Radio-Frequency Susceptibilities of Weak Ferromagnets: $MnCO_3$ and NiF_2

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The rf susceptibilities of weakly ferromagnetic $MnCO_3$ and NiF_2 were calculated and measurements were performed at about 10 and 20 Mc/sec. For both crystals we find that whenever the rf magnetic field is perpendicular to the ferromagnetic component and also in the plane which contains the magnetic moments of the sublattices, the susceptibility, χ', is large provided only small static fields, H, are present. However, when the rf field is perpendicular to the plane which contains the magnetic moments, χ' is small regardless of the magnitude or orientation of H. For $MnCO_3$ a maximum appears in χ' which is a function of the orientation and magnitude of H and which is related to an effective magnetic field in the (111) plane of about 60 G. The experimental results of $MnCO_3$ are in good agreement with the theory and are characteristic of a weak ferromagnet with anisotropic spin-spin interaction and a small anisotropy field in the plane which contains the spins. Similar results were obtained for NiF_2 with a maximum in χ' which occurs when H in the (001) plane is approximately 0.7 kG. This and other results of NiF_2 do not agree with the theory and are not understood at present.

1. INTRODUCTION

CRYSTALS of $MnCO_3$ and NiF_2 are antiferromagnets with a weak ferromagnetic component perpendicular to the antiferromagnetic vector $\mathbf{l}=\mathbf{S}_1-\mathbf{S}_2$.[1-8] These crystals represent two basically different cases of weak ferromagnetism. In $McCO_3$ anisotropic spin-spin interaction is responsible for the ferromagnetic component and in NiF_2 it is the single ion anisotropy. $MnCO_3$ has a rhombohedral crystal structure (Fig. 1) and becomes a weak ferromagnet below approximately 32°K. The magnetic moments of the two sublattices and the ferromagnetic component are in the (111) plane. The latter is due to the anisotropic spin-spin interaction[6] which originates from a term $\sum_{i>k}\mathbf{d}_{ik}\cdot\mathbf{S}_i\times\mathbf{S}_k$ in the Hamiltonian. Due to this ferromagnetic component the magnetic field $4\pi M$ is about 50 G at 20.4°K. The anisotropy in the (111) plane has a sixfold symmetry and is very small. The effective anisotropy field is about 0.3 kG and the preferred direction of the ferromagnetic component in the (111) plane has not yet been found experimentally. For magnetic fields much larger than 0.3 kG in the (111) plane, the ferromagnetic component is parallel to the applied magnetic field.

NiF_2 has a rutile-type crystal structure (Fig. 2) and becomes a weak ferromagnet below about 73°K. The magnetic moments of the two sublattices and the ferromagnetic component are in the (001) plane and the latter is parallel to the a direction (or the b direction) and its value $4\pi M$ is about 200 G. There are two inequivalent cation sites in NiF_2, the body center and the corner sites in Fig. 2. The crystalline electric fields around these sites have an orthorhombic symmetry and are the same except that the principal axes are rotated by 90° in the (001) plane. The crystal field gives then a term $(S_{1a}S_{1b}-S_{2a}S_{2b})$ in the Hamiltonian which is responsible for the ferromagnetic component.[3] According to Moriya,[5] the effective anisotropy field in the (001) plane is approximately 25 kG, and, therefore, we expect that magnetic fields, which are small compared to 25 kG, do not change appreciably the direction of the ferromagnetic moments and the antiferromagnetic ordering.

We are dealing here with two basically different canting mechanisms, and the effective anisotropy fields in the planes of the two crystals which contain the spins are different by two orders of magnitude. Therefore, we expect that the rf susceptibilities are different but typical for the two types of weak ferromagnetism. We have calculated the rf susceptibilities for weak ferromagnets of the $MnCO_3$ and the NiF_2 type as a function of the magnitude and orientation of a static magnetic field with respect to the crystallographic axes. We also performed relative rf susceptibility measurements at about 10 and 20 Mc/sec.

