neighbor sites. In MgO the distortion from cubic symmetry produced a variation in the $g$ values of at most a fraction of a percent, while the largest zero-field splitting (11.5 kMc/sec) resulted from a rhombic field. The effect of the distortion in $Y_2O_3$ as compared to MgO is thus about an order of magnitude larger in the zero-field splitting and of the same order of magnitude in $g$.

In summary, the intense part of the spectrum observed in $Y_2O_3$ is accurately described by the spin Hamiltonian of Eq. (1) when the magnetic $z$ axis is along the symmetry axes of the $A$ sites. The remarkably large zero-field splitting of the Kramers' doublets results when the $Cr^{3+}$ enters the lattice substitutionally at the $Y^{3+}$ sites in the crystal field of point group symmetry $S_6$. The weak isotropic line in the spectrum is notable because of the only slight change in the $g$ value.


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# Magnetic Resonance in Canted Ferrimagnets*

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The classical theory of the uniform ($k=0$) modes of a four-sublattice, planar canted ferrimagnet is developed. Two of these modes should lie in the microwave range for reasonable values of applied field, anisotropy, and exchange constants: the normal low-frequency mode familiar in collinear ferrimagnets and the mode excited by longitudinal rf fields at a frequency depending on anisotropy and angle of cant. Observation of this latter mode should allow analysis of ferrimagnetic structures and phase changes.

IN his original theory of magnetic structures in ferrospinels, Néel[1] conceived of phases either with parallel aligned groups of spins or with spins in random paramagnetic arrangements. Yafet and Kittel[2] extended and partly corrected this idea by pointing out that triangular spin arrangements become energetically favorable in the molecular field model when the interactions between spins that are all on tetrahedral $A$ or all on octahedral $B$ sites become comparable with the intersite $AB$ interaction. Lotgering[3] formulated this model in some detail.

These concepts have been further extended by Kaplan[4] and, for the case of antiferromagnets, by Yoshimori,[5] to include generalized helical spin arrangements. Our present knowledge of the stability ranges for these various structures in the presence of anisotropy and in noncubic spinels is still far from complete.

On the experimental side Gorter[6] and more recently Lotgering,[3] Prince,[7] and Dwight and Menyuk[8] have invoked canted sublattices to account for the relatively low saturation magnetization observed in various spinels. Wickham and Goodenough,[9] on the other hand, interpreted the same observations in terms of a partly reversed but collinear $B$ sublattice. Presumably their structure would differ from the Néel model by a physical distortion identifying the lattice sites for reversed spins.

The high-field susceptibility measurements of Jacobs[10] on $Mn_3O_4$ are consistent with a canted lattice structure but in principle depend only on the existence of low effective exchange fields. Neutron-diffraction data[7] for $CuCr_2O_4$ are also consistent with canted lattices, but the evidence has not uniquely determined individual spin alignments.

It is our purpose here to point out that the magnetic resonance spectrum of canted ferrimagnets differs characteristically from that of ferrimagnets with collinear sublattices, and that this difference should permit the detection and analysis of canted structures. Such experiments appear capable of showing definitely the existence of canting and, together with magnetization data, of indicating the spins involved and their relative orientations.

## RESONANCE CONDITIONS

Some aspects of the canted lattice resonance problem have been treated previously by Eskowitz and Wangs-

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ness,[11,12] who concentrated their attention on the effective gyromagnetic ratio for the usual microwave mode. In the present case we will deal with the modes influenced by exchange fields.

Consider a tetragonally distorted, normal spinel at 0°K, with its tetragonal axis magnetically hard (an idealization of the properties of $Mn_3O_4$). The anisotropy assumption is important because the frequency of the resonant mode characteristic of the canted sublattices will be seen to depend critically on this factor. The $A$ sublattice is assumed to divide by lattice distortion into the groups $A_1$, $A_2$, each with magnetization $M_A$, the $B$ sublattice into the groups $B_3$, $B_4$, with magnetization $|M_B|>|M_A|$. The molecular field constants $\lambda$, $\alpha\lambda$, $\alpha'\lambda$, $\alpha'\lambda$, $\beta\lambda$, $\beta'\lambda$, $\beta'\lambda$, describe the interactions between individual $A-B$, $A_1-A_2$, $A_1-A_1$, $A_2-A_2$, $B_3-B_4$, $B_3-B_3$, and $B_4-B_4$ spins, respectively. They are assumed to be negative numbers indicating antiferromagnetic interactions. The vectors $\mathbf{m}_1$, $\mathbf{m}_2$, $\mathbf{m}_3$, $\mathbf{m}_4$ describe the small dynamic displacements of the sublattice magnetization vectors from their static equilibrium values $\mathbf{M}_{1s}$, $\mathbf{M}_{2s}$ in the $z$ direction, $\mathbf{M}_{3s}$ and $\mathbf{M}_{4s}$ lying in the magnetically easy $zx$ plane (Fig. 1). Effective fields to account for anisotropy torques due to deviations $m_{1y}$, $m_{2y}$, $m_{3y}$, and $m_{4y}$ out of this plane, are described by the constants $N_A{}^e$ and $N_B{}^e$ for the $A$ and $B$ sublattices, respectively. For the sake of clarity we will assume a spherical sample with an external field $\mathbf{H}$ applied in the $z$ direction. With a simple triangular spin arrangement, the canted $B$ lattice vectors are determined by the equation[3]

