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On the possibility of fast vortices in the cuprates: A vortex plasma model analysis of THz conductivity and diamagnetism in $La_{2-x}Sr_xCuO_4$

L. S. Bilbro,[1] R. Valdés Aguilar,[1] G. Logvenov,[2] I. Bozovic,[2] and N. P. Armitage[1]

[1]*The Institute for Quantum Matter, Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218, USA*

[2]*Brookhaven National Laboratory, Upton, New York 11973, USA*

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We present measurements of the fluctuation superconductivity in an underdoped thin film of $La_{1.905}Sr_{0.095}CuO_4$ using time-domain THz spectroscopy. We compare our results with measurements of diamagnetism in a similarly doped crystal of $La_{2-x}Sr_xCuO_4$. We show through a vortex-plasma model that if the fluctuation diamagnetism solely originates in vortices, then they must necessarily exhibit an anomalously large vortex diffusion constant, which is more than two orders of magnitude larger than the Bardeen-Stephen estimate. This points to either the extremely unusual properties of vortices in the underdoped d-wave cuprates or a contribution to the diamagnetic response that is not superconducting in origin.

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Nearly 25 years after the demonstration of high-temperature superconductivity in cuprate superconductors and more than 15 years since the discovery of the anomalous pseudogap in underdoped compounds, the microscopic physics of the superconducting phase and its relationship to the pseudogap remain hotly debated. Due to their low superfluid densities, it is generally agreed that superconducting fluctuations will be large and prominent in these materials.[1] What is less agreed upon is the temperature range above T_c in which superconducting correlations are truly significant and their contributions to the physics of the pseudogap. Experimental probes such as photoemission, tunneling, NMR spin relaxation, heat capacity, the Nernst effect, and diamagnetic susceptibility have shown evidence for a gaplike structure reminiscent of d-wave superconductivity in the density of states, implying a strong connection of the pseudogap to superconductivity and/or superconducting correlations at temperatures well above T_c.[2–6] However, other mechanisms exist that can create such structures in the density of states.[7,8]

Interestingly, perhaps the most essential probe of the electronic properties—charge transport—does not show an extended range of superconducting fluctuations in temperature or field.[9–11] In $La_{2-x}Sr_xCuO_4$ the region of enhanced diamagnetism extends almost 100 K above T_c (Ref. 6) while the THz fluctuation conductivity has an extent limited to 10–20 K above T_c.[12] This is surprising, as one might expect a close correspondence between these quantities.[13] Similarly, it has been argued from Nernst and diamagnetism measurements that H_{c2} may be as high as 150 T,[6] while the resistive transition is essentially complete in optimally and underdoped lanthanum strontium copper oxide ($La_{2-x}Sr_xCuO_4$) by 45 T.[10,11]

In this Rapid Communication we present results of our detailed THz time-domain spectroscopy (TTDS) study of the fluctuation superconductivity in LSCO. The THz fluctuation conductivity shows an onset approximately only 10 K above T_c, which contrasts strongly with measurements such as diamagnetism in which the onset is ∼100 K above T_c. We analyze our data in the context of a vortex plasma model and show, however, that it is not the functional dependences of these data that are in strongest contrast, but their overall scales. Conventional vortex dynamics would predict a much larger fluctuation conductivity given the size of diamagnetism. We demonstrate that if the regime of enhanced diamagnetism originates in vortices, then the vortex diffusion constant D must be anomalously large and in the range of 10–30 cm^2/s above T_c. This is more than two orders of magnitude larger than conventional benchmarks based on the Bardeen-Stephen model.[14] It is then a well-posed theoretical challenge to explain a D this large. This points to either extremely unusual vortex properties in the underdoped d-wave cuprates or a contribution to the diamagnetic response that is not superconducting in origin.

