Brownian Motion of Vortex-Antivortex Excitations in Very Thin Films of $Bi_2Sr_2CaCu_2O_8$

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Low-frequency magnetic noise and mutual inductance measurements indicate that the electromagnetic response of very thin $Bi_2Sr_2CaCu_2O_8$ films is dominated by Arrhenius law diffusion of thermally created vortices and antivortices. We explain the measured diffusion coefficient via pinning of the vortex cores on a single type of line defect. The observed superfluid response differs from simple Kosterlitz-Thouless predictions as it is predominantly controlled by vortex-pin rather than vortex-antivortex interactions.

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Many experiments have now been performed using thin-film heterostructures, particularly those of $YBa_2Cu_3O_{7-y}$ (YBCO) and the nonsuperconducting material $PrBa_2Cu_3O_{7-z}$ (PrBCO) [1-3], to test whether two-dimensional (2D) behavior is an essential aspect of the high-temperature superconductors. While many of these experiments have been interpreted within the framework of the Kosterlitz-Thouless (KT) transition, detailed comparison with KT theory [4] and additional experiments [5] suggest that sample inhomogeneities may mask the intrinsic superfluid response. In particular, the tensile stress, due to the roughly 1.5% lattice mismatch between YBCO and PrBCO, and charge transfer effects [6], may well modify the superconductive behavior of ultrathin YBCO layers even in structurally ideal YBCO-PrBCO heterostructures.

Recently, Eckstein *et al.* [7,8] have reported atomic-layer-by-layer molecular beam epitaxy (ALL-MBE) growth and transport properties of single-crystal thin films from the $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+2}$ family. These materials offer significant advantages over the YBCO-PrBCO system for studying 2D effects in the high-T_c materials: The a and b lattice parameters of the 15-K superconductor $Bi_2Sr_2CuO_6$ (2:2:0:1) and the 85-K superconductor $Bi_2Sr_2CaCu_2O_8$ (2:2:1:2) are nearly identical, thus allowing for excellent heteroepitaxy and the elimination of strain effects. Also, carrier depletion effects should be insignificant in 2:2:0:1-2:2:1:2 due to their more nearly equal carrier densities. Bozovic *et al.* [9] have reported nearly bulk resistive transition temperatures $T_c \approx 75$ K for superlattices composed of half-unit cell layers of 2:2:1:2 between up to 5 unit cells of 2:2:0:1. This high T_c is a vast improvement over the strongly suppressed values observed in single unit cell YBCO-PrBCO superlattices.

In this paper, we present a study of c-axis-oriented, untwinned trilayer heterostructures containing only a *single* unit cell layer (roughly 3.1 nm per unit cell) of 2:2:1:2. Our films are grown by ALL-MBE on (100)-oriented $SrTiO_3$ substrates with the 2:2:1:2 layer stabilized between two much thicker layers of 2:2:0:1. We have used a dc superconducting quantum interference device (SQUID) to measure two related quantities: The first is the frequency and temperature-dependent complex sheet impedance, $Z(\omega,T)=R+i\omega L_k$, where R is the film's sheet resistance and L_k is the kinetic inductance. L_k is related to the superconductive penetration depth, λ, via [10] $L_k=\mu_0\lambda^2/d$, where μ_0 is the permeability of free space and d is the film thickness. The second quantity we measure is the flux noise power spectrum, $S_\phi(\omega)$. By measuring both the linear transport coefficient Z and the noise spectrum S_ϕ we can utilize the fluctuation-dissipation theorem in understanding the superfluid response.

Our dc SQUID is coupled to the film via a 30-turn superconducting solenoid with a mean diameter of 2.8 mm. Located within this solenoid is a concentrically mounted astatic pair of coils, similar to those described by Jeanneret *et al.* [11] for inducing currents in the film. The face of this coil set is roughly 200 μm back from the surface of the film thus allowing the variation of sample temperature from 4.2 to nearly 200 K without affecting the SQUID system. $Z(\omega,T)$ is determined by inversion [11] of the measured SQUID response to known input currents in the astatic pair. S_ϕ is determined by standard Fourier analysis of the SQUID noise in the absence of input current. The local magnetic field strength is reduced to less than 1 mG by two layers of high permeability metal and a PbIn superconducting shield heat sunk to 4.2 K. SQUID system noise at all sample stage temperatures is roughly $1\times10^{-4}\phi_0/\mathrm{Hz}^{1/2}$ referred to the film and is independent of dc magnetic fields in the range of ±0.1 G.

