RESONANT AND NONRESONANT MICROWAVE LOSSES IN $TmFeO_3$

F. B. Hagedorn, E. M. Gyorgy, R. C. LeCraw, J. C. Hensel, and J. P. Remeika
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received 24 May 1968)

Experimental and theoretical results are presented which show that a pair of closely spaced resonant loss peaks at microwave frequencies in $TmFeO_3$ can be transformed into a single nonresonant loss peak by rotation of the applied field through as little as 3 deg. These findings provide the basis for understanding some previously puzzling microwave absorption measurements in the rare-earth orthoferrites.

Recent microwave absorption measurements[1] in several rare-earth orthoferrites have shown loss peaks which have not been adequately explained. Torque measurements[2] on these materials show that the crystalline anisotropy has components of both twofold and fourfold symmetry and that the twofold component changes sign in the temperature range where the loss peaks are observed. The orientation of the spontaneous magnetic moment changes from one crystalline axis to another over the same temperature range. It was tentatively suggested initially[1] that the microwave loss peaks were associated with maxima in the susceptibility arising from the reorientation of the magnetic easy axis.

In a subsequent effort to interpret the microwave results,[1] Shane[3] extended Herrmann's[4] analysis of resonance in canted antiferromagnets by including the fourfold anisotropy terms. While Shane's calculation leads to a reasonable result for the temperature dependence of the applied field required for resonance, two absorption peaks as a function of field are predicted[3] while only a single peak was reported.[1] Moreover, Shane's absorption peaks are associated with resonances, whereas the early observations[1] indicated that the phase relationship between the real and imaginary parts of the susceptibility was not characteristic of resonance. We report in this Letter some new results which resolve these questions and which clarify the nature of the microwave losses reported previously[1] in $TmFeO_3$.

We have found that small changes in the direction of the external magnetic field (H_{dc}) can lead to dramatic changes in the microwave susceptibility of $TmFeO_3$ at temperatures near the spin reorientation range. (In this range the spontaneous moment rotates from the a to the c direction with increasing temperature.) Specifically, we find that rotation of H_{dc} through three degrees transforms a pair of closely spaced resonant absorption peaks into a single, nonresonant absorption peak. This effect is illustrated in Figs. 1(a)-1(c), where we show tracings of the experimentally observed real (χ') and imaginary (χ'') parts of the susceptibility plotted as a function of H_{dc}. A single crystal of $TmFeO_3$ mounted in a TE_{101} rectangular cavity operated at 57.7 Gc/sec was used for these measurements. The temperature was 81°K, and the orientation of H_{dc} in the ac plane was (a) along the c axis, (b) 1.5° away from the c axis, and (c) 3° away from the c axis.

It is seen that the double peaks in χ'' when H_{dc} is parallel to the c axis [Fig. 1(a)] merge into a single peak when H_{dc} is 3° from the c axis [Fig. 1(c)]. The correspondence in Fig. 1(a) of the zero crossings in χ' with the peaks in χ'' identifies these as resonances. On the other hand, χ' and χ'' pass through positive maxima simultaneously in Fig. 1(c), leading to the conclusion that the well-defined loss peak seen there is not due to a simple resonance phenomenon.

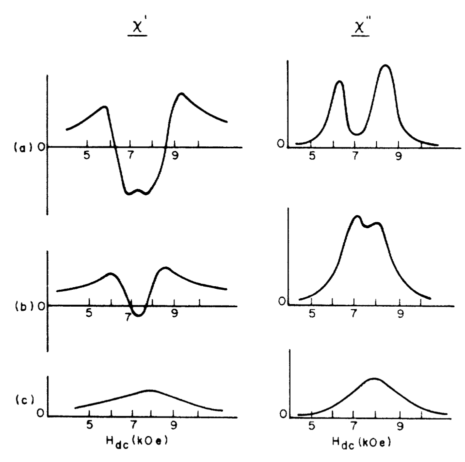

FIG. 1. Plots of the experimentally measured real (χ') and imaginary (χ'') susceptibilities for $TmFeO_3$ at 81°K as a function of field (H_{dc}). In (a) H_{dc} was along the c axis. In (b) and (c), H_{dc} was in the ac plane, 1.5° and 3°, respectively, from the c axis. The ordinate scales for each χ' and χ'' are arbitrary, but remain the same for the different angles.

