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Investigation of ac screening response in the mixed state of Bi₂Sr₂CaCu₂O₈ single crystals

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We have measured the ac screening response and resistance of superconducting $Bi_2Sr_2CaCu_2O_8$ (BSCCO) crystals versus frequency, temperature, and applied dc field. Within the examined field and temperature range, we find the systematic behavior of the observed dissipation peak to be consistent with electromagnetic skin size effects rather than a phase transition. However, a clear contradiction is found between the observed screening response and the response time implied by thermal activation models on the basis of the measured resistance, which raises important questions for the theoretical treatment of the vortex dynamics. We note that a partial resolution of this paradox points to a distribution of pinning energies within the sample.

The mixed state of the oxide superconductors has been the subject of considerable recent study. The combination of a short coherence length, high transition temperature, and anisotropy leads to many interesting predictions for their magnetic properties. The effects of anisotropy, disorder, and reduced dimensionality should be observable in the vortex behavior and new phase transitions have been predicted. The $Bi_2Sr_2CaCu_2O_8$ (BSCCO) system is of particular interest due to its large anisotropy. A detailed understanding of its magnetic properties involves fundamental questions and is essential for many proposed applications.

One measurement that has been widely used to study the magnetic properties of the high-temperature superconductors is the ac screening response. A small ac field is used as a probe and the response is detected inductively. The response can be studied as a function of temperature and external dc magnetic field. It is often assumed that the screening signal is a bulk response, and is thus identified as the ac susceptibility $(\chi = \chi' + i\chi'')$. On cooling, the *apparent* susceptibility shows a step in the real part and a peak in the imaginary part. Much controversy surrounds the origin of this dissipation peak. It was first proposed that it indicated the onset of superconductivity.¹ Later comparison with hysteresis observed in the dc magnetization suggested identification with an "irreversibility line."² Others have considered the vortex dynamics, such as the thermally assisted flux flow³ (TAFF) and critical state⁴ models. These involve microscopic details of the vortex behavior and possess empirical parameters. Similar behavior was observed in mechanical oscillator studies, and was interpreted as a vortex melting transition.^{5,6}

Detailed calculations of the linear response of superconductors to an ac field have recently appeared.⁷⁻⁹ The complex ac penetration depth and susceptibility were shown to depend on the measurement frequency ω relative to the (intrawell) relaxation rate of elastically pinned vortices τ_0^{-1} and the thermally activated (interwell) vortex depinning rate τ^{-1} .

However, it was recently suggested 10 that the screening behavior arises simply from skin size effects within a material of finite resistance. The dissipation peak occurs when the electromagnetic penetration depth δ becomes comparable to the sample size L, yielding a simple relation between the frequency $v = \omega/2\pi$ and dc resistivity ρ at the peak independent of the microscopic details of the vortex behavior. The screening response can only be identified as the bulk susceptibility *above* the peak temperature. Below the peak, the response is limited by the skin size effect and thus is *not* a volume response. Although this treatment ignores microscopic details, detailed vortex calculations^{7,8} yield the identical result for $\omega \tau \ll 1$.

In this work we observe strong frequency dispersion, which contradicts the identification of the dissipation peak as a vortex melting transition within the measured temperature and field range. Instead, both the dependence of the peak and the detailed screening response appear consistent with the skin size effect. However, comparison with thermal activation models which assume a *single* activation energy leads to a serious inconsistency regarding the vortex response time implied by the ac screening response *vis*-à-*vis* that obtained from the resistivity. We believe that at least a partial resolution of this paradox requires the consideration of a *distribution* of pinning energies within the sample, with the imaginary screening response dominated by vortices with response times $\tau \ll \omega^{-1}$.

Measurements were made using a first-order magnetic gradiometer, consisting of two pick-up coils located concentrically within a solenoid that was driven by a sinusoidal current whose frequency and amplitude could be varied. A key feature is the ability to vary the measurement frequency over a wide range. Single crystals of BSCCO $(1 \times 1 \times 0.3 \text{ mm}^3)$ were placed within one coil. The responses from the coils were amplified, connected in opposition, and the in- and out-of-phase components of the output signal were detected. Helium gas flowed over the magnetometer at a controlled temperature, which was reduced at a uniform rate of 0.4 K/min in a constant dc magnetic field. The dc field was produced by a separate electromagnet. In these measurements both ac and dc fields were parallel to the crystalline \hat{c} axis.

Our ac screening response displayed the characteristic steplike transition in the real part (μ') of the output signal, and a dissipation peak in the imaginary part (μ'') . Examination of all the data showed that the ratio of the

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peak in μ'' to the step in μ' was essentially constant and equal to 0.40 ± 0.05 . This has important ramifications, which we return to later.

