

Ferromagnetic Resonance of EuO

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Ferromagnetic and paramagnetic resonance has been observed with single crystals of europous oxide. The frequencies used were 23.8 and 27.5 Gc/sec, and the temperatures ranged from 1.5 to 300°K. The linewidth increased from about 800 Oe at low temperatures to about 1400 Oe at room temperature. The low-temperature anisotropy field is given approximately by $K_1/M = -190$ Oe.

WE have performed preliminary ferromagnetic resonance experiments on single crystals of the insulating ferromagnet EuO.¹ These have led to a measurement of the anisotropy at 1.5°K. Heretofore the lack of suitable single-crystal specimens has barred many fundamental experiments on EuO, though its simple crystal and spin structure make it an appealing experimental material.

The europous oxide was prepared by the reduction of europium sesquioxide using a stoichiometric quantity of europium metal in a sealed tantalum bomb.² The mixture was heated slowly to 1600°C during which a strong exothermic reaction took place. It was then maintained at 1600°C for 12 h with repeated inversion of the crucible to insure complete reaction. Chemical analysis showed the total europous oxide content calculated from the gain in weight on ignition to the sesquioxide to be $(98.0 \pm 0.5)\%$. X-ray diffraction analysis of the material showed it to be cubic, rock-salt structure, $a_0 = 5.143$ Å. The data indicate either that there was oxidation of the powder between preparation and analysis or that there is a homogeneity range in the composition of the monoxide. The x-ray examination did not show the presence of a second phase, but comparison of the x-ray diffraction patterns of this sample with that of another sample prepared in a different way showed differences in lattice parameter which would be consistent with a homogeneity range for the oxide.

Close examination of some of the purple-brown lumps showed that they contained single crystals approaching a cubic millimeter in volume. From the polycrystalline

lumps, it was possible to grind and polish a few single-crystal spheres by the two pipe technique. The use of this technique is important because in a material with such a large magnetization, small deviations from sphericity lead to large variations in H_{res} and to the excitation of unwanted magnetostatic modes.³

The orientation was done by an x-ray goniometric technique, but a Laue photograph was taken in each case to ensure that we were dealing with a single crystal. As is usually done, the sphere was mounted so that it could be rotated about a [110] axis which was perpendicular to the magnetic field.

The field, H_{sat} , required to saturate a sphere of EuO at low temperatures is $4\pi M/3 = 24\,000/3$ Oe = 8000 Oe, and thus a resonance experiment performed at less than $\gamma H_{\text{sat}} = 22\,330$ Mc/sec would be dealing with an unsaturated sample. In addition, the lines encountered are very broad, so the frequency must be somewhat above this lower limit. Our choice of an experimental frequency of about 27 Gc/sec was set by these considerations and by the fact that the maximum field available was about 11 000 Oe. The microwave apparatus, the recording technique, and the procedure for intermediate temperatures have been described before.⁴

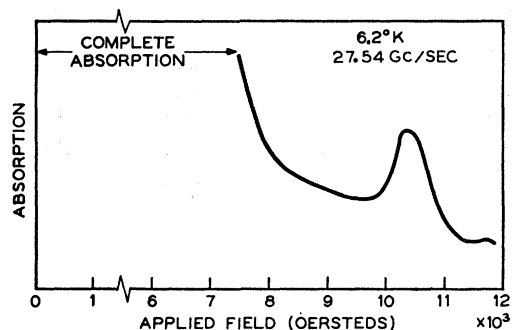


FIG. 1. Typical plot of microwave absorption versus field for EuO single crystal. The temperature was 6.2°K, the frequency 27.54 Gc/sec. Below about 7500 Oe, absorption is so great as to wipe out the signal completely.

³ J. F. Dillon, Jr., Phys. Rev. **112**, 59 (1958).

⁴ J. F. Dillon, Jr., and J. W. Neilsen, Phys. Rev. **120**, 105 (1960); J. F. Dillon, Jr., Phys. Rev. **127**, 1495 (1962).

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¹ B. T. Matthias, R. M. Bozorth, and J. H. Van Vleck, Phys. Rev. Letters **7**, 160 (1961).