These effects were predicted in an analysis[5] of resonance and susceptibility in canted antiferromagnets. The analysis was stimulated by the observation[1] of single absorption peaks in $TmFeO_3$ and was specifically directed toward those canted antiferromagnets whose spontaneous moments (m_S) undergo reorientations with temperature change. (Torque measurements[2] have shown that m_S in $TmFeO_3$ rotates smoothly from the *a* axis at 81.5°K to the *c* axis at 94°K, in the absence of an external magnetic field.) Our analysis includes a fourfold crystalline anisotropy term to account for the reorientation. Otherwise, we use a two-sublattice model similar to that of Herrmann.[4] However, we have extended our analysis beyond that of Herrmann and of Shane[3] (who also used a fourfold anisotropy term) by including phenomenological damping, by allowing H_{dc} to have any orientation in the *ac* plane, and by obtaining complete expressions for the susceptibility.

The method of analysis is similar to that followed by Herrmann[4] and starts from a general expression for the magnetic free energy of the two-sublattice system. Equilibrium orientations for the two sublattices are found by minimizing the total energy. After expanding around the equilibrium orientation, linearized solutions of a phenomenological equation of motion are found. From these solutions, expressions for the susceptibility and resonance frequency are obtained.

The details of the calculation, along with general expressions for the resonance frequency and for the complex susceptibility, will be published elsewhere. We will show here some results obtained by evaluating our expressions with parameter values which are appropriate for $TmFeO_3$ at 81°K. In Fig. 2, we plot the calculated resonance frequency f_r as a function of H_{dc} with the angle (in degrees) between H_{dc} and the *c* axis as a parameter. (Shane's[3] calculation is for the cases where this angle is either 0° or 90°.) It is seen that the minimum f_r increases rapidly from zero as H_{dc} is moved away from the *c* axis, reaching a value of 80 Gc/sec for 5°.

Our analysis shows that the susceptibility changes character when realigning H_{dc} causes the minimum f_r to pass through the fixed operating frequency. From Fig. 2, it is apparent that the minimum f_r is near 60 Gc/sec (the operating frequency for Fig. 1 was 57.7 Gc/sec) when H_{dc} is about 2° from the *c* axis. For larger values of this angle, resonance will not occur at all as the magnitude of H_{dc} is changed, provided that the operating frequency remains unchanged. In such cases, the minimum f_r is always larger than the operating frequency. However, anomalies in χ' and χ'' still occur even though the minimum f_r is always larger than the operating frequency. For this case, the anomalies will occur at the same location on the field axis for any value of the operating frequency less than the minimum f_r. This type of behavior was reported in Ref. 1.

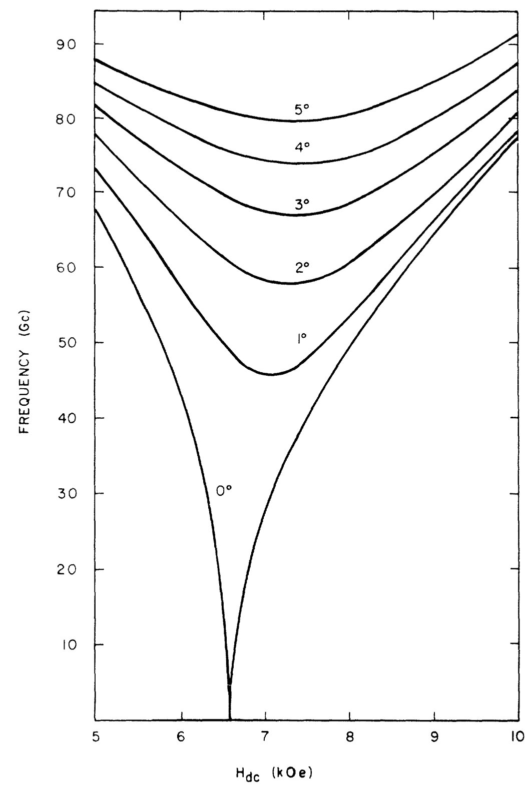

FIG. 2. Calculated resonance frequencies for a material similar to $TmFeO_3$ near 81°K as a function of field (H_{dc}). The parameter is the angle in degrees between H_{dc} and the *c* axis, with H_{dc} in the *ac* plane.

