Experimental observation of high-field diamagnetic fluctuations in niobium

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We have performed a magnetic study of a bulk metallic sample of Nb with critical temperature $T_c = 8.5$ K. Magnetization versus temperature (M vs T) data obtained for fixed magnetic fields above 1 kOe show a superconducting transition which becomes broader as the field is increased. The data are interpreted in terms of the diamagnetic lowest Landau level (LLL) fluctuation theory. The scaling analysis gives values of the superconducting transition temperature $T_c(H)$ consistent with $H_{c2}(T)$. We search for universal three-dimensional LLL behavior by comparing scaling results for Nb and YBaCuO, but obtain no evidence for universality.

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High-field diamagnetic fluctuations are predicted to occur in superconductors in a strong magnetic field H. This constrains the paired quasiparticles to remain in the lowest Landau level (LLL), reducing their effective dimensionality.[1,2] The width of the region around the superconducting temperature transition $T_c(H)$ where the high-field fluctuations occur is given by the field-dependent Ginzburg criterion $G(H) = (8\pi\kappa^2 k_B T_c H/\phi_0 \xi_c H_{c2}^2)^{2/3}$,[3] where κ is the Ginzburg-Landau (GL) parameter, ϕ_0 is the quantum flux, ξ_c is the c-axis coherence length at zero temperature, and H_{c2} is the upper critical field at zero temperature and for fields applied along the c-axis direction. One important effect of the LLL fluctuations is to produce a rounding of various data curves around $T_c(H)$.[4,5] Consequently, the superconducting transition appears continuous, rather than distinct, as expected for a second-order transition $T_c(H)$.[6] High-T_c superconductors (HTSC's), with their small ξ, high κ, and high critical temperatures T_c, display a broad fluctuation region. The LLL fluctuation theory has been invoked to explain the nature of the broad "fan"-shaped transition observed in high-field magnetic measurements in YBaCuO.[6] Predictions of the LLL theory include scaling laws for various physical quantities.[4,5,7] For magnetization in particular, the scaling predicts that M vs T data obtained at different fields H should collapse onto a single curve when the variable $M/(TH)^{(D-1)/D}$ is plotted against $[T - T_c(H)]/(TH)^{(D-1)/D}$. Here, $T_c(H)$ becomes a fitting parameter, and D is the dimensionality of the system. This scaling law has been used to identify LLL fluctuations in a given material and to determine its dimensionality.[5,6,8–13] An important check of the scaling is that it should provide reasonable values of $T_c(H)$.[6,8] For lower-dimensional or layered materials, the LLL analysis also helps to explain the crossing points observed in $M(T)$ curves.[9–11,13,14]

Despite the relevance of LLL fluctuations for HTSC's there are few comparable studies in conventional superconductors. Niobium is one of the most studied type-II superconductors. Observations of an irreversibility line and a broad high-field reversible region in Nb,[15] similar to HTSC's, have renewed interest in this element. Extensive studies of fluctuations have been performed in Nb at low fields.[16] For pure samples, high-field diamagnetic fluctuations were observed in specific heat measurements for $H = 4$ kOe, but only over a narrow temperature range, of the order of 40 mK.[17] It is therefore a matter of interest to gather further evidence for high-field diamagnetic fluctuations in Nb. Our work is motivated by the possibility that a Nb sample with an elevated value of $\kappa > 1$ could show enhanced high-field fluctuation effects.

We address this issue by performing magnetization measurements as a function of field and temperature in an impure but homogeneous Nb sample. The results show that $M(T)$ curves obtained for different fields follow the three-dimensional (3D) LLL scaling. The Nb curves are somewhat more separated than comparable curves in HTSC's, where LLL scaling has been applied.[6,8–11,13,14] The crossing point behavior sometimes observed in HTSC's is not here observed for Nb, as consistent with its expected 3D nature.

