

tion region were not present, it appears that the mixed-state helicon resonance would change rather rapidly to the normal-state value. This is the behavior expected at low frequencies from the work of Caroli and Maki.¹ Because of the presence of the transition region, attempting a fit to their theory does not seem reasonable.

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²Dr. R. W. Meyerhoff prepared the niobium sample used in this investigation. The material is Union Carbide electrolytic niobium that has been rolled into a strip and then annealed and outgassed about 100°C below the melting point in a vacuum of 10^{-10} Torr. Heating was accomplished by passing a current through the niobium. A section of the annealed strip was used for these experiments.

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MÖSSBAUER STUDY OF FERRIMAGNETIC ORDERING IN NICKEL FERRITE AND CHROMIUM-SUBSTITUTED NICKEL FERRITE

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Mössbauer-effect measurements in external magnetic fields show that the magnetic structure of ferrimagnetic NiFe_2O_4 is the collinear Néel type. $\text{NiFe}_{0.3}\text{Cr}_{1.7}\text{O}_4$ is shown to have a triangular structure with $\theta_A = 20 \pm 10^\circ$ and $\theta_B = 50 \pm 4^\circ$.

In a recent Letter,¹ Kedem and Rothem have presented Mössbauer data which they propose is evidence for a Yafet-Kittel triangular spin arrangement² in the ferrimagnetic spinel NiFe_2O_4 . Their conclusions are in sharp disagreement with the results of susceptibility measurements by Jacobs,³ who proposed a Néel collinear model⁴ for this material. We report here Mössbauer measurements in external magnetic fields which provide conclusive evidence for the Néel model and thus support the conclusions drawn by Jacobs. In addition, we show that the chromium substituted ferrite $\text{NiFe}_{0.3}\text{Cr}_{1.7}\text{O}_4$ is consistent with a Yafet-Kittel model.

Many of the magnetic properties of the ferrimagnetic spinel compounds $\text{M}^{2+}\text{N}_2^{3+}\text{O}_4$ are well understood on the basis of the Néel collinear model. However, for spinels with large amounts of chromium, the spontaneous magnetization is lower than the expected from this model and is usually interpreted in terms of

the Yafet-Kittel triangular arrangement in which each tetrahedral *A* and octahedral *B* sublattice is divided into two sub-sublattices; the resultant moments of the two triangular sublattices are antiparallel. Experimental evidence for the Yafet-Kittel model has been established by high-field susceptibility measurements^{3,5} and neutron-diffraction experiments.^{6,7} Previous nmr⁸ and Mössbauer^{9,10} studies of NiFe_2O_4 indicate two different hyperfine fields, corresponding to the iron ions on the *A* sites and the *B* sites. However, Kedem and Rothem¹ have concluded, mainly from the width of the Mössbauer lines, that there are four hyperfine fields and that this observation constituted experimental evidence for the Yafet-Kittel model.

Our samples were made by firing mixed oxides including Fe_2O_3 enriched in Fe^{57} in a platinum crucible at 1200°C in air for ten hours; the resulting product was then ground to a pow-

der, heated in a nitrogen atmosphere at 1200°C for 12 hours, and then allowed to cool slowly. X-ray diffraction analysis confirmed the spinel structure and the absence of other phases.

The Mössbauer experiments were performed using a conventional constant acceleration electromechanical drive system together with a multichannel analyzer for collecting and storing the data. The magnetic field was produced by a Nb₃Sn superconducting solenoid operating in the persistent mode up to 75 kOe.

Most of the experiments were carried out with the magnetic field applied along the γ -ray propagation direction. If the moments are collinear with the field, the polarization conditions require the disappearance of the $\Delta m = 0$ lines in the hyperfine pattern. Figure 1(a) shows the results for NiFe₂O₄ at 4.2°K. The zero field spectrum indicates two hyperfine fields^{1,9,10}; the widths of the lines are greater than those obtained with an α -Fe₂O₃ absorber, but this is expected in a powder

sample. The effect of applying a longitudinal field to the NiFe₂O₄ sample is also shown in Fig. 1(a); one observes the disappearance of the $\Delta m = 0$ lines at about $H_0 = 12$ kOe, and a further increase of the external field splits the outer $\Delta m = \pm 1$ lines into doublets of equal intensity, corresponding to the spin-up and spin-down sublattices. The measured fields at the nuclei H_n for the sublattices $T = 4.2^\circ\text{K}$ and $H_0 = 70$ kOe are -574 ± 5 kOe (A site) and -477 ± 5 kOe (B site), and using the relation

$$H_n(Z) = H_{\text{hf}}(Z) \pm H_0, \quad Z = (A, B), \quad (1)$$

where $H_{\text{hf}}(Z)$ are the hyperfine fields, we find $H_{\text{hf}}(A) = -504 \pm 5$ kOe and $H_{\text{hf}}(B) = -547 \pm 5$ kOe in agreement with the hyperfine fields measured at $H_0 = 0$: -506 ± 5 kOe (A) and -548 ± 5 kOe (B). These observations constitute definitive evidence for the Néel model in NiFe₂O₄. A 0.005-in.-thick NiFe₂O₄ single crystal, the plane of which is perpendicular to the [100], was also studied. Application of a small transverse field (1.25 kOe) in the plane of the absorber fully aligned the moments as evidenced by the relative intensities of the $\Delta m = 0$ and $\Delta m = \pm 1$ hyperfine lines, indicating very low anisotropy. A portion of the crystal was crushed to make a polycrystalline absorber and was studied in high external magnetic fields; the results were identical with the powder spectra.

