anisotropy (K_1) of disordered lithium ferrite. (See A. D. Schnitzler: Thesis, University of Maryland, January 1961). This upper limit is 1/100 of that K_1 value which follows from the D-value assumed by Callen for ferric ions. Thus the contribution of Callen's mechanism to the line-width is negligibly small in this case. Furthermore, it was found by Schnitzler (to be published) that the linewidth of our disordered lithium ferrite samples increases with increasing frequency, whereas the scattering theories predict a decrease of linewidth (because of the decreasing number of degenerate states) with increasing frequency. In regard to ordered lithium ferrite, our experimental results (Schnitzler, Folen and Rado, paper to be presented at the 1961 Phoenix Magnetism Conference) show that in the samples used the linewidth is predominantly due to impurity ions which relax rapidly in the manner postulated by Kittel for the rare earth iron garnets.

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Intrinsic Relaxation of the Uniform Precession in Ferromagnetic Resonance of Yttrium Iron Garnet

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Studies of the intrinsic relaxation of the uniform precession in ferromagnetic resonance of yttrium iron garnet are described. The data, which are obtained by the parallel pump technique, are believed to be characteristic of perfect YIG and are in good agreement with a theory for the intrinsic relaxation of the uniform precession in YIG developed by Kasuya.

Introduction

Recently it has become possible to make meaningful observations of the intrinsic relaxation time, τ_0, of the uniform precession in narrow line width ferromagnetic single crystals. Previously this quantity has been masked by a number of deceptive factors. The new observations have been made possible primarily by the following three developments; (1) understanding and elimination of the effects of surface roughness (2) elimination of rare earth and other impurities, particularly silicon, and (3) separation of spin-lattice from spin-spin relaxation effects.

Intrinsic processes for relaxation of the uniform precession were considered early in the history of ferromagnetic resonance. However, many orders of magnitude existed between these processes and the best experimental data, which yielded line widths of ~100 oe. With the discovery of yttrium iron garnet (YIG), the observed line widths began at ~10 oe and were soon reduced to ~0.5 oe.

At this point the strong effects of surface roughness on relaxation of the uniform precession became apparent. Surface imperfections couple the uniform precession to other degenerate spin waves which then relax by faster processes not available to spin waves with $k=0$. Since it is impossible to achieve a perfect polish and the complete absence of volume imperfections, a method was needed to differentiate between relaxation of the uniform precession by processes outside the degenerate manifold (generally spin-lattice type processes) and those occurring via the degenerate manifold (spin-spin type processes). This was achieved by a series of modulation experiments[1] in which the relaxation of the longitudinal and transverse components of magnetization, M_z and M_t, were independently observed for various degrees of surface roughness. With this technique τ_0 was evaluated even in the presence of small unavoidable amounts of scattering to degenerate spin waves. Note that τ_0 as used here is the relaxation time of the uniform precession energy by all processes *other* than via the degenerate manifold.

Discussion of experiments

For the samples used in Ref. 1, τ_0^{-1} was 7.3×10^6 sec^{-1} at room temperature and 6.2 kMc/sec. After the importance of rare earth impurities was recognized, purer samples were obtained and τ_0^{-1} was decreased to 2.6×10^6 sec^{-1} at the same temperature and frequency. At the same time the low temperature line width maximum was reduced from $\sim$6 oe to $\sim$0.15 oe. The much greater reduction at low temperatures than at room temperature was highly suggestive that it might be possible at room temperature to observe for the first time the intrinsic relaxation of the purely ferric ion lattice in a ferromagnetic insulator.

Recently an important new technique, easier to use than the modulation method, was proposed by Schlömann[2] and Morgenthaler[3] called the parallel pump technique. It can be shown that growing pairs of spin waves

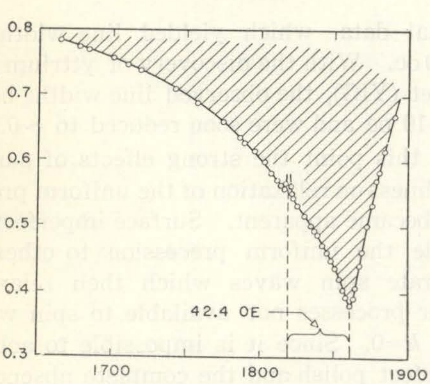

Fig. 1. Values of h_{crit} vs dc field for small single crystal spheres of YIG at room temperature using the parallel pump technique. The pump frequency is 11.4 kMc/sec and the spin-wave frequencies are 5.7 kMc/sec. The dc field is along the [111] axis. The exchange constant D is obtained from the 42.4 oe displacement of the one magnon-one phonon interaction.