We begin with the observation that the ratio $\chi_{2D}/\mu_0 G$ of the two-dimensional (2D) susceptibility over the conductance has units of length squared over time, i.e., diffusion.[15] One can show that in a diffusive vortex plasma this ratio gives a unique measure of the vortex diffusion constant.[16] Using the notation of Halperin and Nelson,[13] but in SI units, the 2D susceptibility and conductance of a conventional thin superconducting film at temperatures above a vortex unbinding transition are

$$\chi_{2D} = -\frac{c_2 \pi^2 \mu_0 k_B T}{\phi_0^2}\xi^2, \quad (1)$$

$$G_S = \frac{1}{\phi_0^2 n_f \mu}. \quad (2)$$

Here ξ is a correlation length, ϕ_0 is the flux quantum, and μ is the vortex mobility. n_f is the areal density of thermally excited free vortices, which is related to the correlation length by the relation $n_f = 1/2\pi c_1 \xi^2$. c_1 and c_2 are small dimensionless constants. It is reasonable to expect that very close to T_c vortices are the principal degrees of freedom in even quasi-2D materials. Note that these are essentially model-free forms constrained only by dimensional analysis, Maxwell equations, and immutable properties of superfluid vortices such as the Josephson relation. Using accepted values for c_1 and c_2,[13] and the Einstein relation $D = \mu k_B T$, the expression

$$D(T) = -\frac{6}{\mu_0}\frac{\chi_{2D}}{G_S} \quad (3)$$

follows[16] and in principle may be used to give a determination of the vortex diffusion constant D using only experimentally determined quantities. Interestingly, this treatment using the analogous equations within the Gaussian approximation and in the dirty limit gives the diffusion constant of the normal-state *electrons*. This is potentially useful as a diagnostic considering that electronic diffusion is proportional to the normal-state conductance while vortex diffusion is conventionally proportional to the normal-state resistance. One may also heuristically motivate Eq. (3) through the fact that correlations in length (diamagnetism in 2D $\propto \xi^2$) probed by a thermodynamic measurement such as susceptibility and the correlations in time ($1/\Omega$) probed by a dynamic measurement such as conductivity are related within diffusive dynamics as $\xi^2 \propto D/\Omega$, where Ω is the characteristic fluctuation rate.

A problem with applying Eq. (3) to real type-II superconductors is that, in general, the motion of vortices is limited by both dissipative (viscous) flux-flow and pinning forces. In 2D, the classical equation of motion for a single vortex is $\dot{x}/\mu + k_p x = K_y \phi_0$, where K_y is a driving sheet current, x is the vortex displacement, and k_p is a pinning constant.[17] Here the complex physics of pinning and flux flow are represented by phenomenological parameters. This leads to an expression for the 2D resistance from moving vortices as $R_v = \phi_0^2 n_f \mu[1/(1 + i\omega_d/\omega)]$, where $\omega_d = k_p \mu$ is the "depinning frequency." This expression shows that at frequencies well above ω_d, viscous forces dominate and the motion of vortices becomes predominately dissipative. This is a considerable simplification. In this limit the expression for R_v reduces to the inverse of Eq. (2) for the vortex conductance. In cuprate superconductors, ω_d is generally of the order of a few GHz.[18] This puts the appropriate frequency regime to probe purely dissipative vortex transport in the range of our TTDS measurements.

We have measured the THz range optical conductivity of molecular beam epitaxy (MBE) grown LSCO films using a homebuilt transmission-based time-domain THz spectrometer. With this technique the complex transmission function can be directly inverted to get the complex conductivity.[19] In Figs. 1(a) and 1(b) we present the real (σ_1) and imaginary (σ_2) THz conductivity of one particular LSCO film ($x = 0.095$, $T_c = 23.5$ K) out of a large series we have recently studied.[12] At high temperature σ_1 is fairly constant in frequency. As the temperature is lowered, σ_1 increases, develops a frequency dependence near T_c, and then decreases as spectral weight is shifted into a delta function at zero frequency. The σ_2 versus frequency data in Fig. 1(b) show a small imaginary part of the conductivity at high temperatures, which is enhanced dramatically as temperature is reduced near T_c. At the lowest displayed temperatures σ_2 shows the $1/\omega$ dependence expected for the superfluid response of a superconductor. While the low- and high-temperature limits are easily understood, we are most interested in the fluctuation regime near T_c.

