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SIMULTANEOUS OBSERVATION OF ABSORPTION AND DISPERSION SIGNALS AND LOW-POWER SATURATION EFFECTS IN THE PARAMAGNETIC RESONANCE OF $CaWO_4$:Fe^{3+}

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We have studied the effect of saturation on the paramagnetic resonance (pmr) absorption and dispersion signals from Fe^{3+} in $CaWO_4$. The striking feature of these studies is the decrease, with decreasing temperature, of the intensity of the absorption without accompanying changes in linewidth or line shape. While such a decrease would also occur for resonances arising from excited states due to depopulation of the excited levels, our experiments rule out this interpretation. In particular, the intensity of the dispersion signal increases when the temperature is reduced from 4.8 to 1.8°K, a fact which cannot be accounted for by an excited-state resonance. This is conclusive proof that the pmr of Fe^{3+} in $CaWO_4$ arises from a ground state as reported earlier,[1] and not from an excited doublet as claimed in a recent Letter.[2]

The distinguishing feature of this type of system is that the spin diffusion time[3] is longer than T_1, the spin-lattice relaxation time, whereas T_1 is itself longer than the inverse of the inhomogeneous linewidth. In this case, the line shape is unaffected by saturation, but the intensity of the saturated absorption line will decrease with decreasing temperature because of the increasing T_1. This behavior of the absorption line was observed for the first time for Cr^{5+} ions in $CaWO_4$,[4] and since then has been seen for Mo^{5+} and Nd^{3+} in the same host material. The present simultaneous observation of absorption and dispersion signals from T_1-limited spin packets[4,5] in Fe^{3+}-doped $CaWO_4$, which shows that the intensity of the dispersion signal is larger than the intensity of the absorption signal ($\chi' > \chi''$), is conclusive proof that the Fe^{3+} absorption resonance is indeed saturated at very low power levels in spite of its unsaturated appearance.

In these experiments, our bridge-type spectrometer[6] was operated in such a way that absorption and dispersion signals were displayed simultaneously on separate chart recorders. Two lock-in amplifiers, having a common 6-Kc/sec reference, were used. For purposes of intensity comparison, the two amplifiers were calibrated by changing the phase of the rf field in the reference arm of the bridge by 90° with respect to the phase of the rf field in the signal arm. This procedure changed the resonance displayed on a recorder from absorption to dispersion and vice versa. The power incident on the cavity (P_i) was measured by both dry calorimetric and thermistor methods. Low powers were obtained by means of a calibrated attenuator. The magnetic field modulation amplitude was slightly larger than the 0.27-G linewidth. Smaller modulation amplitudes increased saturation while larger amplitudes distorted the line shape severely so that the above compromise was desirable.