of equal and opposite wave number and frequency $\omega_k=\omega_p/2$ may be excited when an rf magnetic field of frequency ω_p with sufficient magnitude is applied parallel to the dc magnetic field H. The threshold rf field is

$$h_{\rm crit}=\omega_p/(\gamma^2\tau_k 4\pi M_s \sin^2\theta_k) \tag{1}$$

where τ_k is the relaxation time of a spin wave of wave number k, and θ_k is the angle between the dc field and the spin wave. Thus the threshold is lowest for $\theta_k=\pi/2$. Figure 1 shows $h_{\rm crit}$ vs. H at 5.7 kMc/sec and room temperature for a polished single crystal YIG sphere with the dc field along the [111] axis, the axis of all the data. A microwave spectrometer with a nearly critically coupled reflection cavity was used for these experiments. Each point in Fig. 1 corresponds to the first onset of instability as one moves the dc field into the unstable region, shown by the cross-hatching. The onset of instability is indicated by an abrupt change in the reflection from the cavity. The notch in the curve is caused by the one magnon-one phonon interaction first observed by Turner[4]. From the position of the notch and a knowledge of the transverse sound velocity, 3.87×10^5 cm sec^{-1}, one can determine the magnitude and temperature dependence[5] of the exchange constant D in the dispersion relation

$$\omega_k{}^2=\gamma^2\left(H^i+\frac{Dk^2}{\gamma\hbar}\right)\left(H^i+\frac{Dk^2}{\gamma\hbar}+4\pi M_s\sin^2\theta_k\right). \tag{2}$$

The value of D determined in this manner compares well with D determined from low temperature specific heat data.

Using the data from the low field branch of Fig. 1, together with Eq. (2) and the measured value of D (0.92×10^{-28} erg cm^2), one obtains a plot of τ_k vs. k as shown in Fig. 2. The high field branch in Fig. 1 is not yet well understood. Here the spin waves have $k\sim0$ and $\theta_k\neq\pi/2$. For such long wavelength spin waves the plane wave approximation used in Eq. (1) is invalid and the sample boundary conditions must be considered.

In the derivation of Eq. (1), τ_k was introduced as the spin wave "lifetime" by all available processes, including scattering to degenerate spin waves by sample imperfections. Thus the parallel pump technique cannot differentiate between spin-lattice and spin-spin contributions to τ_k. The modulation method is required for this. However, it has been found that when interpreted properly the two methods yield the same result for τ_0^{-1}. The data in Fig. 2 can be described by

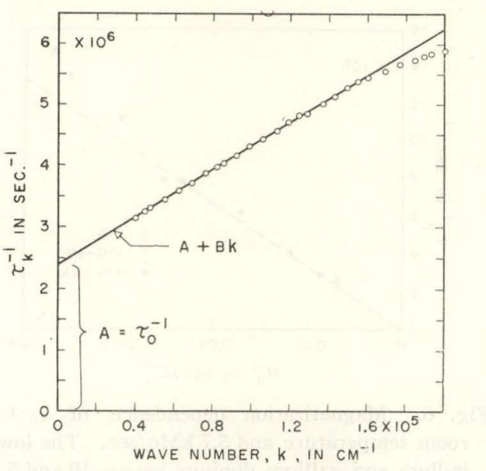

Fig. 2. Plot of τ_k^{-1} vs k obtained from the data in Fig. 1, with the acoustic notch subtracted off.

$$\tau_k^{-1}=A+Bk \tag{3}$$

from $k=0.4\times10^5$ to 1.6×10^5 cm^{-1}. The factors A and B have the following significant properties: (1) A is very nearly equal to τ_0^{-1} as measured on the same sample by the modulation method. (2) A and B are both essentially independent of surface roughness, although for more polished surfaces, the straight line portion extends to lower k values than for less polished surfaces. (3) At 8.2 kMc/sec the quantity A/γ is 0.205 oe which is within 15 per cent of our lowest observed line width at this frequency of 0.235 oe. and (4) the value of B of 20.5 cm sec^{-1} is in reasonable agreement with the three-magnon relaxation calculation of Sparks and Kittel[6]. Their calculation predicts a contribution to τ_k^{-1} which is linear in k and T and with a value for YIG at this temperature and frequency of 23 cm sec^{-1}. (In obtaining this value M_s and D at 0°K were used.) The departure from linearity for $k>1.6\times10^5$ cm^{-1} is probably due to the assumption $Dk^2\ll\hbar\omega$ in their calculation no longer being satisfied. At lower temperatures the agreement for B is not as good, with B decreasing considerably slower than T.