The enhancement of the conductivity in this fluctuation regime is more clear in Figs. 1(c) and 1(d), where we plot σ_1 and σ_2 versus temperature. One can see clearly the slow increase and subsequent decrease in σ_1 as temperature is lowered below T_c. At low frequency there is a well-defined peak around T_c. The location of this peak shifts to lower temperature as frequency is reduced, corresponding to the slowing down of fluctuations as the temperature decreases. Above T_c, we see a sudden onset in σ_2 at a temperature $T \sim 30$ K. In earlier work, we found that the second derivative with respect to temperature of the quantity $\omega\sigma_2$ (which is related to the phase stiffness) showed a clear and dramatic onset from a near-zero high-temperature signal.[12] We denoted this temperature as T_o, and defined it as the onset of superconducting fluctuations in the charge conductivity (for this film $T_o \approx 31$ K). Note that there is no sign of conductivity enhancement at the high temperatures of the Nernst or diamagnetism onset.[4–6]

As mentioned above, in conventional models where vortices are the principal degree of freedom in the region above T_c, one expects that correlations in length and time scale together as a diffusionlike relation with vortex diffusion constant D. Therefore, the large difference in the temperature of the inferred onset of superconducting correlations T_o above T_c between our experiments (10–20 K) and, for instance, diamagnetism measurements ($\approx$100 K)[5,6] begs an explanation. Here we evaluate the relative size of the signals in terms of the diffusion constant derived in the above analysis and show that conventional vortex dynamics would predict a much larger fluctuation conductivity given the size of diamagnetism.

In Fig. 2(a), we plot the magnitude of the conductivity $|\sigma|$. To isolate the superconducting fluctuation contribution σ_S, we define a normal-state contribution that fits the conductivity well at temperatures above the onset of diamagnetism ($T_D \approx$ 75–110 K in this doping range[6]), extrapolate to low temperatures,

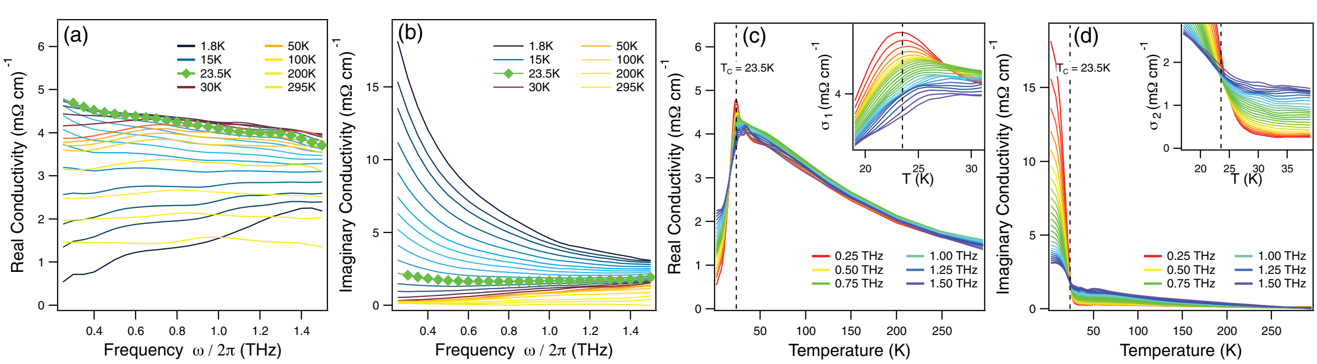

FIG. 1. (Color online) (a) Real and (b) imaginary conductivities as a function of frequency at different temperatures of a $x = 0.095$, $T_c = 23.5$ K LSCO film. (c) Real and (d) imaginary conductivities as a function of temperature at different frequencies. In (a) and (b) the green curve marked with diamonds denotes T_c. In (c) and (d) the vertical lines represent T_c. The insets to (c) and (d) show expanded views of the fluctuation region.

