Temperature and laser power dependence of femtosecond optical pair-breaking rate in optimally doped $YBa_2Cu_3O_{7-\delta}$ epitaxial thin films

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The temperature and laser power dependence of the femtosecond optical pair-breaking rate in optimally doped $Y_1Ba_2Cu_3O_{7-\delta}$ epitaxial thin films are studied. The temperature dependence is consistent with numerical simulations using the kinetic inductance model and the Rothwarf-Taylor theory. The laser power dependence shows strong nonlinear behavior. The magnitude of the nonlinearity, and its significant and systematic temperature and photon energy dependence, cannot be accounted for by the kinetic inductance model and the Rothwarf-Taylor theory, demonstrating the complexity of nonequilibrium superconductivity in high-temperature superconductors.

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Investigations of the ultrafast optical response of hightemperature superconducting thin films have attracted considerable attention ever since high-temperature superconductivity was discovered in 1986.¹ These studies were motivated by the practical interest in developing ultrafast, ultrasensitive, and broadband photodetectors as well as the physics interest in the exploration of nonequilibrium dynamics in strongly correlated electron systems in search of the fundamental mechanism of high-temperature superconductivity. To this end, two experimental methods have been generally employed. One is the pump and probe measurement scheme including the electric-optic (EO) sampling technique²⁻⁶ while the other is the transient photoimpedance response measurement.⁷⁻¹⁰

The pump and probe measurement scheme has the advantage of high temporal resolution and can provide important information about the nonequilibrium guasiparticle dynamics of high-temperature superconductors in the time domain. Using femtosecond pulsed optical radiation and a subpicosecond EO sampling system, electrical transients of singlepicosecond duration from a current biased YBa₂Cu₃O₇₋₈ (YBCO) microbridge have been observed.^{5,6} The nonequilibrium quasiparticle dynamics have been successfully modeled using either the two-temperature model¹¹ for the optical response in the resistive state, or the kinetic inductance model⁷ and the Rothwarf-Taylor theory¹² in the superconducting state. The transient photoimpedance response experiment, despite its lower time resolution, has the advantage of a simpler experimental setup, high signal-to-noise ratio, and, most importantly, direct correlation with electron pairing due to its optical pump and electrical probe measurement scheme. A sharp resonant triplet fine structure was observed in femtosecond optical pair-breaking spectroscopy of optimally doped, underdoped, and Zn-doped YBCO thin films near 1.5 eV⁸, which is consistent with the earlier all-optical measurements.^{4,13–15} It was proposed that the charge transfer excitonic excitation associated with the Cu-O plane might be the origin of this resonant feature. The systematics of the

data strongly suggests the presence of insulating domains even in the superconducting state of the YBCO system over a subpicosecond time scale and is consistent with the elecphase separation model of high-temperature tronic superconductivity.^{16–21} In this article, we present a comprehensive study of laser power and temperature dependence of the femtosecond optical pair-breaking rate in optimally doped YBCO thin films. Upon analysis of the kinetic inductive optical response using the conventional Rothwarf-Taylor theory, it is found that although this simple model can provide adequate description of the temperature dependence measurement, it cannot account for either the strong nonlinear laser power dependence or the systematic variation of the nonlinearity as a function of temperature and photon energy. A modified and improved theory for the nonequilibrium quasiparticle dynamics in YBCO systems is clearly needed.

The optimally doped YBCO thin film was prepared by the pulsed laser deposition technique on a (100)-oriented LaAlO₃ single crystal substrate. The thickness of the film was 100 nm, nearly equal to the penetration depth of the laser beam at wavelength centered around 800 nm.²² The films (with a $T_c \sim 90$ K and $J_c > 10^6$ A/cm² at 77 K) were patterned using standard photolithography to obtain the coplanar waveguide device structure for high-speed electrical signal propagation. The sketch of the experimental setup and the device can be found in our previous paper.⁸ The device was mounted on a cold finger located in a high-vacuum cryogenic system where the substrate temperature can be controlled between 10 and 300 K with ± 0.1 K stability. The dc bias current was 4 mA. The device was illuminated with 100 fs laser pulses at 10 kHz from a Ti:sapphire femtosecond laser system consisting of an argon-ion pumping laser, an oscillator, and a regenerative amplifier. The low repetition rate eliminates accumulation effects of previous pulses and leads to only a fast kinetic inductive optical response with rise and fall times on the order of picoseconds. The corresponding wave forms were monitored by a fast digital sampling oscilloscope with a temporal resolution of ~ 20 ps. The