2. THEORY

$MnCO_3$

In the two sublattice model the free energy of $MnCO_3$ is written approximately as

$$E=\frac{N}{2}[-S^2[J\mathbf{S}_1\cdot\mathbf{S}_2+d\mathbf{k}\cdot(\mathbf{S}_1\times\mathbf{S}_2)+K(S_{1z}^2+S_{2z}^2)-\delta\mathbf{h}\cdot(\mathbf{S}_1+\mathbf{S}_2)]. \quad (1)$$

$\mathbf{S}_1$ and $\mathbf{S}_2$ are unit vectors parallel to the sublattice magnetizations; $\mathbf{k}$ is a unit vector parallel to the [111] direction; $\mathbf{h}$ is a unit vector parallel to the applied

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[2] I. E. Dzialoshinskii, Zh. Eksperim. i. Teor. Fiz. **33**, 1454 (1957) [translation: Soviet Phys.—JETP **6**, 1120 (1958)].
[3] T. Moriya, Phys. Rev. **117**, 635 (1960).
[4] I. E. Dzialoshinskii, Zh. Eksperim. i. Teor. Fiz. **32**, 1547 (1957) [translation: Soviet Phys.—JETP **5**, 1259 (1957)].
[5] T. Moriya, chapter on "Weak Ferromagnetism" (to be published in a book by Suhl and Rado).
[6] T. Moriya, Phys. Rev. **120**, 91 (1960).
[7] A. S. Borovik-Romanov and M. P. Orlova, Zh. Eksperim. i. Teor. Fiz. **31**, 579 (1959) [translation: Soviet Phys.—JETP **4**, 531 (1957)].
[8] A. S. Borovik-Romanov, Zh. Eksperim. i. Teor. Fiz. **36**, 766 (1959) [translation: Soviet Phys.—JETP **9**, 539 (1959)].

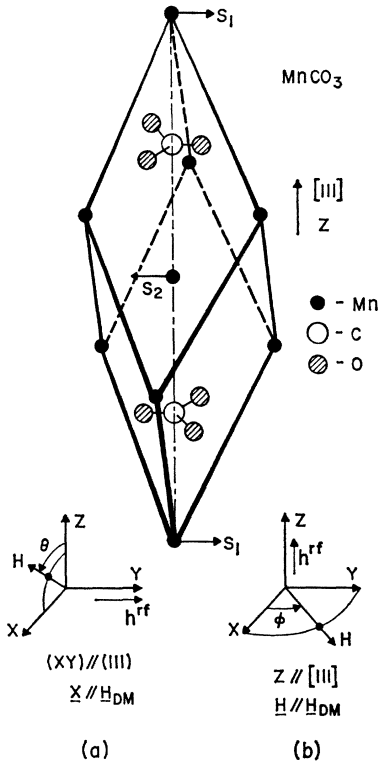

FIG. 1. The crystal structure of $MnCO_3$. Parts (a) and (b) are two experiments where the relation between the static field H and the rf field $h^{(rf)}$ with respect to the crystallographic directions is shown.

magnetic field; J is the effective exchange energy; d is the effective anisotropic spin-spin energy; K is the uniaxial anistoropy energy; δ is equal to $g\mu_B H/S$. For the time being we have neglected the anisotropy in the (111) plane. The rf susceptibility can be calculated from the equations of motion

$$d\mathbf{M}_i/dt=\gamma\mathbf{M}_i\times\mathbf{H}_i, \tag{2}$$

where $\mathbf{H}_i$ is the effective field in the ith sublattice and it is calculated from

$$\mathbf{H}_i=-(\partial E/\partial\mathbf{M}_i). \tag{3}$$

We consider the case for which the external field is always larger than the effective anisotropy field in the (111) plane. The ferromagnetic component is then always parallel to the static field, $\mathbf{H}$, in the (111) plane, the direction of which we call the x direction. The z direction is parallel to the [111] direction and we assume that $\mathbf{H}$ is applied anywhere in the xz plane. The effective fields are

$$\mathbf{H}_1=H_E\mathbf{S}_2-H_{DM}(\mathbf{k}\times\mathbf{S}_2)+H_A S_{1z}\mathbf{k}-\mathbf{H}-\mathbf{h}^{(rf)}, \tag{3a}$$

$$\mathbf{H}_2=H_E\mathbf{S}_1+H_{DM}(\mathbf{k}\times\mathbf{S}_1)+H_A S_{2z}\mathbf{k}-\mathbf{H}-\mathbf{h}^{(rf)}, \tag{3b}$$

