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[14]It is more important to the problem treated by Brinkman and Lee (Ref. 6) since both initial and final momenta are restricted to less than the Fermi momentum.

Linear Electric Field Shift of a Ferromagnetic Resonance: Lithium Ferrite

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The first observation of magnetic resonance field shifts proportional to an applied electric field in a magnetically ordered material is reported. It involves ferromagnetic resonance experiments on lithium ferrite crystals in an applied electric field at temperatures between 4.2 and 77°K. The observed shifts are predicted by using statically measured values of the electric field derivative of the uniaxial anisotropy energy existing in the "skin" region of these crystals.

In several paramagnetic materials the application of an electric field E is known[1] to shift the electronic magnetic resonance by an amount proportional to E. No such linear shift has been observed or predicted hitherto in any magnetically ordered material. The E-induced nonlinear shift observed[2] in gallium iron oxide was shown[3] to be producible by Joule heating and hence does not constitute an intrinsic effect of E.

This Letter reports the first observation and interpretation of an E-induced linear shift, δH_{res}, of the magnetic resonance field H_{res} in a magnetically ordered material. Specifically, we are concerned with ferromagnetic resonance (FMR) where H_{res} is the applied magnetic field H required for resonance at a fixed frequency. The possible existence of such a shift was suggested by the discovery that in at least two materials, namely, magnetite[4] and lithium ferrite,[5] the *macroscopic* magnetocrystalline anisotropy energy contains a portion linear in E. Using lithium ferrite, whose FMR linewidth is considerably narrower than that of magnetite, we measure a shift δH_{res} which is proportional to E over a range of temperatures T extending at least from 4.2 to 77°K. In addition, we predict the measured δH_{res} by using values of the electric field derivative of the anisotropy energy measured statically over this same temperature range with a method[5] reported previously.

It should be noted that the discovery[4,5] of an E-dependent anisotropy energy was based neither on a direct measurement of anisotropy by a torque method nor on some experimental technique (such as FMR) which involves an indirect but reasonably standard measurement. Instead, the E-dependent anisotropy energy was inferred from measurements of the electric polarization induced by an applied H in the absence of an applied E. This inference included a previously untested extension[4,5] of thermodynamic theory. Since the novel phenomenon of E-dependent anisotropy should be established unambiguously, it is important to attempt the observation of δH_{res} or some other quantity whose value can be predicted on the basis of an E-dependent anisotropy. Such a prediction usually requires additional nontrivial assumptions. In the case of δH_{res}, for example, we make the simplest assumption, namely, that the microwave and static values of the anisotropy energy are equal and that all the resonance parameters other than the anisotropy are independent of E. It is worth noting that the linear tuning of a FMR with an applied E, which is demonstrated by our δH_{res} measurements, has an impact on potential microwave applications as well as on the establishment of an E-dependent anisotropy.

The samples are circular disks of 0.215-cm diam cut from the previously described "skin" region[5] of ionically ordered lithium ferrite. The symmetry axis of the disks, labeled z axis, is chosen to be perpendicular to that crystallographic (111) plane (cubic notation) which is parallel to

the large growth faces of the crystal platelets. Both (111) faces of the disks were provided with thin electrodes consisting of either silver paint or evaporated aluminum. The disks were then enclosed in a nylon holder, fitted with fine copper wires through which a static voltage V was applied, and supported in the center of a microwave cavity by a quartz tube. The cavity was operated in a TE_{102} mode at a fixed frequency of about 9.2 GHz. The microwave magnetic field was parallel to the electrodes, whereas the static magnetic field H (about 6 kOe) and the (small) microwave electric field were both perpendicular to the electrodes, i.e., parallel to the z axis. A Varian E-9 spectrometer was used to display the H derivative of the FMR absorption spectrum as a function of H on an XY recorder. Thus the δH_{res} caused by the application of V could be measured directly on any absorption line. We determined δH_{res} on the (2, 2, 0) magnetostatic mode rather than on the uniform mode because the narrower linewidth of the former increases the accuracy of the δH_{res} measurements. At room temperature, the linewidth of the (2, 2, 0) mode and the uniform mode was about 40 and 150 Oe, respectively, in the presence of electrodes whereas each linewidth was only about 18 Oe in the absence of electrodes. These linewidths do not vary strongly with temperature and no attempt was made to decrease the linewidths by finely polishing the samples. Furthermore, the original "mat" surface of the "skin" region[5] was preserved during the fabrication of the disks. Concerning δH_{res}, we found experimentally on various samples that its value for randomly chosen magnetostatic modes of low order equals that for the uniform mode. This is consistent with the theoretical result[6] that in a situation involving cylindrical symmetry around a ⟨111⟩ axis, such as that obtained in our experiment, a change in anisotropy causes all the modes to be displaced by the same change in H_{res}. In calculating δH_{res}, therefore, we can proceed as if δH_{res} were measured on the uniform mode throughout.

Figure 1 shows typical experimental data on δH_{res} vs T at constant V for two samples having different thicknesses t. It is seen that for each sample δH_{res} increases monotonically as T is increased from 4.2 to 77°K. For each sample, moreover, we observed that δH_{res} at constant T is proportional to V provided that $|V|$ does not greatly exceed the value given in Fig. 1. But if the data for the two samples are compared with each other, it is found that at constant T the