Temperature dependence of τ_0^{-1}

The temperature dependence of τ_0^{-1} at 5.7 kMc/sec, obtained by equating A and τ_0^{-1}, is shown in Fig. 3. This data represents the closest approach yet achieved to ideally pure YIG, in that both the rare earth and divalent iron content[7] have been greatly reduced over previously labelled "pure" YIG. Below~125°K, careful extrapolations such as in Figure 2 have not been made, so the data is only an approximation to τ_0^{-1} in this region. It is seen that τ_0^{-1} is proportional to T from~175°K to~325°K. Above 325°K

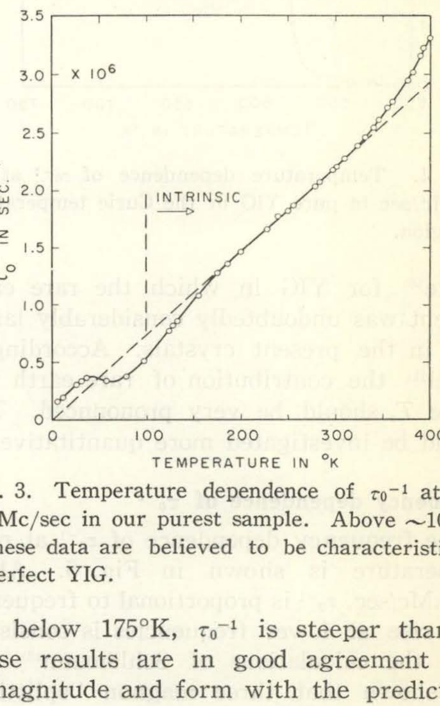

Fig. 3. Temperature dependence of τ_0^{-1} at 5.7 kMc/sec in our purest sample. Above ~100°K these data are believed to be characteristic of perfect YIG.

and below 175°K, τ_0^{-1} is steeper than T. These results are in good agreement both in magnitude and form with the predictions of a theory proposed by Kasuya and Le Craw[8] based on two magnon-one phonon and three magnon processes derived from the uniaxial Hamiltonian. Space does not permit here an adequate discussion of the theory. Only in these ultra pure samples has it been possible to observe the departure from linearity below~175°K as predicted by the theory. The remaining low temperature maximum is not contained in the theory and apparently is caused by residual impurity ions. Theoretical calculations have been made of another process, the four magnon process, which might contribute to τ_0^{-1}, but the contribution appears to be negligible[9].

The temperature dependence of τ_0^{-1} near the Curie point is shown in Fig. 4. It is of interest that in the paramagnetic region the value of τ_0^{-1} (assuming $\tau_0^{-1}=\gamma\Delta H$) is approximately the same as that obtained by

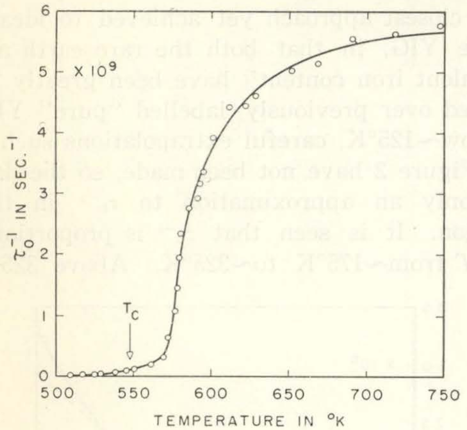

Fig. 4. Temperature dependence of τ_0^{-1} at 5.7 kMc/sec in pure YIG in the Curie temperature region.

White[10] for YIG in which the rare earth content was undoubtedly considerably larger than in the present crystals. According to Kittel[11] the contribution of rare earth ions above T_c should be very pronounced. This should be investigated more quantitatively.

