does not alter the asymptotic edge,[15] there is a shift to lower photon energies of the apparent continuum absorption coupled with the disappearance of all exciton peaks at $\mathcal{S} > \frac{1}{4}$. Published GaAs absorption data[16] have been taken at temperatures which are too high to observe these effects. Preliminary data at liquid-nitrogen temperature[17] confirm the prediction of the shift in the continuum absorption in GaAs. A description of the experiments[18] in Cu_2O requires the selection of $m = 1$ in Eqs. (4). Our numerical calculations have not yet been extended to include this case.

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INTERNAL FIELD OF Fe^{57} IN Ni IN THE REGION OF THE CURIE POINT*

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Recent calculations of the spontaneous magnetization and susceptibility of theoretical ferromagnets emphasize the importance of the region around the Curie point in analyzing specific models. The magnetization near T_C is generally described by the equation

$$M = \text{const} \times (1 - T/T_C)^{\beta}, \qquad (1)$$

and a similar equation with exponent γ is used for the susceptibility. Numerical approximations of infinite series expansions yield $\beta \simeq 0.31$ [1,2] and $\gamma \simeq -5/4$ [1,3] for three-dimensional Ising ferromagnets, and $\gamma \simeq -\frac{4}{3}$ [3-5] for the Heisenberg model. These results, which are relatively insensitive to the details of lattice structure, spin, and range of interaction, differ from the molecular-field model values $\beta = \frac{1}{2}$ and $\gamma = -1$.

In measurements of the insulating ferromagnet EuS, Heller and Benedek[6] found that the zero-field resonant frequency of the Eu nuclei followed a $\frac{1}{3}$-power law (0.33 ± 0.015) over the range $0.9 < T/T_C < 0.99$. Experimental values for γ of metals appear to favor the Heisenberg model: for Fe, $\gamma = -1.37 \pm 0.04$ [7] and -1.33 [8]; for Ni, $\gamma = -1.35 \pm 0.02$ [9] and -1.29 ± 0.03 [10]; and for Co, $\gamma = -1.21 \pm 0.04$.[11] Direct measurements of the spontaneous magnetization of metallic ferromagnets in the critical region have not

been reported. In this Letter we describe Mössbauer-effect measurements of the internal field of Fe^{57} impurities in Ni, which provides a gauge of the magnetization of Ni very close to T_C.

In studying the internal field of Fe^{57} in Ni from 77°K to $0.9T_C$ we found[12] that the field does not follow the magnetization of the Ni exactly, but instead has a molecular-field temperature dependence,

$$h = B_J(\zeta T_C m/T), \qquad (2)$$

where h is the reduced field, m is the reduced magnetization, and ζ is an empirical constant. Equation (2), which was first proposed by Jaccarino, Walker, and Wertheim[13] to describe the behavior of Mn^{55} in Fe,[14] has since received additional theoretical justification by Callen, Hone, and Heeger.[15] For our samples, the parameter $\zeta = 2.8 \pm 0.5$, which presents a more conventional temperature dependence than of Mn^{55} in Fe, where $\zeta < 1$. As T approaches T_C, h according to Eq. (2) becomes more nearly proportional to m, and for $T/T_C > 0.95$, $\zeta = 2.8$, the linearity is within 5%. Although the molecular-field expression could not be tested above $0.9T_C$, it seems reasonable to expect it to continue into the critical region.

Our measurements were carried out on submillicurie sources of Co^{57} in high-purity Ni prepared in the standard manner, except that care was taken to remove absorbed hydrogen introduced by the chemical reduction procedure. The sources were mounted in a small vacuum oven designed to provide good temperature stability and homogeneity, and provided with a Be end window. The temperature of the sample holder was controlled by a servo consisting of a Chromel-Alumel thermocouple, dc comparison amplifier, and auxiliary heater, while the major heating power was supplied by an unregulated ac heater. The temperature of the sample was measured by an independent Chromel-Alumel thermocouple and potentiometer and was monitored by a dc amplifier and recorder. Temperatures could be held constant to within 0.5°C for 8 to 10 hours, while for short times the stability was comparable to the thermocouple sensitivity of 0.05°C. The thermocouples were calibrated at the melting points of Pb and Sn to establish the temperature scale approximately. All temperatures were referred to the thermocouple reading at T_C by direct calibration each day using a procedure described below.

