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Low-Temperature Ferromagnetic Relaxation in Yttrium Iron Garnet

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The ferromagnetic spin lattice relaxation time, τ_0, has been obtained from 4.2° to 300°K in single-crystal yttrium iron garnet in which considerable care was taken to reduce both the extraneous rare earth and ferrous impurity ions. The measurements are shown to be indicative of the purely ferric ion lattice at temperatures considerably lower than we previously had been able to attain. A new feature brought out in these data is the observation that below approximately 150°K, τ_0^{-1} vs T is steeper than proportional to T, which implies an extremely low value at 4.2°K. As a result of this, the nature of the ferric ion relaxation is more clearly defined over the low-temperature region.

PREVIOUS measurements on low-temperature ferromagnetic relaxation in yttrium iron garnet (YIG) led us to conclude[1,2] that the inverse spin lattice relaxation of the ferric ion lattice is proportional to temperature below 300°K. The data presented in this paper have brought out a new characteristic in the temperature dependence of the ferric ion relaxation. Below approximately 150°K the inverse spin lattice relaxation time, τ_0^{-1}, is no longer proportion to temperature but becomes a steeper function of temperature. This implies that a much lower value exists for τ_0^{-1} in the liquid helium temperature range than would be expected by extrapolating from the room temperature data.

Kasuya and LeCraw[3] have shown that spin lattice relaxation mechanisms are available in the room temperature region in which τ_0^{-1} is proportional to T. This theory was originally developed to explain room temperature data. At that time it was recognized that information in the low-temperature region was obscured by unidentified mechanisms. These mechanisms persisted even after considerable efforts to remove rare earth impurities which cause high relaxation rates in this temperature region. We have now determined that extraneous ferrous ions are also strong contributors to τ_0^{-1} in YIG in the temperature range below approximately 150°K.

By increasing the number of ferrous ions in a previously measured sample, the character of τ_0^{-1} vs T due to ferrous ions in YIG was obtained. Then, by using YIG grown from materials such that the ferrous ion content was reduced over our best previous samples, the data of this paper were obtained. From the measured curve the remaining normalized ferrous ion contribution was subtracted. As a result the nature of the ferric ion relaxation has become more clearly defined in the low-temperature region.

The parallel pump technique[4] was used for the measurements. As can be seen in Fig. 1 of reference 3, the inverse relaxation time, τ_k^{-1}, of long-wavelength spin waves ($k \leq 10^4$) is approximately equal to the inverse spin lattice relaxation time, τ_0^{-1}, of the uniform precession. Since the spin waves involved here have wave numbers $k \leq 10^4$, it is convenient to present the data in terms of the linewidths of these spin waves denoted by $\Delta H_{k\rightarrow 0}$. The linewidth is related to the relaxation time by, $\Delta H_{k\rightarrow 0} \approx \gamma^{-1}\tau_0^{-1}$.

Figure 1 gives the values of $\Delta H_{k\rightarrow 0}$ vs T for two particular samples. The upper curve (open circles) represents measurements taken on a sphere of single-crystal YIG, grown using yttrium oxide purified to eliminate the fast relaxing rare earth ions.[5] It became clear that the $\Delta H_{k\rightarrow 0}$ maximum must be identified and, if not intrinsic, eliminated to obtain relaxation times of the purely ferric lattice below about 150°K. The combined yttrium, iron, and lead oxides, however, were found to contain as much as 100 parts per million of silicon.

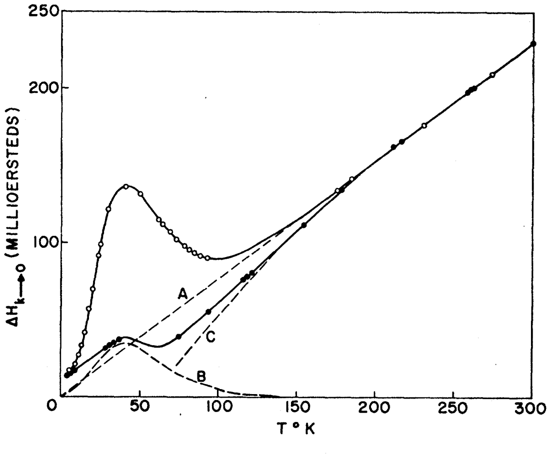

FIG. 1. Upper curve (open circles) is $\Delta H_{k\rightarrow 0}$ vs T for a sphere of YIG grown from Y_2O_3 purified by ion exchange. Bottom curve (solid circles) is $\Delta H_{k\rightarrow 0}$ vs T for a sphere of YIG grown from the same materials but in which silicon has been reduced. Dashed line A shows that from high temperatures $\Delta H_{k\rightarrow 0}$ extrapolates to the origin; B indicates Fe^{2+} mechanism; and C indicates trend of ferric ion relaxation at low temperatures. The frequency of pump is 16 450 Mc/sec, the spin waves k are at 8225 Mc/sec. All measurements are along [111] axes.