where the exchange field H_E is $SJ/g\mu_B$; the canting field H_{DM} is dH_E/J; the uniaxial anisotropy field parallel to the [111] direction H_A is $2KH_E/J$; the static magnetic field is $\mathbf{H}$; and the rf magnetic field is $\mathbf{h}^{(rf)}$. We assume that the magnetic moments make small vibrations around their equilibrium position. The equilibrium position is obtained by minimizing the total free energy with respect to the position of the magnetic moments of the two sublattices. Two coordinate systems are introduced with the z' and z'' axes parallel to the equilibrium position of $\mathbf{S}_1$ and $\mathbf{S}_2$, respectively, and in the equations of motion the components of $\mathbf{S}_1$ are written in the $(x'y'z')$ system and those of $\mathbf{S}_2$ in the $(x''y''z'')$. The equations of motion are linearized and the free-energy parameters d/J, K/J, and $g\mu_B H/JS$ are retained up to the second order. The four equations in $S_{1x'}$, $S_{1y'}$, $S_{2x''}$, and $S_{2y''}$ represent the spin wave motion. These equations are solved for the components of $\mathbf{S}_1$ and $\mathbf{S}_2$ as a function of the applied rf field. The components of the vectors $\mathbf{S}$ of the primed systems are transferred back to the components of the unprimed system and summed, and from this one obtains the real part of the susceptibilities in (xyz) which we call χ_{ij}'.

In order to include a small ansiotropy field in the (111) plane, we add a term $(N/36)g\mu_B SH_A'\cos 6\varphi$ to Eq. (1). The anisotropy field in the (111) plane, H_A', is almost parallel to the sublattice magnetization in each sublattice. In our case it is parallel and antiparallel to the y axis. When this is done one obtains for the susceptibilities:

$$\chi_{xx}'=\chi_0\frac{\omega_-^2}{\omega_-^2-\omega^2}, \tag{4}$$

$$\chi_{yy}'=\chi_0\frac{H_{DM}+H_x}{H^*+H_x}\frac{\omega_+^2}{\omega_+^2-\omega^2}+\chi_0\frac{H_z^2}{2H_EH_A+H_z^2}\frac{\omega_-^2}{\omega_-^2-\omega^2}, \tag{5}$$

$$\chi_{zz}'=\chi_0\frac{\omega_+^2}{\omega_+^2-\omega^2}, \tag{6}$$

$$\chi_{xy}'=\chi_{yx}'^*=i\chi_0\left(\frac{\omega}{\gamma}\right)\frac{H_z}{2H_EH_A+H_z^2}\frac{\omega_-^2}{\omega_-^2-\omega^2}, \tag{7}$$

$$\chi_{yz}'=\chi_{zy}'^*=i\chi_0\left(\frac{\omega}{\gamma}\right)\frac{1}{H^*+H_x}\frac{\omega_+^2}{\omega_+^2-\omega^2}, \tag{8}$$

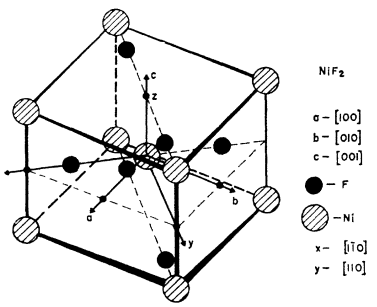

FIG. 2. The crystal structure of NiF_2 and the orientation of (xyz) with respect to the crystal axes.

$$\chi_{xz}'=\chi_{zx}'=\chi_0\gamma^2H_xH_z\frac{\omega^2}{(\omega_+^2-\omega^2)(\omega_-^2-\omega^2)}, \quad (9)$$

with

$$\chi_0=M/H_E, \quad (10)$$

$$\omega_+^2=\gamma^2[2H_EH_A'+H_x(H_x+H_{DM})], \quad (11)$$

$$\omega_-^2=\gamma^2[2H_EH_A+H_{DM}(H_{DM}+H_x)+H_z^2], \quad (12)$$

$$H^*=2H_EH_A'/H_{DM}, \quad (13)$$

where ω_+^2 and ω_-^2 are quoted for the $k=0$ mode. For $\omega\ll\omega_+$ and ω_- the diagonal elements of the susceptibility tensor are independent of frequency. For H^* and H_x small compared to H_{DM}, χ_{yy}' is large. This result is characteristic of a weak ferromagnet with anisotropic spin-spin interaction and an effective anisotropy field in the plane which contains the spins which is small compared to the canting field, H_{DM}. The resonance frequencies have been calculated in part previously,[5,8–10] and similar susceptibilities for $\alpha-Fe_2O_3$ were obtained by Turov and Gusseinov[10] for $H_z=H^*=0$.

