 9.7 kOe which was applied parallel to the light beam. The principle of operation of our polarimetric system is as follows[12]: The light beam is linearly polarized and passes through a magneto-optic modulator to the sample, then through an analyzer on to a photomultiplier. The modulator causes the polarization plane of the light reaching the analyzer to rotate cyclically a few degrees either side of the mean position. When this oscillation occurs symmetrically about the crossed position of analyzer and polarizer, the fundamental cyclic frequency vanishes from the phototube output signal. Using phase-sensitive detection of this unbalance signal, the crossed position can be located very precisely. When the sample induces a rotation of the polarization plane, the analyzer follows this rotation automatically. The analyzer position is displayed directly on a recorder.

The modulator consists of a 10-cm-long flint glass rod placed into a coil producing an ac magnetic field with 50-Hz frequency and 600-Oe amplitude. The Faraday effect caused by this oscillating field in the glass rod provides the cyclic rotation of the polarization plane. The polarizer and analyzer are Glan prisms. The dc field magnetizing the sample is produced by an electromagnet with axial holes which allow optical access parallel to the field direction.

The sample, a single-crystal GdIG platelet 0.49 mm thick, was thermally insulated by vacuum and a radiation shield. The shield together with the crystal-holder block could be cooled down with dry ice. While the whole unit warmed up slowly to room temperature, the rotation of the polarization plane induced by the sample was recorded as a function of temperature. Temperature measurements were made by means of a thermocouple attached to the crystal.

Three typical recorder traces are shown in Fig. 3. A few degrees above and below the compensation temperature – which is 285.7 K in our sample – the rotations are about equal in magnitude but have opposite signs and vary little with temperature and field. Close to the compensation temperature, the rotation angle changes rapidly in a temperature interval which is about 2° wide at 9.7 kOe and 1° at 4.6 kOe. In lower fields the interval boundaries are less clearly marked in the curves, since at temperatures close to the compensation point the field fails to saturate the sample.

In Fig. 3(c) the Faraday rotation varies nearly linearly with temperature in the reorientation region, but with some reproducible superimposed oscillations. Without analyzing the motion of the magnetization vectors in detail, we attribute these oscillations to the small magnetocrystalline anisotropy. Since the applied field is weak compared to the exchange fields acting on the magnetic moments, the sublattice moments will remain nearly collinear during their reversal, and the Faraday effect will reflect the change of their projection on the field direction. The easy directions are [111],[5] and our sample was cut normal to [211]. The sequence of hard and easy directions which the moments sample during their rotation can account qualitatively for the observed pattern.

Our measurements cover an extremely small portion of the phase diagram, but they yield the slopes of the two phase boundaries at the compensation point. The experimental values $dT_{crit}/dH_0 \approx \pm 0.1$ deg/kOe compare favorably with the calculated ones of ± 0.094 deg/kOe. This agreement is encouraging, since the values of the molecular-field coefficients were not fitted to our results but were taken from magnetization measurements. Note also that anisotropy effects, though they are small, could play a role in our experiments since the Zeeman energy in our weak fields may be equally small.

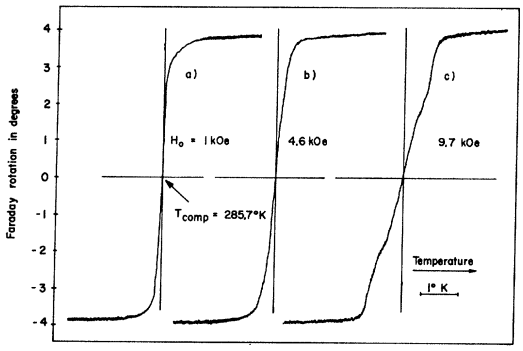

FIG. 3. Temperature dependence of the Faraday rotation in GdIG. Sample thickness $d = 0.49$ mm, wavelength $\lambda = 1.15$ μm.

Further valuable information could be obtained from measurements in considerably higher fields.

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[8]Anderson obtained the coefficients by fitting the magnetization curve. Due to an error in the nearest-neighbor numbers ($z_{23}=2$, and not 4), however, his values are not symmetric ($\lambda_{23}=2\lambda_{32}$). This error already occurs in Pauthenet's paper [R. Pauthenet, Ann. Phys. (Paris) 3, 424 (1958)], while the correct nearest-neighbor numbers can, e.g., be found in a paper by Nowik [I. Nowik, Phys. Rev. 171, 550 (1968)]. We choose $\lambda_{23}=\lambda_{32}=-4075$ and compensate the so-caused shift of the compensation temperature by changing λ_{13} from -800 to -900. The corrected set still provides a very good fit for the total and the sublattice magnetizations given in a paper by Myers *et al.* [S. M. Myers, R. Gonano, and H. Meyer, Phys. Rev. 170, 513 (1968)].

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PHYSICAL REVIEW B VOLUME 3, NUMBER 3 1 FEBRUARY 1971

Activation Volumes of Interstitial Diffusion in Ferromagnetic Metals and Alloys*

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The disaccommodation technique was used to determine the activation volumes of carbon diffusion ΔV of a Fe–1.4 wt% Si alloy, of hcp cobalt, and of nickel at the temperatures of −21, 75, and 110 °C, respectively. The obtained values of ΔV are $+0.3 \pm 0.3$ and -0.6 ± 0.9 $\mathrm{cm^3\,mole^{-1}}$ for the two relaxations observed in the Fe-Si alloy, -1.7 ± 2.0 and -0.5 ± 0.2 $\mathrm{cm^3\,mole^{-1}}$ for the two relaxations observed in hcp cobalt, and 1.2 ± 0.1 $\mathrm{cm^3\,mole^{-1}}$ for nickel. These results are discussed in terms of a continuum model that interprets the activation energy of diffusion as a sum of the strain and exchange energies. It is found that in ferromagnetic solids the activation volume of diffusion is essentially controlled by the ferromagnetic anharmonicity.

I. INTRODUCTION

The activation volume of diffusion ΔV is the volume change of a crystal that occurs as the diffusing species performs one elementary diffusive jump. In the case of substitutional and self-diffusion, ΔV is the sum of the activation volumes of formation ΔV_f and migration ΔV_m, whereas $\Delta V = \Delta V_m$ for impurity interstitial diffusion and diffusion by an exchange-type mechanism. On the basis of a hard-sphere model,[1] ΔV would be expected to be positive and of the order of the atomic volume of the diffusing species Ω. While negative activation volumes are inconsistent with the hard-sphere model, it is conceivable that $\Delta V_f < 0$ for vacancy-type defects if the lattice relaxation around the defect is sufficiently large.[1] The activation volume of migration, on the other hand, should not be negative, as this would require that an inward relaxation is associated with the elementary dif-