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Nonlinear optical response of granular Y-Ba-Cu-O films

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The nonlinear optical response of a highly granular film of $YBa_{2.1}Cu_{3.4}O_{7-x}$ has been interpreted on the basis of the two-dimensional characteristics of the superconducting transition in this film. The behavior of the current-voltage characteristics is consistent with a dissipation mechanism which is dominated by vortex motion as in a Kosterlitz-Thouless transition. The optical response at temperatures near the transition to zero film resistance cannot be interpreted satisfactorily in terms of a realistic thermal model. We believe that nonequilibrium effects contribute to the photoresponse, particularly at low bias currents and low temperatures.

There has been recent interest[1-4] in the photoresponse of high-T_c superconducting films, especially with regard to their possible use as infrared sensors. However, the details of the physics of the photoresponse have not been very well established. This report presents evidence that very granular films of Y-Ba-Cu-O can be described by a quasi-two-dimensional (2D) model,[5-7] where the transition to the zero-resistance state and the dynamics of the photoresponse are dominated by vortex motion.

The photoresponse of a film of Y-Ba-Cu-O (i.e., the increase in voltage induced by visible light) has been measured and correlated with the temperature dependence of the resistance (R), dR/dT, and the current-voltage (I-V) characteristics. We have found that in the transition region, where R and dR/dT are vanishing, the photoresponse (PR) becomes a strong nonlinear function of current, in contrast to the linear dependence observed in the more resistive part of the transition at higher temperatures. There are two distinct transition temperatures for a granular film: T_{c0}, the superconducting transition temperature of the Y-Ba-Cu-O grains, and T_c, the temperature where the entire film resistance $R \rightarrow 0$. In the region around T_c the behavior of the I-V curves of the Y-Ba-Cu-O film are nearly identical to those which have been observed previously[7] for two-dimensional, highly granular, NbN/BN films. In the latter case the transition to the zero-resistance state was topological, involving the condensation of vortex-antivortex pairs.[5] The PR of the NbN/BN films also depended[3] nonlinearly on bias current, very much as is reported here for Y-Ba-Cu-O. We propose that the nonlinear response observed for Y-Ba-Cu-O is due to a quasi-two-dimensional granular microstructure in the Y-Ba-Cu-O films. This conclusion is consistent with recent kinetic inductance measurements[8] in which the Kosterlitz-Thouless (KT) transition[9] was observed for a high-quality film of Y-Ba-Cu-O, indicating 2D superconducting behavior.

The Y-Ba-Cu-O film was prepared by radio-frequency magnetron sputtering deposition onto a MgO substrate using a $YBa_2Cu_3O_{7-x}$ target. Elastic backscattering analysis data indicated a spatially averaged film composition of $YBa_{2.1}Cu_{3.4}O_{7-x}$. After oxygen annealing (900 °C, 10 min; 450 °C, 5 h) the 2500-Å-thick film had a metallic black color, but weakly transmitted visible light. (Optical measurements on 2000-Å-thick films indicated[10] that for light energies near 2 eV, transmittance and reflectance are each about 10%; hence, most of the light is absorbed in the film.) The film was scribed to allow four-probe resistance measurements. Electrical contact was made through evaporated Ag pads. The conductivity at high temperatures was weakly activated, with $T_{c0} \sim 82$ K and $T_c \sim 11$ K—such features are characteristic of granular films. The PR measurements were carried out in a variable temperature Dewar with optical access. Light sources were a 1-mW HeNe laser (632.8 nm), which was chopped from 1 Hz to 20 kHz, and an attenuated pulsed dye laser (584 nm) with an 8-ns pulse duration. The optical beams were weakly focused to a 2 mm diameter—the width of our device.