NiF_2

For simplicity, we assume that the static field is applied in the (010) plane. The ferromagnetic component of the crystal is assumed to be perpendicular to the b direction. When Moriya's[3] calculations are extended, one finds that χ_{aa}' can be replaced by χ_{xx}' [Eq. (4)]; χ_{cc}' by χ_{zz}' [Eq. (6)]; χ_{ab}' by χ_{xy}' [Eq. (7)]; and χ_{ca}' by χ_{zx}' [Eq. (9)], where ω_-^2 is identical to Eq. (12) and ω_+^2 is replaced for the $k=0$ mode by

$$\omega_+^2=\gamma^2(H_a+H_M)(4H_M+H_a). \quad (14)$$

For the a, b, and c directions see Fig. 2. The other terms are

$$\chi_{bb}'=\chi_0\frac{H_M+H_a}{4H_M+H_a}\frac{\omega_+^2}{\omega_+^2-\omega^2}+\chi_0\frac{H_z^2}{2H_EH_A}\frac{\omega_-^2}{\omega_-^2-\omega^2}, \quad (15)$$

$$\chi_{bc}'=\chi_{cb}'^*=i\chi_0\left(\frac{\omega}{\gamma}\right)\frac{1}{4H_M+H_a}\frac{\omega_+^2}{\omega_+^2-\omega^2}, \quad (16)$$

where in Eq. (15) H_z^2 was neglected with respect to $2H_EH_A$.

The static field H_a is parallel to the [100] direction. The exchange field, H_E, the anisotropy field parallel to the [001] direction, H_A, and H_M are related to Moriya's notation by

$$H_E=8J_1/g\mu_B, \quad (17)$$

$$H_A=H_ED/8J_1, \quad (18)$$

$$|H_M|=2H_EE/8J_1, \quad (19)$$

where H_M is responsible for the weak ferromagnetism.

[9] P. Pincus, Phys. Rev. Letters **5**, 13 (1960).

[10] E. A. Turov and N. G. Gusseinov, Zh. Eksperim. i. Teor. Fiz. **38**, 1326 (1960) [translation: Soviet Phys.—JETP **11**, 955 (1960)].

3. EXPERIMENT

A single crystal of $MnCO_3$ or NiF_2 was placed into a cylindrical coil of the resonance circuit of a Colpitts oscillator similar to that used by Baker *et al.*[11] Measurements of the resonance frequency of the oscillator were performed when a static magnetic field was applied in a plane perpendicular to the axis of the sample coil. An increase in the induced rf magnetization parallel to the axis of the sample coil can be observed as a decrease in the resonance frequence of the LC circuit or vice versa. One obtains

$$\chi'=\frac{1}{4\pi\eta}\frac{|\Delta f|}{f_0} \quad (20)$$

for $|\Delta f|\ll f_0$, where f_0 is the frequency of the oscillator without the crystal in the sample coil and η is the filling factor. Experiments were performed at about 10 and 20 Mc/sec which is well below the natural resonance frequencies of the above samples. The frequency stability and the accuracy of the equipment was about ±100 cycles/sec, the exact value of η was unknown, and the accuracy of the alignment of the crystals was about $\pm2°$ or better. The operating temperature was 20.4°K. Three natural crystals of $MnCO_3$ and one synthetic[12] crystal of NiF_2 were investigated.

4. RESULTS AND INTERPRETATION

$MnCO_3$

Figure 3, curve a, shows the experimental results when a constant static magnetic field is applied in a

