probability of its occurrence is rather small and the attempted atomic interchanges may be largely unsuccessful. In other words, diffusion would be highly correlated as envisioned in the free-volume model by Turnbull and Cohen[13] with an effective frequency much smaller than the normal Debye atomic frequency thus lowering D_a^0 by orders of magnitude. The precise value of D_a^0 would depend on the number of atoms involved in the cluster, the thermal vibrations, and the availability of the free space; it is not yet possible to quantify them at this stage. Energetically, the diffusion process would be easier in view of the looser atomic packing in the amorphous phase and a relaxed state at the saddle point through which atomic jumps are made.

Finally, as seen in Fig. 2, the Ag^{110m} profiles show spatial shifts which were quite unexpected. Electron microprobe and Auger spectroscopic studies indicated a presence of ~1 at.% dissolved oxygen in the as-splat-quenched specimens. During diffusional annealing the oxygen diffused out as evidenced by formation of progressively thicker silicon oxide layers on the specimen surface and oxygen depletion in the interior. While a precise explanation of the observed shifts must await detailed studies, it may be remarked that the presence of the dissolved oxygen in the splat-quenched amorphous Pd–19-at.%-Si alloy has been characterized for the first time.

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New Method for Investigating Magnetic Tricritical Points*

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A new method, based on the observation of hysteresis effects at the first-order phase transition, has been used to study the behavior of dysprosium aluminum garnet near one of its tricritical points. The magnetization-temperature phase diagram and the exponents β_u, δ_+, δ_-, and γ_u have been determined and found to be in good agreement with the present theory.

Recent theoretical studies[1] have given clear predictions for the asymptotic behavior near tricritical points (TCP's). The exponents turn out to be the same as those given by classical Landau theory, in sharp contrast to the situation at ordinary critical points, and it is clearly of interest to test this prediction. TCP's have been identified experimentally in a wide variety of physical sys-

tems,[2-6] but so far relatively complete results are available only for ^{3}He-^{4}He mixtures.[2] These results are in good agreement with the theory but the rather unique nature of the ^{3}He-^{4}He system makes it desirable to extend the study to other materials. Careful neutron-scattering experiments have been reported on the structural phase transition in ND_4Cl, but only the tricritical exponent β_t was measured.[3,7] More recently, there have been reports of a number of detailed experiments on a magnetic system, $FeCl_2$, in which the exponents $\beta_\perp$ and β_t were measured.[4,5] However, the results obtained for $\beta_\perp$ by two different techniques were in serious disagreement and so far this difficulty has remained unresolved. Experiments on magnetic systems are generally difficult to analyze since there are intrinsic uncertainties in detecting the onset of the first-order phase separation, which become very large near the TCP. In this paper we shall describe a new experimental method which overcomes some of these difficulties and the application of the method to the Ising-like antiferromagnet dysprosium aluminum garnet (DyAlG).[8]

Previous experiments on magnetic systems have located the first-order phase boundaries close to the TCP by measuring either the magnetization or the sublattice magnetization as a function of the externally applied field, correcting for demagnetizing effects and observing the discontinuities in the gradient (kinks) which occur at the phase boundaries.[9] In the case of the upper-phase-boundary determination using neutron scattering this gives a good estimate,[4] but in all other cases one has the problem that the kinks become less and less distinct as one approaches the TCP since the susceptibility on either side of the phase boundary diverges at the TCP. The problem is compounded by the fact that the demagnetizing corrections also become more important as the susceptibility increases. To overcome these difficulties, we have developed a method which identifies the region of coexistence without any reference to the corresponding values of the magnetization.

The method is based on the recognition of the fact that dynamic magnetization processes in the coexistence region are basically different from those of either the antiferromagnetic or paramagnetic phases. In single-phase regions, magnetization proceeds via individual spin reversals, while in mixed-phase regions, macroscopic processes involving domains take place. Based on our understanding of such domains[10,11] one might expect a relatively slow response to changes in the applied field, in contrast to more rapid relaxation processes in the single-phase regions. Changes in the low-frequency response should therefore signal the onset of the first-order transition.

The sample was a 4.658(3)-mm-diam sphere of DyAlG, aligned with the field parallel to a $\{110\}$ axis. We note that the complication of an induced staggered field[12,10] which is present in DyAlG for other orientations [e.g., $\{111\}$] should be rigorously zero for fields along $\{110\}$.[12] The sample was immersed in superfluid ^{4}He, to ensure good thermal equilibrium, inside a simple mutual-inductance coil consisting of two secondaries connected in opposition, surrounded by a primary coil. The coil was located inside a superconducting magnet. The temperature and field were controlled and measured with relative accuracies of ± 0.5 mK and ± 0.2 Oe, respectively. The absolute errors were approximately ± 5 mK and ± 25 Oe, respectively.