Calculated plots of χ' and χ'' as a function of H_{dc} are shown in Fig. 3, where we have used the same material parameter values as in Fig. 2. A damping constant was selected to approximate the experimental linewidths in Fig. 1. All the essential features of the experimental results shown in Fig. 1 are seen in Fig. 3. Most of the detailed structure, such as the local maximum in χ' between the two zero crossings in Fig. 1(a), can also be identified with similar structure in Fig. 3. It should also be noted in Fig. 3(d) that the calculated χ' and χ'' simultaneously exhibit single peaks for sufficiently large misalignment

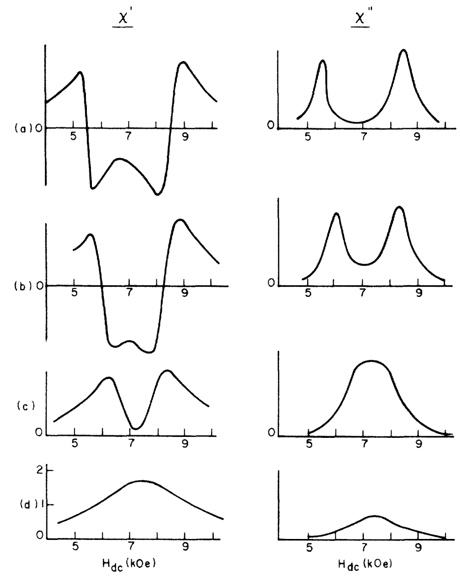

FIG. 3. Calculated values of χ' and χ'' for the parameter values used in Fig. 2. The angles in the *ac* plane between H_{dc} and the *c* axis are (a) 0°, (b) 1°, (c) 2°, and (d) 3°. The ordinate scales for each χ' and χ'' are arbitrary, but remain the same for the different angles.

between H_{dc} and the *c* axis, anomalies which are definitely nonresonant.

The major source of the discrepancies which do appear between Figs. 1 and 3 is probably inhomogeneity in the direction of the *c* axis of the crystal, which was a flat plate with the *c* axis normal to the surface. The crystal was probably mechanically strained either during mounting or during the cooling process, thereby introducing some bending. With inhomogeneity one would expect the experimentally observed plots of χ' and χ'' to be weighted averages of the calculated values that occur within the distribution of angles. As a result the very sharp features of Fig. 3(a) are not observed experimentally. Another important result of this averaging process is that one will not be able to resolve the double peaks in χ'' below a certain operating frequency, depending on the degree of inhomogeneity.

The crystal used in the present work has a linewidth[1] of 0.37°K at 17 Gc/sec. No evidence of splitting in χ'' is observed in this crystal at 17 Gc/sec, while only a small notch in χ'' appears with the optimum-field orientation at 35 Gc/sec. Most crystals exhibit broader absorption peaks, thus exhibiting only single peaks even at 57.7 Gc/sec. These latter observations indicate the presence of crystal inhomogeneities and provide a basis for a satisfactory understanding of the initial results,[1] where only single absorption peaks were reported.

One important conclusion we draw from these results is that the locations on the field axis of the loss peaks in Ref. 1 do not uniquely determine the material constants as a function of temperature. This point is clearly illustrated in Figs. 3(a) and 3(b), where the separation between the double peaks in χ'' is decreased by moving H_{dc} away from the *c* axis by 1°. A similar decrease in separation occurs if one calculates χ'' using a larger value for the symmetric exchange constant of $TmFeO_3$. As a consequence, detailed knowledge about angular inhomogeneity in the crystal will be required in order to use the microwave results for making accurate determinations of the material constants.

A second important aspect of the results contained in Fig. 1 is that we exhibit there a transformation between gyromagnetic resonance response and essentially quasistatic response to an alternating magnetic field at microwave frequencies. This transformation is brought about by a small reorientation of H_{dc}. Very high resonance frequencies are typical in antiferromagnetic resonance because of the dominating role played by the exchange interaction. In the canted antiferromagnets which exhibit spin reorientation as a function of temperature, the several large terms which make up the resonance frequency expression[3-5] can be made to cancel each other entirely under certain very specialized conditions with quite modest values of H_{dc}. The cancellation is extremely sensitive to the alignment of H_{dc}, however, and allows one for the first time to show the connection between resonant and nonresonant magnetic losses in the same materials by simply rotating H_{dc} through a small angle.

---

[1]R. C. LeCraw, R. Wolfe, E. M. Gyorgy, F. B. Hagedorn, J. C. Hensel, and J. P. Remeika, J. Appl. Phys. 39, 1019 (1968).

[2]E. M. Gyorgy, J. P. Remeika, and F. B. Hagedorn, J. Appl. Phys. 39, 1369 (1968).

[3]J. R. Shane, Phys. Rev. Letters 20, 728 (1968).

[4]G. F. Herrmann, J. Phys. Chem. Solids 24, 597 (1963).

[5]F. B. Hagedorn and E. M. Gyorgy, Bull. Am. Phys. Soc. 13, 389 (1968). A complete report will be published elsewhere.