It has been previously demonstrated[7] that the transition to the superconducting state of thin granular NbN/BN films is very well described by a vortex-antivortex pairing transition[5,6] of the type described by Kosterlitz and Thouless.[9] This transition exhibits the following temperature dependence of the I-V characteristics:[5-7]

$$T \ll T_c \begin{cases} V = 0 \quad \text{for } I < I_c \\ V \sim (I - I_c)^n, \; n > 3, \quad \text{for } I > I_c \end{cases} \tag{1a}$$

$$T = T_c, \quad V \sim I^3 , \tag{1b}$$

$$T > T_c, \quad V \sim I \text{ for small } I . \tag{1c}$$

Thus, there is a dramatic change in the current-voltage characteristics in a narrow temperature range near T_c. Figure 1 shows I-V characteristics (measured in the dark) of the granular Y-Ba-Cu-O film in the temperature interval around T_c, where the resistance vanishes for low bias currents ($\sim 20\ \mu$A). The behavior of this film is very much as predicted by Eqs. (1a)–(1c): It exhibits a distinct critical current at low temperature (consistent with a flux pinning mechanism), the I-V curves follow a power law which is cubic over a reasonable current interval for $T_c \sim 11.5$ K, and the I-V curves have a significant linear region for $T > T_c$ for small bias currents (~ 0.2 mA). Therefore, on the basis of the I-V behavior, the transition to the superconducting state for this granular film is consistent with a vortex pairing interaction.

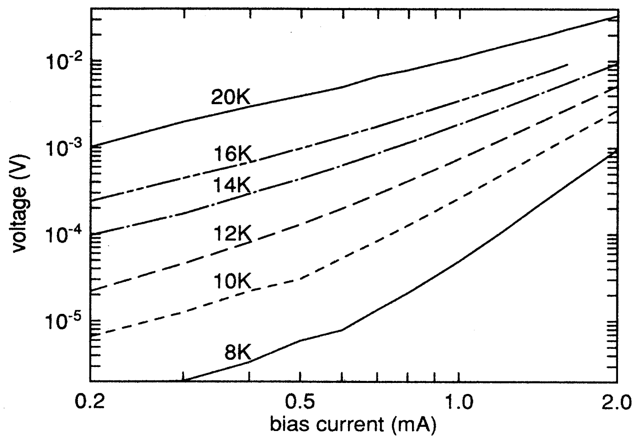

FIG. 1. Film voltage vs bias current measured for six temperatures with no visible light incident on sample.

A further test of the 2D nature of the transition observed for the above Y-Ba-Cu-O film is to compare the measured resistance for very low bias currents with the theoretically predicted form:[7]

$$R/R_{\max}=a\exp[-2(b/\tau)^{1/2}]\,, \quad (2)$$

where $\tau=(T-T_c)/(T_{c0}-T)$. In this relation a and b are constants, and $R_{\max}$ is the maximum film resistance for T just above T_{c0}. Figure 2 shows measurements of the resistance of the Y-Ba-Cu-O film made as functions of temperature. A plot of $\log_{10}(R/R_{\max})$ vs $\tau^{-1/2}$ yields a straight line for $T_c=10.5\pm1$ K. The appropriate values for the constants in Eq. (2) are $a=3.2$ and $b=0.8$ for $T_c=10.5$ K, which in turn yield a vortex dielectric constant[7] $\epsilon_c\sim2.3$. Slightly larger values of T_c ($\sim$11.0 K), which are more in agreement with our I-V characteristics, yield larger values of ϵ_c, consistent with the value of $\epsilon_c=4.6$ deduced by Fiory *et al.*[8] For a film which exhibits 2D character, it is also expected that a weak magnetic field applied perpendicular to the film surface will have a significantly larger effect on the resistance than an in-plane applied field. This is in fact observed, as shown in the inset of Fig. 2. Our results are consistent with the resistance in these films, especially near T_c, being due to vortex dissipation as has been proposed by Fiory *et al.*[8] to explain the shape of the transition of a thin ($\sim$500 Å) high-quality Y-Ba-Cu-O film. We observe 2D behavior in a thicker (2500 Å) granular film. Because the granularity of the film results in a broader transition, the characteristic dependences of voltage on current and resistance on temperature can be measured over a broader temperature range.

