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Flux expulsion and reversible magnetization in the stripe phase superconductor $La_{1.45}Nd_{0.40}Sr_{0.15}CuO_4$

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Magnetization and free energy surfaces have been studied for superconducting $La_{1.45}Nd_{0.40}Sr_{0.15}CuO_4$ in order to determine whether this stripe-phase material has a thermodynamic critical field curve, H_c, similar to the classical superconductors. A large region of thermodynamic reversibility is found so that magnetization and free energy surfaces can be determined reliably over much of the H-T plane. In these stripe-phase materials there is evidence from neutron scattering that the holes collect in the domain walls of the antiphase domain structure, so the superconducting order parameter might be space dependent and the vortex lattice might be different from ordinary type-II superconductors. Although some uncertainty is introduced because a substantial background magnetization from the magnetic Nd ions must be subtracted, it can be stated that the H_c vs T curve is found to be comparable to Nb. The material is a good bulk superconductor, but the shape of the magnetization curves (M_s vs H) is quite different from the predictions of conventional type-II superconductor theories. [S0163-1829(97)01530-0]

I. INTRODUCTION

Neutron scattering studies[1,2] have shown stong evidence for a stripe phase in the cuprate superconductor $La_{1.45}Nd_{0.40}Sr_{0.12}CuO_4$, and there have been several theoretical studies showing how the charge might phase separate into these structures.[3,4] As the sample is cooled, there are charge-ordering peaks that appear at about 60 K, then spin-ordering peaks of the Cu that appear at about 50 K.[1] Detailed study shows that the peaks are caused by an antiphase magnetic domain material with charge ordering in which the mobile holes reside in the domain walls. These stripes of holes run along the (l00) and (010) directions in successive planes of the CuO_2 layers. Neutron scattering measurements[2] that also have been done for a Sr concentration of 0.15 show evidence for charge stripes. Low-field magnetization measurements show that this Sr 0.15 material has a superconducting transition temperature, T_c, of about 10.5 K. If the material is a true bulk superconductor, then one might expect that this spatial separation of charge might cause a situation in which the order parameter is space dependent and the vortex structures might differ from classical superconductors.

The purpose of this work is to determine whether these stripe phase materials have magnetization curves and a free energy surface similar to classical superconductors. First, it is necessary to demonstrate that the magnetization is thermodynamically reversible over a large portion of the H-T plane so that reversible thermodynamics can be used. Second, we wish to show that these stripe-phase materials are truly bulk superconductors that can expel flux at high fields to the same extent as a classical type-II superconductor such as Nb. Third, if $La_{1.45}Nd_{0.40}Sr_{0.15}CuO_4$ is a bulk superconductor, then it is important to determine whether the H_c vs T curve of this stripe-phase compound is similar to all of the classical superconductors[5] so that a law of corresponding states is obeyed. Fourth, if the sample is a stripe-phase bulk superconductor, then it would be important to determine whether the magnetization curves (M_s vs H) obey a Ginsburg-Landau-Abrikosov-Gorkov[5] or a Hao-Clem[6] M vs H curve. With stripes having a periodicity of a few lattice spacings, one would expect the vortex lattice to be distorted and, hence, different from classical type-II superconductors.

The central difficulty in carrying out this program is to account for the background magnetization of the magnetic Nd ions, M_n, well enough that it can be subtracted from the total to give an accurate superconducting contribution, M_s. The free Nd ion has a spin of 9/2 and a Lande g factor of 0.7273. Hence, with 20% of the La sites occupied by Nd, the magnetization from the Nd is a major factor. In addition, the Nd ions order[2] at about 3 K so that the modeling of M_n cannot be done with a simple Brillouin function for a spin of 9/2. At present, there are no measurements of the crystal field splittings that occur. Once M_s is known by subtracting M_n from the total magnetization, M ($M_s = M - M_n$), then the free energy difference between the normal and superconducting state, $G_n - G_s$, can be determined from the area under the magnetization curve (M vs H). The thermodynamic critical field is given by $H_c^2/8\pi = G_n - G_s = \int -M dH$ where the limits of integration are from $H=0$ to the normal state.