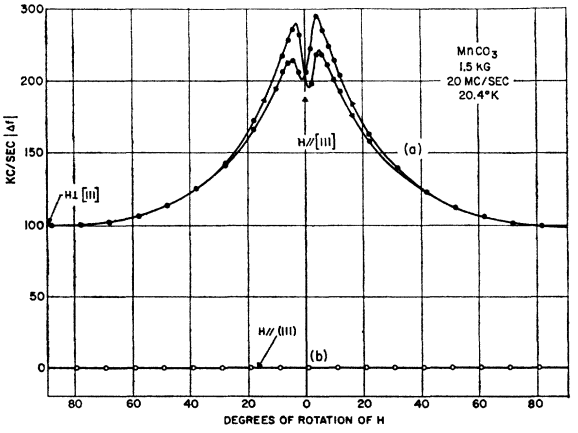

FIG. 3. The change in the resonance frequency of an LC circuit which contains $MnCO_3$ in the sample coil. A constant magnetic field, H, is rotated (a) in a plane perpendicular to the (111) plane and the rf field is perpendicular to the plane which is swept out by H [Fig. 1(a)]; (b) H is in the (111) plane and the rf field is perpendicular to (111) [Fig. 1(b)]. An increase in the ordinate corresponds to an increase in χ'.

[11] J. M. Baker, J. A. J. Lourens, and R. W. N. Stevenson, J. Phys. Soc. (Japan) **17** (B-I), 478 (1961).

[12] This crystal was grown by H. J. Guggenheim, Bell Telephone Laboratories, Murray Hill, New Jersey.

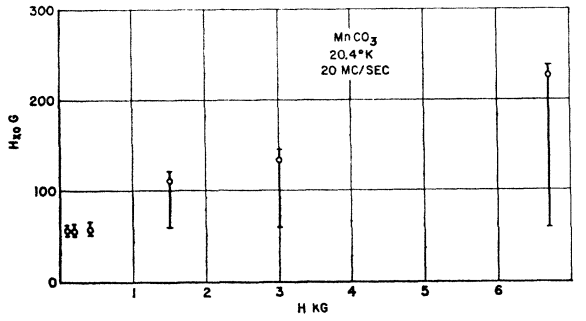

FIG. 4. The position of the maximum of χ' of $MnCO_3$ (see Fig. 3 curve *a*) is plotted as a function of the applied static field. θ_0 is measured from the [111] direction. $H_{x0}=H\sin\theta_0$ is plotted vs H.

plane perpendicular to the (111) plane which is assumed to be the xz plane. The ferromagnetic component is assumed to be parallel to the x direction, which is true as long as the static field parallel to the x axis is large compared to the anisotropy field in the (111) plane [compare Fig. 1(a)]. The rf magnetic field is applied parallel to the y direction, which is perpendicular to the plane in which the static field is rotated. An increase in the ordinate corresponds to an increase in the rf susceptibility parallel to the y direction. When the static field, H, is applied along the x direction which is an arbitrary direction in the (111) plane χ' is a minimum. It increases rapidly when H approaches the [111] direction (z direction) and reaches a maximum close to the [111] direction; then χ' decreases and reaches a minimum when the magnetic field is parallel to the [111] direction. The position of the maximum is a function of the orientation and magnitude of the applied static field and Fig. 4 shows the experimental relation between θ_0 and H, where θ_0 is the angle between the maximum (Fig. 3) and the [111] direction. For H large, θ_0 was small and therefore a small error in the alignment of the crystal gives an appreciable error in $H\sin\theta_0$. It appears that the magnetic field which corresponds to the peak in Fig. 3 has approximately a constant value in the (111) plane of about 60 G if one allows for an error in alignment of the crystal of about 2°. Figure 5, curve *a*, shows χ' parallel to the y direction when the magnitude of the static field is varied parallel to the x direction. At small fields there is a rapid increase in χ'. The experimental results were fitted in the high-field region to an expression of the form

$$\Delta f=C_0(H_{DM}+H_x)/(H^*+H_x), \qquad (5a)$$

with $C_0=4\pi\eta f_0\chi_0=34.6$ kc/sec, $H^*=0.32$ kG, and $H_{DM}=3.62$ kG at 20.4°K. The value of H^* is calculated from $2H_EH_A'/H_{DM}$, where $2H_EH_A'=(1.07)^2\,\mathrm{kG}^2$ is taken from the extrapolation of the resonance of the low-frequency branch to zero field at 20.4°K[13,14] and H_{DM} is obtained for susceptibility measurements.[7,8] This agrees well with χ_{yy} calculated from Eq. (5) for magnetic fields larger than about 1.5 kG. Below this value χ_{yy}' follows the same trend as the measured values but the agreement is not as good. The reason for this is that below 1.5 kG the crystal is not any longer a single domain but divides itself into domains which keep on growing when the field is reduced. The measured values of χ' follow the calculated values of χ_{yy}' only for those domains which are still perpendicular to the rf field. The contribution of the domains which are not perpendicular to the rf field is smaller, thereby reducing the total contribution to χ'. This can be seen readily by comparing Eqs. (5) and (4). Above approximately 60 to 70 G the division into domains seems only partially effective. When H is decreased below this value, the crystal divides itself completely into domains and χ' does not agree any longer with χ_{yy}'.

