Phase Diagram of the Isotropic Antiferromagnet $RbMnF_3$: Test of Scaling and Renormalization-Group Calculations*

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The variation of the ordering temperature $T_c(H)$ of the (very nearly) isotropic antiferromagnet $RbMnF_3$ with magnetic field H was determined from thermal expansion measurements in fields up to 180 kOe. With increasing H, $T_c(H)$ first increases, then reaches a maximum, and finally decreases. The bow-shaped phase boundary, in the T-H plane, is well described by recent theories based on scaling and renormalization-group calculations.

Recent theories, based on scaling and renormalization-group calculations,[1-3] have stimulated a detailed re-examination of multicritical points in antiferromagnets. The new experimental investigations focus on the recently predicted departures from the results of mean-field theory. Thus far, the experimental tests have been confined to easy-axis antiferromagnets.[4-7] In this Letter we present experimental data which confirm, for the first time, the striking predictions of the new theories for isotropic antiferromagnets.

In mean-field theory, the ordering temperature $T_c(H)$ of an isotropic antiferromagnet decreases quadratically with increasing H, for low H. In contrast, the new theories[2,3] indicate that as H is increased from zero, $T_c(H)$ first increases, then reaches a maximum, and decreases only later. The predicted phase boundary, in the temperature–magnetic-field (T-H) plane, is a bow-shaped curve which (for low H) is given by the equation

$$\Delta T \equiv T_c(H) - T_N = AH^{\tilde{\Psi}} - BH^2, \qquad (1)$$

where $T_N \equiv T_c(H=0)$ is the Néel temperature, A and B are positive constants, and $\tilde{\Psi} = 1.60$ for a three-dimensional isotropic Heisenberg antiferromagnet. The Néel point of such an antiferromagnet is a multicritical point.[2]

To test these predictions we have measured the H dependence of T_c for the cubic antiferromagnet $RbMnF_3$, which closely approximates the ideal isotropic Heisenberg antiferromagnet.[8] For this material $T_N = 83$ K, and the exchange and anisotropy fields at $T = 0$ are $H_E(0) = 8.9\times10^5$ Oe and $H_A(0) = 4$ Oe, respectively.[9,10] For $T \lesssim T_N$, spin reorientation (analogous to the spin-flop transition in a uniaxial antiferromagnet) is expected to occur[10] at fields of order $[H_E(0)H_A(0)]^{1/2} \sim 10^3$ Oe. At much higher fields, the phase transitions of $RbMnF_3$ are expected to approximate closely those of an ideal isotropic antiferromagnet. Therefore, our conclusions, which are based on data taken in steady fields up to 180 kOe, are not expected to be influenced significantly by the anisotropy.

The ordering temperature $T_c(H)$ of a $RbMnF_3$ single crystal was obtained from thermal expansion measurements. The volume of the sample is equal to the partial derivative of the thermodynamic potential $\Phi(H,T,P)$ with respect to the pressure P The thermal expansion coefficient is proportional to $\partial^2\Phi/\partial P\,\partial T$ and, therefore, exhibits a λ anomaly at the order-disorder transition, as do other second derivatives of Φ. The λ anomaly in the thermal expansion coefficient at $H = 0$ was observed earlier by Teaney, Moruzzi, and Argyle[11] and by Golding.[12]

In the present experiments, the length $l(H,T)$ along the [100] direction was measured as a function of T at various fixed $\vec{H}$ applied along [100]. A three-terminal capacitance dilatometer, made of copper, was used.[13] The capacitance cell was enclosed by a copper can filled with helium exchange gas. A second (outer) copper can surrounded the first (inner) can. The vacuum space between the two cans was pumped continuously to less than 10^{-6} mm Hg. The only physical contact between the inner can and the outside environment was a thin tube of "nonmagnetic" stainless steel (No. 304). Temperatures were measured with a thermistor resistance thermometer[14] attached to the capacitance cell. The thermistor was calibrated *in situ*, at $H = 0$, against an adjacent platinum resistance thermometer. The small magnetoresistance of the thermistor was determined in a series of independent measurements.[15] This magnetoresistance was quadratic in H and (for temperatures near T_N) was equivalent to -40 ± 9 mK at 180 kOe.

