Specific Heat of a Superconducting Multilayer: 2D Fluctuations and 2D-0D Crossover

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We have measured the specific heat of multilayers of superconducting amorphous $Mo_{77}Ge_{23}$ layers separated by insulating amorphous germanium. We observe a fluctuation regime in quantitative agreement with predictions for two-dimensional superconductivity. The fluctuation peak is rapidly suppressed by the application of small magnetic fields perpendicular to the layers, and the transition becomes extremely broad as the field is increased. The transition widths scale as expected for a field-induced 2D to 0D crossover, and are in excellent agreement with the exact result for 0D fluctuations.

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Conventional bulk superconductors are extremely well described by BCS mean-field theory. The size of the coherence length and the dimensionality determine the region around T_c in which corrections driven by thermodynamic fluctuations must be considered, and it is normally too narrow to be observed in specific-heat measurements. High-temperature superconductors (HTSC) possess a short coherence length and are very anisotropic, so the fluctuation region is greatly enhanced and is observable in the bulk material [1]. Specific-heat fluctuations in these materials have been fitted with 3D Gaussian fluctuations and critical fluctuations, but the analysis is complicated by a large phonon background as well as uncertainties in many of the relevant material parameters and concern about sample homogeneity [2]. Fluctuation effects are also enhanced in reduced dimensions, particularly two-dimensional thin films. Unfortunately, experimental difficulties have thus far limited specific-heat measurements to films much thicker than the zero-temperature coherence length, where no fluctuation peak was observed [3]. In this Letter we report the first observation of a zero-field fluctuation peak for a conventional superconductor. The data are well described by theoretical predictions for 2D superconductivity using known materials parameters.

The application of a magnetic field produces an effective reduction in dimensionality [4]. Close to H_{c2}, fluctuations of the superconducting order parameter are confined to the lowest Landau level, thus eliminating the degrees of freedom perpendicular to the applied field. Measurements on bulk niobium produced excellent agreement with scaling laws based on a 3D to 1D crossover [5,6]. We present results of two-dimensional layers in an applied field, and the data are well described by scaling laws for a 2D-0D crossover. In addition, the scaled data agree very well with the exact result for a 0D superconductor. The results are remarkably similar to observations in Y-Ba-Cu-O.

Specific-heat measurements on small samples are limited by the ability to separate out the contribution of the addenda, typically a substrate, heater, and thermometer. We have used multilayers to increase the volume of superconductor, while maintaining the 2D character. Bulk $Mo_{77}Ge_{23}$ is well described [7] by weak-coupling BCS theory in the dirty limit, with $\xi(0) \cong 50$ Å and $k_F l \cong 5$. The multilayers were grown by multitarget magnetron sputtering described previously [8]. The samples for specific-heat measurements were sputtered onto sapphire substrates that had been prepared with a doped silicon heater and thermometer on the back side. The addenda, primarily the sapphire, account for (70–90)% of the total specific heat. The data presented are from ac calorimetry, and the general features have been confirmed by measurements using the relaxation technique. We have measured two multilayers, each with 100 superconducting layers of thickness $d_s = 55$ Å (approximately equal to the zero-temperature coherence length, resulting in an $R_\square$ per layer $\cong 300$ Ω), one separated by insulating layers of $d_i = 65$ Å and the other by $d_i = 95$ Å. We have also measured a thick (1.1 μm) film of pure MoGe to check the bulk properties. Transport measurements [8] of similar multilayers with thinner insulating layers have shown that the dependence of the fluctuation conductivity on layer spacing is well described by the Lawrence-Doniach model for layered superconductors, with mass ratios in good agreement with those predicted by the measured tunneling decay length in amorphous Ge (8.1 Å). Extrapolating those results to the 55/95 multilayer would predict a 2D-3D crossover at $|T - T_c| < 10$ μK. Since layer-to-layer variations in T_c introduced by the growth process are certainly greater than this, we expect that the 3D crossover will never occur. We will show that this conclusion is supported by both transport and specific-heat measurements.