Figure 1 is a plot of $L_k^{-1}(T)$ at three different frequencies for a film consisting of a single unit cell of 2:2:1:2 sandwiched between two 2:2:0:1 layers of 10 unit cells each. For this film and the three other films measured to date, the superfluid response shows two major jumps, one at roughly 15 K associated with the 2:2:0:1 superconducting transition and one at higher temperature, in this case near 35 K, associated with the thin 2:2:1:2 layer. Further, the single unit cell 2:2:1:2 superfluid rise shows an

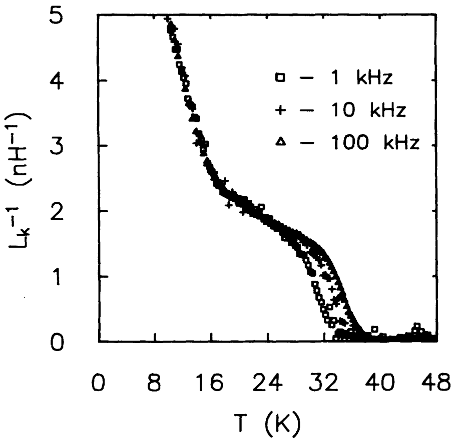

FIG. 1. $L_k^{-1}(T)$ vs T and at three different frequencies for a single unit cell 2:2:1:2 film sandwiched between two 10 unit cell thick layers of 2:2:0:1.

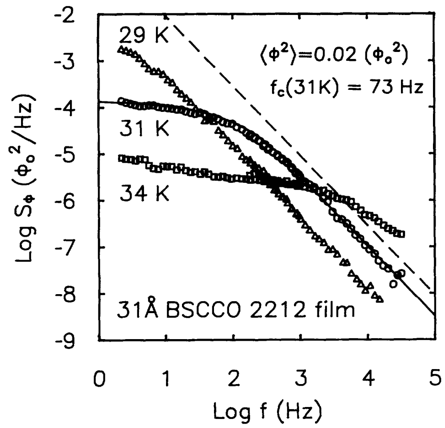

FIG. 2. Log$S_\phi(t)$ vs log$f$ at 29, 31, and 34 K in the 2:2:1:2 transition region. The dashed line shows the $1/f^{3/2}$ power-law characteristic of diffusion noise. The solid line through the 31-K data is a fit to a simple diffusion spectrum from Ref. [20] yielding the parameters listed in the figure.

obvious frequency dependence which is absent in the thick 2:2:0:1 films and which is also absent for thick 2:2:1:2 films. The frequency dependence shows that the high-temperature superfluid response of the thin 2:2:1:2 is a function of the time scale of the measurement; i.e., supercurrents induced for short periods of time will decay to zero over longer periods. This type of decay is predicted to arise for 2D superconductors due to the diffusive motion of thermally created vortex-antivortex pairs [12–15]; it represents initial evidence that 2D effects are controlling the 2:2:1:2 superfluid response.

We next consider the behavior of S_ϕ. Figure 2 is a plot of log$S_\phi(f)$ vs log$f$ at three different temperatures in the region of the 2:2:1:2 superfluid rise for the film of Fig. 1. Early measurements [16] of S_ϕ for films of YBCO and crystals of 2:2:1:2 showed an S_ϕ with a $1/f$ frequency dependence and amplitude which is strongly peaked near T_c. In contrast, for the single unit cell 2:2:1:2 we find a spectrum which is nearly frequency independent below a characteristic frequency, f_c, and which follows a $1/f^{3/2}$ behavior above f_c. Further, f_c is observed to be temperature dependent, moving to lower frequencies with decreasing temperature.

