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[2] A. Bril and W. L. Wanmaker, *J. Electrochem. Soc.* **111**, 1363 (1964).

[3] For comparison: the most efficient Ce^{3+}-activated phosphor $Ca_2Al_2SiO_7$ has an efficiency of 4% (ref. 1).

HIGH TEMPERATURE RESONANCE LOSSES IN SILICON-DOPED YTTRIUM–IRON GARNET (YIG)*

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The ferrimagnetic resonance linewidth of silicon-doped YIG, measured as a function of temperature at 13.4 kMHz, is found to show a pronounced peak at 105°C. The anisotropic behavior of this peak is in good agreement with the four-level valence-exchange model proposed by Clogston. The model yields for the electron ordering energy a value 5×10^{-4} eV which agrees closely with the energy deduced from magnetic anneal studies. The activation energy for electron transfer (0.25 eV) is virtually identical with values reported in investigations of electrical conductivity and acoustic loss.

It is known that the introduction of small concentrations of silicon into the YIG lattice leads to a significant increase in magnetic loss. There is general agreement, however, that silicon itself plays only an incidental role and that the losses are actually due to the presence of divalent iron created in response to the addition of quadrivalent silicon.

In YIG samples containing Fe^{2+} two peaks have been reported in the plot of microwave resonance linewidth vs temperature. Typically, one peak occurs at low temperature below ca. 100°K, and the other above ca. 300°K. It was suggested, at the time of its discovery, that the low temperature peak was due to valence exchange,[1,2] i.e., electron transfer between Fe^{2+} and Fe^{3+} cations. More recently, Tchernev[3] has argued that the electron-hopping mechanism is, in all likelihood, "frozen out" at temperatures below 100°K, and therefore, it is more reasonable to ascribe the peak to the localized Fe^{2+} ion acting, more or less,[4] as a slow relaxer. The high temperature peak has been looked at to a lesser extent[5,7] and the suggestion has been made that it is due to valence exchange.[7]

In our work the temperature dependence and anisotropy of the upper peak have been examined in some detail. Our results provide strong evidence that the high temperature resonance losses are, indeed, due to valence exchange, a conclusion that would seem to exclude this mechanism as the one also responsible for the low temperature peak.

We have measured the linewidth of the uniform precession at 13.4 kMHz for several single crystals of silicon-doped YIG ($Y_3Fe_{5-\delta}Si_\delta O_{12}$) between room temperature and the Curie point. The crystals were grown from a PbO-PbF_2 flux using high purity starting materials to which controlled amounts of SiO_2 were added. Silicon concentration was determined from spectrographic analysis of several crystals (not the actual samples) selected from the growth run. Measurements were made on samples prepared as well polished spheres, 0.025″ in diameter. These were mounted in a TE-108 transmission cavity and linewidth measurements were made using the half-power method.[8]

Our results for the composition $\delta = 0.06$ are shown in Fig. 1. A sample containing a smaller silicon content ($\delta = 0.04$) showed the same temperature dependence but with the magnitude of the losses reduced roughly in proportion to the doping.

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A pure sample showed no peak but merely a monotonic increase in linewidth that became extremely rapid near the Curie point.[9,10] It is evident from Fig. 1 that the "background losses" in the doped samples show this same type of behavior. At $T = 105°C$ the pure sample had a linewidth slightly less than 1 Oe.

The anisotropy in linewidth is in excellent agreement (Fig. 2) with Clogston's four-level model for valence exchange.[11] In this scheme the valence-exchange electrons are assumed to order over four energetically inequivalent sites, each characterized by an energy

$$E_i = -\epsilon \cos^2 \theta_i; \quad i = 1, 2, 3, 4 \tag{1}$$

where ϵ is the electron ordering energy and θ_i is the angle between the magnetic axis and one of the four body diagonals of the cubic lattice. Clogston also examined the consequences of a three-level scheme, where each level has the same angular dependence as in Eq. (1) but where the angle θ_i is taken with respect to the i^{th} cube-edge direction.

The three-level assumption predicts a valence-exchange contribution to the linewidth that is a maximum in the [111] direction and zero along [100]. The linewidth anisotropy of nickel ferrous ferrite[12,13] appears to conform to this energy scheme.[11]