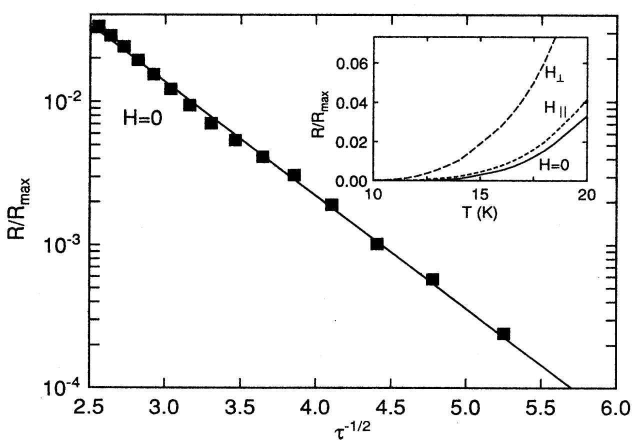

FIG. 2. Plot of $R/R_{\max}$ vs $\tau=(T-T_c)/(T_{c0}-T)$, where R is film resistance and $R_{\max}$ is the maximum film resistance just above the superconducting transition temperature T_{c0} of the Y-Ba-Cu-O grains. T_c is the temperature where R vanishes for small bias currents. Bias current of 20 μA used in measurement (solid squares). Solid line is a fit to Eq. (2). Inset shows same data ($H=0$), as well as additional measurements with 580-G field applied parallel and perpendicular to the film plane.

The response to optical radiation of the granular Y-Ba-Cu-O film is shown in Fig. 3. These data were obtained using a 1-mW HeNe laser at a chopping rate of 3 kHz at four different bias currents. The film resistance R was measured simultaneously with the PR. The dotted curve in Fig. 3 is dR/dT for a 2-mA bias current. The peak in dR/dT near 80 K is identified with T_{c0}. The small peak near 60 K may be due to the presence of a second superconducting phase. The PR generally reflects the shape of the three highest dR/dT peaks with a linear dependence on bias current. The most striking feature of the data is the appearance of a peak at the lowest temperatures near T_c, where dR/dT is rapidly approaching zero. The intensity of this peak increases approximately quadratically with bias current, which is readily explained by the 2D character of the granular film. From Eq. (1b) $V\sim I^3$, and therefore the PR $\sim\delta V/\delta I\sim I^2$ for $T\sim T_c$.

The observed temperature dependence of the PR may be due to a variety of processes, including the heating of quasiparticles, electron pair breaking, and the breaking of

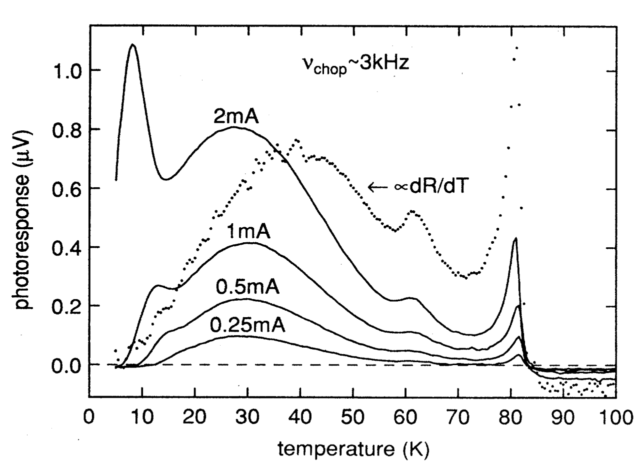

FIG. 3. Photoresponse of film to 6328-Å light chopped at 3 kHz for four bias currents. Dotted curve is dR/dT calculated from film resistance R for 2-mA bias current. Photoresponse and R were measured simultaneously.