The low-frequency response of the system was measured by reversing a small dc current in the primary coil and recording the induced voltage across the secondaries on a multichannel analyzer. This procedure could be repeated several times to improve the signal-to-noise ratio, and the corresponding measurement with the sample removed from the coil was used to allow for the small signal due to the empty coil. It was thus possible to measure $\partial M/\partial t$ as a function of t, where M is the magnetization and t is the time.

The $(\partial M/\partial t)$-t traces for all fields that were investigated were found to be qualitatively similar in form and approximately exponential with time constants of the order of a few tenths of a second. However, a more detailed study showed that the traces for fields in the mixed-phase region were in fact functions of the past magnetic history of the sample, in contrast to the behavior in the single-phase regions. The hysteresis could be measured quantitatively by taking the difference between corresponding sets of $(\partial M/\partial t)$-t traces following two particular field cycles, using the multichannel analyzer, and integrating the resulting difference curve over an appropriate time interval with an integrating digital voltmeter.

A typical plot of integrated hysteresis as a function of applied field is shown in Fig. 1(a). It can be seen that the hysteresis goes to zero quite rapidly at both phase boundaries, locating them to better than ± 4 Oe. The primary field, h, used

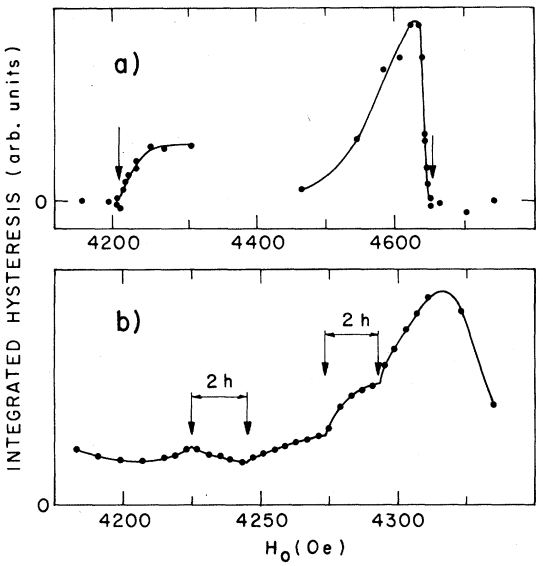

FIG. 1. (a) Integrated hysteresis versus applied field, H_0, at $T = 1.640$ K ($\epsilon = 0.093$). The arrows mark the location of the phase boundaries. (b) Integrated pseudohysteresis versus H_0 at $T = 1.791$ K ($\epsilon = 0.009$). Note that there are two pairs of kinks with both the upper and lower pair separated by 20 Oe, twice the primary field used.

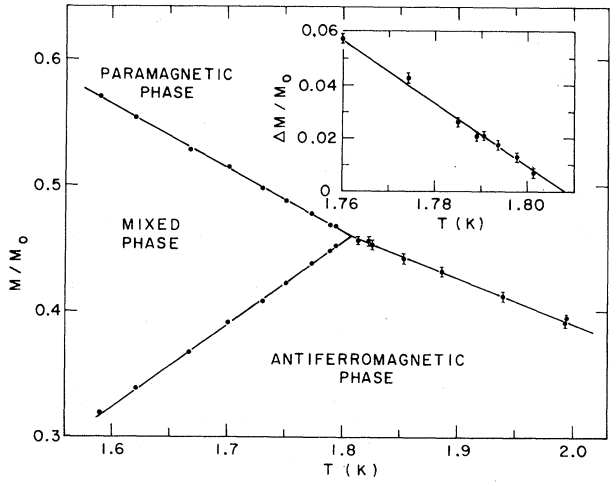

FIG. 2. M-T phase diagram. M is here plotted in terms of $M_0 = 545$ emu/cm^3, saturation magnetization calculated from Ref. 8. The inset shows data for the discontinuity close to the TCP.

here and for most of the measurements was 10 Oe, but similar experiments with different primary fields gave the same locations for the edges of the mixed phase.