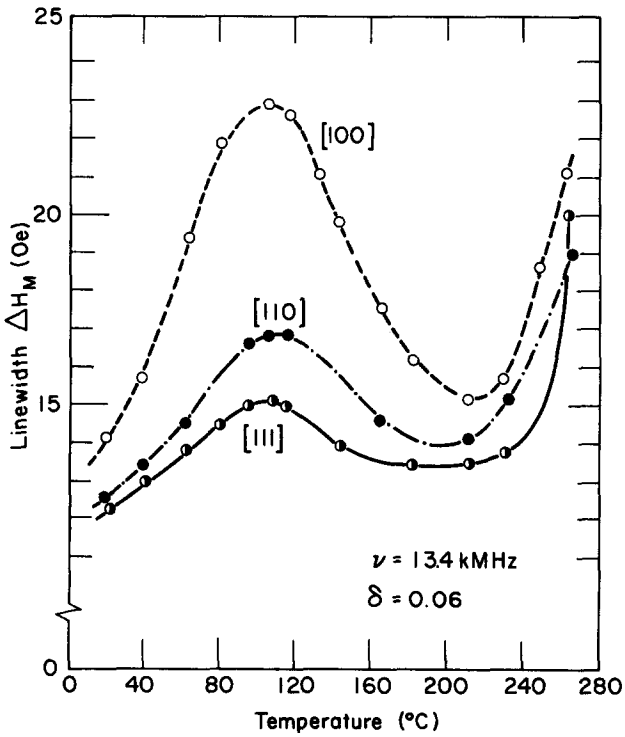

Fig. 1. Resonance linewidth as a function of temperature for silicon-doped YIG, $Y_3Fe_{5-\delta}Si_\delta O_{12}$, $\delta = 0.06$.

For YIG, Hunt[14] has carried out a calculation which supports the four-level scheme. For this level structure Clogston's theory leads to a linewidth that is maximum along [100], as observed in our experiments. In the (110) plane Clogston finds that the valence-exchange contribution has the form

$$\Delta H_{VE} = \Delta H^0_{VE}\left[1 - 2\left(\frac{1}{4}\sin^4\phi + \sin^2\phi\cos^2\phi\right)\right] \tag{2}$$

with

$$\Delta H^0_{VE} = \frac{8}{9M}\left(\frac{N\epsilon^2}{kT}\right)\frac{\omega\tau}{1+(\omega\tau)^2}\,. \tag{3}$$

Here, N is the concentration of Fe^{2+} ions, M is the saturation magnetization, ϕ the angle between the magnetization and the [001] direction and τ the relaxation time for electron transfer between Fe^{2+} and Fe^{3+}.

In Fig. 2 the linewidth ΔH_M measured in the (100)-plane has been fitted to the expression $\Delta H_M = \Delta H_{VE} + \Delta H_B$, where ΔH_B has been introduced to account for a background loss that is assumed to be isotropic. At the temperature of the peak ($T = 105°C$) our data yield $\Delta H^0_{VE} = 11.8$ Oe and $\Delta H_B = 11.4$ Oe for the composition $\delta = 0.06$.

It should be noted that the background loss is about a factor 10 larger than in the undoped crystal. Without having additional information about the temperature and frequency dependence of this loss term one can only speculate on its origin. Quite possibly it is due to an enhancement of the Kasuya–Le Craw two-magnon one-phonon process caused by the large local uniaxial anisotropy[14] of the Fe^{2+} ion.

At the peak, $\omega\tau = 1$ and, therefore, $\Delta H^0_{VE} = (4/9M)(N\epsilon^2/kT)$. Assuming that each silicon atom produces a valence electron, we have $N = N_0\rho\delta/A =$

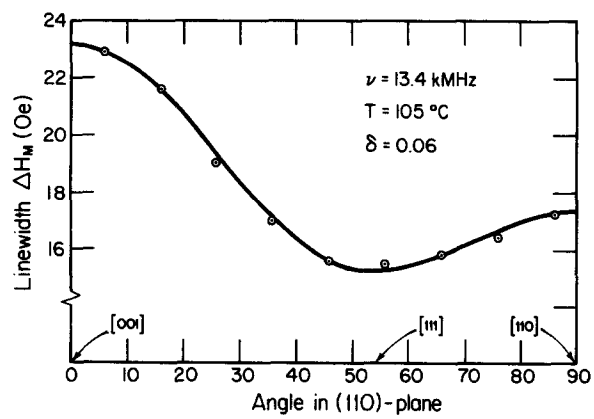

Fig. 2. Linewidth anisotropy in the (110) plane at $T = 105°C$. The points are experimental, the curve is a plot $\Delta H_M = \Delta H_B + \Delta H_{VE}$, where ΔH_{VE} is given by Eq. (2).

4.32×10^{21} (cm^{-3}) where N_0 is Avogadro's number, ρ the density (5.17 gm/cm^3) and A the molecular weight of YIG (738.01 gm/mole). At $T = 105°C$, $M = 115$. From our data fit in Fig. 2 it follows that $\epsilon = 5 \times 10^{-4}$ eV.

Hunt[14,15] has used the four-level valence-exchange model to explain field-induced anisotropy effects observed in Si-doped YIG under quasistatic conditions at low temperature. From analysis of his data at 15°K he finds an ordering energy $\epsilon = 6 \times 10^{-4}$ eV, in very good agreement with our result.

Clogston's theory also predicts a change in both the effective anisotropy constant and in the effective field for resonance. In the three-level scheme the valence-exchange mechanism contributes a negative term to K_1, whereas for the four-level structure the contribution is positive. For both energy schemes there is an identical depression of the effective field for resonance.

From measurements carried out at the loss peak on our sample $\delta = 0.06$ we find that $K_1/M = -12.1$ Oe, compared with the value -17.6 Oe for an undoped sample measured at the same temperature and frequency. Thus the valence-exchange contribution to K_1 is *positive*, consistent with our contention that we are dealing with the four-level structure. However, the magnitude of the observed change in K_1/M (5.5 Oe) is about a factor four larger than the theoretical prediction based on the value of ϵ obtained from the linewidth anisotropy. There is less inconsistency in our result for the change in effective field for resonance. We observe a downward shift of 5 Oe compared with a predicted value of about 3.5 Oe.