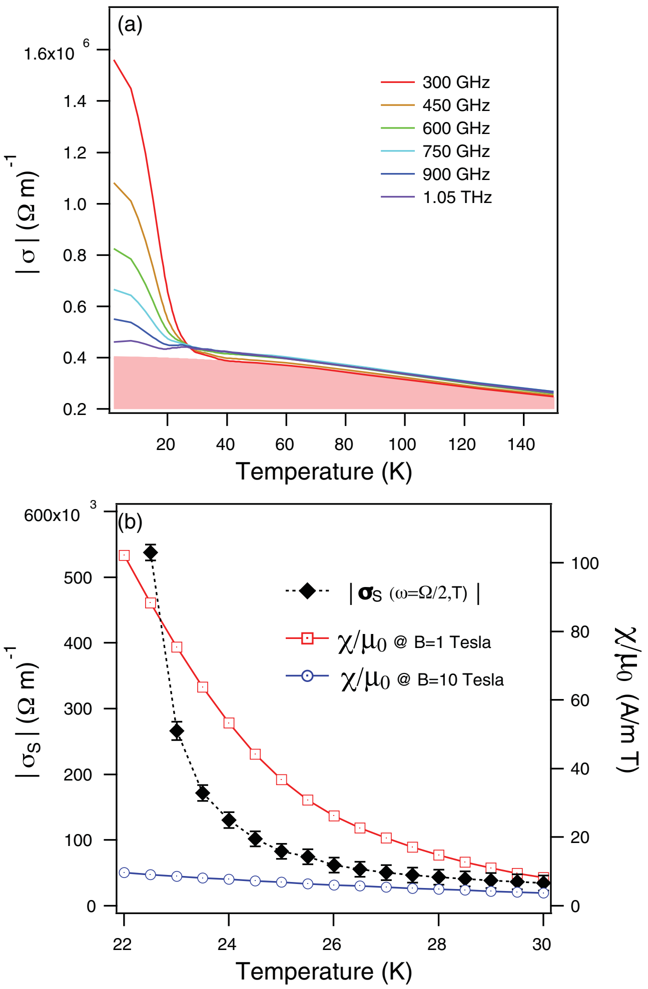

FIG. 2. (Color online) (a) Magnitude of the conductivity ($|\sigma|$) as function of temperature. The filled region is a fit of the normal-state background conductivity at 300 GHz (Ref. 19). The fluctuation conductivity σ_S is obtained by subtracting this background from $|\sigma|$. (b) A comparison of fluctuation conductivity with the diamagnetism in similarly doped $La_{2-x}Sr_xCuO_4$ crystals (Ref. 20).

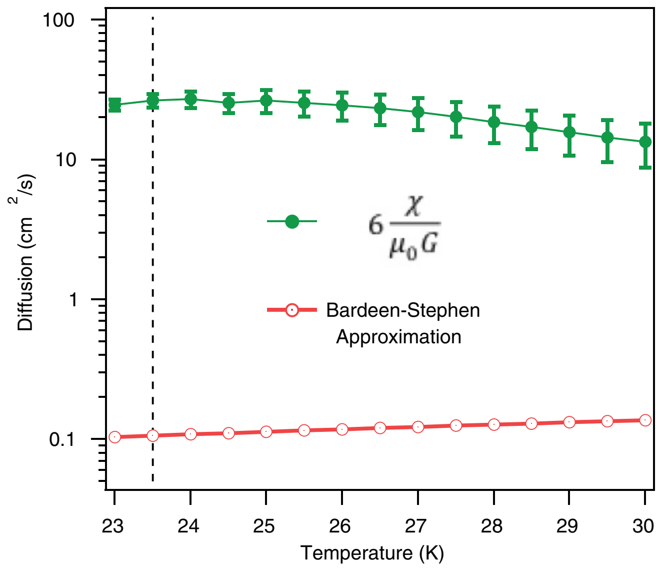

FIG. 3. (Color online) Comparison of the Bardeen-Stephen estimation and calculated diffusion constant using our measured fluctuation conductivity, the estimated normal-state background and the 1-T magnetic susceptibility.

and take the difference. Although we fit the background to a temperature-dependent Drude model,[19] our final conclusions are not sensitive to the precise background choice as we are only concerned with the temperature region up to ~10 K above T_c, where the fluctuations are obvious.