It is known [17,18] that any system which relaxes to equilibrium via a diffusion equation will display a power spectrum of the type we observe. f_c is then related to the diffusion coefficient D and a characteristic length scale of the system L via $f_c = D/2L^2$. For $f > f_c$, the spectrum has a $1/f^{3/2}$ power law, while for $f < f_c$ the spectrum can have a frequency dependence anywhere from constant to $1/f^{1/2}$ depending on system dimensionality and the measurement configuration. To map this picture onto our system, we hypothesize that a thermally created vortex-antivortex population exists and undergoes diffusive Brownian motion about equilibrium with a single diffusion coefficient D. The natural length scale in this case is the typical vortex-antivortex spacing, $\xi_+(T)$, i.e., the spatial granularity scale of the vortex system.

The solid line through the 31-K data is a simple diffusion spectrum calculated by Scofield and Webb [19] which fits the data well and allows us to determine accurate values of both f_c and the total integrated area under S_ϕ, i.e., the thermodynamic variance, $\langle\phi^2\rangle$. The variance is a particularly interesting quantity as it is easy to make an estimate of its value for a gas of vortex-antivortex pairs: If $\phi_v(\mathbf{R})$ is the flux at the SQUID due to a vortex at position $\mathbf{R}$, then for a vortex-antivortex pair of separation $\mathbf{r}_p$, the associated flux ϕ_p at the SQUID is

$$\phi_p = [\phi_v(\mathbf{R}) - \phi_v(\mathbf{R}+\mathbf{r}_p)] \approx (d\phi_v/dR) r_p \cos\theta$$
$$\approx -\phi_M (r_p/R_{\rm coil})\cos\theta ,$$

where ϕ_M is the maximum flux coupled into the SQUID by a single vortex, $R_{\rm coil}$ is the pickup coil radius, and θ is the angle between $\mathbf{R}$ and $\mathbf{r}_p$. The flux, ϕ, due to adding δN pairs is $\phi \approx \phi_p \delta N$. Thus, in terms of mean values, we find $\langle\phi^2\rangle \approx \phi_M^2 (\xi_+/R_{\rm coil})^2 \langle(\cos\theta)^2\rangle\langle\delta N^2\rangle$. Taking the angular average and using the standard thermodynamic result which relates the mean square fluctuations to the total number of pairs inside the coil area, i.e., $\langle\delta N^2\rangle = N_{\rm tot} \approx R_{\rm coil}^2/\xi_+^2$, leads to the result $\langle\phi^2\rangle \approx \phi_M^2/2$ independent of temperature and pair density. The known value of $\phi_M \approx 0.1\phi_0$ for our SQUID system leads to the prediction $\langle\phi^2\rangle \approx 0.005\phi_0^2$. As is shown in Fig. 2, we find $\langle\phi^2\rangle \approx 0.02\phi_0^2$ independent of temperature in good agreement with the arguments above.

Figure 3 shows that f_c follows an Arrhenius law, $f_c = f_0 \exp(-E_0/k_BT)$ with $E_0/k_B \approx 1200$ K and $f_0 \approx 2 \times 10^{19}$ Hz. The temperature dependence [12,15] of $\xi_+(T)$ by itself fails to explain that observed for $f_c = D/2\xi_+^2$. Indeed, in the temperature range which we consider, between the mean-field transition temperature T_{c0} and the KT transition temperature T_c, ξ_+ is a rather weak function of temperature. Thus, most of the varia-

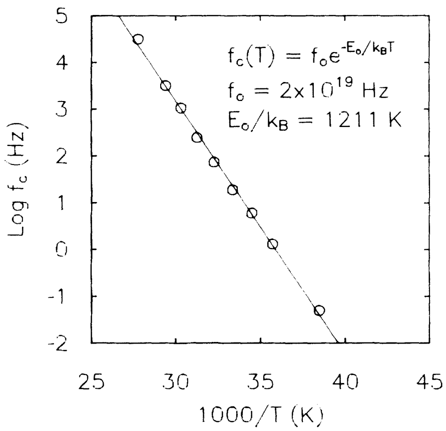

FIG. 3. Plot of $\log f_c$ vs $1/T$ determined by diffusion noise fits. The solid line is a fit with an Arrhenius law with the parameters listed in the figure.