² The preparative procedure was described in more detail in N. G. Nereson, C. E. Olsen, and G. P. Arnold, Phys. Rev. **127**, 2101 (1962). Quantitative spectrographic analysis of the powder used showed the following: Be <0.002%; Sr <0.005%; Al, Sc, Ti, Cr, Mn, Fe, Y, Zr, Ba, and Yb each <0.05%; Co, Ni, Tm, and Lu each <0.1%; V, Gd, Tb, Dy, Ho, Er, Pb, and Th each <0.2%; Ce, Pr, Nd, and Sm each <0.5%; Mg and Ca each 0.005%; La 0.05% and Ta 1%. The symbol < represents the limit of detection, and also that these elements were below the limits of detection.

FIG. 2. H_{res} of EuO single crystal in the ferromagnetic and paramagnetic temperature ranges. The sphere was 0.63 mm in diameter. At the low-temperature end of the data, anisotropy leads to a spread of field for resonance along different crystal directions. A magnetostatic mode interacts with the uniform precession between 40 and 80°K. Data above and below 90°K were taken at two different frequencies. In this plot they have been matched at $g=2.00$.

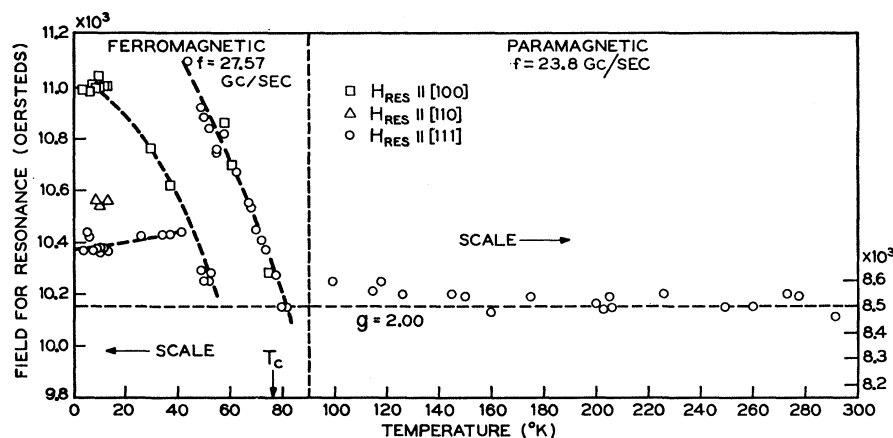


Figure 1 shows the response of the spectrometer with a 0.63-mm-diam sphere of EuO at 6.2°K. Below saturation, the large losses associated with the domain structure dominate the absorption trace. One can follow roughly the magnetization variation with temperature by observing the field at which these low-field losses disappear.

LINewidth

At 1.5°K, the resonance linewidth appears to be about 700 or 800 Oe. The uncertainty arises from our inability to establish a satisfactory baseline. The width does not show any violent orientation dependence. It increases roughly linearly with temperature; there is no particular anomaly at the Curie temperature. Above this, the paramagnetic resonance linewidth increases approximately linearly with temperature from about 1100 Oe at 80°K to about 1400 Oe at 300°K.

It is our presumption that the large ferromagnetic resonance linewidths are associated with spurious imperfections, impurities, or composition gradients within the samples. The early experiments on oxidic ferrimagnets containing Fe^{+++} showed comparably large linewidths, but improvements in sample preparation spread over a decade led to samples with very narrow lines. Hopefully, the development of techniques to produce better samples of EuO will lead to narrower lines which can be compared with fundamental theory. Note that the magnetization here is six or ten times larger than that of the common iron containing ferrimagnets (e.g., YIG and MnFe_2O_3). The Curie temperature of EuO is approximately one eighth that of the garnets, and its exchange field is correspondingly small. The paper by Clogston, Suhl, Walker, and Anderson⁵ leads to an expression for that part of the linewidth which arises from impurity, disorder and other scattering of the uniform precession into frequency degenerate spin wave modes. On examination of the principal

⁵ A. M. Clogston, H. Suhl, L. R. Walker, and P. W. Anderson, *Phys. Chem. Solids* **1**, 129-136 (1956).

result of that paper, we see that the linewidth due to certain kinds of imperfections including voids and non-magnetic inclusions would be about $\sim 10^3$ times larger in EuO than in a geometrically equivalent crystal of, for example, yttrium iron garnet. From this point of view the linewidths we have observed are not as disparate as they might at first appear.

FIELD FOR RESONANCE, g VALUE, AND ANISOTROPY

The most satisfactory sample on which measurements have been made thus far is about 0.63 mm in diameter, much too large for a 27-Gc/sec experiment. From past experience, we would expect the use of such a large sample to involve some displacement of the field for resonance and the appearance of magnetostatic modes other than the uniform precession. Though analytical expressions are available for the shift, their use does not seem warranted in this case with such a large sphere and magnetization. From the experimental point of view, meaningful g determinations must await the preparation of narrow linewidth samples of several diameters down to perhaps 0.1 mm, and perhaps measurements at two frequencies, say 25 and 35 Gc/sec.