Figure 1 shows the specific-heat data on the 55/95 multilayer in various magnetic fields applied perpendicular to the layers. The zero-field results show an upward curvature below the transition, as well as a long tail at high temperature. It is important to note that while inhomogeneities could produce a tail above T_c, they cannot produce the observed curvature below the transition. Also evident is a sharp change in slope at 5.1 K. The inset shows the zero-field data for the multilayer and the thick film as a function of reduced temperature after subtraction of the normal-state specific heat measured in high magnetic field. The mean-field T_c of the multilayer is re-

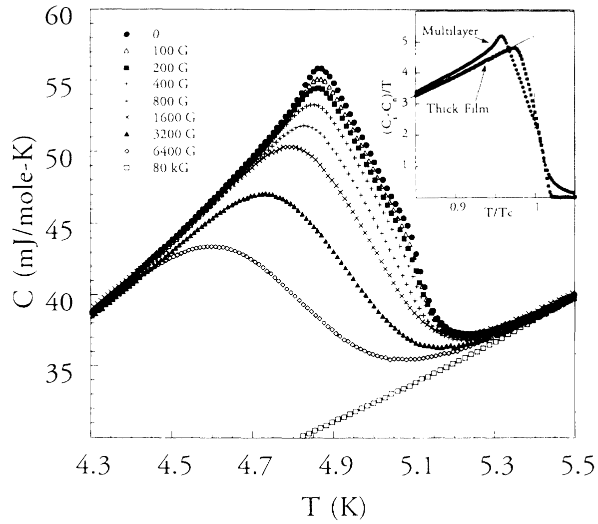

FIG. 1. Specific heat of the (55 Å)/(95 Å) superconducting multilayer with a magnetic field applied perpendicular to the layers. Inset: Specific heat of the superconducting state minus the normal-state specific heat for the multilayer and for a thick film. The solid line represents the mean-field behavior.

duced from the thick film ($T_c=6.1$ K) by dimensional effects [7]. The vertical scale for the thick film has been reduced by 15%. The uncertainties in composition and sample mass are too large to determine whether this reduction is related to dimensional effects. The specific heat of the 55/65 multilayer is qualitatively similar to the 55/95, showing both the fluctuation peak and the high-temperature tail, but the transition is about 50% broader, indicating lower homogeneity.

The application of a magnetic field perpendicular to the layers has two dramatic effects. At low fields, both the peak at 4.85 K and the knee at 5.1 K are rapidly suppressed. The features are completely eliminated by an applied field of 800 G, while $H_{c2}(0) \approx 120$ kG. In higher field, the entire transition broadens, while the onset temperature shifts only slightly. In the thick film, by contrast, the application of a magnetic field does not produce a measurable broadening ($H<5$ kG), in accordance with the mean-field result. The primary source of inhomogeneity is most probably compositional drift during the deposition process. $[d(H_{c2})/dT]_{T_c}$ is only weakly dependent on composition [7], so any field-induced broadening should be small compared to the T_c shift, as observed in the thick film.

By contrast, a magnetic field applied parallel to the layers has very little effect. As a result of limitations imposed by the experimental technique, as well as unevenness in the multilayer itself, the field cannot be aligned to better than a few degrees. The effect of the parallel field over the measured range from 100 G to 25 kG is almost identical, both in the initial reduction of the peak and the knee and then the broadening of the transition, to that measured for a perpendicular field reduced in magnitude by a factor of 20. It seems highly probable, therefore, that we are observing the effect of a perpendicular component resulting from an average misalignment of 3°, and that the parallel component has no measurable effect. This result strongly suggests that the observed effects are the result of orbital effects occurring within the plane of the layers.

To shed light on the multilayer specific-heat results, we have measured fluctuation conductivity on a sample grown alongside the specific-heat sample. Well above T_c, the fluctuation conductivity has the expected $|T-T_c|^{-1}$ dependence, with $T_c \approx 4.95$ K. In contrast to single-film results, however, the fluctuation conductivity begins to grow faster than $|T-T_c|^{-1}$ several hundred mK above T_c, finally diverging at 5.10 K. These results together with the specific-heat measurements can be understood by considering the unusual inhomogeneity produced by the growth process.