II. EXPERIMENTAL TECHNIQUE

A 140 mg single-crystal sample, which was a portion of the same sample used for the neutron scattering, was oriented using x rays and mounted with the c axis parallel to the magnetic field of the Quantum Design magnetometer. The precision of magnetization measurements is better than 0.1% and the accuracy of the temperature measurement is better

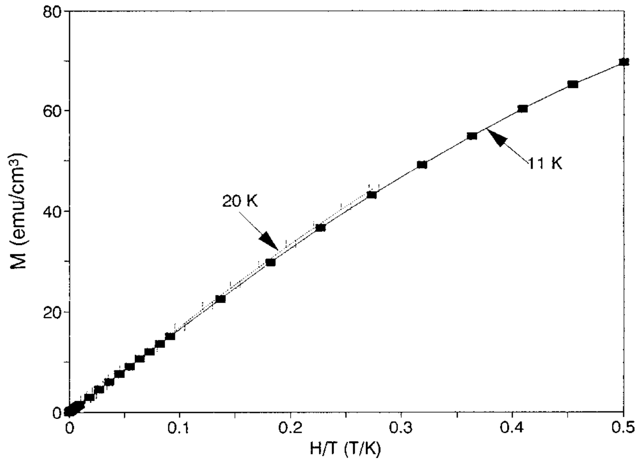

FIG. 1. Normal state magnetization data at 11 K (solid curve) is compared to data at 20 K (dotted curve) on an M_n vs H/T plot. This positive magnetization arises from the Nd ions.

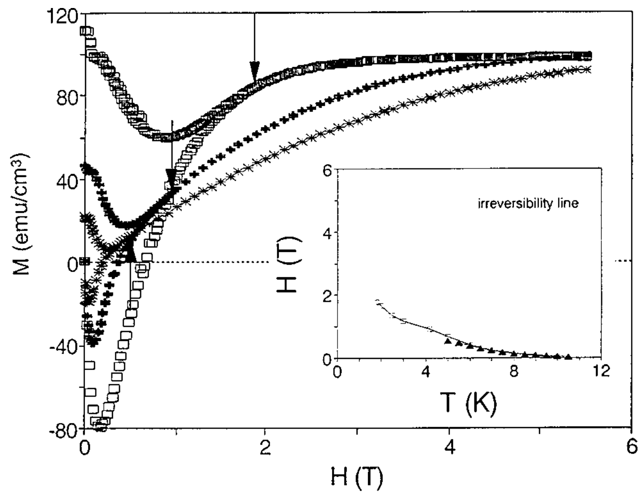

FIG. 2. Magnetization data below the superconducting transition temperature showing the saturation of the magnetization at 1.85 K and the onset of irreversible behavior. The open squares are 1.85 K; the pluses are 4.20 K; the asterisks are 6.00 K. The inset shows H_{irr} vs T.

than 0.010 K. For each magnetization vs field run, the sample was warmed to 20 K and cooled to the desired temperature in zero field. The magnetic field was then swept from zero to +5.5 T to −5.5 T and back to zero field so that the sample had been swept through a full magnetization cycle. Then the magnetization data were taken from zero field to 5.5 T and back to zero field to map out both the reversible region and the hysteretic region. The dominant source of error in determining the superconducting magnetization arises from uncertainty in subtracting the magnetization on the Nd ions.

III. RESULTS AND DISCUSSION

The normal state magnetization data were taken at 1.0 K intervals from 11 K to 20 K. If the Nd ions were free spin 9/2 objects, the data would follow a universal Brillouin function curve on an M vs H/T plot. As shown in Fig. 1, the data at 11 K (solid squares) lie somewhat below the 20 K data (open squares). After several attempts to fit the data with crystal field split levels, it was found that a Curie-Weiss-like approach in which T is replaced by $T-\theta$ fits the data rather well. Fitting the data with the relation

$$M_n=\frac{N[\Sigma_a\mu_a e^{\mu_a\cdot H/k(T-\theta)}]}{[\Sigma_a e^{\mu_a\cdot H/k(T-\theta)}]}, \tag{1}$$

where $\mu=gm_j\mu_B$, m_j is the projection of the angular momentum along H, μ_B is the Bohr magneton, N is the number of Nd ions per cubic centimeter, and a is an index that runs over the m_j quantum numbers. The solid line of Fig. 1 shows a fit to the 11 K magnetization data with $g=0.7029$ and $\theta=3.94$ K. Hence, the date fit Eq. (1) to rather good accuracy with a g value close to the free ion value (0.7029 compared to 0.7273) and a θ value close to the ordering temperature seen by neutrons.[2]

Typical data taken below the superconducting transition are shown on Fig. 2 for 1.85 K, 4.20 K, and 6.00 K. At 1.85 K, the initial field increasing data (open squares) show strong Meissner-like screening at fields below 1000 Oe. At higher fields, the data bend over and approach a saturated behavior as the magnetic field approaches 5 T. If we define the irreversibility field, H_{irr}, as the field where the difference between the field increasing and field decreasing magnetization differ by about twice the noise in the data (ranging from 0.05 to 0.10 emu/cm^3), then H_{irr} is about 1.8 T at 1.85 K, 0.92 T at 4.2 K, and 0.45 T at 6.0 K as shown by the vertical arrows on Fig. 2. The full irreversibility line, shown by the inset in Fig. 2, indicates that there is a wide portion of the H-T plane where magnetization is a well-defined quantity and reversible thermodynamics can be used.