$$\mathbf{H}+\lambda(\mathbf{M}_{1s}+\mathbf{M}_{2s})+\beta\lambda(\mathbf{M}_{3s}+\mathbf{M}_{4s})=0. \tag{1}$$

It follows that the four equilibrium vectors are given by

$$\begin{aligned} \mathbf{M}_{1s}&=\mathbf{i}_z M_A, \quad \mathbf{M}_{3s}=\mathbf{i}_z M_{Bz}+\mathbf{i}_x M_{Bx},\\ \mathbf{M}_{2s}&=\mathbf{i}_z M_A, \quad \mathbf{M}_{4s}=\mathbf{i}_z M_{Bz}-\mathbf{i}_x M_{Bx}. \end{aligned} \tag{2}$$

The equations of motion for the individual sublattices,

$$d\mathbf{M}_i/dt=\gamma_i\mu_0(\mathbf{M}_i\times\mathbf{H}_i), \quad i=1,2,3,4, \tag{3}$$

are now solved with the effective fields

$$\begin{aligned} \mathbf{H}_1&=\mathbf{H}+\lambda(\mathbf{M}_3+\mathbf{M}_4)+\alpha\lambda\mathbf{M}_2+\alpha'\lambda\mathbf{M}_1-\mathbf{i}_y N_A{}^e m_{1y},\\ \mathbf{H}_2&=\mathbf{H}+\lambda(\mathbf{M}_3+\mathbf{M}_4)+\alpha\lambda\mathbf{M}_1+\alpha'\lambda\mathbf{M}_2-\mathbf{i}_y N_A{}^e m_{2y},\\ \mathbf{H}_3&=\mathbf{H}+\lambda(\mathbf{M}_1+\mathbf{M}_2)+\beta\lambda\mathbf{M}_4+\beta'\lambda\mathbf{M}_3-\mathbf{i}_y N_B{}^e m_{3y},\\ \mathbf{H}_4&=\mathbf{H}+\lambda(\mathbf{M}_1+\mathbf{M}_2)+\beta\lambda\mathbf{M}_3+\beta'\lambda\mathbf{M}_4-\mathbf{i}_y N_B{}^e m_{4y}. \end{aligned} \tag{4}$$

Eight independent linear equations for small-angle excitation can be chosen from the ten that are available and, written in terms of the convenient variables $m_{1x}-m_{2x}$, $m_{1y}-m_{2y}$, $m_{1x}+m_{2x}$, $m_{1y}+m_{2y}$, $m_{3x}+m_{4x}$, $m_{3y}+m_{4y}$, $m_{3y}-m_{4y}$, and $m_{3z}+m_{4z}$, lead to the secular determinant

$$\begin{vmatrix} \Omega_A & H+2\lambda M_{Bz}+2\alpha\lambda M_A+N_A{}^eM_A & 0 & 0 & 0 & 0 & 0 & 0\\ -H-2\lambda M_{Bz}-2\alpha\lambda M_A & \Omega_A & 0 & 0 & 0 & 0 & 0 & 0\\ 0 & 0 & \Omega_A & H+2\lambda M_{Bz}+N_A{}^eM_A & 0 & -2\lambda M_A & 0 & 0\\ 0 & 0 & -H-2\lambda M_{Bz} & \Omega_A & 2\lambda M_A & 0 & 0 & 0\\ 0 & 0 & 0 & -2\lambda M_{Bz} & \Omega_B & (N_B{}^e-2\beta\lambda)M_{Bz} & 0 & 0\\ 0 & 0 & 2\lambda M_{Bz} & 0 & 2\beta\lambda M_{Bz} & \Omega_B & 0 & 0\\ 0 & 0 & 0 & 0 & 0 & 0 & \Omega_B & -2\beta\lambda M_{Bx}\\ 0 & 0 & 0 & 0 & 0 & 0 & -N_B{}^eM_{Bx} & \Omega_B \end{vmatrix}, \tag{5}$$

