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PARAMAGNETIC RESONANCE TRANSMISSION IN GADOLINIUM*

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Using the spin transmission or selective transmission resonance method previously applied to conduction-electron paramagnetic resonance,[1] we have observed rather unexpected but unique and characteristic resonance lines transmitted through 99.9% purity gadolinium foil at temperatures above the Curie temperature of 289°K. In this type of resonance technique, the sample forms the common wall between two microwave cavities tuned to the same frequency and so arranged that the external magnetic field may be oriented either perpendicular or parallel to the foil with the microwave magnetic field perpendicular to the static field. One cavity is used for excitation of the resonance, and the other cavity, into which leakage must be kept to a minimum, is connected to a sensitive superheterodyne receiver which is coherent with the excitation generator and therefore provides phase-sensitive detection.

The sample foil is 75 μ thick, and the skin depth at the frequency used in this experiment (9200 MHz) is about 1 μ. Transmitted paramagnetic resonance under these conditions is surprising. If it is assumed that a signal is carried by independent motion of conduction electrons, a conventional estimate of spin relaxation due to spin-orbit[2] or spin-ion core[3] interaction leads to too short a relaxation time to make such a resonance observable. If one assumes that the paramagnetism above the Curie temperature is due only to statistically oriented paramagnetic ion cores with atomic-range order, it is clearly impossible to transmit a coherent phase resonance through a sample thicker than a skin depth. The alternative,

which the shape of the resonance seem to corroborate, is to invoke superexchange or itinerant magnetic order with longer range than might be expected.

Figure 1 shows the experimental phenomena when the static magnetic field is parallel to the surface of the gadolinium sample. The bottom two traces show a paramagnetic-resonance absorption signal observed by reflection from the excitation cavity only. Trace (d) shows this strong signal by direct power measurement on a crystal detector, and trace (c) shows the same resonance using field modulation and a lock-in amplifier which gives the derivative. [The signal-to-noise of (c) is poorer than (d)
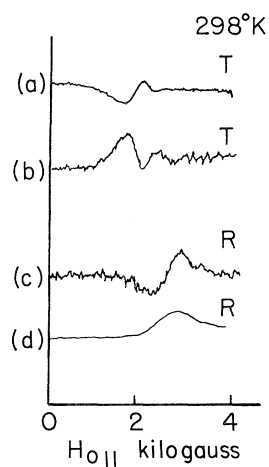

FIG. 1. Resonance traces in 3-mil gadolinium foil at 298°K with static field parallel to sample. (a) and (b) Transmission resonances at two phase settings. (c) Derivative of reflection signal. (d) Direct reflection signal.

because of a field-modulation amplitude which is very small compared to the wide lines.] Traces (a) and (b) show the transmission signal at two different phase settings. One notices the apparent shift of the resonance to low fields and the phase wiggles at the high-field tail which become more closely spaced as they become smaller. The transmitted resonances are not derivatives since the magnetic field was not modulated, but instead the modulation for lock-in detection was supplied by the insertion of a chopper modulator between the receiver cavity and the receiver.

The line shape is highly suggestive of a simple phenomenological model. Coherent long-range spin-wave excitations are propagated through the sample and have phase shifts of many times 2π from the excitation to the transmission side. This phase shift increases as the magnetic field increases. Figure 2 illustrates this conjecture more clearly. The reflection signal, which involves only the incoherent absorption of energy at one surface, is shown by the heavy dashed lines. The coherent transmitted signal starts out in phase at low field with a small $\vec{k}$ vector or small phase shift, but the phase shift increases as the field increases. In an idealized perfect section through

the sample, the signal might appear as shown in the fine solid line. However, in an actual sample with thickness variations and other sources of nonuniformity, the observed line appears as a phase-averaged collection of many such excitations which destructively interfere at higher fields, and this is shown by the heavy solid line. The transmitted line appears at lower fields as a consequence. The magnitude of such a large transmitted signal in the low-field tail of the absorption line is compatible with the experimental conditions if the reader takes note of the fact that the sensitivity of the apparatus to transmitted signals is about 10^{-19} W, a figure which is perhaps 100 dB smaller than the sensitivity of the crude but adequate detector which we have used to observe reflected signals from the excitation cavity.