To determine the value of the normal state magnetization arising from the Nd ions at temperatures between 6 and 11 K, we adopt a strategy based on the observation that the Nd moment saturates at 1.85 K for fields above 5.0 T. If M were truly constant, then the sample would be normal and the upper critical field would be less than 5.0 T at 1.85 K and also presumably at all higher temperatures. This would be a very useful fact in analyzing the higher temperature superconducting data if it were true because M_n could be fit to the measured M in the 5.0 to 5.5 T region to determine g and θ. A more detailed look at these 1.85 K data, however, show that the curve is still rising slightly by about 0.1 emu/cm^3 between 5.0 and 5.5 T with a scatter in the data of about 0.03 emu/cm^3. This rise could occur because the Nd ions are not totally saturated, or it could occur because vortices are being expelled. Because this rise in magnetization is so small at 1.85 K, the strategy for determining the Nd ion magnetization between 6 K and 11 K will be to assume that M_s is too small to measure above 5.0 T so that Eq. (1) can be fit to the magnetization data, M, for all temperatures above 6.0 K and fields above 5.0 T.

In this analysis, we assume that M_s is negligible above 5.0 T and seek values of g and θ that make Eq. (1) match the measured M at both 5.0 T and 5.5 T. To determine M_s, the M_n fit data are subtracted from the total so that $M_s=M-M_n$. The superconducting magnetization derived in this way is shown in Fig. 3(a) by the asterisks for the 6.0 K data.

To test the size of the errors in M_s resulting from a misfit to the background magnetization from the Nd ions, three different ways of treating the background have been used. In

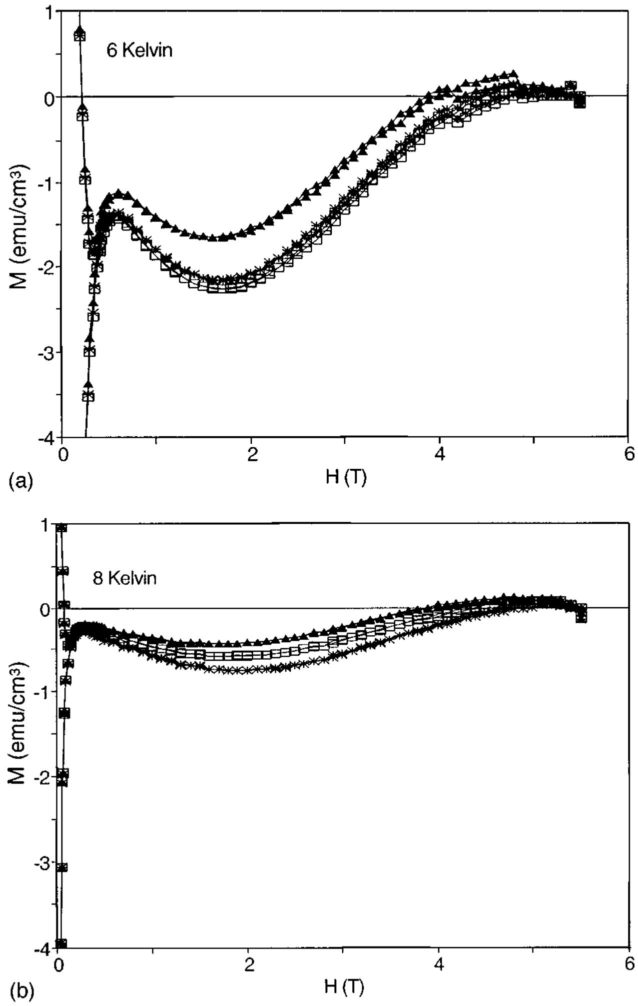

FIG. 3. (a) Superconducting magnetization at 6 K derived with three different assumptions about the background magnetization. The open squares are fit with $M = M_n$ at 4.5 and 5.5 T; the asterisks are fit with $M = M_n$ at 5.0 and 5.5 T; the solid triangles are fit with $M = M_n$ at 4.0 and 5.5 T. (b) Similar data at 8 K.