The niobium sample adopted in the present study is an ellipsoid with axes $2r_1 = 4.7$ mm and $2r_2 = 5.3$ mm, mass $= 0.6487$ g, and $T_c = 8.5$ K with $\Delta T = 0.3$ K determined at low fields. The sample was manufactured in an arc melt furnace from 99.9% Nb wire. X-ray diffraction shows the metallic Nb phase. Magnetization data were always taken after cooling the sample in zero field. A commercial Quantum Design superconducting quantum interference device (SQUID) magnetometer (7 T) with 3 cm scans was utilized in the measurements. Hysteresis loops of M vs H were obtained at fixed temperatures with values running from 3 K to 6 K. Isofield magnetization curves, M vs T, were also obtained for fixed applied magnetic fields with values running from 1 kOe to 10 kOe. The value of $T_c = 8.5$ K, lower than 9.2 K found for pure Nb,[18] indicates the possible existence of magnetic impurities. Neverthless, the corresponding magnetic signal is very small and could not be resolved from the background magnetization. The corrected background magnetization is found to be field dependent but not temperature dependent, as expected for Pauli paramagnetism in the stud-

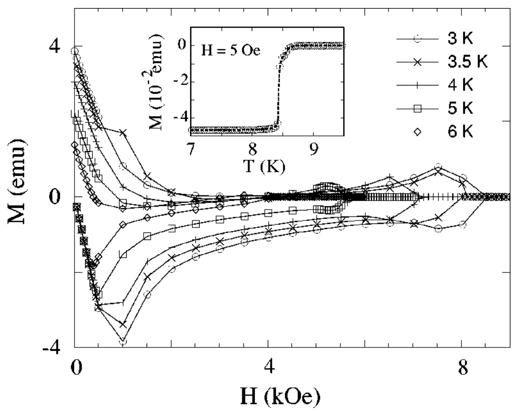

FIG. 1. M vs H curves obtained for fixed temperatures ranging from 3 K to 6 K. Inset shows the superconducting transition for an applied magnetic field of 5 Oe.

ied temperature range. The demagnetization factor of the sample is obtained from the Meissner region in hysteresis magnetization curves M vs H and is very close to 1/3 as expected for a sphere. A previous study performed in our sample[19] determined the value of the Ginzburg parameter $\kappa = 4$. Since the temperature width of the high-field fluctuations can be expressed as $\Delta T_{\rm fluct} \simeq G(H) T_c \propto \kappa^{4/3}$, large values of κ can be expected to enhance fluctuations effects. High-purity Nb, with $T_c = 9.2$ K, has $\kappa \simeq 1$.[18]

Figure 1 shows zero-field-cooled M vs H hysteresis curves. The inset shows the superconducting transition T_c. The entire transition occurs within a temperature window of less than 0.3 K. However, the step, which accounts for 80% of the transition, has a 80 mK width, between 8.42 and 8.5 K. Most of the $M(H)$ curves in Fig. 1 display a pronounced peak effect occurring near the irreversible field $H_{\rm irr}$, reminiscent of HTSC's.

Figure 2 shows a detail of the reversible region of the hysteresis M vs H curve obtained at 3.5 K. The reversible diamagnetic region extends more than 2 kOe above $H_{\rm irr}$. Figure 2 shows the standard linear extrapolation of the reversible magnetization down to $M = 0$, used to estimate H_{c2}.[20] The linear extrapolation is clearly an approximation, since there is a large region of the curve which deviates from

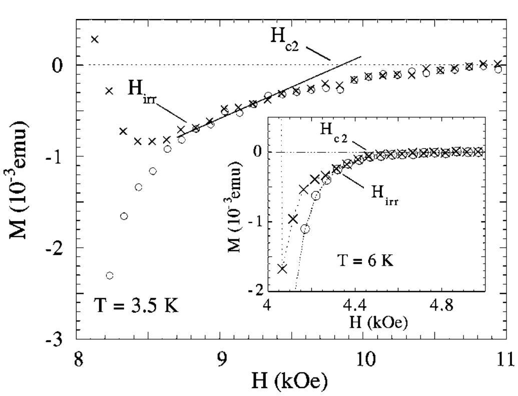

FIG. 2. Detail of the M vs H curve obtained at 3.5 K for fields in the region of H_{c2}. Inset shows detail of the M vs H curve obtained at 6 K in the region of H_{c2}.