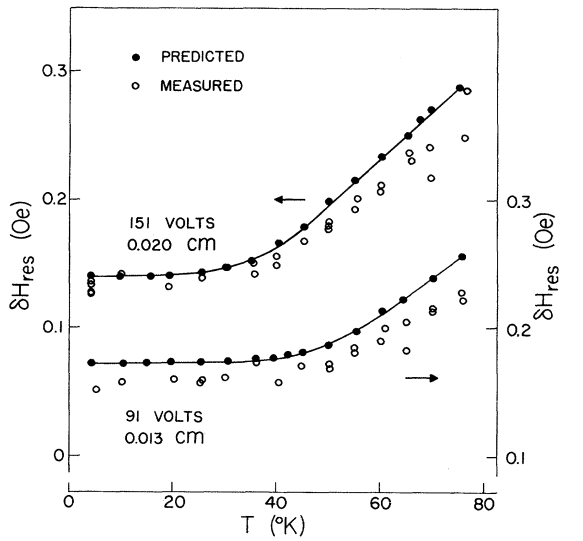

FIG. 1. Comparison of the measured and predicted values of the electrically induced magnetic resonance field shift δH_{res} as a function of temperature T for two samples of lithium ferrite. The thickness and applied voltage for each sample are also given.

ratio of δH_{res} to V/t is not independent of t. This result is to be expected because in separate experiments we established that both the resistivity ρ and the electric field derivative $\partial K_{uc}'/\partial E$ of the uniaxial crystalline anisotropy energy[5] are z dependent. Thus δH_{res} may depend not only on V/t and T but also on the fraction of the disk thickness occupied by the skin region. As shown below, however, δH_{res} can be predicted even if the detailed z dependences of ρ and of $\partial K_{uc}'/\partial E$ are unknown.

In our experiment the directions of $\vec{H}$ and $\vec{E}$, the symmetry axis of the disks, the axis of the uniaxial anisotropy, and the easy axis of the cubic anisotropy are all parallel to one another and to the z axis. For this situation, the ferromagnetic resonance condition for the uniform mode at a fixed frequency is easily shown to give

$$\delta H_{res} = -(2/M_s)(\partial K_{uc}'/\partial E)E, \quad (1)$$

where M_s is the saturation magnetization and Gaussian units are used. Equation (1) assumes that (a) all the resonance parameters (e.g., the g factor) other than K_{uc}' are independent of E and that (b) the sample is homogeneous. Attempts to avoid assumption (a) do not seem to be justified unless the accuracy of the experimental data can be improved. Assumption (b), on the other hand, will now be modified by replacing Eq. (1) with the

spatial average

$$\delta H_{res} = -(2/M_s t)\int_0^t (\partial K_{uc}'/\partial E)E\,dz \quad (2)$$

and assuming that the previously mentioned theoretical result[6] on the mode displacements due to anisotropy remains valid if the sample is inhomogeneous. The requirement that the conduction current density arising from the application of V be continuous then leads to

$$\delta H_{res} = -(2V/M_s t)(\partial K_{uc}'/\partial E)_{meas}, \quad (3)$$

where $(\partial K_{uc}'/\partial E)_{meas}$ is, at this point, merely an abbreviation for the quantity

$$\left(\frac{\partial K_{uc}'}{\partial E}\right)_{meas} = \frac{\int_0^t (\partial K_{uc}'/\partial E)\rho\,dz}{\int_0^t \rho\,dz}. \quad (4)$$

Since the z dependences of $\partial K_{uc}'/\partial E$ and ρ are unknown, it is fortunate that appropriate static methods[4,5] enable us to measure $(\partial K_{uc}'/\partial E)_{meas}$ directly. In fact, the quantity labeled $\partial E_{uc}'/\partial E$, which we measured in our static magnetoelectric experiments on lithium ferrite,[5] actually represents $(\partial K_{uc}'/\partial E)_{meas}$. We prove this identification and thus derive Eq. (4) by applying to that experimental situation the requirement of continuity of the total (conduction-plus-displacement) current and then making an approximation which amounts to neglecting the sample capacity in comparison to the capacity of the cable plus electrometer.

Using the method described previously,[5] we measured $(\partial K_{uc}'/\partial E)_{meas}$ over a temperature range from 4.2 to 77°K on the same samples as those used for the δH_{res} measurements. Since V, t, and M_s ($\approx$ 330 emu) are known, Eq. (3) enables us to predict δH_{res} without requiring any free parameters. For the $t = 0.020$ cm sample at 4.2°K, for example, the values $V = 0.503$ statvolt and $(\partial K_{uc}'/\partial E)_{meas} = 0.918$ statcoulomb/cm^2 lead to a predicted δH_{res} of 0.140 Oe. This is to be compared with measured δH_{res} values ranging from 0.125 Oe to 0.136 Oe. The predicted values of δH_{res} as a function of T for two samples are shown in Fig. 1 and compared with the corresponding measured values. It appears that the agreement between the predicted and measured temperature dependences is as good as can be expected; the accuracy of the measured small δH_{res} is limited not only by the FMR linewidth and crystal variations, but also by the stabilities of the magnet power supply, the spectrometer, and the temperature. It is presently impractical, therefore, to determine whether the discrepancies between the measured and predicted δH_{res} values are within the experimental error. We note that in some cases these discrepancies are smaller than those shown in Fig. 1. For example, the agreement between the predicted and measured δH_{res} values at 4.2°K is within 4% for a $t = 0.011$ cm sample and within 1% for a $t = 0.0074$ cm sample.

Presently underway are measurements at higher temperatures and attempts to reduce the FMR linewidths by polishing the samples. Moreover, we hope to clarify the atomic origin[7] of these magnetoelectric phenomena by investigating the composition of the skin region.

In summary, we have (I) reported the first observation of magnetic resonance shifts δH_{res} proportional to an applied electric field E in a magnetically ordered material, (II) demonstrated linear tuning of a ferromagnetic resonance with an applied electric field, (III) predicted the observed δH_{res} by using statically measured values of the E derivative of a uniaxial anisotropy energy, and (IV) confirmed our previous interpretation[5] that certain magnetically induced electric polarizations are caused by an E-dependent anisotropy energy.

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