The data presented below were obtained with liquid nitrogen (boiling point 77 K) surrounding

the outer copper can. For each fixed H, the capacitance cell was first heated to a temperature above $T_c(H)$, and the data for l vs T were taken subsequently as the cell cooled slowly (~10 mk/min) toward 77 K. Because high-precision thermometry at high fields was an important requirement in these experiments, several checks on the thermometry were made. In one check, the thermal expansions at $H=0$ and $H=100$ kOe were also measured with liquid argon (boiling point 87 K) surrounding the outer copper can, which caused the capacitance cell to warm slowly. Since all the measurements were made near 83 K, the replacement of liquid nitrogen by liquid argon produced a (near) reversal of the temperature gradient in the cell. The results of this check indicated the existence of a temperature difference of several millidegrees Kelvin between the sample and the thermistor thermometer. However, between $H=0$ and 100 kOe this difference did not change by more than 6 mK. As a further check, the thermal expansions at five fixed values of H, up to 174 kOe, were measured with a second thermistor thermometer attached directly to the sample with G.E. 7031 varnish. Liquid nitrogen surrounded the outer copper can. The maximum deviation between any of the values of $T_c(H)-T_N$ obtained in this check and the results presented below was 7 ± 6 mK. The limited data obtained in this check led to conclusions which agreed with those based on the more extensive data below.

The data for $l(H,T)$ vs T were recorded on a punch tape and were differentiated numerically by a computer to obtain $\partial l(H,T)/\partial T$. Results for $\partial l/\partial T$ vs T at three fixed values of the applied field are shown in Fig. 1. For each H, the transition is marked by a λ peak in $\partial l/\partial T$. The small rounding of the peak is caused, in part, by the differentiation procedure.

Two different criteria for defining $T_c(H)$ were used. In one procedure, $T_c(H)$ was identified as the temperature at which $\partial l/\partial T$ was maximum. In the second procedure, we estimated the temperature at which $\partial l/\partial T$ would have been maximum if the peak had not been rounded. These two criteria led to values of $T_c(H)$ which differed by ~3 mK. However, the difference $\Delta T=\Gamma_c(H)-T_N$ was the same to within 2 mK. As a final value for for ΔT we chose the average of the values obtained using the two procedures. The uncertainty in the final values of ΔT can be divided into three parts: (1) A random error which did not exceed 5 mK. (2) A possible systematic error, proportional to H^2, due to the uncertainty in the determination of the magnetoresistance of the thermistor thermometer. The estimated maximum error due to this cause is 9 mK at 180 kOe; the main effect of such an error would be to change the experimentally derived value of the parameter B in Eq. (1) by less than 1%. (3) Other systematic errors, if any, are expected to vary monotonically with H and with ΔT, and are estimated to be smaller than 7 mK. None of these experimental uncertainties in ΔT are expected to have a significant effect on the results or conclusions of the present work. The Néel temperature of our sample was $T_N=83.13\pm0.04$ K, where the uncertainty in this case represents the absolute accuracy for T (rather than the precision for changes in T).

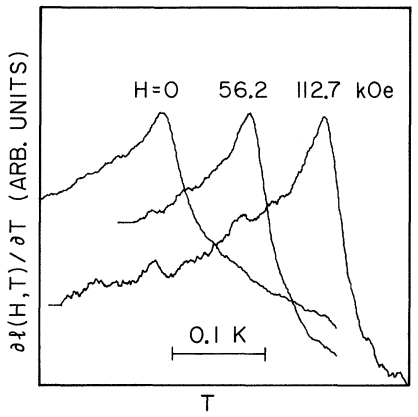

FIG. 1. Temperature dependence of $\partial l/\partial T$, where l is the length of the $RbMnF_3$ sample, at $H=0$, 56.2, and 112.7 kOe. The ordinate scale is not exactly the same for all three curves.