Frequency dependence of τ_0^{-1}

The frequency dependence of τ_0^{-1} at room temperature is shown in Fig. 5. Above ~3 kMc/sec, τ_0^{-1} is proportional to frequency. The rise at lower frequencies is consistent with the calculations of Schlömann[12] who has shown that three magnon "splitting"

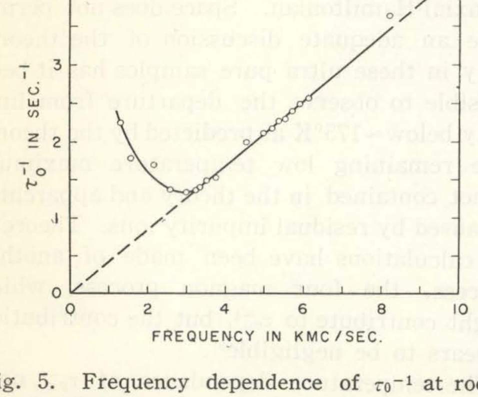

Fig. 5. Frequency dependence of τ_0^{-1} at room temperature.

processes contribute to τ_0^{-1} in this region. Above ~3 kMc/sec in YIG this process vanishes. The variation of τ_0^{-1} directly with frequency above~3 kMc/sec is in agreement with the theory outlined in Ref. 8. The linear dependence should continue at least until $\hbar\omega/kT$~0.1.

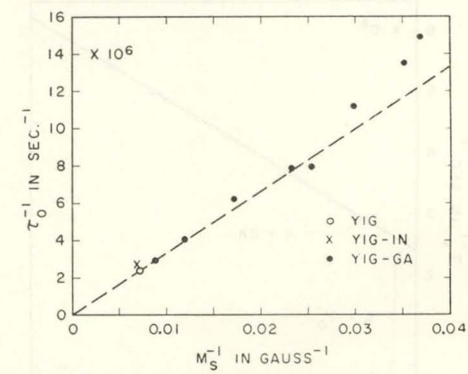

Fig. 6. Magnetization dependence of τ_0^{-1} at room temperature and 5.7 kMc/sec. The lowest indium and gallium dopings have ~10 and 5 At. % of the iron replaced, respectively.

Magnetization dependence of τ_0^{-1}

The magnetization dependence of τ_0^{-1} at room temperature is shown in Figure 6. The reduction of M_s was achieved by adding gallium. The addition of indium raises M_s at 0°K, but leaves M_s at room temperature relatively unchanged[13]. These data are also in agreement with the theory of Ref. 8, which predicts that τ_0^{-1} should vary nearly as M_s^{-1}. It may be noted from these results (particularly the indium doping) that "vast" amounts of nonmagnetic impurity ions may be added to YIG without causing appreciable line broadening. The pertinent perturbation field here is of course not a pseudodipolar field of order 10^5 oe, as was assumed for certain ferrites[14], but is more likely to be a term associated with the uniaxial anisotopy of order 10^3 oe.

Conclusions

The information in Figures 3-6 represents the present state of our knowledge about the relaxation of the uniform precession in single crystal YIG. It is believed that these data are characteristic of perfect YIG (except below~100°K). The results are in good agreement with a theory proposed by Kasuya and LeCraw and a detailed treatment being prepared by Kasuya. YIG is perhaps the only ferromagnetic material in which such a reasonable state of agreement exists between theory and experiment for the intrinsic relaxation of the uniform precession.

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DISCUSSION

E. SCHLÖMANN: It seems to me that the theory, as far as I understand it, makes one believe that the dependence of τ_0^{-1} on $1/M_s$ in gallium and indium substituted YIG should be expected if nothing is changed except the saturation magnetization. If you make a substitution, I presume you also change the exchange constant. Would this be an appreciable effect?

R. C. LECRAW: Experimentally it appears that the magnetization effect does predominate over the effect of the exchange as well as the scattering which is introduced. The theory is rather complicated on this point.

C. KITTEL: How do you distinguish between the scattering effects of an impurity and the effects which enter through M_s?

R. C. LECRAW: In ten percent indium-doped YIG the line width barely increases at all, whereas ten percent gallium-doped YIG yields a much bigger increase. This indicates that magnetization effects predominate over the scattering. (See Fig. 6)