the first method, the g and θ values are adjusted to match the measurements, M, at 5.0 and 5.5 T with the results shown by the asterisk points in Fig. 3(a). This fit gives $g = 0.6319$ and $\theta = 3.096$ K. Note that these data represent both field increasing and field decreasing data, so the reversibility is quite good. In the field range from 4.3 T to 5.5 T, there is a glitch in the field increasing data of about 0.1 emu/cm^3. This probably reflects the level of the accuracy of the data. In the second method, the g and θ values are adjusted to match the M measurements at 4.5 and 5.5 K with the results shown by the open squares. This fit gives $g = 0.6403$ and $\theta = 2.9332$ K. Within the accuracy of the data, forcing Eq. (1) to fit at 4.5 K and 5.5 K produces an M_n that fits M throughout the whole interval from 4.5 to 5.5 T. In the third method, the g and θ values are forced to fit M at 4.0 and 5.5 T with the M_s results shown by the solid triangles of Fig. 3(a). This fit gives $g = 0.6460$ and $\theta = 2.822$ K. With this fit, there is a positive M_s region well above the noise between 4.0 and 5.5 K. This positve magnetization cannot physically represent flux expulsion, so we assume that this M_n is not satisfactory. Of these methods, the second for which M_n is fit at 4.5 and 5.5 T [open squares in Fig. 3(a)], is probably closest to the correct magnetization curve at 6 K with the asterisks and solid triangles representing an interval of probable error. Similar results are shown for 8.0 K data in Fig. 3(b).

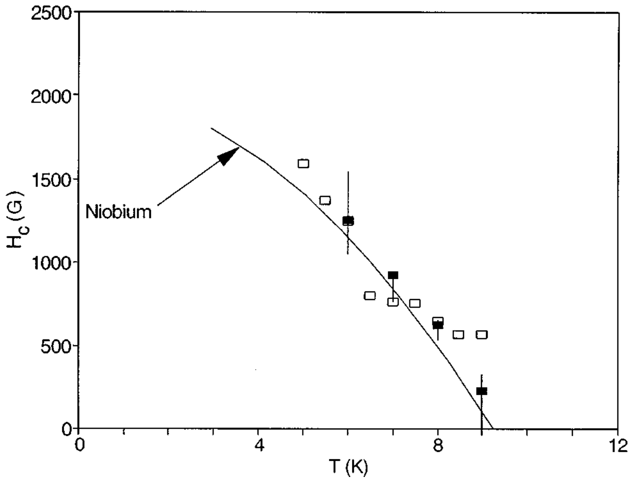

FIG. 4. Comparison of H_c for $La_{1.45}Nd_{0.40}Sr_{0.15}CuO_4$ with Nb. Solid squares are the first run and the open squares are the second run.

In some ways the data of Fig. 3 are very ordinary but in other ways they are rather unusual. If we estimate the thermodynamical critical field by taking the area under reversible magnetization, the H_c vs T data of Fig. 4 (solid squares) are very close to a classical superconductor like Nb,[7] shown by the solid curve. The error bars on the data represent the uncertainty of fitting M_n as described above and shown in Fig. 3. There is some error in estimating the area for fields lower than H_{irr}. A linear extrapolation to $H = 0$ was used. After this analysis was complete, a full set of data was taken again giving the open squares in Fig. 4. Hence, in spite of the uncertainties on the order of 30%, we can conclude that this material is a good bulk superconductor that reversibly expels flux roughly as strongly as the classical type-II superconductor Nb.[7]

The general shape of the magnetization curves, shown in Fig. 3, is very robust and is preserved regardless of the assumptions about M_n. Shifting M_n changes the magnitude of M_s and the area under the curve, but leaves the shape roughly the same. Furthermore, the shape is quite different from Nb and it is quite different from the cuprates Y(123) (Refs. 8,9) and Y(124).[10] In the vortex regime of thermodynamical reversibility, Nb,[5] Y(123),[8,9] and Y(124) (Ref. 10) have a monotonic decrease in the absolute magnitude of M_s as the field increases. For this $La_{1.45}Nd_{0.40}Sr_{0.15}CuO_4$ sample, however, starting at high field, the exclusion of flux goes through a maximum in a midfield range in the vortex state [about 1.5 T at 6.0 K in Fig. 3(a)] and then falls. There even seems to be another upturn, close to the point where the sample goes irreversible (about 0.5 T at 6.0 K). These nonmonotonic curves are very common in irreversible magnetization where different regimes of flux pinning occur.[11] Reversible magnetization curves that have this general shape recently have also been reported for Bi(2212) that has been irradiated with heavy ions.[12] In the case of a dense array of heavy ion defects, the order parameter is suppressed in the region of the channels and the peak in the reversible magnetization occurs at approximately the magnetic field where there is one vortex per ion channel. Both the stripe-phase and irradiated Bi(2212) are similar in that there is a space dependence of the order parameter. The details of the stripe phase case, however, are different. If an average spacing of vortices is defined as $[\phi_0/H]^{1/2}$, then the average spacing of vortices would be 45 nm at 1.0 T, 26 nm at 3.0 T, and 20 nm at 5.0 T. All of these distances are much larger than the spacing of the stripes seen by neutrons[1,2] so it is not clear that the nonmonotonic behavior arises from any matching effect. With the stripes rotated by 90° in every successive copper oxide layer, it is a bit difficult to picture the structure of vortices in these materials. It would, however, not be surprising that vortex structure and the magnetization curves for stripe-phase materials are quite different from Nb.