T_c measurements of samples grown during a variety of sputtering runs, as well as the specific-heat results on thick films, show that the composition of Mo_xGe_{1-x}, varies slightly in the course of a long run due to drifts in the relative sputtering rates of the elements. All of the properties of Mo_xGe_{1-x}, including T_c, vary smoothly with composition around the target composition of $x=0.77$. This drift will not broaden the individual layer T_c's, since each layer is 2D, but will produce layer to layer variations. The observed behavior can be understood in the following way: As T_c is approached from below, the fluctuation specific heat of all of the layers will add to produce an upward curvature. When the lowest T_c layers start to go normal, their contribution to the total specific heat will drop rapidly, resulting in a downward slope. This process will continue until the last layer goes normal, at which point the specific heat will cross over to the average fluctuation specific heat. Thus the layer T_c's are distributed from the peak in the specific-heat data (4.85 K) to the knee (5.10 K). The fluctuation conductivity well above T_c will extrapolate to the average of the layer T_c's (≈ 4.95 K), while diverging at the highest T_c (5.1 K). This T_c distribution requires a 1% drift in the relative sputtering rate of Mo and Ge over the course of the 2-h film deposition, consistent with observed drift rates. The data suggest that the T_c's are fairly uniformly distributed from 4.85 to 5.10 K, implying that the drift rate is constant. This model T_c distribution, shown in the inset of Fig. 2, will be assumed in what follows. The features observed in the specific heat cannot be reproduced with a Gaussian distribution of T_c's, or a distribution with a significantly different width.

Figure 2 shows the specific heat of a single layer (the lowest T_c layer) deconvolved [9] from the data using the model T_c distribution. Also plotted is the result of an approximate solution of the Ginzburg-Landau Hamiltonian for the 2D case using the screening approximation to include higher-order corrections to Gaussian fluctuations [10]. This approximation includes an interaction between

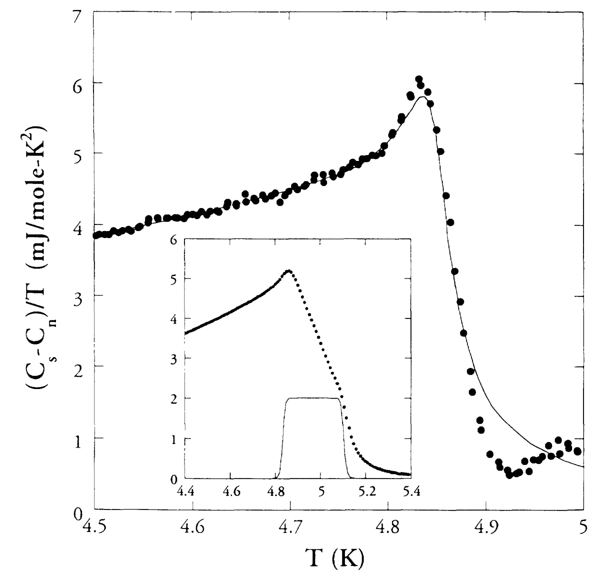

FIG. 2. Single-layer specific heat in zero field determined from the measured data using the T_c distribution shown in the inset. The solid line is the prediction of the screening approximation.

Gaussian fluctuations of the order parameter that is screened by virtual fluctuations, and provides good agreement with the exact solution in 0 and 1 dimension. The only free parameters are the peak temperature and an amplitude scaling required to account for uncertainty in the volume of the superconductor and the mean-field specific-heat discontinuity. The peak temperature depends on the mean-field T_c and the momentum cutoff used in the theory. The overall shape, including the width and the peak height, is determined from the theory in terms of $k_F^2 l$ (7.7 Å^{-1} [7]) and the layer thickness (55 Å). It is difficult to assess the validity of the approximations used in the theoretical calculation, but the result provides a reasonable interpolation between the Gaussian fluctuation regimes above and below T_c. The data can also be well described by 2D Gaussian fluctuations with an arbitrary cutoff of the divergence and a 40-mK T_c distribution. Again the amplitude of the fluctuation contribution is set by materials parameters, but the cutoff and the T_c distribution are adjustable parameters. A more detailed study of the specific-heat shape will require samples free of the distribution of T_c's.

Figure 3 shows the result of applying the deconvolution to the data taken when a magnetic field is applied. The simultaneous disappearance of the knee and the peak observed previously is now understood as the broadening of the fluctuation peak. A similar field dependence has been observed in HTSC [11], as well as anisotropic organic superconductors [12] and Monte Carlo calculations of 2D superconductors [13]. These results can be understood as a field-induced dimensional crossover, in our case from 2D to 0D. The volume of the 0D particle is given by the volume of the film divided by the degeneracy of the Landau levels, $V_H = a_H^2 d$, where the magnetic length $a_H^2 = \Phi_0/H$ and d is the film thickness. The width of the transition for a small particle scales as $\sqrt{V}$, so the magnetic-field-induced broadening is proportional to $\sqrt{H}$. Figure 4 shows the data from 400 to 6400 G scaled by the 0D Ginzburg criterion [14], $\zeta_G(H) = [k_B/(\Delta C)V_H]^{1/2}$. The vertical axis is scaled $\Delta C_{\rm bulk}(H)/\Delta C(0)$, where $\Delta C_{\rm bulk}(H)$ is the field-dependent bulk specific-heat discontinuity determined from the thick film, and $\Delta C(0)$ is determined from the zero-field fit described above. $T_c^{\rm MF}(H)$ is determined from $d(H_{c2})/dT$ measured on the thick film and a $T_c(0)$ of 4.88 K. The only adjustable parameter is a small (<0.1) constant that has been subtracted from some of the curves to ac-

