Effective Gyromagnetic Ratio for Triangular Ferrimagnetic States*

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It is shown from the semiclassical equations of motion and the molecular-field approximation that the triangular ferrimagnetic state can also be described by an effective gyromagnetic ratio. This effective ratio depends explicitly on the ratio of the inter- to intra-sublattice molecular field constants rather than on the static sublattice magnetizations as in the antiparallel state. It is estimated that the shift in g values for $FeCr_2O_4$ should be observable if there is a transition from a triangular to an antiparallel state.

I

AN extension of the two-sublattice models of ferrimagnetism was made by Yafet and Kittel[1] to take account of large interactions within the sublattices. In so doing, they introduced the concept of triangular ferrimagnetic states in which the original sublattices are further subdivided and the resulting magnetization vectors are no longer completely parallel (or antiparallel) to each other or to the net magnetization. This theory has been elaborated by Lotgering,[2] and he has found some experimental evidence for the existence of triangular states in the chromium spinels, $MnCr_2O_4$ and $FeCr_2O_4$, from the values of their low-temperature magnetizations.

In this paper, we shall be concerned with the question as to whether the existence of triangular states will be reflected in the properties of these materials in magnetic resonance experiments, as is the case with sublattices generally in the more usual ferrimagnetics. In particular, one would like to know whether they can also be usefully described by an effective gyromagnetic ratio[3] in spite of the fact that there may exist large magnetization components transverse to the direction of the applied field.

We shall consider the three-sublattice system illustrated in Fig. 1. If we let λ_{ij} be the molecular-field coefficient between the ith and jth sublattices, and $\mathbf{H}$ the applied field, then the total field on the ith sublattice is

$$\mathbf{H}_i=\mathbf{H}-\Sigma_j\lambda_{ij}\mathbf{M}_j.$$

In this equation, we have used the minus sign for convenience in discussing the predominant cases of antiferromagnetic couplings. Using the general relation $\lambda_{ij}=\lambda_{ji}$, we can write the fields explicitly as

$$\mathbf{H}_1=\mathbf{H}-\lambda_{11}\mathbf{M}_1-\lambda_{12}\mathbf{M}+(\lambda_{12}-\lambda_{13})\mathbf{M}_3, \quad (1a)$$

$$\mathbf{H}_2=\mathbf{H}-(\lambda_{22}-\lambda_{23})\mathbf{M}_2-\lambda_{12}\mathbf{M}_1-\lambda_{23}\mathbf{M}, \quad (1b)$$

$$\mathbf{H}_3=\mathbf{H}-(\lambda_{33}-\lambda_{23})\mathbf{M}_3-\lambda_{13}\mathbf{M}_1-\lambda_{23}\mathbf{M}, \quad (1c)$$

where $\mathbf{M}=\mathbf{M}_2+\mathbf{M}_3$.

In the equilibrium case, when $\mathbf{H}=0$, the sublattice magnetizations must be parallel to the total field on them. If we designate the static magnetizations by $\mathbf{M}_i^0$, we see from Eqs. (1) that

$$(\lambda_{12}-\lambda_{13})\mathbf{M}_3^0=0,$$
$$\lambda_{12}\mathbf{M}_1^0+\lambda_{23}\mathbf{M}^0=0,$$
$$\lambda_{13}\mathbf{M}_1^0+\lambda_{23}\mathbf{M}^0=0.$$

Thus, we see that $\lambda_{12}=\lambda_{13}=\lambda$, and that a condition on the static magnetizations is

$$\mathbf{M}_1^0+\beta\mathbf{M}^0=0, \quad (2)$$

where

$$\beta=\lambda_{23}/\lambda. \quad (3)$$

These results agree, of course, with those of Lotgering.

If we choose the direction of the net magnetization (which is the same as that of $\mathbf{M}_1^0$) to be the z direction and write $M_{iz}^0=M_i$, then, in terms of the components, (2) yields

$$M_1+\beta M=M_1+\beta(M_2+M_3)=0, \quad (4)$$

$$M_{2x}^0+M_{3x}^0=M_{2y}^0+M_{3y}^0=0. \quad (5)$$

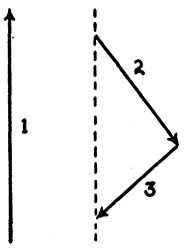

FIG. 1. Relative orientations of the sublattice magnetizations for the case discussed in the text.

II

If $\mathbf{H}\neq 0$, and is in the z direction, the sublattice magnetizations will deviate slightly from their static values, so that, taking account of (5), we can write:

$$M_{1x}=m_{1x}, \quad M_{1y}=m_{1y}, \quad M_{1z}=M_1+m_{1z}, \quad (6a)$$

$$M_{2x}=\bar{M}_x+m_{2x}, \quad M_{2y}=\bar{M}_y+m_{2y}, \quad M_{2z}=M_2+m_{2z}, \quad (6b)$$

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[1] Y. Yafet and C. Kittel, Phys. Rev. 87, 290 (1952).

[2] F. K. Lotgering, Philips Research Repts. 11, 190, 337 (1956).

[3] R. K. Wangsness, Phys. Rev. 91, 1085 (1953); 93, 68 (1954); 98, 1200 (1955).

$$M_{3x}=-\bar{M}_x+m_{3x},\ M_{3y}=-\bar{M}_y+m_{3y},\ M_{3z}=M_3+m_{3z}, \tag{6c}$$

where $\bar{M}_x$ and $\bar{M}_y$ are large static components.

The undamped equations of motion of this system are

$$d\mathbf{M}_i/dt=\gamma_i\mathbf{M}_i\times\mathbf{H}_i, \tag{7}$$

where γ_i is the gyromagnetic ratio of the ith sublattice and the $\mathbf{H}_i$ are given by (1).