FIG. 1. A typical wave form of the femtosecond optical response signal for optimally doped YBCO thin film in the superconducting state.

laser beam was focused onto the device by a cylindrical lens, resulting in a spot size of ~ 5 mm by 200 μ m. The wavelength of the laser system was tunable in the range of 750 to 850 nm.

A typical wave form of the femtosecond optical response signal in the superconducting state is shown in Fig. 1. The strongest peak denoted by the fast optical response (FOR) is the primary signal and of main interest in this article. The overall oscillatory features of the transient are characteristic for the nonequilibrium kinetic inductive optical response of superconductors in the superconducting state,^{5,23} with the negative part representing the Cooper pair-breaking process and the positive part corresponding to quasiparticle recombination. According to the kinetic inductance model,^{7,23} when an optically thin superconducting thin film device biased with constant current *I* is illuminated by a laser pulse, the incoming photons break Cooper pairs, leading to an abrupt change in the Cooper pair density n_s and a voltage transient *V* across the device given by

$$V = I \left[-\frac{ml}{2e^2 w d} \right] \frac{1}{n_s^2} \frac{dn_s}{dt},\tag{1}$$

where *m* and *e* are the mass and the charge of an electron, and *l*, *w*, and *d* are the length, width, and thickness of the thin film device. The nonequilibrium dynamics of n_s can be obtained numerically from the Rothwarf-Taylor rate equations¹²

$$-\frac{dn_s}{dt} = i_{qp} - R(n_0 - n_s)^2 + \frac{2}{\tau_b} n_{\omega}, \qquad (2)$$

$$\frac{dn_{\omega}}{dt} = \frac{1}{2}R(n_0 - n_s)^2 - \frac{1}{\tau_b}n_{\omega} - \frac{n_{\omega} - n_{\omega T}}{\tau_{es}},$$
(3)

where⁶ n_0 (~2×10²¹ cm⁻³), n_{ω} , and $n_{\omega T}$ are the number density of all carriers, phonons, and phonons at thermal equilibrium, respectively. Further, i_{qp} is the external quasiparticle generation rate per unit volume, *R* is the recombination rate for quasiparticles back to Cooper pairs, τ_b is the Cooper pair-breaking time by phonons, and τ_{es} is the phonon escaping time. *R* is related to the quasiparticle recombination time τ_r by

$$R = \frac{2}{\tau_r n_{qpT}} \tag{4}$$

where n_{qpT} is the number density of quasiparticles at thermal equilibrium given by²⁴

$$n_{qpT} = n_0 \left(\frac{T}{T_c}\right)^2.$$
(5)

 $n_{\omega T}$ can be obtained by solving the Rothwarf-Taylor equations in the steady state with zero laser power to give

$$n_{wT} = \frac{\tau_b}{\tau_r} n_{qpT}.$$
 (6)

In optimally doped YBCO thin films, both τ_b and τ_r are found to be on the order of a picosecond²⁵ while τ_{es} is on the order of a nanosecond.⁹

It is important to point out that due to the limited temporal resolution of the digital sampling oscilloscope used in this experiment, the signal actually observed on the oscilloscope is related to both the amplitude and the shape of the voltage transient generated across the superconducting device, as well as the impulse response function of the electrical circuit of the oscilloscope. Generally speaking, the duration and the shape of a voltage transient with a time scale shorter than the temporal resolution will not be reproduced faithfully by the oscilloscope. However, it was shown²⁶ that the maximum signal amplitude measured on the oscilloscope (corresponding to the fast optical response signal in Fig. 1) is proportional to the maximum voltage transient, with a constant proportionality factor depending on both the pulse shape of the voltage transient and the impulse response function of the oscilloscope which is invariant under the current experimental condition. In this article, only the laser power and the temperature dependence of the FOR signal are of interest. Therefore, although it is difficult to compare the numerical simulation results of the kinetic inductance model and the Rothwarf-Taylor theory directly to the experimental ones, it is meaningful and instructive to compare the dependence of the FOR signal on the external parameters in these cases.