where $\Omega_A=-i\omega/\gamma_A\mu_0$ and $\Omega_B=-i\omega/\gamma_B\mu_0$. Setting the determinant to zero yields four resonant frequencies, listed here in their probable order of magnitude in low fields:

$$\begin{aligned} \omega_1&=-\gamma_e\mu_0H+\omega_a,\\ \omega_2&=-\gamma_B\mu_0M_{Bx}[2|\beta\lambda|N_B{}^e]^{\frac{1}{2}},\\ \omega_3&=(\gamma_e-\gamma_A-\gamma_B)\mu_0H-2\lambda\mu_0(\gamma_BM_A+\gamma_AM_{Bz})+\omega_a',\\ \omega_4&=\gamma_A\mu_0[H+2\lambda(M_{Bz}+\alpha M_A)]^{\frac{1}{2}}\\ &\quad\times[H+2\lambda(M_{Bz}+\alpha M_A)+N_A{}^eM_A]^{\frac{1}{2}}, \end{aligned} \tag{6}$$

where $\gamma_e=\gamma_A\gamma_B(M_A+M_{Bz})(\gamma_BM_A+\gamma_AM_{Bz})^{-1}$, and $\omega_a$, $\omega_a'$ are terms depending on the anisotropy constants. Their precise forms are complicated in general, as Wangsness[13] has shown, and are not of great importance in the present development.

In the familiar microwave mode $\omega_1$, the sublattice vectors precess essentially in their equilibrium configuration. In the normal infrared mode $\omega_3$, the combined $B$ lattice precesses in the negative sense and at an angle with respect to the $A$ lattice precessing in the same sense. The new intermediate mode $\omega_2$ is of particular interest. It corresponds to the two $B$ sublattices, both precessing symmetrically in the positive sense but in phase opposition, while the $A$ sublattice remains immobile (Fig. 2). For convenience, this mode will be referred to as a "$Y$ mode." In analogy with degenerate antiferromagnetic resonances,[14] its frequency is determined by a combination of a molecular field torque alternating in time with an anisotropy torque as the magnetization vectors swing in and out of the $zx$ plane.

The anisotropy factor $N_B{}^e$ is given by

$$N_B{}^e=-2K_{1B}/\mu_0M_B{}^2, \tag{7}$$

where $K_{1B}$ is the axial anisotropy constant relating to the $B$ sublattices alone. This is to be distinguished from the over-all anisotropy constant $K_1$, determined

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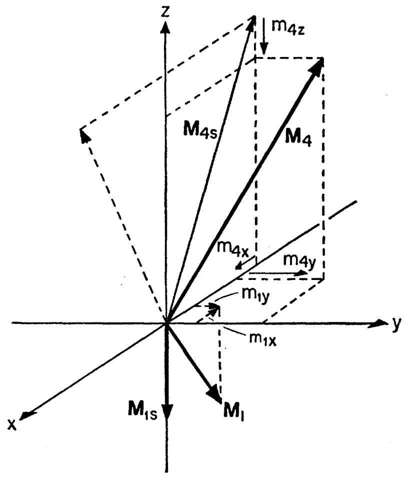


FIG. 1. Definition of dynamic deviations of $\mathbf{M}_1$, $\mathbf{M}_4$, two of the four magnetization vectors, from their equilibrium values $\mathbf{M}_{1s}$, $\mathbf{M}_{4s}$.

from torque measurements on the net magnetization. Using the values $|\beta\lambda|=300$, $M_{Bx}=2.5\times10^5$ amp/m, $N_B^e=1$, and the free-spin value for $\gamma_B$, gives a figure of about 7 cm$^{-1}$ for the $Y$-mode frequency.

The final mode $\omega_4$ corresponds to the two $A$ sublattices precessing in phase opposition. In this mode, molecular field torques are brought into play for both $x$ and $y$ displacements.

### FIELD DEPENDENCE

Figures 3(a) and 3(b) indicate the frequency versus field behavior of all the sublattice modes. Here the sense of each resonance mode is defined by the sense of rotation of the individual moments projected onto the $xy$ plane. For example, the normal microwave mode projections move in a positive sense for $\gamma_e<0$, corresponding to a clockwise precession seen by looking along the field direction. The broken lines indicate mode branches that cannot be excited (compare next section).