If we substitute (6) into (7), use (3) and (4), and keep only first-order terms in the small deviations from the static values, we get

$$m_{1z}\simeq 0, \tag{8a}$$

$$\dot{m}_{2z}/\gamma_2=-\dot{m}_{3z}/\gamma_3 =-\lambda[\bar{M}_x(m_{1y}+\beta m_y)-\bar{M}_y(m_{1x}+\beta m_x)], \tag{8b}$$

$$\dot{m}_{1+}/i\gamma_1=-Hm_{1+}+\lambda(Mm_{1+}-M_1m_+), \tag{8c}$$

$$\dot{m}_{2+}/i\gamma_2=-H(m_{2+}+\bar{M}_+) -\lambda[M_2(m_{1+}+\beta m_+)-\bar{M}_+(m_{1z}+\beta m_z)], \tag{8d}$$

$$\dot{m}_{3+}/i\gamma_3=-H(m_{3+}-\bar{M}_+) -\lambda[M_3(m_{1+}+\beta m_+)+\bar{M}_+(m_{1z}+\beta m_z)], \tag{8e}$$

where $m_{1+}=m_{1x}+im_{1y}$, etc., and we have defined $\mathbf{m}=\mathbf{m}_2+\mathbf{m}_3$. From these equation and (4), one easily finds the general relation:

$$[(\dot{m}_{1+}/\gamma_1)+(\dot{m}_{2+}/\gamma_2)+(\dot{m}_{3+}/\gamma_3)] +iH(m_{1+}+m_{2+}+m_{3+})=0.$$

If the γ_i are all different, the determination of the normal frequencies of the system of essentially eight variables described by (8) is quite tedious and we will not discuss it further. However, we can discuss very easily a simpler case which should give us many of the essential features.

If we now assume that $\gamma_2=\gamma_3=\gamma$, and add (8d) and (8e), we find that the system of equations becomes

$$\dot{m}_z\simeq 0,$$
$$\dot{m}_{1+}/i\gamma_1=-Hm_{1+}+\lambda(Mm_{1+}-M_1m_+),$$
$$\dot{m}_+/i\gamma=-Hm_+-\lambda(Mm_{1+}-M_1m_+),$$

with the use of (4). But these equations, together with (8a), are exactly those describing a system with two antiparallel sublattices and small components transverse to the applied field. Therefore, we can make use of previous results,[3] and say at once that in the limit of strong molecular fields the normal frequency of the lower branch is given by

$$\omega_0=(\gamma^a)_{\text{eff}}H,$$

where $(\gamma^a)_{\text{eff}}$ is the effective gyromagnetic ratio for this example of a triangular state. It is given by

$$(\gamma^a)_{\text{eff}}=(M_1+M)/[(M_1/\gamma_1)+(M/\gamma)], \tag{9}$$

and depends only on the static components of the magnetization which are parallel to the applied field, the large transverse components $\bar{M}_x$ and $\bar{M}_y$ having dropped out of the final result.

The triangular case is, however, significantly different from the usual antiparallel case in that the static magnetization components must satisfy the constraining Eq. (4). If we use (4), we can rid (9) of its apparent dependence on M_1 and M. The final expression for the effective gyromagnetic ratio of the triangular state is then given by

$$(\gamma^a)_{\text{eff}}=(\beta-1)/[(\beta/\gamma_1)-(1/\gamma)]. \tag{10}$$

III

We can profitably compare (1) with γ_{eff} for the usual antiparallel case which, in a corresponding notation and with $\rho=|M_1/M|$, becomes

$$\gamma_{\text{eff}}=(\rho-1)/[(\rho/\gamma_1)-(1/\gamma)].$$

It is easily shown that

$$R=(\gamma^a)_{\text{eff}}/\gamma_{\text{eff}}=(\beta-1)/\{\beta-1+\Delta[(\gamma_{\text{eff}}/\gamma_1)-1]\} =(\beta-1)[(\gamma/\gamma_1)\rho-1]/\{(\rho-1)[(\gamma/\gamma_1)\beta-1]\}, \tag{11}$$

where $\Delta=\beta-\rho$. The case we are considering will generally correspond to $\beta>\rho$.[2] We see from (11) that in general $(\gamma^a)_{\text{eff}}$ can be expected to differ the most from the corresponding antiparallel value when β is much different from ρ and also when the two sublattice gyromagnetic ratios are quite unlike.

Lotgering has found $\beta\simeq 1.3$ for iron and manganese chromites. We can get an estimate of the value of R for these cases if we use the spin-only values of ρ. Using g values of 2.1, 2.0, 1.97 for the Fe, Mn, and Cr sublattices, respectively,[4] we find that,

$$\text{for}\quad FeCr_2O_4,\quad \rho=\tfrac{2}{3},\quad R=1.5;$$
$$\text{for}\quad MnCr_2O_4,\quad \rho=\tfrac{5}{6},\quad R=1.1.$$

Thus, we see that if $FeCr_2O_4$ has a transition from a triangular to an antiparallel state, the corresponding change in g value should be observable.

By using methods similar to those used before for the limit of large molecular fields,[3] one can easily show that the equation of motion for the total magnetization can be taken to be the usual macroscopic equation but with the effective gyromagnetic ratio being used. In other words, one can use

$$d\mathbf{M}/dt=(\gamma^a)_{\text{eff}}\mathbf{M}\times\mathbf{H}$$

for further analysis of the response of the system to applied fields rather than being forced to deal with the more unwieldy set of sublattice equations.

[4] Greenwald, Pickart, and Grannis, J. Chem. Phys. **22**, 1597 (1954); E. W. Gorter, Philips Research Repts. **9**, 295, 321, 403 (1954).