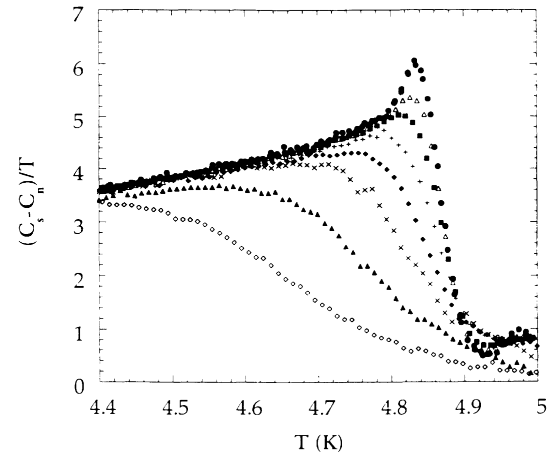

FIG. 3. Single-layer specific heat with a magnetic field applied perpendicular to the layers.

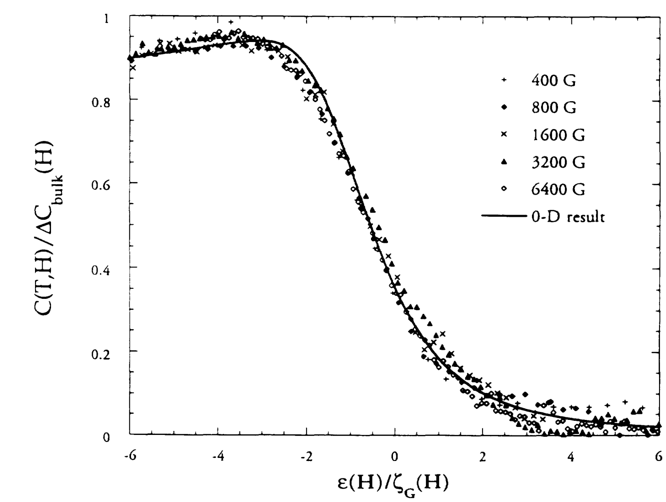

FIG. 4. Scaled specific-heat data for magnetic fields from 400 to 6400 G. $\varepsilon(H) = 1 - T/T_c^{\rm MF}(H)$ is scaled by the 0D Ginzburg criterion, $\zeta_G(H) = [k_B/(\Delta C)V_H]^{1/2}$, where $V_H = (\Phi_0/H)d$. The vertical axis is scaled by the bulk field-dependent specific-heat discontinuity measured in a thick film. The solid line is the exact result for a 0D superconducting particle.

count for systematic errors that vary slowly with temperature in the ac calorimetry. The scaling fails for fields below 400 G, where the data show significant upward curvature. The scaled data rise above the high field curves on the low-temperature side of the transition. The 2D fluctuations that induce the upward curvature and zero-field broadening are not included in the scaling relation. Also shown is the exact result for a 0D particle of size V_H [15]. The agreement is quite surprising, given the limited range of the validity of the lowest-Landau-level assumption. 1D scaling behavior has been suggested for Y-Ba-Cu-O [16], although the specific-heat amplitude scaling must be introduced in an arbitrary way.

The excellent quantitative agreement observed between our results and theoretical predictions further solidify the analysis that has been used to explain the magnetic-field-induced broadening of the thermodynamic and resistive transitions in thin films and anisotropic superconductors. A better theoretical understanding of the low-field behavior is necessary, as well as theoretical and experimental study of the critical regime in zero field. While the zero-field data are well described by the screening approximation, more quantitative results are necessary. A 2D superconducting film should have a Kosterlitz-Thouless transition [17,18]. The specific heat is expected to be smooth at $T_{\rm KT}$, and the transition at the mean-field T_c should be quite broad. The entropy of the vortices that mediate the KT transition is quite small, however, and their effect may only be seen very close to T_c. We are attempting to improve our measurement sensitivity to allow us to study the region close to T_c on a single film.

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