The form of these curves can be understood by considering the magnetic phase changes produced by an external field. In a canted ferrimagnet, the over-all magnetization $M_s=(2M_A+2M_{Bz})$ in the field direction may be either parallel or antiparallel to the resultant of the two canted sublattices, depending on whether $\beta$ is less or greater than $(1+H/2\lambda M_A)$. Assuming that the Yafet-Kittel model[2] of planar spin arrangements is valid, it follows from Eq. (1) that for a $Y$ configuration in low fields [Fig. 4(a)], an increase in $z$ field will decrease the angle between the canted vectors until at $H=2|\lambda|(|\beta M_B|-|M_A|)$ all vectors are collinear. For $H>2|\lambda|(|M_B|-|\alpha M_A|)$, the $A$ sublattice vectors become canted to form an inverted $Y$ configuration, producing antiparallel $A$ vectors at $H=2|\lambda M_B|$ and passing to a completely collinear structure for $H>2|\lambda|(|M_B|+|\alpha M_A|)$. If an inverted $Y$ configuration

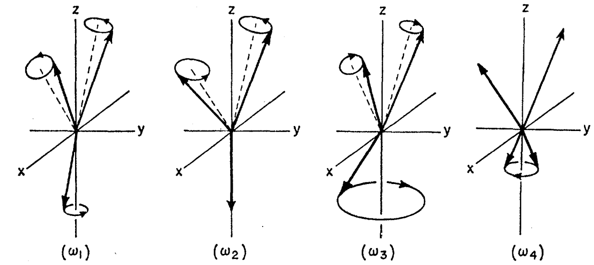


FIG. 2. The four resonant modes characteristic of canted ferrimagnets in a $Y$ configuration.

exists in low fields [Fig. 4(b)], the $B$ lattice vectors will become antiparallel at $H=2|\lambda M_A|$ and give a completely collinear structure at $H=2|\lambda|(|M_A|+|\beta M_B|)$. In each case the normal microwave resonance frequency $\omega_1$ increases almost linearly with field. For an initial $Y$ configuration the infrared mode frequency $\omega_3$ varies linearly with field, with negative slope if a canted phase is stable and positive slope in collinear phases. For the inverted case, the slope is always positive with the frequency passing through zero near $H=2|\lambda M_A|$.

The field dependence of the $Y$ mode resonances is implicit in Eq. (1), which determines the canting angle and hence the equilibrium value of $M_{Bx}$:

$$M_{Bx}=(M_B^2-M_{Bz}^2)^{\frac{1}{2}},\quad \frac{dM_{Bx}}{dH}=\frac{1}{2\beta\lambda}\frac{M_{Bz}}{(M_B^2-M_{Bz}^2)^{\frac{1}{2}}}. \quad (8)$$

Thus, if the anisotropy torque for $y$ displacements is independent of the canting angle, as our simplified model has supposed, a $Y$ resonance frequency will decrease monotonically with increasing $z$ field, tending to zero for collinear $B$ sublattices. At higher fields, this mode takes on a form analogous to $\omega_4$ of Fig. 2, with a frequency

$$\omega_2'=\gamma_B\mu_0(H+2\lambda(M_{Az}+\beta M_B))^{\frac{1}{2}} \times(H+2\lambda(M_{Az}+\beta M_B)+N_B^eM_B)^{\frac{1}{2}}. \quad (9)$$

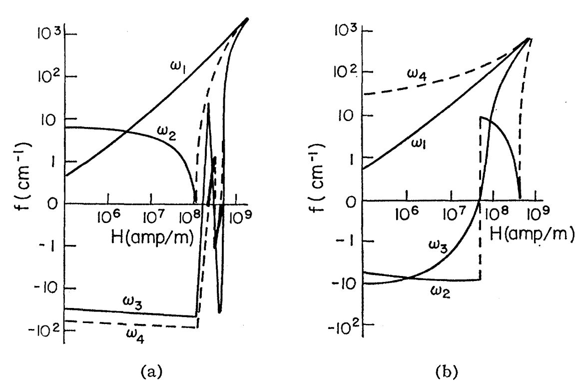


FIG. 3. Frequency vs field for resonances in canted ferrimagnets (schematic); (a) $Y$ configuration in zero field ($\alpha=0.6, \beta=0.8$); (b) inverted $Y$ configuration in zero field ($\alpha=0.6$, $\beta=1.2$). In each case $M_A=1.0\times10^5$ amp/m, $M_B=3.0\times10^5$ amp/m, $|\beta\lambda|=300$.