The issue of amplitude dependence in the screening response has been widely discussed. Some authors found no dependence^{3,11,12} while others observed a large depression of the response curve for increased driving amplitude. $^{13-15}$ Our measurements were made with a driving field of ≈ 5 mOe, and appear to be in the linear-response regime. No amplitude dependence in the position of the dissipation peak was observed for such low driving fields. However, at substantially larger driving fields (≥ 1 Oe) the peak did shift to lower temperature, signalling nonlinear response. Similar behavior was also observed with mechanical oscillators, where the oscillation amplitude was kept small (≤ 150 nm) to ensure linear response.^{5,6} For our data, a simple estimate of the vortex displacement near the sample edge gives a distance of order 10 nm for diffusive motion.

Figure 1 shows the dissipation peak temperature T_P obtained as a function of different ac drive frequencies v. Note the strong frequency dependence for the magnetic fields studied, with significant shifts of T_P even at low frequencies.

A small flake of the crystal used for the ac screening measurements was used for direct four-point resistance measurements in field. The temperature dependence of the resistance in three different values of dc magnetic field is shown on an Arrhenius plot in Fig. 2. The pinning energy U_0 obtained from the slope lies in the range 600-1200 K for the field range examined, commensurate with other reported values.¹⁶

For constant values of the dc field, the frequency of the dissipation peak and the directly measured in-plane resistivity ρ at the same temperature were used to calculate the ratio of the sample size L to the electromagnetic skin depth δ , where $\delta = [(2/\mu_0)(\rho/\omega)]^{1/2}$. The ratio L/δ calculated from the values of ρ and ω at the dissipation peak



FIG. 1. Drive frequency v and temperature T_P of the dissipation peak shown on an Arrhenius-like plot. The data were taken at four values of the dc magnetic field, with ac and dc fields both parallel to the c axis of the crystal. The dashed lines are fits to the data.



FIG. 2. Arrhenius plot of the directly measured resistivity of a crystal for three magnetic-field values. The normal-state resistivity ρ_N (100 K) was 600 $\mu \Omega$ cm. The squares, diamonds, and circles show the resistivity inferred from ac screening response measurements in fields of 400, 1600, and 3715 Oe, respectively, according to skin size effects. A *single* value for the ratio L/δ of 1.9 was used for *all* the screening response data.

was found to be 1.9 ± 0.2 for magnetic fields of 400, 1600, and 3715 Oe. Using the single criterion of $L/\delta = 1.9$, the corresponding resistance values were calculated for the ac screening response data from Fig. 1. These points are shown in Fig. 2 for comparison with the directly measured resistance. There is clearly good agreement between the derived and measured values, with the screening data allowing extrapolation to lower temperatures where the resistance became too low to be measured by direct means. We note the somewhat steeper slope for the 400 Oe screening response data compared to the measured resistivity, although there is still good agreement with the value of $L/\delta = 1.9$.

According to the skin size effect hypothesis, the dissipation peak occurs when the electromagnetic penetration depth is of the order of the sample size. Thus the ratio of skin depth to sample size obtained in this experiment is in good agreement with this explanation over a substantial range, with a geometrical factor of order unity. (For a given geometry there will be a factor relating sample size and skin depth at T_P , calculated to be 2.25 for a slab geometry with the ac field perpendicular to \hat{c} .¹⁰ This factor likely differs for the ac field parallel to \hat{c} , as in our case.) Note that the frequency dependence of the dissipation peak does *not* arise from strong dispersion of ρ , thus justifying the use of a single resistivity value in the skin depth determination.

The skin size effect also suggests comparison to mechanical oscillator experiments, as sample oscillations within a magnetic field are analogous to the application of an ac field to the sample.⁸ The features observed (a step in μ' and a peak in μ'') are identical to our observations.^{5,6,17} Surprisingly, ultrasonic attenuation measurements show similar behavior.¹⁸

Let us reflect on the results so far. First, as a true phase transition is defined in the zero-frequency limit, our observed strong frequency dispersion seriously calls into question the identification of a dissipation peak obtained at *fixed* frequency as a phase transition. This contrasts sharply to the interpretation of the high-Q mechanical oscillator results, based upon observations at a single frequency.^{5,6} Furthermore, within the skin depth interpretation our results imply that the resistance is finite and does not markedly deviate from the extrapolated approximately activated behavior. Our observed finite resistance also contradicts a transition to a vortex glass or Bose glass phase.¹⁹ Finally, the observed "peak-to-step ratio" (ratio of the peak in the imaginary part to the step in the real part) is larger than that expected for the nonlinearresistance vortex glass phase.⁹ Thus, within our measured range, we observe no evidence of any phase transition which would manifest itself in the resistance behavior. Of course, this does not rule out transitions at lower temperatures and/or higher fields.