This method was used to map out the phase boundaries for $T \leq 1.7$ K, corresponding to $\epsilon = |T - T_t| / T_t \sim 0.06$, where T_t is the tricritical temperature. Unfortunately for temperatures closer to T_t the hysteresis measured in this way was too small to observe. However, we did discover a second related effect, which we shall term a "pseudohysteresis" effect, which persisted to higher temperatures. We found that the $(\partial M/\partial t)$-t traces also depended upon the initial polarity of the primary field (i.e., parallel or antiparallel to the field of the superconducting magnet) and that this effect was different in the single-phase and mixed-phase regions. To get a quantitative measure of this effect, we took the differences between the $(\partial M/\partial t)$-t traces for different polarities and integrated the resulting curves. Typical results are shown in Fig. 1(b). It can be seen that the curve has four characteristic "kinks," the kinks occuring in pairs with the members of each pair spaced twice the primary field apart. We interpret the kinks as occuring whenever the field of the solenoid plus or minus the primary field just overlaps a phase boundary and this interpretation is supported by experiments with different primary fields. The kinks thus serve to locate the phase boundaries and we estimate the accuracy as typically ± 2 Oe.

The detailed interpretation of the pseudohysteresis effect is quite complicated, and we shall discuss this in more detail elsewhere, but for the present we can conclude that pseudohysteresis effects serve to locate the phase boundaries quite accurately. In our experiments the pseudohysteresis effect was observable very near the TCP and using it in conjunction with the regular hysteresis we were able to map out the phase boundaries below the TCP over the range $0.2 > \epsilon > 0.003$.

Measurements of the magnetization corresponding to the phase boundaries could be made *in situ* using the same apparatus. The sample was simply pulled out of the coil and the induced voltage across the secondary was integrated using the digital voltmeter. The relative accuracy in $\Delta M/M_t$ was about 6×10^{-4}, where M_t is the tricritical magnetization.

In order to locate the second-order phase boundary (the λ line) for $T > T_t$, we estimated the gradient $\partial M/\partial H$. Sharp peaks were found in the expected region, and these served to locate the λ line to better than ± 2 emu/cm^3 in M (± 2 Oe in H_i, the internal field). The phase diagram combining all our results is shown in Fig. 2.

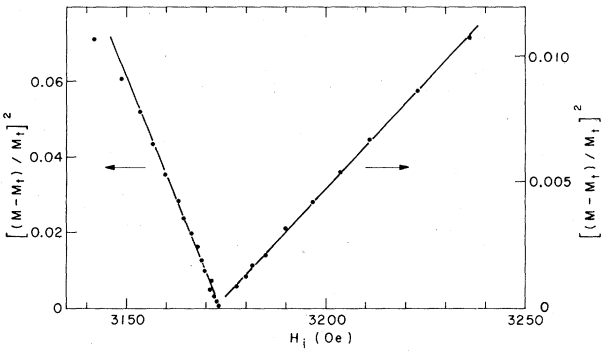

FIG. 3. $[(M-M_t)/M_t]^2$ versus internal field, H_i, for $T=1.808$ K$\simeq T_t$. $M_t=250.6$ emu/cm^3 was determined independently from the data of Fig. 2. Note the different scales for $H_i > H_t = 3173$ Oe and $H_i < H_t$.

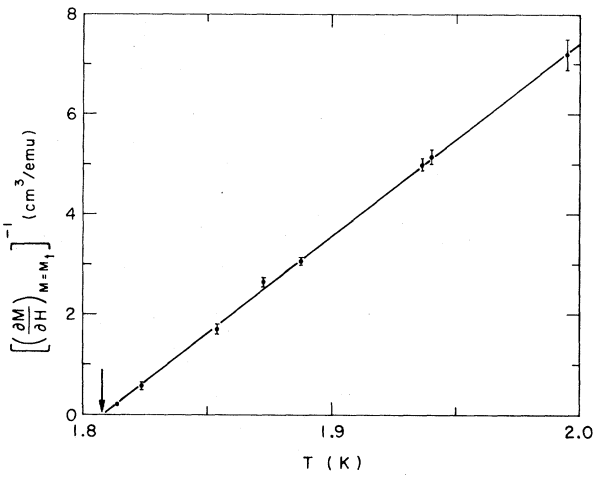

FIG. 4. $[(\partial M/\partial H)_{M=M_t}]^{-1}$ versus T for $T > T_t$. The arrow indicates the tricritical temperature as determined from the data of Fig. 2.