Mössbauer spectra obtained with a multichannel spectrometer were resolved into six-line patterns at temperatures below $0.98T_C$. The width of the resolved hyperfine components of the best source was the same as the width of the single line of the source several degrees above T_C, 0.21 mm/sec versus a thin iron absorber. The nearly natural linewidth indicates good homogeneity, which was confirmed by the

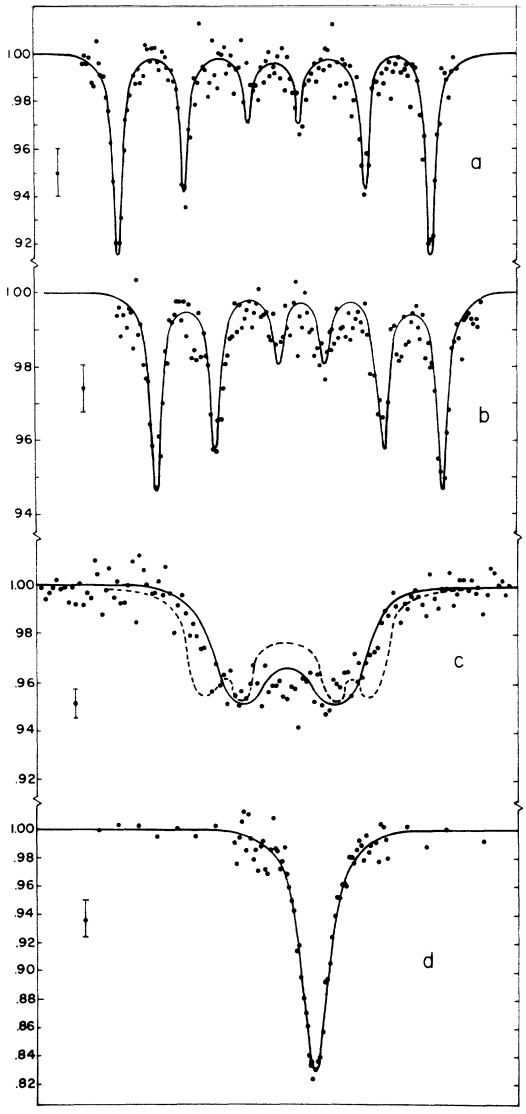

FIG. 1. Doppler spectra at various temperatures. Spectra are shown with different velocity scales. Curves are drawn according to Lorentzian component line shapes. (a) 295°K. (b) 565°K. (c) $T_C - 0.7$°K. The solid curve is drawn according to 3:3:1 intensities and $h = 0.11$. The dashed curve is for 3:3:1 intensities and $h = 0.14$, which field corresponds to an extrapolation of the $\frac{1}{3}$ power law. (d) $T_C + 0.6$°K.

measurements of the spectra just below T_C. Analysis of the unresolved spectra above $0.98T_C$ indicated that normal intensity ratios and relative separations continued to hold, and there was no evidence of the mixed and seven-line patterns obtained by Preston, Hanna, and Heberle[16] just below the Curie point of Fe. Several examples of the experimental patterns are given in Fig. 1. The apparent Curie point was located by a thermal scanning procedure similar to the method used for Fe,[16] by measuring the total 14-keV intensity transmitted by a stationary stainless-steel absorber as the source temperature was changed slowly. The changes, shown in Fig. 2, were an extremely sensitive indicator of the changing hyperfine splitting, and provided a thermocouple reference point defined to ±0.05°C, against which all other thermocouple readings were referred. Internal fields could be measured to ±5 kOe and were based on a calibration versus an Fe absorber.[16]