It can be seen in Figs. 1 and 2 that both the dissipation peak frequency and the resistance show approximately exponential dependences in (1/T). The graphs show similar behavior, emphasizing the connection between the two measurements that was formally expressed in the treatment of the skin depth δ . For both cases, exponential behavior has been interpreted as thermal activation of vortices across pinning barriers. ^{16,20}

This dependence motivates more detailed correlation of the resistance to our ac screening response within thermal activation models, i.e., to better connect the macroscopic response to microscopic parameters. The ac response has been calculated in three regimes,^{7,8} assuming a single activation energy U_0 . In the high-frequency regime ($\omega \tau_0 \gg 1$) vortices oscillate within the pinning potential wells. In the intermediate regime ($\tau^{-1} \ll \omega \ll \tau_0^{-1}$) the magnetic permeability is predominantly real with the peak-tostep ratio $\propto (\omega \tau)^{-1}$. Additionally, the penetration depth for the ac field $\lambda_{ac} = (\lambda^2 + \delta^2 \omega \tau/2)^{1/2} \cong \delta(\omega \tau/2)^{1/2} \gg \delta$, as the superconducting magnetic penetration depth λ is smaller than δ for our case.

For $\omega \tau \ll 1$, the vortex motion consists of thermally activated hopping between pinning wells. The vortex response is diffusive, with diffusivity $D = \rho_{TAFF}/\mu_0$ where ρ_{TAFF} is the thermally activated flux flow resistivity. It is this regime which yields the results of Ref. 10, with a penetration depth $\lambda_{ac} = 2^{-1/2}\delta$ and the ac linear response set by the skin size effect (with $\rho = \rho_{TAFF}$). The peak-tostep ratio is *independent* of ω and equal to ≈ 0.4 . Recall that the observed dependence of the dissipation peak was consistent with the skin size effect with field penetration set by δ , and the observed peak-to-step ratio was 0.40 ± 0.05 for all data. Hence, our observed ac screening response argues strongly that we are in the $\omega \tau \ll 1$ regime.

According to these models the depinning time τ can be obtained from the relaxation time τ_0 via $\tau \approx \tau_0 \exp(U_0/kT)$, assuming a *single* value for U_0 as obtained from the resistance. Values of τ were calculated using the U_0 obtained from the measured resistance, assuming an accepted value for τ_0 of order 10^{-10} sec. For the screening measurements described here, we thus obtain $4 \le \omega \tau \le 10^3$. This stands in complete contradiction with the detailed behavior of the ac screening response, and is not readily resolved by a physically meaningful choice for the time τ_0 .

Consequently, we face the following paradox. Detailed examination of the ac response is consistent with the regime $\omega \tau \ll 1$. However, the activation energy U_0 as obtained from the resistivity points to the regime $\omega \tau \gg 1$, where one expects a reduced imaginary response and field penetration on the scale of $\lambda_{ac} \gg \delta$. The implications of this paradox run deep. In particular, our skin depth δ was calculated from this very same resistivity, and consequently our ratio $L/\delta \approx 1.9$ at the peak reflects U_0 .

We believe that the physically reasonable allowance for a distribution in activation energies within the crystal at least partly resolves this paradox. Such a distribution was previously observed in flux noise measurements of individual vortices.²¹ For a given experiment, the observed behavior would be determined by those vortices which dominate the appropriate response. For our case, the ac screening response might be dominated by those vortices with $\omega \tau \ll 1$, which provide the largest contribution to the imaginary part of the response and their shorter $\lambda_{ac} \approx \delta$ may set the field decay length. However, this simple treatment does not address the important question of why the resistivity displays the higher activation energy U_0 . Note that we have only considered the energy distribution, and have not addressed the important spatial components which are intertwined with the dynamics.

We take a model-independent route to infer the width of the distribution. To explain the data, there must be sufficient density of low pinning energies within the distribution in order to yield a diffusive response. An upper bound on the pinning energies probed by the ac screening measurements can be simply obtained from the condition $\omega \tau \lesssim 0.1$, where μ'' beings to roll off as a function of $\omega \tau$.⁸ This criterion implies that the imaginary part of the screening response is determined by a range of pinning energies U less than 440-730 K for our data, i.e., below an appreciable fraction of U_0 .