In previous work[12] we have performed a scaling analysis that allowed us to extract the characteristic frequency scale Ω of the fluctuation superconductivity in the region above T_c.[19] In the analysis that follows we evaluate $|\sigma_S(\omega,T)|$ at a frequency $\omega = \Omega(T)/2$ for each temperature. This conductivity differs formally from the conductivity in Eq. (2) by a constant of order unity, which we set to one below. The use of THz frequencies eliminates the effects of pinning and the scaling analysis essentially connects the response of the system at finite frequency to the dc response that the system *would* have had in the absence of vortex pinning. In Fig. 2(b), on the left-hand axis, we plot the magnitude of the fluctuation conductivity contribution, evaluated at $\omega = \Omega(T)/2$, versus temperature. On the right-hand axis, we include diamagnetic susceptibility χ/μ_0 at 1 and 10 T of a single-crystal sample LSCO sample[20] with a similar doping and T_c ($x = 0.9$ and 23 K, respectively). In this data, one can see how the larger field suppresses the susceptibility near T_c. Although there is some correspondence between the form of the lower-field susceptibility with the conductivity, we now show that in fact it is the relative scale of these quantities which is particularly remarkable.

We now apply Eq. (3) with the data in Fig. 2(b) to extract D for a small range of temperatures above T_c. As shown in Fig. 3, we find that D is of the order of tens of cm^2/s throughout the range above T_c. This is at least two orders of magnitude larger than a simple Bardeen-Stephen (BS) estimate $D = (2k_BTe^2\xi_c^2)/(\pi\hbar^2\sigma_n t)$ (Ref. 14) (here σ_n is the extrapolated normal-state background conductivity, t is the spacing between CuO_2 layers, and ξ_c is the vortex core size[21,22]). The BS approximation appears to work well to model flux-flow dissipation in conventional s-wave materials,[23–25] where the majority of dissipation occurs through quasiparticle motion in the vicinity of the vortex cores. Note that the magnetic susceptibility appears to become singular as $B \to 0$ near T_c ("fragile London rigidity"),[6] so that evaluating D at lower fields (corresponding to our $B = 0$ TTDS experiment) will only increase the ratio of χ/G and the discrepancy with the BS estimate. Although there is an expectation that due to their d-wave nature, short coherence lengths, gapped vortex core, and proximity to the Mott insulator, the cuprate vortices may be "fast" as compared to the BS estimate,[26–32] the discrepancy we find is extreme. It is an open question whether a diffusion constant as large as we have found can be reconciled. We have currently performed this analysis for one underdoped sample due to difficulty in obtaining compatible diamagnetism data. However, we anticipate similar behavior for the entire underdoped part of the phase diagram, since signals of conductivity and diamagnetism vary smoothly as a function of doping.[6,12]

There are two obvious possible conclusions from our data and analysis. If in fact the large diamagnetic response in the cuprates comes entirely from superconducting correlations, then we have shown that their vortex motion must be anomalously fast and their dissipation anomalously small to reconcile the behavior with charge transport. One expects that above T_c an effective two-fluid model may apply where the total conductivity has contributions from both normal electron and superconducting degrees of freedom in the form of $\sigma_T = \sigma_N + G_S/t$, where G_S is given by Eq. (2) in the vortex regime. Our results show the manner in which superconducting correlations may persist far above T_c but be invisible to the charge response; the fast vortices are shorted out by the normal electrons. It is a separate but well-posed theoretical challenge to explain vortex motion this fast. Although detailed calculations must be performed, it is possible that such anomalously fast diffusion may arise as a consequence of the cuprates' d-wave nature and small gapped cores,[31,33] inhomogeneities,[34] the existence of a competing state nucleated in the vicinity of a vortex,[28] or the proximity to a Mott insulator.[30,32] Alternatively, if calculations show that vortex dissipation must always be at least parametrically related to the BS estimate by numbers of order unity, then our analysis shows that there must be another large contribution to the diamagnetic response that is not superconducting in origin (see Ref. 35 for one such possibility).

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