The phase boundary of $RbMnF_3$, in the T-H plane, is shown in Fig. 2. These results were corrected for the demagnetizing field which was equal to ~0.07% of the applied field. The overall shape of the phase boundary is in agreement with recent theoretical predictions,[2,3] but is in complete disagreement with mean-field theory. The solid curve in Fig. 2 is a least-squares fit to all the data with Eq. (1), where $\tilde{\Psi}$ is set equal to 1.60, and A and B are treated as adjustable parameters. For this fit, $A=5.818\times10^{-9}$ and $B=4.173\times10^{-11}$, where ΔT is in units of K and H is in oersteds. The agreement between the data and the fit is excellent. The rms deviation between the observed values of ΔT and the fit in Fig. 2 is $\sigma=1.48$ mK; and the maximum deviation is 3.6 mK. Similar fits with Eq. (1) were also made, each with a different fixed values of $\tilde{\Psi}$. The fits showed clear systematic deviations from the data

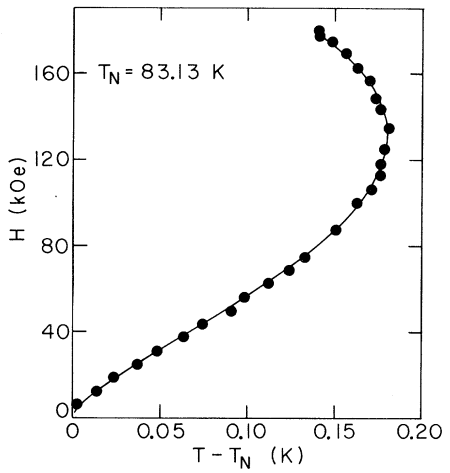

FIG. 2. Dependence of the order-disorder transition temperature $T_c(H)$ of $RbMnF_3$ on magnetic field H. The solid curve is a least-squares fit to the data of Eq. (1) with $\tilde{\Psi}$ set equal to 1.60.

whenever $\tilde{\Psi}$ was outside the range $1.4 < \tilde{\Psi} < 1.8$. The rms deviations for the fits with $\tilde{\Psi} = 1.4$ and $\tilde{\Psi} = 1.8$ were both equal to 3.6 mK, i.e., a factor of 2.4 larger than for $\tilde{\Psi} = 1.6$. A best fit with Eq. (1) in which $\tilde{\Psi}$ as well as A and B were allowed to vary gave $\tilde{\Psi} = 1.590$, $A = 6.366 \times 10^{-9}$, and $B = 4.044 \times 10^{-11}$, with $\sigma = 1.47$ mK. This fit was practically indistinguishable from the curve in Fig. 2 for which $\tilde{\Psi}$ is held fixed at its theoretically predicted value 1.60.

In mean-field theory, $\Delta T = -B^* H^2$, where B^* is a positive constant. Using the mean-field expression for B^* in terms of T_N [Shapira and Foner,[16] Eq. (4)], one obtains $B^* = 3.2 \times 10^{-11}$ K/Oe^2, which is comparable to the values for B derives from fits of the present data by Eq. (1) (with $\tilde{\Psi} = 1.60$, or with an adjustable $\tilde{\Psi}$). This supports the statement in Ref. 3 that the term $-BH^2$ in Eq. (1) represents the usual depression of T_c by a magnetic field, present even in mean-field theory.

Equation (1) may be rewritten in terms of a reduced temperature $t = [T_c(H) - T_N]/T_N$ and a reduced field $h = H/2H_E(0)$, namely,

$$t = ah^{\tilde{\Psi}} - bh^2. \qquad (2)$$

The normalization of H by the field $2H_E(0)$ is motivated by the observation that the order-disorder transition at $T = 0$ should occur[8] at $H = 2H_E(0)$. A least-squares fit of our data by Eq. (2) with $\tilde{\Psi} = 1.60$ gives $a = 0.70$ and $b = 1.59$. If $\tilde{\Psi}$ is also allowed to vary then $\tilde{\Psi} = 1.59$, $a = 0.67$, and $b = 1.54$. Thus, both a and b are of order unity.

In conclusion, the experimentally determined phase diagram of the (very nearly) isotropic antiferromagnet $RbMnF_3$ confirms the predictions of the theory by Fisher, Nelson, and Kosterlitz.