If we assume that the relaxation time τ in Eq. (3) varies as $\tau = \tau_\infty \exp(w/kT)$, we can extract from the temperature variation of the linewidth a value for the activation energy w. To eliminate the temperature dependence of the background loss ΔH_B we take the difference between the [100] and [111] linewidths, thereby obtaining

$$T\,[\Delta H_{M\text{-}[100]} - \Delta H_{M\text{-}[111]}] \propto \frac{\omega\tau}{1 + (\omega\tau)^2}. \quad (4)$$

Analysis of the temperature behavior contained in this functional relationship gives $w = 0.25$ eV. Graczyk[16] has studied the electric conductivity of a similar composition ($\delta = 0.05$) in the same temperature range and has found very nearly the same activation energy (0.26 eV). Le Craw and Comstock[17] have reported a similar activation energy (0.27 eV) for acoustic losses observed in the MHz region on a sample with $\delta \simeq 0.1$.

Judy's measurements at 9 kMHz show a resonance loss peak at 350°K. Extrapolating our data to this frequency, using 0.25 eV as an activation energy, we would expect a peak at 355°K.

Although interpretation of our data has been in terms of valence exchange, it must be pointed out that the observed loss peak and linewidth anisotropy can be explained in another way. Teale and Tweedale[18] have noted that Clogston's treatment may be used formally to describe the slow-relaxation-impurity mechanism.[19] In this process the losses are associated not with electron transfer between Fe^{2+} and Fe^{3+}, as in valence exchange, but, instead, with electronic transitions occurring between the two lowest levels of the magnetic "impurity" ion. However, if this were the operative mechanism, we would not expect agreement between the activation energy for magnetic loss and that for electric conduction, except accidentally. Since we must allow for accident, we cannot definitively rule out the possibility of Fe^{2+} acting as a slowly relaxing impurity. However, the weight of our present data seems clearly to favor the four-level valence-exchange model as the most likely explanation for the high temperature resonance losses. To test further the validity of this conclusion we intend to study the frequency dependence of this loss peak.

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THE FORMATION OF THE MACROVORTEX STRUCTURE IN HARD SUPERCONDUCTORS*†

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Measurements of the field at the surface of a magnetized hard superconducting wire have shown that the application of a local perturbation precipitates the sequential formation of a spatially periodic magnetization (macrovortex structure) along the axis of the wire.

Direct evidence of the existence of a spatially periodic structure in the magnetization of a hard superconducting wire has been presented in a previous Letter.[1] The existence of such a structure was suggested by Walker and Hulm[2] who called it the "macrovortex" structure.

In this Letter we report observations which show that this "macrovortex" structure is the result of the sequential collapse of an initially uniform magnetization at discrete points, equally spaced along the wire and that the collapse can be triggered by a small local pulse of magnetic field. Further we conclude that the train of voltage pulses observed by Walker and Hulm[2] is generated by this sequential formation of macrovortices.

The experimental arrangement, similar to that described in a previous Letter,[1] consisted of a 2″ length of 0.02″-diam, heat-treated Nb–25% Zr wire. This wire was embedded in the grooved end of a phenolic tube which was located in the bore of a superconducting magnet. The wire filled a 300° arc of the groove leaving a 60° arc unfilled.

The wire was subjected to a local magnetic field pulse generated by a small coil attached to the end of the phenolic tube. The pulse field had a peak amplitude of 50 G and a duration of 125 μsec and augmented the ambient field which was transverse to the axis of the wire. The pulse field locally initiated a collapse of magnetization (a flux jump).

A magnetoresistance probe was used to explore the distribution of the surface field along the axis of the wire. The probe was 0.004″ thick and was 0.01″ wide in the direction of the axis of the wire. The resistance of the probe was measured by a bridge circuit with an accuracy of 5 parts in 10^5 while the current in the ambient-field superconducting magnet was stable to one part in 10^5.

The experimental procedure was as follows: The wire was magnetized in a suitable ambient field and the field strength at the surface of the wire was measured at intervals of $2\frac{1}{2}°$ of arc. The field pulse was then applied and the surface field again measured at each $2\frac{1}{2}°$ interval. The experimental results are shown in Figs. 1 and 2 in which the resistance of the probe, normalized to the value measured at the unfilled part of the groove, is plotted against the angular position of the probe. The results shown in Fig. 1 were obtained from a sample of wire which consisted of copper-clad and non-copper-clad portions. A 150° arc of the grooved phenolic was filled with the unclad portion of the wire and the other 150° arc was filled with the copper-clad portion. The ambient field strength in this case was 22.5 kG. The open triangles and open circles correspond respectively to the paramagnetic and diamagnetic states of the wire before the application of the pulse. The resistance of the probe increases sharply between the empty part of the grooved phenolic and the unclad portion of the wire in the paramagnetic state, indicating the presence of magnetization in the wire. Similarly

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