vortex-antivortex pairs. At sufficiently high temperatures, $T_{c0} > T \gg T_c$, a significant population of these excitations will be thermally unbound, so that light absorption is most likely dominated by direct heating of the film. At lower temperatures $T \sim T_c$ depairing processes are more likely to occur. However, all such processes will eventually lead to phonon generation and hence a thermal model must be included in the analysis of the data. In a purely thermal model[11] the PR can be expressed as $\delta V = I(dR/dT)\delta T$, where δT is the temperature increase of the film; our measurements of δV and $R(T)$ imply that $\delta T \sim 1$ mK for steady-state laser excitation and δT is ~ 2 K for pulsed laser excitation. We have measured in detail the dependence of the PR on light chopping frequency from 1 Hz to 20 kHz. We find that the data shown in Fig. 3 for $T > 20$ K are well described by the thermal model, where δT is dominated by the thermal conductivity of the MgO substrate. On the other hand, the thermal model cannot describe the strong PR peak observed near the KT transition (~ 11 K). The thermal model is also shown to fail for the 8-ns pulsed light excitation, for which we observe only the peak near the KT transition, but none of the bolometric features at higher temperatures. In this faster time regime the temperature rise δT is inversely proportional to the thermal conductance K of the Y-Ba-Cu-O film MgO interface.[11,12] Using our measured values of dR/dT and the predicted T^3 dependence of K in this low-temperature regime, we are unable to account for the PR peak observed near the KT transition on the basis of a bolometric interpretation.

We conclude from the above considerations that the PR peak observed near the KT transition, for light chopped or pulsed on time scales ranging from 1 to 10^{-8} s, is due in part to nonequilibrium effects associated with an optically induced nonthermal distribution of unpaired electrons and/or unbound vortices. This conclusion is supported by the observed dependence of the pulsed PR on bias current, which is shown in Fig. 4 for three temperatures. The inset shows schematically the time dependence. The rise time of the PR was comparable to that of the incident light pulse. In order to examine the decay of the PR, two time windows have been chosen. The solid (dashed) lines in Fig. 4 reflect the average response over the 0–40 ns (40–70 ns) time window, respectively. The relative magnitudes of the two time window components are indicated by the normalization factors in Fig. 4. Several important features are observed for these data. Well into the resistive region, for $T = 20$ K the observed PR depends linearly on current and the PR line shape is independent of current, as expected for a bolometric mechanism. For $T < T_c$ the PR depends nonlinearly on current. For $T = 5$ K there is a critical current (0.5 mA) below which there is no long-time (40–70 ns) component. At $T = 11$ K this critical current approaches zero. The long-time component can be interpreted as due to "excess heating" of the film to a temperature greater than T_c. These observations are consistent with nonequilibrium effects associated with creating a nonthermal distribution of quasiparticles that can be described to first order by an effective temperature T^*, which is higher than the temperature that the same input optical power would produce if the electrons and the lattice were in thermal equilibrium.[13]

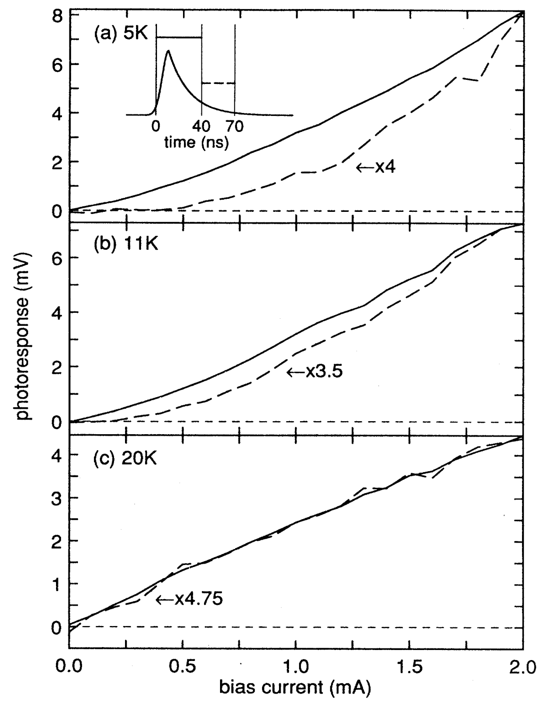

FIG. 4. Pulsed photoresponse of film vs bias current for two time windows and three temperatures. Long-time components have been normalized by indicated multiplying factors. Laser wavelength, 584 nm; pulse energy, 2 μJ.

In summary, we have presented evidence that the nonlinear current dependence of the response to light observed near the transition to zero resistance of a highly granular film of Y-Ba-Cu-O can be interpreted in terms of a quasi-two-dimensional phase-transition model, where the dissipation mechanism is dominated by vortex motion. The contributions of nonequilibrium effects to the low-temperature photoresponse have been considered.

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