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[13]The basic dilatometer design described by G. K. White [Cryogenics 1, 151 (1961)] was modified to accommodate an insulating sample. The dilatometer measures the change in the sample's length relative to that of the copper cell. Over the narrow temperature interval at which our measurements were performed, the monotonic thermal expansion of the copper cell contributed an essentially constant background to the temperature derivative of the signal. This background had a negligible effect on the determination of $T_c(H)$.

[14]Manufactured by Keystone Carbon Co., St. Marys,

Pa.; Model No. L0904-125-H-T2.

[15]The magnetoresistance of the thermistor was measured at the boiling points of liquid nitrogen and liquid argon, with the thermistor in each case immersed in a short column of the boiling liquid. The magnetoresistance near T_N was then obtained by interpolating between the two sets of data. Additional measurements of the magnetoresistance near T_N were made with the same arrangement (with the two cans) as in the thermal expansion measurements. Each data point near T_N was taken by first stabilizing the temperature and then changing H from zero to a given value and then back to zero. The time in which the field was changed was short compared to the time in which the temperature drifted by several millidegrees Kelvin, but was sufficiently long to avoid significant eddy-current heating. The results of the two methods of determining the magnetoresistance near T_N were in agreement with each other.

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Quasiparticle Propagation and Recombination in Bulk, Superconducting Pb

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The propagation characteristics of quasiparticles in bulk superconducting Pb single crystals is studied. A transition from quasiparticle diffusion to diffusion of the combined gas of quasiparticles and phonons is observed as the temperature is increased. The *intrinsic* quasiparticle recombination time, as well as the decay time of the quasiparticle density, is determined. The latter is found to be at least an order of magnitude longer than this quasiparticle recombination time.

Recently there has been considerable activity in the study of nonequilibrium superconductivity.[1] Most of the experimental studies to date have involved a study of the superconducting transition of a thin film or the I-V characteristics of thin-film tunnel junctions under the influence of laser excitation. In this Letter, we report on the propagation characteristics of photoexcited quasiparticles in bulk, single-crystal, superconducting lead using a tunnel junction as a quasiparticle detector and time-of-flight techniques with nanosecond resolution. The combined use of *bulk* samples and time-of-flight techniques makes possible a *direct* determination of the spatial and temporal behavior in the nonequilibrium state in a heretofore unexplored regime.

We present data which show a transition from diffusive heat propagation (in the combined gases of quasiparticles and phonons) to a pulse of diffusing quasiparticles as the temperature is lowered below $T \sim 2.8$ K. The quasiparticle scattering rate, in the high-temperature regime, is shown experimentally to be equal to the quasiparticle recombination time as determined from calculations using tunneling data. The low-temperature pulse reflects the decoupling between the local temperature and the quasiparticle number.

The experiments were performed on high-purity (99.9999%) Pb single crystals with propagation direction [111] and thicknesses 4.5, 2.2, and 0.87 mm. The resistance ratio ($R_{300\ \mathrm{K}}/R_{2\ \mathrm{K}}$) of these samples is of the order of 20 000. The samples were polished using a combined mechanical and chemical technique and suspended from a single point to avoid straining them during the cooldown in the liquid helium cryostat. One surface of the sample was cleaned by back-sputtering in argon to remove undesirable surface contaminants. A controlled oxide layer was then grown on this surface and a thin lead film was evaporated on top to form the tunnel junction. The size of the junction was $\sim 0.25 \times 0.45$ mm^2 and the normal-state resistance was ~ 20–50 mΩ. Silicon oxide and photoresist were used to insulate the lead film from the lead crystal outside the junction area. The photoexcited quasiparticles were generated at the opposite surface of the crystal by means of a dye laser which was in turn pumped by a nitrogen laser. The laser had a peak power of ~ 1 kW (only a fraction of which was absorbed by the crystal) with a rise time of 1.5 ns and a pulse width of 5 ns. The beam was about 0.2 mm in diameter. The laser and power supply were located outside a shielded room in which the cryostat was placed to eliminate undesirable electrical pickup. Great care was also taken to eliminate scattered light from reaching the detector. The voltage signals from the tunnel detector (biased in the region of thermal quasiparticle tunneling) were amplified by means of a B&H amplifier with 2-GHz bandwidth. In the initial experiments a PAR 162 boxcar integrator with a 400-ps gate