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Measurement of the Fluctuation Heat Capacity of Niobium in a Magnetic Field

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The heat capacity of niobium at constant magnetic induction has been measured in the immediate neighborhood of $T_c(B)$ with millikelvin temperature resolution. An enhanced heat capacity attributable to fluctuations is observed both above and below $T_c(B)$. The temperature dependence throughout the transition region can be described over a wide temperature range by a universal curve with a temperature scale uniquely determined by measured thermodynamic parameters.

We present some measurements of the heat capacity of a very pure niobium crystal measured at constant magnetic induction B in the immediate neighborhood of the second-order phase transition at $T_c(B)$. With millikelvin temperature resolution we can distinguish a peak in the heat capacity just below $T_c(B)$ and a transition width which increases markedly for transitions in finite fields. We attribute both effects to the presence of critical fluctuations of the superconducting order parameter, which assume a one-dimensional (1D) character in the presence of a magnetic field.[1] A similar peak in the heat capacity below $T_c(H)$ was first observed by Barnes and Hake[2] for a type-II superconductor in the extreme dirty limit, though they were unable to study the behavior in the critical region because of additional sample broadening of the transition. In addition to confirming the existence of the peak in a clean superconductor, we are also able to make a detailed study in the critical region where sample broadening in the bulk of our crystal is shown to be negligible.

In a recent calculation Thouless[3] has extended the theory of the fluctuation specific heat of a type-II superconductor in a magnetic field. Unlike previous calculations,[1,4,5] predictions are made which are expected to be applicable for all values of $T_c(B)$ and on both sides of the transition. Thouless has also suggested that the appropriately scaled temperature dependence of the fluctuation heat capacity is likely to be closely related to the exact solution for a 1D problem,[6] not only well away from $T_c(B)$ but also in the critical region itself. Using the scaling factors proposed by Thouless, which are calculated independently from measured experimental parameters, we are able to plot our results at all temperatures on a single curve almost exactly of this form.

The single crystal used in these experiments was cylindrical (length ~ 2 cm, diam ~ 5 mm) and had a resistance ratio, extrapolated to zero temperature and field, of order 4000. All surfaces of the crystal were roughened with emery powder until they acquired a uniform matt appearance.

A constant and uniform magnetic induction field was trapped inside the crystal by allowing it to cool from a temperature well above $T_c(0)$ in a magnetic field homogeneous to one part in 10^4 over the sample volume. The field was directed along a $\langle 100\rangle$ symmetry direction perpendicular to the cylindrical $\langle 001\rangle$ axis of the crystal. Magnetic measurements showed that no flux was expelled from the crystal either during the initial cooling or during any subsequent thermal cycling in the neighborhood of the bulk transition temperature. This extreme form of magnetic irreversibility, resulting from the surface damage, enabled us to make measurements of the heat capacity for the bulk crystal, at a constant and uniformly distributed magnetic induction, which were ideally reversible when measured as a function of sample temperature.

Measurements of the heat capacity were based on an ac-calorimetric technique described by Sullivan and Siedel.[7] The crystal was weakly coupled to the bath temperature with a resulting thermal time constant of order 1 sec. The mean specimen temperature could be raised above that of the bath and swept slowly through the transition region in both directions. The specific heat was determined from measurements of the small temperature excursions induced by an additional periodic square-wave heating pulse. Standard carbon-resistance thermometry and phase-sensitive detection (PSD) were used to monitor the temperature excursions. The frequency of the periodic heating and the phase of the detector were chosen so that the recorded amplitude was inversely proportional to the heat capacity.[7]

The amplitude of the temperature excursions was chosen to be much less than the width of the transition. In practice this limited our measurements to below 7 K, as above this temperature the intrinsic fluctuation width was too sharp to resolve with any accuracy. Below 2 K the difference between the superconducting and normal-state heat capacities becomes too small to measure very accurately.

In Fig. 1, typical xy-recorder traces of the PSD output (inversely proportional to heat capacity) are shown both for the sample undergoing a transition from the mixed state to the normal state at constant magnetic induction and for the sample in a field much larger than $B_{c2}(T)$. The

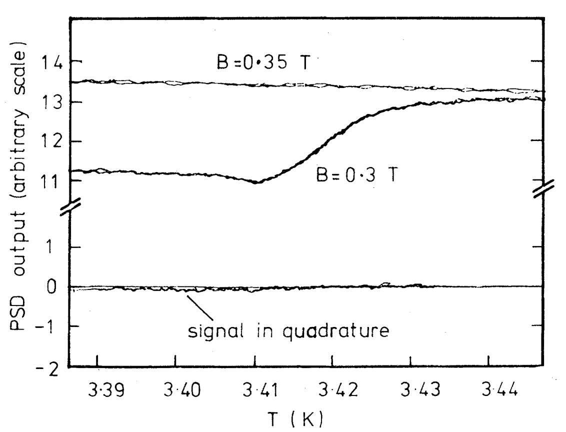

FIG. 1. A typical xy-recorder plot for the sample undergoing a transition showing both the signal in phase and the signal in quadrature. Also shown is a trace for the sample in the normal state. These measurements were obtained using a 6-Hz heating pulse, a 10-sec PSD time constant, and a 30-min temperature sweep. The signal amplitude is inversely proportional to the heat capacity. Each trace is plotted twice to check reproducibility.