The variation of field for resonance with crystal orientation, however, would seem to give us a valid measure of the anisotropy. Figure 2 is a plot of the temperature variation for the steady field parallel to the crystallographic directions. In the region from 40 to 60°K, a second magnetostatic mode is evident and it interacts strongly with what appears to be the uniform precession. At the low-temperature end of the range H_{res} versus angle in (110) appears to vary as if the anisotropy field can be accounted for by a $K_1/M = -190$ Oe. This corresponds to $K_1/\text{ion} = 0.093$ $\text{cm}^{-1}/\text{ion}$. The corresponding figure for the Gd^{+++} ion in gadolinium iron garnet is 0.10 $\text{cm}^{-1}/\text{ion}$. Thus, the anisotropy energy associated with an Eu^{++} ion in EuO is virtually identical to that associated with the iso-

electronic Gd^{+++} ion in gadolinium iron garnet.⁶ The anisotropy field seems to decrease to a very low value by about 40°K.

In the temperature interval from 160 to 300°K $g=2.00\pm 0.01$. We believe the displacement from 2.00 below that has to do with magnetostatic modes or with a size effect.

⁶ G. P. Rodrigue, H. Meyer, and R. V. Jones, *J. Appl. Phys.* **31**, 3765 (1960).

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Expansions for Magnetization Characterization for Polycrystalline Orthorhombic and Cubic Magnetite at Low Temperatures

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Taylor series expansions are obtained for the reduced magnetization and crystalline anisotropy energy which do not require that the magnetization vector lie in a plane formed by the easy axis and the applied field. The free energy which is assumed to be composed of magnetostatic and crystalline anisotropy energy is minimized with respect to the spherical coordinates of the magnetization vector. The expansions for the magnetization in terms of the crystalline anisotropy constants are used to find the mean magnetization at different magnetic field intensities for polycrystalline magnetite, which undergoes an order-disorder transition at -155°C . Experimental results for the magnetization of polycrystalline magnetite are compared with those predicted by the expansions and good agreement is observed.

INTRODUCTION

PRACTICALLY all the research reporting the order-disorder transition observed in magnetite at -155°C is concerned with monocrystalline properties.¹ This does not mean that polycrystals of the same material do not exhibit a like effect.² This researcher studied the change in magnetization as a result of the transition for natural and synthetic magnetite with varying degrees of doping and oxidation and found rather surprising results which will be reported at a later time. Because the material that was being studied was polycrystalline a direct comparison with other results was not possible.

To obtain the polycrystalline magnetic behavior one must be able to average the magnetization or the crystalline anisotropy energy over all possible directions with an appropriate weighting function. Even before one reaches the averaging process, one must be able to represent the total energy of the system in a form which will make the magnetization process progress in a fashion which will make the total energy a minimum.

Three assumptions are usually made^{3,4}: that the free energy is predominantly magnetostatic and magnetoelastic; that it can be expanded as a series in the direction cosines and strains, consistent with the symmetry of the system; and that the magnetization vector remains in the plane of the applied field and of a direction of easy magnetization.

All of these assumptions are applicable in the studies on magnetite as verified by the comparison of experimental results with predicted results based on the above assumptions.⁵ The first two assumptions will be used in the analysis to follow and the third one will be shown to be a good first-order approximation. In order to facilitate the averaging process, the energies to be considered will be expressed in spherical coordinates rather than direction cosines. The minimization of the total energy will also be carried out in spherical coordinates. A Taylor series with two variables is obtained both for the magnetization and the crystalline anisotropy energy. The mks system of units will be used.

¹ There have been numerous investigations into the order-disorder transition for magnetite and references are too numerous to detail. For an excellent listing of references and experimental results see, C. A. Domenicali, *Phys. Rev.* **78**, 458 (1950).

² D. C. Ray, doctoral dissertation, The University of Michigan, 1962 (unpublished).

³ R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., New York, 1951).

⁴ E. W. Lee, *Proc. Phys. Soc. (London)* **72**, 249 (1958).

⁵ H. J. Williams, R. M. Bozorth, and M. Goertz, *Phys. Rev.* **91**, 1107 (1953).