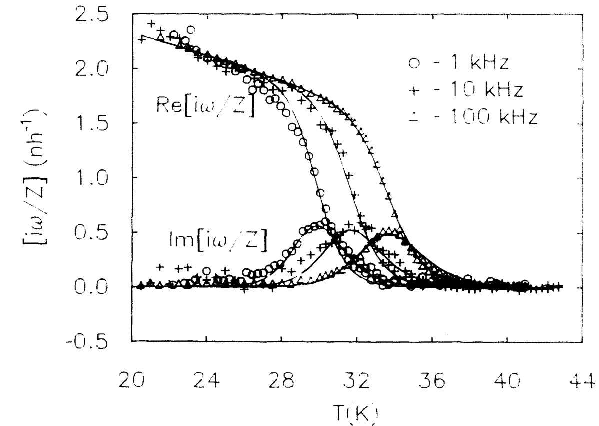

FIG. 4. Real and imaginary parts of $i\omega/Z(\omega,T)$ vs f and T. Solid lines are fits to the data using $f_c(T)$ from the noise spectrum and assuming $\lambda(T)=\lambda(0)/[1-(T/T_{c0})^2]$ with $\lambda(0)=935$ nm and $T_{c0}=65.4$ K.

tion in $f_c(T)$ can be ascribed to an Arrhenius form for $D(T)$. As has been pointed out by Fisher [20] the introduction of a high density of vortex pinning centers with pinning energy $E(T)$ leads to a vortex diffusion coefficient, $D=D_0e^{-E(T)/k_BT}$, where D_0 is the free vortex diffusion coefficient, and the Boltzmann factor represents that fraction of the vortices free to move.

A simple model for $E(T)$ based on pinning of the vortex core explains our data well: By placing the core on a previously existing defect which affects a volume, V_c, a free energy savings occurs of roughly $H_c^2V_c/2\mu_0$, where H_c is the thermodynamic critical field. For line defects running the full thickness of the film, $V_c\approx\pi\xi^2d$. With $H_c=\phi_0/2\sqrt{2}\pi\lambda\xi$ and using the nearly linear temperature dependence of λ^{-2} obtained near T_{c0}, we find $E(T)\approx\phi_0^2L_k^{-1}(0)(1-T/T_{c0})$. This result leads to a diffusion coefficient $D(T)=D_0\exp(E_0/k_BT_{c0})\exp(-E_0/k_BT)$ of the observed Arrhenius form, where $E_0=\phi_0^2/L_k(0)$. Using the extrapolated value of $1/L_k(0)\approx2.8$ nH^{-1} yields $E_0/k_B\approx800$ K in quite good agreement with experiment considering the simplicity of the model. We have verified the predicted scaling of E_0 with $1/L_k$ for 1, 1.5, and 2 unit cell 2:2:1:2 films to date. As all these films have nearly the same value of $\lambda\approx1$ μm, our result for E_0 can be written as E_0/k_B(K) $\approx260d$(nm). Recently, Brunner *et al.* [21] found a magnetic-field-dependent pinning energy for very thin YBCO which extrapolates to zero field result of E_0/k_B(K) $=450d$(nm) very similar to our result for thin 2:2:1:2. Thus, core pinning effects may be general for few unit cell films of the cuprate superconductors.

Given S_ϕ, we can now *quantitatively* understand the mutual inductance measurements within the same vortex-antivortex hypothesis. Several groups have shown [12,14,15] that the largely inductive response expected for a bulk superconductor, i.e., $Z(\omega,T)=i\omega L_{k0}(T)$, where $L_{k0}(T)$ is the kinetic inductance in the absence of 2D fluctuation effects, is modified by vortex-antivortex pairs to be $Z(\omega,T)=i\omega\epsilon(\omega,T)L_{k0}(T)$, where $\epsilon(\omega,T)$ is a complex response function for pair motion. Minnhagen [15] has shown that in the case where pair motion is purely diffusive, ϵ is a function only of $Y(f,T)=f_c(T)/f$, i.e., ϵ is directly related to the *same characteristic vortex response frequency, f_c, as is measured in the noise spectrum.* This result is a direct consequence of the fluctuation-dissipation theorem as applied to the vortex-antivortex system.