The hyperfine spectra of NiFe_{0.3}Cr_{1.7}O₄ are shown in Fig. 1(b). It has been generally supposed^{11,12} that Fe³⁺ ions are situated on the A sites. However, the 70-kOe spectrum [Fig. 1(b)] indicates that approximately $\frac{1}{3}$ of the Fe³⁺ ions are on the B sites. The hyperfine fields for the two sites overlap at 4.2°K in zero external field and have the value -498 ± 5 kOe.¹³ Unlike the NiFe₂O₄, at $H_0 = 70$ kOe the $\Delta m = 0$ lines [lines α in Fig. 1(b)] are still relatively intense and the $\Delta m = \pm 1$ lines (β) do not correspond to Eq. (1). Assuming no large magnetocrystalline anisotropy,³ this may be explained by a Yafet-Kittel arrangement in which the spins on each sublattice Z ($Z = A, B$) make an angle θ_Z with the external field direction. For this model, the field at the nucleus $H_n(Z)$ in an external field H_0 is given by

$$H_n(Z) = (H_0^2 + H_{\text{hf}}^2 - 2H_0 H_{\text{hf}} \cos \theta_Z)^{1/2}. \quad (2)$$

From the observed spectrum we find $\theta_A = 20 \pm 10^\circ$ and $\theta_B = 50 \pm 4^\circ$.¹⁴ Using these values of

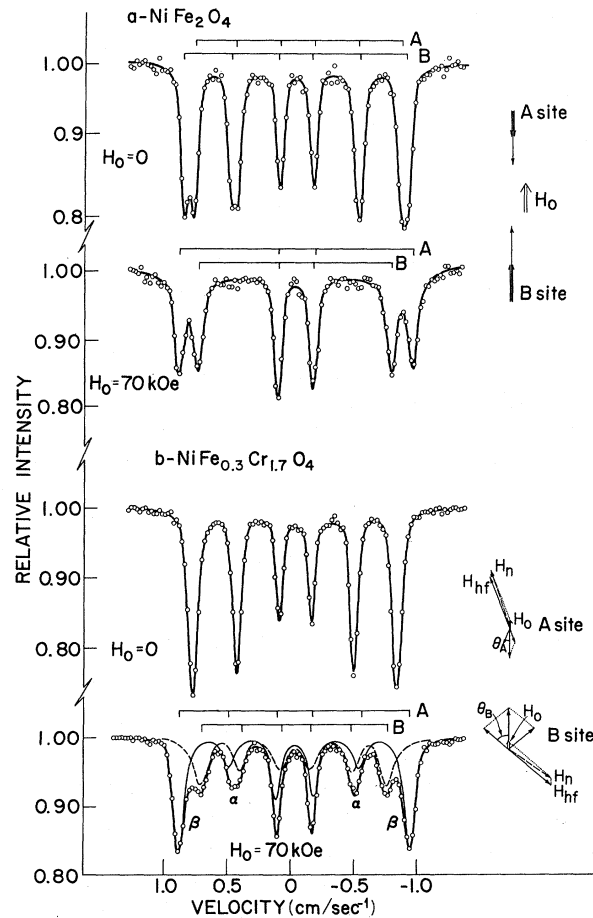


FIG. 1. Mössbauer spectra at $T = 4.2^\circ\text{K}$. (a) NiFe₂O₄. (b) NiFe_{0.3}Cr_{1.7}O₄.

θ_A and θ_B we are able to calculate the expected values of the relative intensities of the $\Delta m = 0$ lines. These values ($\alpha = 0.3$ for the A site, 1.6 for the B site) are in good agreement with the fit indicated for the 70-kOe spectrum of Fig. 1(b). Taking into account the presence of iron in the B site, one finds that the spontaneous moment calculated for such a model is equal to $1.3 \pm 0.5 \mu_B$, which is significantly lower than the moment expected from a Néel model ($3.2 \mu_B$).¹⁵ Furthermore, the B site moment is dominant in agreement with susceptibility data.³ We note that the observation of only two hyperfine fields is consistent with both the Yafet-Kittel as well as the Néel model and is therefore not sufficient to distinguish between these models.

We conclude that the magnetic structure of ferrimagnetic NiFe_2O_4 is of the collinear Néel type. An example of a Yafet-Kittel structure is shown for the chromium substituted $\text{NiFe}_{0.3}\text{Cr}_{1.7}\text{O}_4$.

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SPIN POLARIZATION NEAR LOCALIZED MOMENTS IN METALS*

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Following the treatment by Ruderman and Kittel¹ of magnetic interactions between nuclear magnetic moments in metals, Yosida² showed that localized moments in a metallic environment couple with a long-range oscillatory interaction. This interaction arises from the conduction-electron spin polarization

$$\vec{S}_e(r) = A \frac{\sin 2k_F r - 2k_F r \cos 2k_F r}{(2k_F r)^4} (g-1) \vec{J}, \quad (1)$$

caused by the magnetic ions, with A a positive constant and g the Landé g factor for the moment with angular momentum \vec{J} .³ Blandin⁴ has suggested a different form from the spin polarization: Rather than being caused by the exchange interaction between conduction electrons and magnetic ions, the spin polarization results from the different screening requirements imposed on spin-up and spin-down conduction electrons by the localized moment.