Thus we see that due to the exponential dependence of τ upon U, the required distribution width is actually rather modest. However, it is important to note that even a modest distribution appears sufficient to invalidate any simple classification of the sample response into *either* intrawell or interwell motion.

In conclusion, we have shown that the strong frequency dependence of the peak in the imaginary part of the ac screening response for BSCCO argues strongly against a vortex phase transition in the examined temperature and magnetic-field range. Detailed comparison of ac screening response and direct resistance measurements is consistent with the behavior being due to electromagnetic skin size effects. However, the vortex diffusion, as measured by the ac screening response, is inconsistent with the behavior predicted by thermal activation using a single pinning energy as derived from the resistance. We point out that at least one part of this paradox can be solved by the inclusion of a distribution of pinning energies within the material. More theoretical consideration of the consequences of such a distribution upon the vortex dynamics is necessary.

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- ¹T. K. Worthington, W. J. Gallagher, D. L. Kaiser, F. H. Holzberg, and T. R. Dinger, Physica C **153–155**, 32 (1988).
- ²K. Heine, J. Tenbrink, and M. Thöner, Appl. Phys. Lett. 55, 2441 (1989).
- ³J. van den Berg, C. J. van der Beek, P. H. Kes, J. A. Mydosh, M. J. V. Menken, and A. A. Menovsky, Supercond. Sci. Technol. 1, 249 (1989).
- ⁴J. Z. Sun, M. J. Scharen, L. C. Bourne, and J. R. Schrieffer, Phys. Rev. B **44**, 5275 (1991).
- ⁵P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. J. Bishop, Phys. Rev. Lett. **61**, 1666 (1988).
- ⁶C. Duran, J. Yazyi, F. de la Cruz, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. B 44, 7737 (1991).
- ⁷M. W. Coffey and J. R. Clem, Phys. Rev. Lett. **67**, 386 (1991); and (unpublished).
- ⁸E. H. Brandt, Phys. Rev. Lett. **67**, 2219 (1991). For $\omega \ll \tau^{-1}$ and $d \gg \lambda$, the results [e.g., Eq. (18)] reduce to those of Ref. 10 [Eq. (7)].
- ⁹A. E. Koshelev and V. M. Vinokur, Physica C 173, 465 (1991); and (private communication).
- ¹⁰V. B. Geshkenbein, V. M. Vinokur, and R. Fehrenbacher, Phys. Rev. B 43, 3748 (1991).
- ¹¹D. G. Steel and J. M. Graybeal, Physica C **185–189**, 2227 (1991).
- ¹²P. H. Kes, J. Aarts, J. van den Berg, C. J. van der Beek, and J. A. Mydosh, Supercond. Sci. Technol. 1, 242 (1989).
- ¹³J. H. P. M. Emmen, G. M. Stollman, and W. J. M. De Jonge,

Physica C 169, 418 (1990). Their data show the same inconsistency of time scales shown here.

- ¹⁴Ch. Heinzel, Ch. Neumann, and P. Ziemann, Europhys. Lett. 13, 531 (1990).
- ¹⁵C. J. van der Beek, M. Essers, P. H. Kes, M. J. V. Menken, and A. A. Menovsky, Supercond. Sci. Technol. 5, S260 (1992).
- ¹⁶T. T. M. Palstra, B. Batlogg, R. B. van Dover, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. B 41, 6621 (1990).
- ¹⁷A. Gupta, P. Esquinazi, H. F. Braun, and H.-W. Neumüller, Phys. Rev. Lett. 63, 1869 (1989).
- ¹⁸J. Pankert, G. Marbach, A. Comberg, P. Lemmens, P. Fröning, and S. Ewert, Phys. Rev. Lett. **65**, 3052 (1990). Simple manipulation of their TAFF expression for the attenuation peak temperature eliminates model parameters, yielding a similar relation for δ with the wavelength identified as the sample size. Such correspondence suggests induced electromagnetic fields may play a role in the presence of mobile vortices.
- ¹⁹D. R. Nelson and V. M. Vinokur (unpublished).
- ²⁰M. Inui, P. B. Littlewood, and S. N. Coppersmith, Phys. Rev. Lett. **63**, 2421 (1989).
- ²¹M. J. Ferrari, M. Johnson, F. C. Wellstood, J. Clarke, D. Mitzi, P. A. Rosenthal, C. B. Eom, T. H. Geballe, A. Kapitulnik, and M. R. Beasley, Phys. Rev. Lett. 64, 72 (1990).