The data for the region $0.84 < T/T_C < 0.99$ follow an approximate $\frac{1}{3}$-power law. They are plotted in Fig. 3(a) in terms of the reduced field $h = H(T)/H(0)$, using the value $H(0) = 280$ kOe.[12] The empirical exponent is 0.33 ± 0.03 over the stated range. If the exponent is assumed to be $\frac{1}{3}$ exactly, a least-squares fit to the data has an intercept 0.75° below the temperature of the apparent Curie point determined by the thermal scanning calibration. We attempted to determine h closer to T_C indirectly, by analysis of transmission curves such as Fig. 2. The detailed shape of the transmission curves

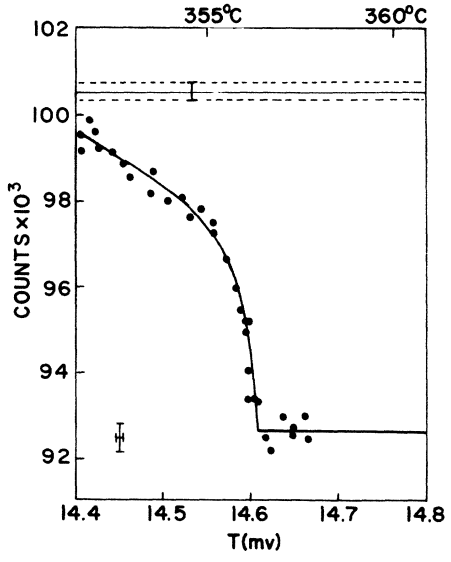

FIG. 2. Intensity transmitted by a stationary stainless-steel absorber as a function of Ni source temperature.

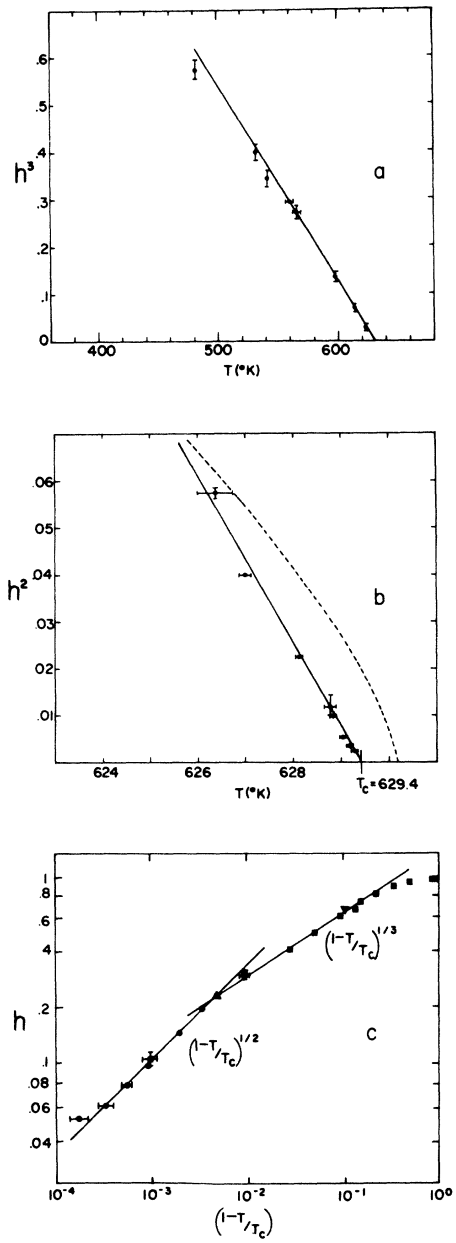

FIG. 3. Internal field at Fe^{57} impurities in Ni as a function of temperature. ■ Doppler measurement; ▼ thermal scan, FeF_2 absorber; ▲ thermal scan, Fe metal absorber; ● thermal scan, stainless steel. Due to the manner in which the transmission curves were analyzed, the uncertainty in field values is reflected as a temperature uncertainty only. (a) h^3 versus T. (b) h^2 versus T. The dashed line is a continuation of the $\frac{1}{3}$-power law found from the cooler region. (c) log-log plot of the entire region below T_C.