When the magnetic field is varied in magnitude parallel to the z direction and χ' parallel to the y axis is measured, one obtains curve *b* in Fig. 5. From Eq. (5) one would expect that χ_{yy}' is almost not affected by the static field for $H_x=0$ and $H_z\neq 0$, because $H_z^2\ll 2H_EH_A$, and $2H_EH_A$ is 1135 kG^2 at 20.4°K.[15] The value of χ' should be about the same for $H_z\neq 0$ as for $H_z=0$, because a magnetic field parallel to the z direction should have no effect on the ferromagnetic components in the (111) plane. However, an error in alignment of the crystal of about 2° is sufficient to explain the observed values, because the first term in Eq. (5a) is very large

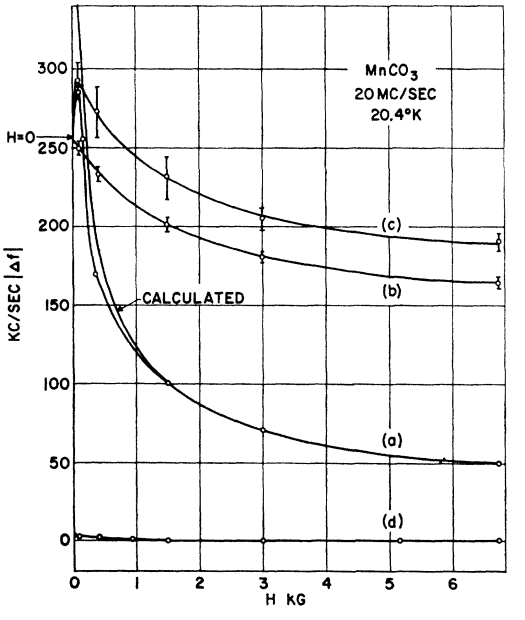

FIG. 5. The change of the resonance frequency of an *LC* circuit which contains $MnCO_3$ in the sample coil for various orientations of the static magnetic field and the rf field. For details see text. An increase in the ordinate corresponds to an increase in χ'.

[13] H. J. Fink, unpublished results; $(2H_EH_A')^{1/2}=1.07$ kG at 20.4°K.

[14] M. Date, J. Phys. Soc. (Japan) **15**, 2251 (1960).

[15] H. J. Fink and D. Shaltiel, Phys. Rev. (to be published).

and changes rapidly for a small value of H_x. Curve c in Fig. 5 is the observed maximum of Fig. 3 plotted as a function of the applied magnetic field. Similar arguments apply to curve c as to curve b.

From the above explanation it is clear that the maximum appearing in Fig. 3(a) is basically the same as the peak in Fig. 5(a). When the magnetic field in the (111) plane reaches about 60 G (see Fig. 4) the crystal divides itself effectively into domains and a field smaller than 60 G does not change appreciably the orientations of the ferromagnetic components in the (111) plane. For magnetic fields below about 1.5 kG hysteresis effects occur which displace the maxima and the minimum in

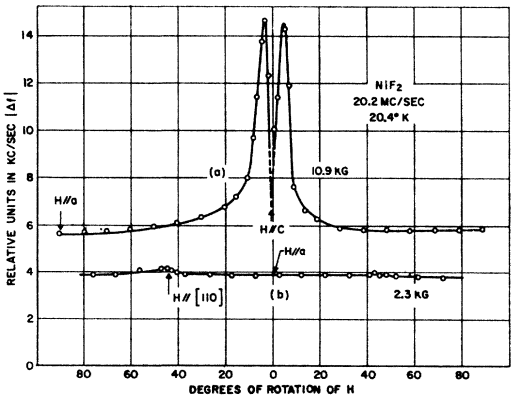

FIG. 6. The change in the resonance frequency of an LC circuit which contains NiF_2 in the sample coil. A constant magnetic field, H, is rotated (a) in the (010) plane and the rf field is parallel to the [010] direction; (b) H is in the (001) plane and the rf field is perpendicular to it. An increase in the ordinate corresponds to an increase in χ'.