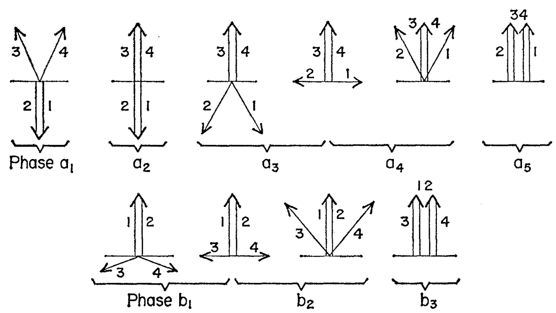


FIG. 4. Equilibrium sublattice configurations in an external field, according to the Yafet-Kittel model[2]: (a) $M_A/M_B<\beta<1$; (b) $\beta>1$.

An inverted $Y$ resonance, on the other hand, will increase in frequency as the $z$ field increases and the sublattices diverge, reach a maximum at $H=2|\lambda M_A|$, and then decrease to zero. The sign of the initial slope of $|\omega_2|$ vs $H$ is sufficient, therefore, to distinguish between the two configurations. For the $Y$ configuration that changes to an inverted $Y$ structure at intermediate fields, the original mode frequency $\omega_4$ will take the form

$$\omega_4'=\gamma_A\mu_0 M_{Az}(2|\alpha\lambda|N_A{}^e)^{\frac{1}{2}} \quad (10)$$

as the $A$ sublattice vectors diverge.

## RESONANCE EXCITATIONS

In collinear ferrimagnets the usual microwave and infrared modes are excited by circularly polarized rf fields whose magnetic field vectors rotate in the $xy$ plane in the positive and negative senses, respectively. It follows from symmetry, however, that the $Y$ and inverted $Y$ resonances will respond only to an oscillating rf field in the $z$ direction. The excitation symmetry will thus characterize the resonance. Its intensity is related to the intrinsic static susceptibility $\chi_{ms}$ by the Kramers-Krönig formula

$$\chi_{ms}=\frac{dM_s}{dH}=\frac{1}{|\beta\lambda|}=\int_0^\infty \chi_m''d\ln\omega. \quad (11)$$

For typical values $|\beta\lambda|=300$, $H=10^6$ amp/m, and $M_s=5\times10^4$ amp/m, the intensity ratio of a $Y$ mode to the normal microwave mode is predicted to be of the order of 0.06. This should be detectable with present microwave techniques, provided the line width is manageable, i.e., $<5000$ oe ($0.4\times10^6$ amp/m). (In this connection it must be realized that the resonance linewidth $\Delta H$, determined at fixed frequency, is a simply defined measure of a relaxation controlled linewidth $\Delta\omega$ at fixed field only over linear portions of the frequency-vs-field diagram.) From symmetry again, the high-frequency mode $\omega_4$ and its analogue $\omega_2'$ cannot be excited in a perfect crystal by uniform fields.

The application of high fields to a canted ferrimagnet produces magnetic phase changes with corresponding changes in mode descriptions, as we have indicated. Changing the temperature can produce even greater variations[2,3] that could be followed, in principle, by careful intensity measurements on both the normal microwave mode and the $Y$ modes. As an example, the transition from a canted to a collinear phase at a temperature $T_Y$ would reduce the $Y$ mode frequency to zero, leaving only the normal mode between $T_Y$ and the Curie temperature $T_C$ with an intensity proportional to the net magnetization. On the other hand, the simultaneous disappearance of the over-all magnetization, the normal mode, and the $Y$ mode resonances would indicate a transition to a mixed antiferromagnetic, paramagnetic state.

## CONCLUSION

The $Y$ mode resonances predicted for canted ferrimagnets are unusual both in their manner of excitation and in the dependence of their frequency on field and temperature. In addition to their potential use in the analysis of magnetic sublattice structures, they offer a possibility for resonances that could be manipulated by variations of anisotropy, field, or temperature to lie at frequencies in the microwave-to-infrared region. An extension of this theory to cover helical spin arrangements would be of great interest. From symmetry alone it follows that in a four-sublattice helical array two modes exist that are the analogues of the $Y$ modes of the present case. These modes, too, will be excited by longitudinal rf fields at frequencies determined by anisotropy-exchange products. Consideration of relaxation effects, which can be expected to depend on the type of mode and hence on the particular energy terms involved, suggests a further means of probing the structure of ferrimagnets.