finite slope of the normal-state trace is largely due to the changing thermometer sensitivity. The ratio of the mixed-state and normal-state heat capacities can be derived simply by comparing the ratio of the two signal amplitudes. To obtain absolute measurements, we have taken the values for the normal-state heat capacity given by Ferreira da Silva, Burgemeister, and Dokoupil.[8]

Between 2 and 7 K we always observed a relatively broad transition like that shown in Fig. 1, with a decrease in signal corresponding to an enhancement of the heat capacity just before the sample undergoes the major transition to the normal state. We attribute both the width of the transition and the enhancement to the presence of fluctuations. No features corresponding to the fluctuation enhancement were observed in the signal in quadrature over a wide range of frequencies, so alternative explanations in terms of a change in the relevant thermal time constants appear to be ruled out.

Before discussing these finite-field measurements in further detail, we will first consider the transition in zero field, where the bulk transition appears to be discontinuous with an experimental width limited only by the amplitude of the temperature excursion used in making the measurements (1–2 mK). Any rounding on the low-temperature side of the transition due to specimen inhomogeneities was certainly less than 1 mK. Above the transition the heat capacity was, however, slightly higher than the normal-state value. This extra contribution decreased slowly above the transition, suggesting that about 5% of our sample (presumably those regions damaged by the surface treatment) had a rather broad transition extending up to 30 mK above the bulk transition temperature. At lower temperatures we would expect these regions to remain superconducting throughout the relatively sharp transition to the normal state of the bulk crystal.

Although the transition close to $T_c(0)$ appears to be free of any additional sample broadening, local variations in purity could, in principle, produce additional broadening at low temperatures, even if H_c itself remains constant, through the dependence of κ, and hence H_{c2}, on the electronic mean free path. Any such broadening would lead to a temperature-dependent transition width varying as $(dB_{c2}/dT)^{-1}\Delta B_{c2}$, where $\Delta B_{c2} \sim (\xi \Delta l/l^2) B_{c2}$. In the clean limit a very pure crystal is required if any such broadening is to be negligible in comparison with the intrinsic

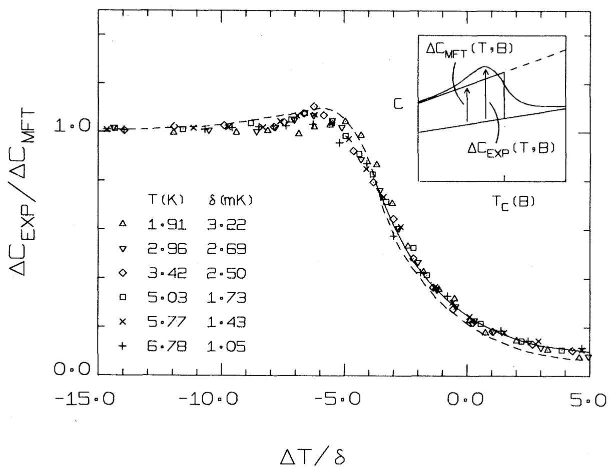

FIG. 2. Representative measurements for $\Delta C_{EXP}/\Delta C_{MFT}$ for a range of transition temperatures, where ΔC_{EXP} and ΔC_{MFT} are defined in the inset. The dashed curve is the temperature dependence expected for an ideal 1D transition, while the solid curve (which would be indistinguishable from the dashed curve at low temperatures) is obtained on the assumption that 5% of the sample remains superconducting throughout the transition (see text).

fluctuation width.

The temperature dependence of the heat capacity in the transition region at finite fields is illustrated schematically in the inset of Fig. 2. $\Delta C_{EXP}(T,B)$ represents the measured difference between the mixed- and normal-state heat capacities and $\Delta C_{MFT}(T,B)$ represents the difference in heat capacities expected from mean-field theory (MFT) ignoring the influence of fluctuations. ΔC_{MFT} has a small but significant temperature variation in the transition region. We have therefore chosen to normalize our measurements to ΔC_{MFT}, rather than to the MFT specific-heat discontinuity, as here we are only concerned with that part of the temperature dependence attributable to fluctuations. To determine ΔC_{MFT}, we made measurements of the heat capacity sufficiently below T_c where the fluctuation contribution predicted by Thouless[3] would be $\leq 1\%$. By subtracting this small theoretical contribution, we found the temperature dependence of ΔC_{MFT} and assumed a linear extrapolation throughout the transition region.