On the assumption that such a form of excitation would undergo less anisotropic demagnetization and dephasing when the magnetic field is applied normal to the sample, the resonance was observed in this mode with the results shown in Fig. 3. The absorption resonance in reflection is moved to higher applied field because of internal-field demagnetization, but indeed the transmitted signal shows considerably greater strength and/or homogeneity of phase. In order to illustrate the approximate effect of temperature, Fig. 4 shows a parallel-field resonance at a temperature of 24° above the Curie point. It indicates that the signal is not degraded by much more than a factor of 2, and also shows that the reflection resonance occurs at a slightly higher applied field, closer to $g=2$, than the line in Fig. 1. We have not yet been able to make a careful study of the resonance in the immediate vicinity of the Cu-
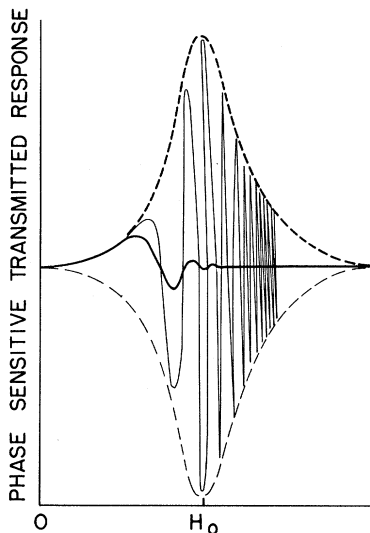

FIG. 2. Phenomenological origin of the resonance line shape. The heavy dashed line is the envelope of response corresponding to absorption. The light line is an idealized transmission response from a homogenous section of sample. The heavy continuous line is the effectively transmitted line after averaging over phase disorder from many idealized excitations propagated through a nonuniform sample.
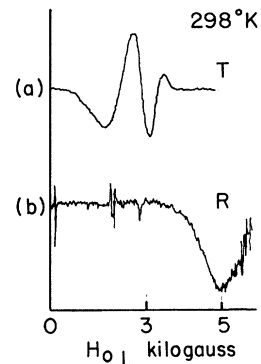

FIG. 3. (a) Transmission and (b) reflection signals from the sample of Fig. 1 when the static field is oriented perpendicular to the sample foil.
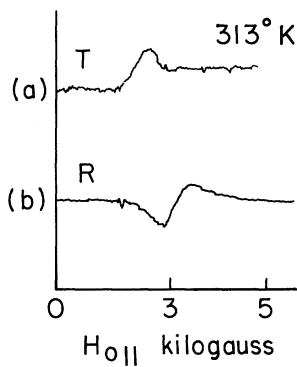
FIG. 4. (a) Transmission and (b) reflection signals at a temperature of 24° above the Curie point.

rie point, but the indications are that the described behavior extends comfortably above this point, and is not a critical-point phenomenon.

At temperatures below the Curie temperature there is no observable indication of this type of resonance. Figure 5 shows two characteristic traces, (b) and (c), for parallel-field ferromagnetic-resonance absorption, but trace (a) indicates the absence of such a signal in transmission to our limits of sensitivity. No transmission signal is seen when H_0 is perpendicular either. It should be remarked that we have not been able to observe a ferromagnetic-resonance signal transmitted through foils of comparable thickness of iron which, like gadolinium, is also a fairly hard ferromagnet. On the other hand, foils of easily permeable materials like Supermalloy (Conetic AA in our case) show transmission ferromagnetic-res-
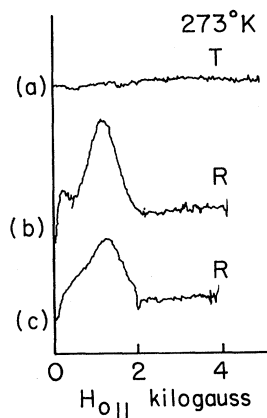

FIG. 5. (a) Transmission and (b) and (c) reflection traces at temperatures below the Curie point with static field parallel to the sample.

onance signals which are unappreciably different, except for attenuation, from the direct absorption resonance in reflection.