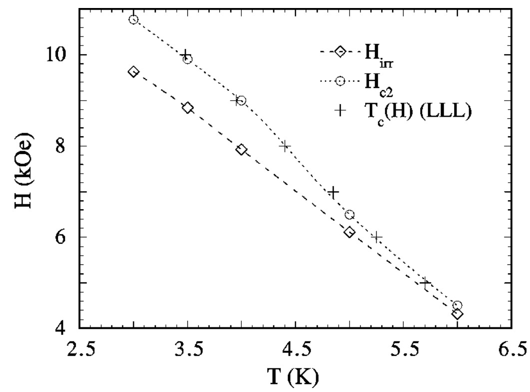

FIG. 3. Magnetic phase diagram for our Nb sample. The plotted values of $H_{\rm irr}$ and H_{c2} are determined as in Fig. 2. The values of $T_c(H)$ are obtained from the 3D LLL scaling analysis.

the linear behavior as the magnetization approaches zero. To this level of accuracy, H_{c2} is located 1.2 kOe above $H_{\rm irr}$. The inset of Fig. 2 shows a detail of the M vs H curve obtained at 6 K, for which the reversible diamagnetic region extends only approximately 300 Oe above $H_{\rm irr}$. The results for H_{c2} and $H_{\rm irr}$ obtained from Fig. 1 are plotted in Fig. 3.

Figure 4 shows zero-field-cooled $M(T)$ curves obtained for fields $H = 10$, 9, 8, 7, 6, 5, 3, 2, and 1 kOe. For $H = 10$, 9, and 8 kOe the field-cooled data are also plotted to show the extent of the reversible regions at high fields. [We mention that values of $(T_{\rm irr}, H)$ obtained from M vs T curves agree with values of $(T, H_{\rm irr})$ obtained from M vs H curves.] The temperature interval between magnetization data in each curve is 50 mK. The rounding of the curves at high fields makes it difficult to extract a value for the critical temperature $T_c(H)$. To illustrate the rounding effect, we mark the transition temperatures, as interpolated from Fig. 3, with arrows for the $H = 1$, 2, and 10 kOe curves. By visual inspection of Fig. 4, it is possible to see a sharp transition for $H = 1$ kOe, which evolves into a broader transition as the field increases.

We now consider possible explanations for the rounded curves in Fig. 4. In Ref. 17, high-field diamagnetic fluctuations were observed for Nb at $H = 4$ kOe, suggesting that the

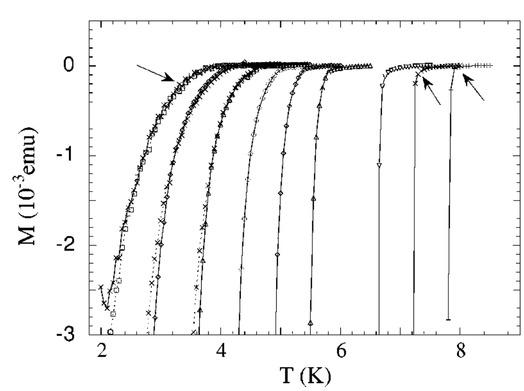

FIG. 4. Zero-field-cooled M vs T curves obtained at fixed magnetic fields. From left to right, curves were obtained for $H = 1$, 2, 3, 5, 6, 7, 8, 9, and 10 kOe. For $H = 8$, 9, and 10 kOe the corresponding field-cooled data are also plotted.