Figure 4 shows both the real and imaginary components of $i\omega/Z(\omega,T)$. The solid lines are fits to the data using only the Minnhagen response function, the temperature-dependent f_c already determined from the noise spectra, and the additional assumption that L_{k0} is well described by using $\lambda(T)=\lambda(0)/[1-(T/T_{c0})^2]$ with $\lambda(0)=935$ nm and $T_{c0}=65.4$ K. This $\lambda(T)$ occurs for superconductors with a high density of elastically pinned vortices [22] and thus represents the most natural description for $L_{k0}(T)$ in these films where most of the vortices and antivortices are pinned and only a Boltzmann distribution is free to move. We find that $\lambda(0)$ varies by roughly 20% for all the films measured to date, while T_{c0} ranges from the 65.4 K of Fig. 4 up to nearly 85 K depending on film. These values for T_{c0} are in good qualitative agreement with those from resistivity measurements of 2:2:1:2-2:2:0:1 superlattices [9] and for our own films. Unfortunately, parallel conduction in the 2:2:0:1 layers prevents our utilizing a standard Azlamozov-Larkin fit to the resistivity for a quantitative cross check on T_{c0}. $\lambda(0)$ is directly related to the density of pinned vortices, N_d, by [22] $\lambda(0)=4(\pi N_d)^{1/2}\xi(0)\lambda_L(0)$, where λ_L is the London penetration depth. Since we assume the entire high-temperature vortex-antivortex system becomes pinned as we cool, $N_d\approx\xi_+^{-2}(30\text{ K})$. Using the relation [12,15] between ξ_+ and $\xi(0)$, we find $\lambda(0)\approx2.5\lambda_L(0)$ or $\lambda_L(0)\approx400$ nm for our films. This value is close to the bulk result [23].

The quality of the fit shows that the electromagnetic response of these single unit cell 2:2:1:2 films is well described by hypothesizing a traditional 2D vortex-antivortex gas which moves diffusively and fluctuates according to a Brownian motion picture. However, a high density of a single type of vortex pinning site causes $D(T)$ to follow an Arrhenius behavior. Therefore, the observed increase in L_k^{-1} arises predominantly from a kinetic effect, indicative of increased vortex viscosity, rather than from vortex-antivortex pair binding as happens for a simple KT transition. The question remains whether the vortex pins arise from a well-defined omnipresent defect or whether, perhaps, they are intrinsic to the single unit cell in 2:2:1:2. The enormous density of pins and the extraordinary fact that only one type seems to exist suggest the latter [24], but the issue remains unresolved.

Finally, we note that the particular $D(T)$ which we observe results in a superfluid response with scaling behavior which mimics that expected from a KT transition: Consider any arbitrarily defined T_c, based on, e.g., where $L_k^{-1}(T)$ rises above the system noise floor. This T_c actually only implies that $D(T)$ has crossed some system-dependent value D_c. Setting $D(T_c)=D_c$ and using the Arrhenius law yields a relation between T_c and E_0 of the form $k_B T_c \approx C\phi_0^2/L_k(0)$, where C is nearly constant, depending only logarithmically on D_c. This is to be compared with the KT universal superfluid jump relation [12-15] $k_B T_c = C_0\phi_0^2/L_k(T_c)$, where $C_0 = 1/16\pi^2$. Both equations will display the same scaling of T_c with, e.g., film thickness and could easily lead to the identification of a KT transition where none exists. Rather, the specific temperature dependence observed for L_k and S_ϕ allows us to determine that for single unit cell 2:2:1:2, 2D vortex-antivortex excitations do indeed exist, but that it is the vortex-pin interaction, as opposed to the vortex-antivortex interaction, which suppresses free vortex motion and allows robust superfluid response at low temperatures.

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