The results of experiments performed at

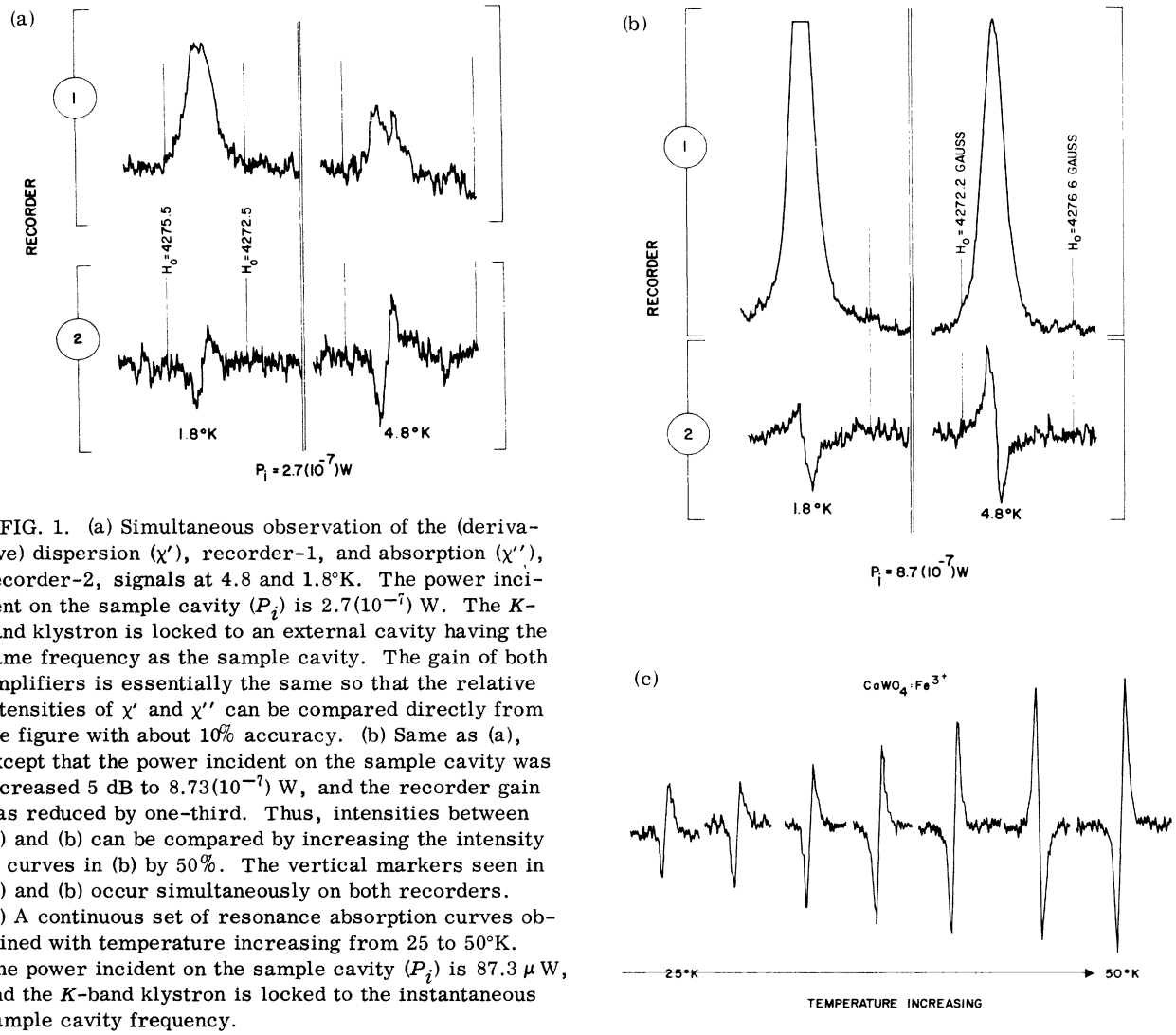

FIG. 1. (a) Simultaneous observation of the (derivative) dispersion (χ'), recorder-1, and absorption (χ''), recorder-2, signals at 4.8 and 1.8°K. The power incident on the sample cavity (P_i) is 2.7(10^{-7}) W. The K-band klystron is locked to an external cavity having the same frequency as the sample cavity. The gain of both amplifiers is essentially the same so that the relative intensities of χ' and χ'' can be compared directly from the figure with about 10% accuracy. (b) Same as (a), except that the power incident on the sample cavity was increased 5 dB to 8.73(10^{-7}) W, and the recorder gain was reduced by one-third. Thus, intensities between (a) and (b) can be compared by increasing the intensity of curves in (b) by 50%. The vertical markers seen in (a) and (b) occur simultaneously on both recorders. (c) A continuous set of resonance absorption curves obtained with temperature increasing from 25 to 50°K. The power incident on the sample cavity (P_i) is 87.3 μW, and the K-band klystron is locked to the instantaneous sample cavity frequency.