Fig. 3(a) toward the direction in which the magnetic field is rotated. There is also a hysteresis in the absolute value of the minimum and the maximum in Fig. 3(a). Within the experimental accuracy identical results were obtained when the xz plane (the xz plane contains H; the rf field is perpendicular to xz) was rotated by any arbitrary amount around the z direction. Therefore, the preferred direction in the (111) plane could not be detected.

Figures 3(b) and 5(d) correspond to the rf susceptibility perpendicular to the (111) plane when the static magnetic field is rotated and varied in magnitude in the (111) plane [compare Fig. 1(b)]. From Eq. (6) one expects χ_{zz} to be a constant which is in good agreement with the experiment. When the resonance frequency was changed from 20 to 10 Mc/sec, all the above measurements were the same.

NiF_2

Similar experiments were performed for NiF_2 as for $MnCO_3$. Figure 6 shows the relative change in χ' along the b axis (the rf field is parallel to the b axis) when a static magnetic field of 10.9 kG is rotated in the (ac)

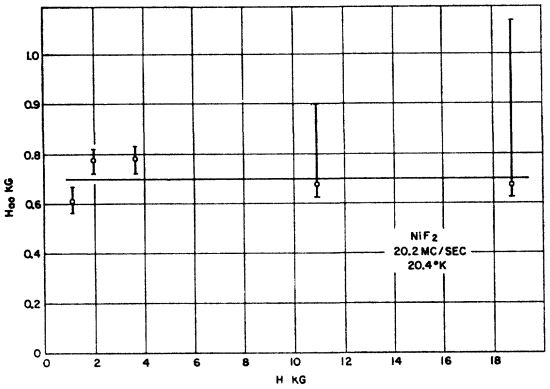

FIG. 7. The position of the maximum of χ' of NiF_2 (compare Fig. 6) is plotted as a function of the applied static field. θ_0 is measured from the [001] direction. $H_{a0}=H\sin\theta_0$ is plotted vs H.

plane. We observe again a maximum which is close to the [001] direction and a minimum when the static field is parallel to the [001] direction. Up to about 4 kG hysteresis effects occurred which displaced the maxima and minimum toward the direction in which the field was rotated. The magnetic field dependence of the maximum is shown in Fig. 7 from which one concludes that the maximum occurs for a constant magnetic field of about 0.7 kG in the (001) plane. Figure 8 shows the relative change in χ' when (a) the static field is increased in magnitude parallel to the c direction and the rf field is applied along the b direction; (b) the static field is decreased in magnitude along the c direction and the rf field is applied along the b direction; (c) the static field is applied along the x direction and the rf field is parallel to the y direction (see Fig. 2 for x and y); and (e) the static field is parallel to the a direction and the rf field is along the b direction. From Figs. 6(b) and 8(e) it is

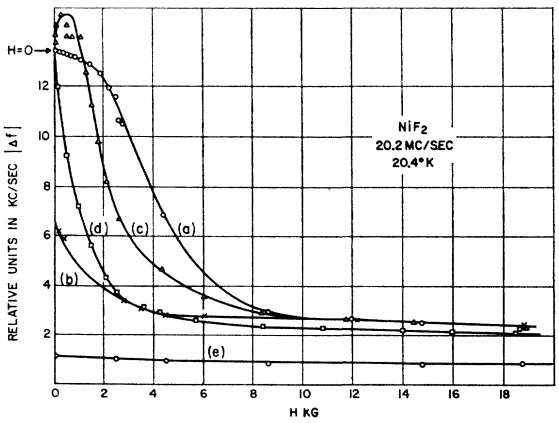

FIG. 8. The change of the resonance frequency of an LC circuit which contains NiF_2 in the sample coil for various orientations of the static magnetic field and the rf field. For details see text. An increase in the ordinate corresponds to an increase in χ'.

[16] M. Peter and J. B. Mock, Phys. Rev. 118, 136 (1960).
[17] P. L. Richards, Suppl. J. Appl. Phys. (to be published).

concluded that the rf susceptibility parallel to the c direction (the rf field is parallel to the c direction) is a constant and independent of the magnitude and orientation of a static magnetic field in the (ab) plane. This is in agreement with the calculated value χ_{cc}' [Eq. (6)]. At 10 Mc/sec one obtained identical results.