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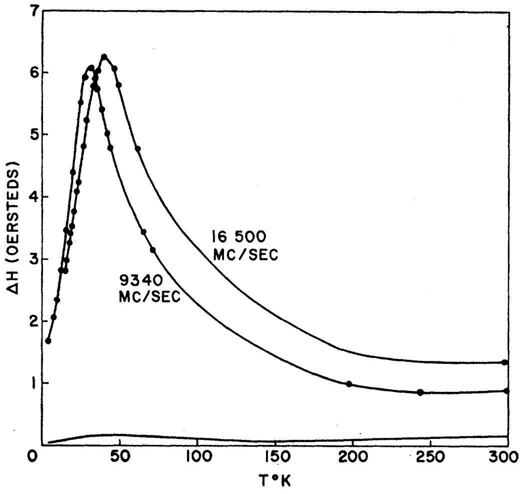

FIG. 2. $\Delta H(T)$ along the [111] axis for YIG in which Fe^{2+} ions have been created by an oxygen defect method. The lowest curve is $\Delta H(T)$ at 9340 Mc/sec taken on the sphere before removing oxygen.

Verweel and Roovers[6] have studied conductivity in ceramic YIG concluding that, because of valence compensation, each Si^{4+} ion gives rise to one Fe^{2+} ion. Geller[7] has shown that the Fe^{2+} ions in YIG (associated with the Si^{4+} ions) have a strong preference for octahedral sites. Dillon[8] has studied the effects of Si^{4+} impurities on the resonance properties of YIG by working with crystals intentionally doped with silicon.

In order to identify the $\Delta H_{k\to 0}$ maximum at 40°K as being caused by Fe^{2+} ions, we were able to create additional Fe^{2+} ions in a previously measured YIG sphere by introducing oxygen defects. This was accomplished by heating the sphere to 1025°C for 12 hr in a vacuum oven at a pressure of 10^{-5} mm of Hg. Figure 2 shows ΔH vs T before treatment (lowest curve) and after treatment (upper curves). Measurements were taken at two frequencies to determine the nature of the ferrous ion relaxation. These data, including the frequency independence of the maximum value of ΔH, are consistent with the valence exchange mechanism as studied by several investigators[9] in ferrites.

There is the other possibility that the ferrous ion effect involves the fast relaxing mechanism suggested by White,[10] based on the theory of Kittel *et al.*[11] In this case, Kittel's equations indicate that the magnitude of $\Delta H_{\max}$ is proportional to frequency which is in opposition to the results of Fig. 2. Thus we are unable to reconcile Kittel's equations with the experimental results on ferrous ions in YIG.

By avoiding growth conditions favorable to magnetite inclusions and by purifying the oxides with respect to silicon,[12] results have been obtained as shown in the bottom curve (solid circles) of Fig. 1. $\Delta H_{k\to 0}$ vs T again is proportional to T in the room temperature region. This further confirms that the two magnon—one phonon process or the three magnon process, both associated with the uniaxial anisotropy, of reference 3, hold above 150°K. Although the calculations are not complete, Kasuya has stated that the deviation from proportionality to T below 150°K is contained in the theory. The dashed line, A, is a straight line through the origin and the high-temperature points. Curve B is an estimate of the Fe^{2+} ion contribution, which is subtracted from the measured curve to give curve C. (The temperature dependence of curve B is obtained from the data of Fig. 2.) From the measured curves on the two samples of Fig. 1, it appears that even if all Fe^{2+} ions were eliminated $\Delta H_{k\to 0}$ would still be approximately 15 moe at 4.2°K. The above two mechanisms, described by Kasuya and LeCraw, are expected to vanish at 0°K. If curve C is a measure of these mechanisms, then it will approach the origin asymptotically. The 15-moe value at 4.2°K appears to be caused by an additional unidentified mechanism.

Mention may be made of the three magnon dipolar relaxation mechanism discussed by Sparks and Kittel[2] and by Schlömann.[13] For the spin waves involved here, $k \leq 10^4$, the contribution of this mechanism at 300°K is less than 10 percent of that due to k independent processes. At 4.2°K the contribution is less than 1 percent.

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[12] We wish to express our appreciation to C. H. Love and M. Huffman of the C. K. Williams Company for their purification of the Fe_2O_3 used in these experiments.

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