incident power levels $P_i = 2.76(10^{-7})$ and 8.73(10^{-7}) W and at temperatures 4.8 and 1.8°K are shown in Figs. 1(a) and 1(b). The top traces (recorder 1) are (derivative) dispersion (χ') signals, and the bottom traces (recorder 2) are (derivative) absorption (χ'') signals. At both power levels ($\Delta P_i = 5$ dB) it is seen that χ'' decreases by about a factor of two as the temperature decreases, as reported in reference 2. But the fact that χ' increases as the temperature decreases is decisive evidence against the interpretation of reference 2 that the resonance arises from an excited doublet approximately 5 cm^{-1} above the ground state. For if the latter were true, both χ' and χ'' would decrease with decreasing temperature below 5°K. However, the observed increase in χ' agrees well with that calculated for a ground-state resonance, while the decrease in χ'' can be accounted for by an increase in T_1 with decreasing temperature. It is important to note, in addition, that at the lowest temperature (1.8°K) the intensity of the dispersion signal is larger than the absorption signal, a fact which clearly demonstrates the presence of saturation.

Further evidence in support of our interpretation is provided by our experiments in the temperature range 25 to 50°K, performed at a power level (87 μW) presumably comparable to that of Kirton and Newman[2] (reported to be less than 100 μW). In this temperature range, the intensity of the absorption line should rise with decreasing temperature, provided the line is unsaturated. This is true whether the resonance arises from an isolated ground doublet

or from an excited doublet ~5 cm^{-1} above the ground state. As seen from Fig. 1(c), however, the absorption falls instead of rising with decreasing temperature. We conclude that this resonance is saturated and hence the resonance observed at the considerably lower temperatures used by Kirton and Newman is a fortiori saturated.

We are grateful to Dr. J. R. Shane for many helpful discussions, to Dr. R. Newman and Dr. R. Damon for their interest and suggestions, and to Mr. R. J. Brophy for performing the resonance experiments.

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OBSERVATIONS ON THE FERMI SURFACE OF ALUMINUM BY NEUTRON SPECTROMETRY

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We have determined the dispersion relations for phonons in aluminum at 80 and 300°K by slow-neutron spectrometry, using a three-axis crystal spectrometer at the 30-MW research reactor R2 in Studsvik.[1] Initial detailed measurements on a single dispersion curve failed to reveal Kohn anomalies of the kind reported by Brockhouse, Rao, and Woods[2] for lead, but later supplementary measurements in selected regions and an improved analysis of data have revealed such effects. They are too small to be obvious by direct inspection of a series of points on a dispersion curve, but the data are sufficiently accurate to allow the variation of slope along a curve to be followed, utilizing average values of the slope in the interval between two successive points, and to see irregularities which are significant in relation to the errors involved. This improved analysis has been applied to all data. Even on curves of slope versus wave number the effects are still not large, but we have two independent series of measurements (at 80 and 300°K) and the shapes of observed one-phonon resonances to refer to, and on this basis feel justified in assigning eight points on the Fermi surface of aluminum.

Figure 1 shows the expected sites of Kohn anomalies in aluminum. The construction is to draw the intersections of spheres with the triangular surface indicated; each sphere is of radius $2\rho_F$ and has its center at a reciprocal lattice point. The radius ρ_F of the Fermi sphere for free electrons is 1.13 (on the same scale as the figure; this scale is used for all wave vectors here). Circles derived from lattice points outside the plane of the figure are dashed. The intersection of two circles is "rounded off" if the centers of the spheres concerned are separated by twice a lattice vector – corresponding to Bragg reflection of electrons and gap formation. The rounding-off is schematic, and does not attempt to follow theoretical descriptions of the Fermi surface. The

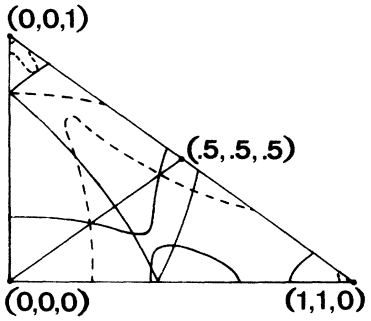

FIG. 1. Estimated positions of Kohn anomalies for a near-spherical Fermi surface.