From paramagnetic resonance experiments[16] of Ni^{2+} in ZnF_2 we estimate a value of H_M in Eq. (15) of about 50 kG and of H_A of about 40 kG. $(2H_EH_A)^{1/2}$ is obtained from antiferromagnetic resonance experiments and its value is about 270 kG.[17] From inspection of χ_{bb}' and χ_{cc}' for NiF_2 [Eqs. (15) and (4)] one would expect that χ' does not change appreciably for magnetic fields smaller than H_M, especially for $H \ll H_M$. This, however, does not seem to agree with the experiments. The results of NiF_2 appear to be similar in character to those of $MnCO_3$ but they do not agree with the calculated values and the detailed behavior of the measured values of χ' is not understood at the present.

5. CONCLUSION

The rf susceptibilities of $MnCO_3$ and NiF_2 were calculated and relative susceptibility measurements were performed at 10 and 20 Mc/sec. These frequencies are well below the high- and low-frequency branch of these crystals. For $MnCO_3$ it was calculated that the rf susceptibility perpendicular to the ferromagnetic component in the (111) plane is one order of magnitude larger than the rf susceptibility parallel to the ferromagnetic component or perpendicular to the (111) plane for $H<H_{DM}$. Relative rf susceptibility measurements confirm this. For $\omega \ll \omega_+$ and ω_- this susceptibility is

$$\frac{\chi_{yy}'}{\chi_0}=\frac{H_{DM}+H_x}{H^*+H_x}+\frac{H_z^2}{2H_EH_A+H_z^2}, \tag{5b}$$

where $H^*=0.32$ kG and $H_{DM}=3.62$ kG at 20.4°K. When a static magnetic field is applied parallel to the [111] direction, there remains always a constant ferromagnetic component in the (111) plane which originates from the anisotropic spin-spin interaction. The effective anisotropy field $H^*(=2H_EH_A'/H_{DM})$ is small compared to H_{DM} and therefore the contribution to χ_{yy}' is large for H_x approximately smaller than 1.5 kG. This result is characteristic of a weak ferromagnet with anisotropic spin-spin interaction and a small anisotropy energy in the plane which contains the spins. From the experiment we conclude that there exists for the investigated crystals an effective field in the (111) plane of about 60 G below which an applied static magnetic field has almost no effect on the formation of domains. This manifests itself in a peak in the rf susceptibility which is observed when a static magnetic field is rotated in a plane perpendicular to the (111) plane (xz plane) and the rf field is applied perpendicular to the xz plane. This field is probably related to the coercive force[18] of the particular crystals which were all broken from the same rock.

The rf susceptibility perpendicular to the (111) plane and parallel to the ferromagnetic component is a constant for frequencies small compared to the resonance frequencies of $MnCO_3$, and it is independent of the orientation and magnitude of the applied field. Its value is M/H_E, which is the same as susceptibility perpendicular to the spins of a pure antiferromagnet.

For NiF_2 we obtain similar experimental results as for $MnCO_3$, but there is no agreement between the calculated and observed values. The reason for this discrepancy can be seen by comparison of Eqs. (5) and (15). The canting field and the anisotropy field in NiF_2 are of the same origin, namely, the crystalline electric field. In order to obtain a ferromagnetic component the anisotropy field must be large and this in turn makes χ' perpendicular to the ferromagnetic component almost independent for H for $H \ll H_M$. The experiments show, however, a peak in χ' when the rf field is parallel to the b direction and the static field is in the (ac) plane. This peak occurs when the static magnetic field in the (001) plane is approximately 0.7 kG. For NiF_2 we find that whenever the rf field is in the plane which contains the spins [(001) plane], and if in addition the rf field has a component perpendicular to the ferromagnetic moment, χ' is large provided the static magnetic field is smaller than approximately 5 kG. For large magnetic fields χ' saturates. However, when the rf field is applied perpendicular to the (001) plane and a static magnetic field is applied in the (001) plane, χ' is a constant and independent of the orientation and magnitude of the applied static field. The latter experimental fact is in agreement with the calculated value. Most of the experimental results in NiF_2 do not agree with the theory. They are probably related to domains and to domain wall motion but they are not understood at present.

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