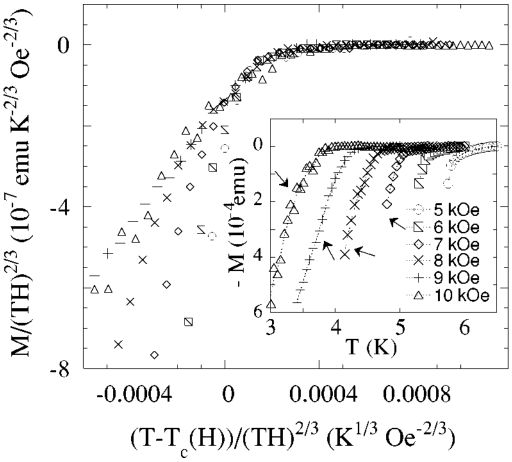

FIG. 5. Magnetization data for Nb are plotted after performing a 3D lowest-Landau-level scaling analysis. The inset shows only the unscaled data collapsed in the main figure.

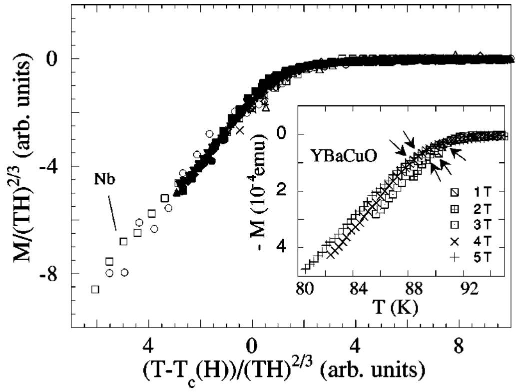

FIG. 6. Simultaneous 3D LLL scaling of Nb and YBaCuO data. The *x* and *y* axes of YBaCuO data are multiplied by 1.5 and 1/150, respectively. Open symbols are used for Nb data and solid symbols for YBaCuO data. The inset shows the unscaled *M* vs *T* data of YBaCuO used in the main figure.

rounding above 5 kOe in our data may also be due to LLL-type fluctuations. An alternative explanation for the broadening could be surface superconductivity, which may occur above $T_c(H)$. However, the surface superconductivity signal for a sphere is expected to be rather small[21] as confirmed for our sample in the *M* vs *T* curve for $H=1$ kOe, where no diamagnetic signal was detected above $T_c(H)$. Additionally, rounding effects in the magnetization due to surface superconductivity would only occur above $T_c(H)$,[21] while in Fig. 4 for $H=10$ kOe the rounding also occurs below $T_c(H)$. Based on these observations, and the fact that the data follow the 3D LLL scaling, we disregard the influence of surface superconductivity in the present measurements. Another possible explanation for the rounding in *M* vs *T* curves is sample inhomogeneity due to the impurities. We believe that our sample is homogeneous based on three points: (1) the sample shows only one superconducting transition at low fields which is relatively sharp, (2) there is no rounding above the transition for the $H=1$ kOe *M* vs *T* curve, and (3) the data follow the 3D LLL scaling.

We now perform a lowest-Landau-level scaling analysis on the high-field data of Fig. 4, with $D=3$, as appropriate for Nb. $M/(TH)^{2/3}$ is plotted versus $[T-T_c(H)]/(TH)^{2/3}$. The transition temperature $T_c(H)$ becomes a fitting parameter, chosen to make the data collapse onto a single curve. The resulting values are plotted in Fig. 3, with a significant correspondence to the values of $H_{c2}(T)$ estimated from the isothermic *M* vs *H* curves. The results of the scaling analysis are presented in Fig. 5. The inset shows the unscaled data while the collapsed data are shown in the main figure. Note that only high-field data in the field range $H=5$–10 kOe have been scaled. The arrows in the inset identify the scaling values of $T_c(H)$, evidencing the breadth of the LLL fluctuation range. A visual inspection of this inset suggests that the LLL fluctuations occur over a large region in the 9 and 10 kOe curves, including data below $T_c(H)$. However, for 7, 6, and 5 kOe, the LLL scaling seems appropriate only above $T_c(H)$. We may compare the observed temperature width of the scaling range to its expected width, $\Delta T_{\rm fluct} \simeq G(H)T_c(H)$. $\Delta T_{\rm fluct}$ is expected to grow with field as $H^{2/3}$. We observe the predicted $H^{2/3}$ behavior for $H=10$, 9, and 8 kOe, but not for $H=7$, 6, and 5 kOe. It is possible that the high-field conditions—namely, the paired quasiparticles lying in the lowest Landau level—are not fully met for the latter data.