The most striking feature of Fig. 2 is the fact that all three phase boundaries appear to approach the TCP *linearly*, as predicted. In addition, we see that the slope of the λ line and the upper phase boundary below T_t are not equal, also as predicted by recent theories,[13] but in contrast to Landau theory. Similar discontinuities in slope have previously been observed also in other systems.[2,4,5]

In Fig. 3 we show results for the tricritical isotherm, where we have plotted $[(M-M_t)/M_t]^2$ versus H_i, using the value of M_t derived from the data of Fig. 2. We see that the data are in good agreement with the prediction, $\delta_\pm = 2$, although the asymptotic region below the tricritical field, H_t, is quite small ($\Delta H_i < 6\times10^{-3}H_t$). We note that the data above and below H_t were taken on different days, and their good agreement with respect to the value of H_t is a gratifying check on our technique.

Finally in Fig. 4, we plot the inverse susceptibility $[(\partial M/\partial H)_{M=M_t}]^{-1}$ as a function of T. The data are again seen to be consistent with the theoretical prediction, $\gamma_u = 1$.

Numerical analysis of the data shown in Figs. 2–4 assuming simple power-law singularities yields the following estimates for the four TCP exponents which we have determined:

$$\beta_u = 0.98 \pm 0.05 \quad (0.2 > \epsilon > 0.003),$$

$$\gamma_u = 1.01 \pm 0.07 \quad (0.1 > \epsilon > 0.003),$$

$$\delta_+ = 2.12 \pm 0.24 \quad (0.02 > \Delta H_i/H_t > 0.002),$$

$$\delta_- = 2.14 \pm 0.26 \quad (0.006 > \Delta H_i/H_t > 0.0002),$$

where we have also indicated the ranges of temperature and field over which the fits were made. We are not able to detect any evidence for logarithmic correction terms[1] within the limits of our accuracy and resolution. Further details of the experiments and analysis will be given elsewhere together with a discussion of the application of scaling theory to our results and the form of the H-T phase diagram.

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Measurement of Spin-Flip-Raman-Scattering Cross Section and Exchange Effects for Donors in CdS by Faraday Rotation

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The simple connection between spin Faraday rotation and spin-flip-Raman-scattering (SFRS) cross section is used to deduce the SFRS cross section for donors in CdS from Faraday-rotation measurements. The Faraday rotation also reveals an antiferromagnetic exchange interaction between donors.

We have measured the Faraday rotation (FR) due to the donor spins in n-CdS at several different optical wavelengths and donor concentrations. These rotations become huge at near-resonant light excitation. In view of the very simple connection between the spin-flip-Raman-scattering (SFRS) cross section ($d\sigma/d\Omega$) and spin FR, which we express directly in terms of a "Raman dipole," one can thus determine the SFRS cross section by the measurement of FR. The latter is uncomplicated by the necessity for corrections for surface and bulk absorption, reflectivity, geometrical factors, etc., which trouble any cross-section measurement as evidenced by the orders-of-magnitude disagreement in $d\sigma/d\Omega$ quoted in the literature.[1,2] In addition, the near-resonant spin FR provides a very sensitive means of measuring interactions between electrons.

In a previous communication, we showed that for cubic symmetry, Raman scattering of a light field $\vec{E}_L \cos(\omega_L t)$ from any two time-reversed states $|a\rangle$ and $|b\rangle$ separated by a Zeeman energy $\hbar\omega_{ba}$ is conveniently represented by matrix elements between states $|a\rangle$ and $|b\rangle$ of an effective Raman dipole[3]

$$\vec{D}^{(2)} = \vec{\sigma}\times\vec{E}_L[\alpha\exp(-i\omega_L t)+\text{c.c.}] + [\beta\vec{E}_L\tilde{I}\exp(i\omega_L t)+\text{c.c.}]. \quad (1)$$

Here $\vec{\sigma}$ is the Pauli spin operator, $\tilde{I}$ is the unit matrix, and β and α are second-order matrix elements of the electric dipole operator $e\vec{r}$ of the type

$$\alpha = \sum_n \tfrac{1}{2} i \langle a|ex|n\rangle\langle n|ez|b\rangle/(E_n - E_a - \hbar\omega_L). \quad (2)$$

β is the usual polarizability associated with the dielectric constant and forward Rayleigh scattering. Spontaneous Raman scattering between $|a\rangle$ and $|b\rangle$ is given by the Raman electric dipole, $\langle b(t)|\vec{D}^{(2)}|a(t)\rangle$. By appropriate insertion of this off-diagonal dipole into the classical radiation formula, the spontaneous differential SFRS cross section is given by

$$(d\sigma/d\Omega)_{SF} = 4|\alpha|^2(\omega_L \mp \omega_{ba})^4/c^4. \quad (3)$$

However, in addition to the component of $\vec{D}^{(2)}$ connected with β which radiates at the unshifted frequency ω_L, there are additional diagonal components associated with α which give rise to a