Changes in temperature in the transition region were measured on a reduced temperature scale in units of δ, where δ was evaluated from the following expression given by Thouless[3]:

$$\frac{\delta}{T}=\frac{2e}{\hbar}B_{c2}(0)\left(\frac{k_B}{8\pi\Delta C}\right)^{2/3} \times\left|\frac{T}{B_{c2}(0)}\frac{dB_{c2}(T)}{dT}\right|^{1/3}\left(\frac{B}{B_{c2}(0)}\right)^{2/3},$$

where $\Delta C=\Delta C_{MFT}(T,B)$ evaluated at $T_c(B)$ and all the other parameters required were obtained straightforwardly in the course of these measurements.

In Fig. 2 we have plotted results for several representative temperatures which have been scaled in the manner described. Simply by choosing the origin of the temperature scale appropriately, all our measurements can be plotted so that they lie on a common curve. This agreement extends over a wide range of transition temperatures and strongly supports the general validity of the scaling law proposed by Thouless.[3] It is important to emphasize that, had the transition width been associated with either specimen or field inhomogeneities, we would have expected it to be almost 3 times as wide at 2 K as that measured at ~ 7 K, when plotted on the above temperature scale. In contrast, we observe no evidence for any additional broadening at low temperatures, which confirms our belief that any sample broadening is negligible in these measurements.

Thouless[3] has argued that the temperature dependence throughout the transition region is likely to be similar in functional form to the exact solution for the 1D superconductor problem.[6] This temperature dependence is indicated by the dashed curve in Fig. 2, where the temperature scale is uniquely determined by the requirement that this curve is of the form $(T/\delta)^{-3/2}$ at high temperatures; $T=0$ corresponds to the mean-field critical temperature. Although our results are described remarkably closely by this curve in the transition region itself, they tend to be slightly too high above $T_c(B)$ and too low below it.

Above $T_c(B)$ this difference has a straightforward explanation, as the small fraction of crystal damaged by our surface treatment is expected to remain superconducting, while the remaining bulk undergoes a transition of the ideal 1D form. The solid line in Fig. 2 represents such a model with 5% remaining superconducting throughout. Although this describes our measurements extremely closely, further measurements, using crystals with less surface damage, are clearly necessary in order to make a critical study of the expected $(T-T_c)^{-3/2}$ variation above $T_c(B)$.

Below $T_c(B)$ the finite κ values of niobium are expected to reduce the difference between the heat-capacity maximum and the mean-field-theory value by the factor $1.16(2\kappa^2-1)/[1.16(2\kappa^2-1)+1]$. Between 2 and 7 K this factor varies from 0.91 to 0.75.[9] This may partially account for the depression of our experimental results below the 1D curve, but cannot explain why the fluctuation peak appears to be rather more sharply peaked than expected. Further experiments are clearly necessary both to establish whether this feature is apparent in other samples and to verify the predicted κ dependence below $T_c(B)$.

In none of our measurements were we able to observe any structure near the center of the transition corresponding to a singularity or discontinuity which might be expected in the small region of temperatures where the system can no longer be considered one dimensional.

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Critical Fluctuations of a Type-II Superconductor in a Magnetic Field

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Ginzburg's theory of critical fluctuations is used to calculate the specific heat of a type-II superconductor in a magnetic field near the upper critical field. A scaling law for the specific heat is found, and the scaling temperature is given in terms of measurable quantities. Some distance from T_c the fluctuation contribution is proportional to $|T-T_c|^{-3/2}$, with known coefficients. The dependence on κ is explored. The diamagnetic susceptibility should have the same temperature dependence as the specific heat.

Various authors[1-4] have pointed out that close to the upper critical field of a type-II superconductor, fluctuations in the number of superconducting electron pairs are essentially one-dimensional (1D), and Lee and Shenoy[5] have argued that they give rise to a specific heat well above $T_c(B)$, the transition temperature in the presence of a magnetic field, proportional to $[T-T_c(B)]^{-3/2}$, which is characteristic of 1D behavior. Various calculations have been made, for example by Hassing, Hake, and Barnes[6] and by Bray,[7] which agree with one another for T well above $T_c(B)$ and for weak fields, but which all involve approximations of doubtful validity in other regions. The measurements by Farrant and Gough[8] of the specific heat of Nb near $T_c(B)$, described in the preceding paper, make it important to have a theory valid below $T_c(B)$ and for fairly strong fields.

Eilenberger[9] has studied the behavior of type-II superconductors near the upper critical field using Ginzburg's[10] theory of critical fluctuations, and I use a very similar approach here, except that I consider a constant external field instead of a constant flux. The free-energy density for a superconductor in a uniform magnetic flux density $\vec{B}$ has the form

$$F=\alpha(T)|\varphi|^2+\tfrac{1}{2}\beta(T)|\varphi|^4+(1/2m)|(-i\hbar\nabla-e\vec{B}\times\vec{r})\varphi|^2+B^2/2\mu_0. \qquad (1)$$

Here φ is the complex superconducting wave function, proportional to the gap parameter, whose