From our very preliminary data we have been unable to determine whether the dispersion in the propagation wave vector is proportional to the magnetic field H_0 or to $\omega - \gamma H_0$, or whether the phase shift may not arise from the more rudimentary process of increased damping as the absorption increases near resonance. One may make the estimate, however, from the fact that the sample is 75 μ thick and is probably pitted and irregular in thickness to 5 or 10% of this thickness, that the wavelength characterizing the phase of the transmitted excitation is at least 5 or 10 μ at low field. Since the peak of resonance transmission occurs at a considerably lower field than the peak of absorption, it is not easy from our preliminary data to estimate an attenuation length, especially since attenuation at the center of the line is presumed to be due to phase averaging. However, an estimate based on the extrapolation of the absorption-line tail to low fields, assuming that it is a Lorentzian shape, and on the known resonance signal strengths of each type of line, shows that the attenuation length is about the same magnitude, 5 μ. Although such an attenuation length is characteristic of conduction-electron resonance transmission in sodium or lithium at room temperature, it is inconceivably large for gadolinium on the basis of a model of independent electron motion through such a high-Z paramagnetic material.

This characteristic resonance shape suggests many possibilities and questions, in particular the two following: Is this a characteristic of all ferromagnetic systems above the Curie point? Is there a similar situation at or above the antiferromagnetic transition point in other materials? It is clear, in any case, that the experimental transmission technique is a most sensitive probe of this type of long-range order, and that the existence of such a long range of phase order is not what one normally expects of a paramagnet. We naturally hope that it may shed further light on the nature of the fundamental ferromagnetic interaction.

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ELECTROABSORPTION IN RUTILE (TiO₂)†

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The bands accounting for electrical conductivity in rutile have been of continuing interest to many investigators. The low carrier mobility has rendered as useless cyclotron resonance, de Haas–van Alphen measurements, and associated experimental techniques which have proven so valuable in the analysis of other materials. Conductivity and Hall-effect measurements[1-3] and, recently, piezoresistance measurements[4] have suggested multivalley conduction and high electron effective mass in this material, but because of the rather involved relationship such data have to possible band structures and various scattering mechanisms, the evidence supporting these conclusions cannot be regarded as having a unique interpretation. Optical measurements show the absorption edge of rutile is dichroic and has a strong temperature dependence.[5,6] It is, in general, difficult to resolve the different processes accounting for absorption from the measurement of the absorption spectrum in the vicinity of the edge. This is especially true of rutile because of the strong temperature dependence of its absorption tail.

Electroabsorption measurements have provided a more detailed understanding of the processes involved in the absorption of light at the edge in a number of materials, e.g., germanium[7] and silicon.[7,8] The results of such measurements on rutile, their interpretation, and their implications concerning the band structure and dichroic nature of this material at the absorption edge are reported herein.

Electroabsorption measurements were performed on rutile plates of approximately 100 μ thickness, each having the c axis and one of the a axes within the plane of the sample. Thin conducting platinum films vacuum-deposited on both sides of the Formvar-coated sample served as contacts for application of the perturbing electric field but did not interfere substantially with the transmission of the light. A description of the measuring technique appears in a previous publication.[8]

Two crystals were studied in considerable detail. The first, which shall be referred to as crystal A, was fired in oxygen at 20 atm at 700°C for six hours; the second, crystal B, was obtained from another source and was fired in atmospheric air at 800°C for 12 h.[9] Two samples were obtained from crystal A and one from crystal B.

The electroabsorption spectrum of rutile was measured with light polarized with E parallel to c ($E_\parallel$) and E perpendicular to c ($E_\perp$), with perturbing electric fields ranging from 1.5×10^4 V/cm to 6×10^4 V/cm at temperatures ranging from 77 to 200°K. Difficulties encountered in calibration procedures limited the accuracy of the magnitude scaling of the various electroabsorption spectra to $\pm 30\%$, but relative accuracy within a given spectrum was limited only by noise. All results are similar to those presented in Fig. 1, which were obtained with an electric field of 2×10^4 V/cm at 77°K. Stronger fields enhance and broaden all peaks, both positive and negative, but their magnitudes do not have the same dependence on field. As the temperature is raised, the effect fades in intensity and the peaks broaden. For instance, at 168°K the effect has faded to approximately one third in intensity and the peaks have broadened considerably. At room temperature the effect has diminished to the point that reasonable integration times are not adequate for detection. This dependence of intensity and peak width on temperature and the relationship of these results to the Urbach rule[10] will be dealt with in a future publication.

Several aspects of the data presented in Fig. 1 should be pointed out. Comparison of the results from the two crystals shows that the large low-energy positive (LEP) peak exhibited by