depends on the temperature dependence of all of the parameters of source and absorber spectra, including component line shapes, relative intensities and splittings, f factors, and relative line shifts. If all properties other than the internal field are known or constant, the transmission curves can be analyzed to yield field values at temperatures closer to T_c than we were able to maintain for the long periods required for complete Doppler spectra. By measuring the temperature shift at lower temperatures and above T_c, we established that there was no abrupt change in shift greater than 0.025 mm/sec. This limiting value, which was about twice the actual change found near T_c in Fe,[16] was small enough to cause no noticeable change in the transmission curve or the calculated field. We measured the f of the source, using a wide opaque absorber and the area method,[17] and found that the areas were constant to within 4% from 10° below to 3° above T_c. The absence of any large change in area near T_c in our work is in agreement with the findings of Preston, Hanna, and Heberle[16] that the Mössbauer fraction of Fe^{57} in Fe does not change markedly in the neighborhood of the Curie point.

Our analysis of the stainless-steel transmission curves was accordingly based on the measured line shapes of the absorber and of the single line of the source above T_c, the constancy of f, the measured temperature shift, and the assumption that the component line shapes, relative intensities and relative splittings of the source remained constant. A transmission curve versus an iron absorber was also obtained; and this curve yielded an important point at $T_c-3°$, at which the splitting of the Ni source could be compared to the splitting of Fe at room temperature. Field values calculated from the iron and stainless-steel transmission data were found to lie on a smooth curve which agreed with the Doppler-shift measurements at $T_c-3°$, and gave $h=0$ at T_c. These data conform to a $\frac{1}{2}$-power law fairly closely and are shown plotted in that form in Fig. 3(b). The empirical exponent for the range $0.996<T/T_c<0.9995$ is 0.51 ± 0.04. If $\beta=\frac{1}{2}$ exactly, a least-squares fit to the data yields the same thermocouple intercept as that given by the raw data, and corresponds to $T_c=629.4°K$.

After obtaining the transmission data, we improved the oven temperature control to enable us to examine the Doppler line shape in the $\frac{1}{2}$-power region. With the improvements, which included a completely new dc bridge, fully controlled dc heater, and better radiation shielding, we were able to stabilize the temperature to 0.1° at $T_c-0.7°$ for a 17-hour run. This Doppler spectrum, shown in Fig. 1(c), could be analyzed in terms of a six-line pattern having normal relative separations, but with a 3:3:1 intensity ratio rather than the 3:2:1 formerly obtained. The new ratio, which persisted at lower temperatures, seems to have been a result of the change from ac to dc of the main heater current, which causes a field of 3 Oe at the source. Analysis according to either ratio yields $h=0.11\pm0.01$ for this measurement, and it agrees with the $\frac{1}{2}$-power law. All data are shown on a single log-log plot, Fig. 3(c).

The appearance of a $\frac{1}{2}$-power law at temperatures extremely close to the transition is not completely unexpected theoretically. Landau's theory of second-order phase transitions[18,19] predicts that $\beta=\frac{1}{2}$ independent of model, although the result rests on assumptions that are not universally valid.[20] Calculations of Ising ferromagnets all yield $\beta\simeq0.31$, but the exponent for a Heisenberg ferromagnet has not been calculated. Susceptibility measurements on Ni and Fe are more closely described by the exponent $\gamma=-\frac{4}{3}$ corresponding to the Heisenberg model. Recently, Callen and Callen[21] reported two-spin cluster calculations of the magnetization of Heisenberg ferromagnets below T_c. Their results for various first- and second-neighbor interactions indicate an exponent of approximately $\frac{1}{3}$ over a wide range of temperatures below T_c, with an increase toward the value $\frac{1}{2}$ of the Landau theory and the molecular-field model at the Curie point itself.

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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