The recent interest in LLL scaling has been stimulated by high-temperature superconductors, which exhibit extensive high-field fluctuations.[4–14] It is of fundamental interest to ask whether scaling in low- and high-temperature superconductors could exhibit universal behavior. To investigate this point we obtain magnetization measurements from the sample of YBaCuO (mass = 1.7 mg, $T_c=91.7$ K) used in Ref. 11. Although YBaCuO is known to be more anisotropic than Nb, it is still expected to behave three dimensionally, justifying the use of 3D LLL scaling. It is possible that the 3D scaling form could break down away from $T\simeq T_c(H)$, owing to the layered nature of the material.[7] (This is the same regime where the simple scaling forms used in this work become inaccurate, due to the more complicated nature of LLL scaling theory in 3D.[7]) We perform simultaneous scaling of YBaCuO and Nb in analogy to Fig. 5; however, we now incorporate sample-dependent scaling factors along both the *x* and *y* axes. Specifically, the YBaCuO data have been multiplied by the relative factors of 1.5 and 1/150, along the *x* and *y* axes, respectively. The results are shown in Fig. 6. The collapse of data from the two different samples is good and may give support to the notion of universality.

We can take this comparison a step further by analyzing the collapse in Fig. 6. Because of the different geometries of the two samples, it is not possible to make a direct comparison of the magnetizations (*y* axis). However, Ullah and Dorsey[5] provide an analytical expression for the full 3D LLL temperature scaling variable along the *x* axis: $x=[\gamma/(2\kappa^2-1)TH]^{2/3}(\partial H_{c2}/\partial T)[T-T_c(H)]$, where the anisotropy parameter is given by $\gamma=\xi_c(0)/\xi_{ab}(0)$. The ratio of the respective YBCO and Nb scaling factors, $r=[(\gamma/(2\kappa^2-1))^{2/3}\partial H_{c2}/\partial T]_{\rm YBaCuO}/[(\gamma/(2\kappa^2-1))^{2/3}\partial H_{c2}/\partial T]_{\rm Nb}$, can therefore be used to rescale the YBCO data to make it collapse onto the Nb data. Using typical values of $\gamma=0.2$,

$\kappa=55$, and $\partial H_{c2}/\partial T=15$ kOe/K for YBaCuO, and $\gamma=1$, $\kappa=4$, and $\partial H_{c2}/\partial T=1.5$ kOe/K for our Nb sample, we calculate $r=0.1$, which is more than an order of magnitude different than the measured value $r=1.5$. Thus, independently, the Nb and YBCO samples appear to scale as expected. A naive universal collapse of the data sets appears outwardly successful. However, the detailed scaling results cannot be reconciled with Ref. 5.

There are two possible explanations for these mixed results, which do not necessarily invalidate the observed 3D LLL scalings: (1) The temperature range of 3D LLL scaling could be so narrow for YBCO, due to its layered nature, that the observed 3D LLL collapse is either rendered invalid or belongs to a different universality class. (2) The theory of Ullah and Dorsey,[5] employed here to determine r, may not be applicable to isotropic superconductors such as Nb, because of its origins in the layered Lawrence-Doniach model. The expression we use for r would therefore be inaccurate, although the basic scaling theory (ignoring scaling factors) is expected to be independently valid for the two types of superconductors.