IV. CONCLUSIONS

Magnetization data for H parallel to the c axis of a single crystal of $La_{1.45}Nd_{0.40}Sr_{0.15}CuO_4$ and temperatures above 6.0 K can be fit very well with a Brillouin-like function with a Curie-Weiss modification of the temperature as in Eq. (1). Within a few degrees of the Curie-Weiss θ, say below 5 K, this approximation begins to fail. For temperatures of 6.0 K and up, the resulting g values are reasonably close to the free ion values and the Curie-Weiss temperatures agree with the ordering that appears in the neutron scattering data. The sample shows thermodynamic reversibility of the magnetization over a wide range of the H-T plane with values of $H_{\rm irr}$ ranging from 0.02 T at 8 K to 1.0 T at 4.2 K. By assuming that the sample is normal above 4.5 T and by taking just the area under the curve in the reversible regime, the values of H_c are found to be similar to Nb. Hence, $La_{1.45}Nd_{0.40}Sr_{0.15}CuO_4$ is a bulk superconductor and excludes flux to a degree comparable to Nb. There is no doubt that this sample is superconducting, even though some earlier phase diagrams indicated that it might not be.[13] The detailed shape of the M_s vs H curves for this stripe-phase material are different from both classical type-II superconductors and different from the cuprates such as Y(123) and Y(124).

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1 J. M. Tranquada, J. D. Axe, N. Ichikawa, Y. Nakamura, S. Uchida, and B. Nachumi, Phys. Rev. B **54**, 7489 (1996).

2 J. M. Tranquada, J. D. Axe, N. Ichikawa, A. R. Moodenbaugh, Y. Nakamura, and S. Uchida, Phys. Rev. Lett. **78**, 338 (1997).

3 V. J. Emery and S. A. Kivelson, Physica C **209**, 597 (1993).

4 U. Low, V. J. Emery, K. Fabricius, and S. A. Kivelson, Phys. Rev. Lett. **72**, 1918 (1994).

5 P. G. deGennes, in *Superconductivity of Metals and Alloys*, edited by D. Pines (Benjamin, New York, 1966).

6 Z. Hao and J. R. Clem, Phys. Rev. Lett. **67**, 2371 (1991).

7 D. K. Finnemore, T. F. Stromberg, and C. A. Swenson, Phys. Rev. **149**, 231 (1966).

8 J. Gohng and D. K. Finnemore, Phys. Rev. B **46**, 398 (1992).

9 J. W. Loram, K. A. Mirza, and P. F. Freeman, Physica C **171**, 243 (1990).

10 J. Sok, M. Xu, W. Chen, and B. J. Suh, J. Gohng, D. K. Finnemore, M. J. Kramer, L. A. Schwartzkopf, and B. Dabrowski, Phys. Rev. B **51**, 6035 (1991).

11 M. Daeumling, J. M. Seuntjens, and D. C. Larbalestier, Nature (London) **346**, 332 (1990).

12 C. J. van der Beek, M. Konczykowski, T. W. Li, P. H. Kes, and W. Benoit, Phys. Rev. B **54**, R792 (1996); T. W. Li, R. J. Drost, P. H. Kes, H. W. Zandbergen, N. T. Hien, A. A. Menovsky, and J. J. M. Franse, Physica C **274**, 197 (1997); A. Wahl, V. Hardy, J. Provost, Ch. Simon, and A. Buzdin, *ibid.* **250**, 163 (1995).

13 B. Buchner, M. Breuer, A. Freimuth, and A. P. Kampf, Phys. Rev. Lett. **73**, 1841 (1994).