There is also a third possible explanation for the failure of universal scaling. It has been shown in Ref. 12 that 3D LLL scaling in YBaCuO is obeyed better for data in the high-field region ($H>5$ T) than in the 1–5 T field region. Unfortunately we do not have high-field data for YBaCuO to test this possibility. The inset of Fig. 6 shows YBaCuO unscaled data used in the main figure. The arrows identify the scaling values of $T_c(H)$, as in Fig. 5. Comparison of Figs. 5 and 6 shows that LLL scaling in Nb is all the more spectacular due to the wide separation of the unscaled data.

Finally, we point out that the M vs T curves of Fig. 4 exhibit no crossing points. In HTSC's the M vs T curves for which LLL have been successfully applied show crossing points[6–11,13,14] which are related to the layered structure (2D character) of those systems. We also mention that we tried to scale our niobium data using a 3D XY critical scaling theory[22] without success. The 3D XY theory has been used to explain fluctuations of HTSC's in the reversible regime.[12,22]

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[1] G. Bergmann, Z. Phys. **225**, 430 (1969).

[2] P. A. Lee and S. R. Shenoy, Phys. Rev. Lett. **28**, 1025 (1972).

[3] R. Ikeda, I. Ohmi, and T. Tsuneto, J. Phys. Soc. Jpn. **58**, 1377 (1989); **59**, 1397 (1990).

[4] R. A. Klemn (unpublished).

[5] S. Ullah and A. T. Dorsey, Phys. Rev. Lett. **65**, 2066 (1990); Phys. Rev. B **44**, 262 (1991).

[6] U. Welp *et al.*, Phys. Rev. Lett. **67**, 3180 (1991).

[7] Z. Tesanovic and A. V. Andreev, Phys. Rev. B **49**, 4064 (1994).

[8] S. Salem-Sugui, Jr. and E. Z. da Silva, Solid State Commun. **90**, 647 (1994).

[9] Z. Tesanovic *et al.*, Phys. Rev. Lett. **69**, 3563 (1992).

[10] L. N. Bulaevskii, M. Ledvij, and V. G. Kogan, Phys. Rev. Lett. **68**, 3773 (1992).

[11] S. Salem-Sugui, Jr. and E. Z. da Silva, Physica C **235**, 1919 (1994).

[12] K. Moloni, M. Friesen, Shi Li, V. Souw, P. Metcalf, L. Hou, and M. McElfresh, Phys. Rev. Lett. **78**, 3173 (1997).

[13] B. Rosenstein *et al.*, Phys. Rev. B **63**, 134501 (2001).

[14] S. Salem-Sugui, Jr., M. Friesen, A. D. Alvarenga, V. Vieira, P. Pureur, S. M. Weinschutz, and M. M. Doria, J. Magn. Magn. Mater. **226-230**, 304 (2001).

[15] M. Suenaga *et al.*, Phys. Rev. Lett. **66**, 1777 (1991).

[16] M. Tinkham, *Introduction to Superconductivity*, 2nd ed. (McGraw-Hill, New York, 1996).

[17] S. P. Farrant and C. E. Cough, Phys. Rev. Lett. **34**, 943 (1975).

[18] D. K. Finnemore *et al.*, Phys. Rev. **149**, 231 (1966).

[19] M. M. Doria, S. Salem-Sugui, Jr., I. G. de Oliveira, L. Ghivelder, and E. H. Brandt, Phys. Rev. B **65**, 144509 (2002).

[20] A. Abrikosov, Zh. Éksp. Teor. Fiz. **32**, 1442 (1957) [Sov. Phys. JETP **5**, 1174 (1957)].

[21] P. G. de Gennes, *Superconductivity of Metals and Alloys* (Addison-Wesley, Reading, MA, 1989).

[22] M. B. Salamon *et al.*, Phys. Rev. B **47**, 5520 (1993).