The Rothwarf-Taylor theory is valid at relatively low temperatures and for weak to moderate external perturbations, i.e., not very far from equilibrium when the percentage of Cooper pair breaking is small. In our case, the fraction of the absorbed optical energy density per pulse is given by $F(1 - R)(1 - e^{-d/\delta})/d$ where *F* is the laser fluence, *R* is the reflectivity, δ is the optical penetration depth, and *d* is the thickness of the film. This is clearly an overestimation, since the superconducting device is assumed to be thermally isolated and the absorption at the window of the Dewar has been ignored. Taking $R \sim 0.1$, $^7 d \sim \delta \sim 100$ nm, 22 a typical laser fluence of $F \sim 10 \mu J/cm^2$, 8 a superconducting gap energy of ~ 20 meV, 27 and assuming that all the absorbed optical energy goes into pair breaking, the total number of Cooper pair broken by a single laser pulse is 8.9×10^{19} cm⁻³, which



FIG. 2. Temperature dependence of the fast optical response for optimally doped YBCO thin film in the superconducting state. The solid line is the numerical simulation based on the kinetic inductance model and is normalized to the experimental data at 80 K.

is less than 20% of the number density of Cooper pairs at 80 K. More importantly, it has been shown that the fraction of the absorbed optical energy that actually goes into pair breaking is very small, on the order of 1% or even less.²⁸ Therefore, in this experiment, the number of Cooper pairs broken by incoming photons is only a very small fraction and the superconducting system is not very far away from equilibrium.

Figure 2 shows the temperature dependence of the FOR signal for an optimally doped YBCO thin film in the superconducting state. The average laser power and the photon energy are kept fixed at 2 mW and 1.54 eV, respectively, throughout the measurement. The amplitude of the FOR signal decreases as the temperature decreases. This can be understood qualitatively by considering Eq. (1). If we assume that dn_s/dt is temperature independent or weakly temperature dependent, the temperature dependence of the FOR signal is determined by $1/n_s^2$. At lower temperatures, n_s is larger, and hence smaller FOR signal results. Quantitatively, Eqs. (1)-(6) can be used to numerically simulate the temperature dependence of the FOR signal, taking into consideration the full temperature dependent dynamics of n_s and dn_s/dt . In this simulation, the incoming laser pulse is assumed to be a Gaussian with pulse width of 100 fs. The Cooper pair-breaking time by phonons τ_b , the quasiparticle recombination time τ_r , and the phonon escaping time τ_{es} are taken to be 1.0 ps, 0.6 ps,²⁵ and 3.5 ns,⁹ respectively. The result is shown as the solid line in Fig. 2 which has been normalized to the experimental data at 80 K. As discussed earlier, this normalization is necessary due to the limited temporal resolution of the oscilloscope and is irrelevant to the physics. The match between the numerical simulation and the experimental data is almost perfect.

Figures 3(a)-3(c) shows the laser power dependence of the fast optical response signal at three different photon energies (a) E=1.62 eV, (b) E=1.54 eV, and (c) E= 1.50 eV, and at three different substrate temperatures (80 K, 60 K, and 20 K), respectively. The signal level for photon energy around 1.62 eV at 20 K is too low to be included here. The laser power dependence of the FOR signal is then fitted to a simple power law as $FOR \propto P^{\alpha}$ where P is the average laser power and α is the exponent. The solid lines are the fitted curves and Table I summarizes the results. Note that these three photon energies correspond to the photon energies of the sharp triplet fine structure [redrawn in Fig. 3(d) for comparison] observed in the photon energy dependence of the FOR signal in an optimally doped YBCO thin film.⁸ The FOR signal is clearly a nonlinear function of the laser power in all cases. As can be seen in Table I, at each photon energy the fitted nonlinear exponent increases as temperature is lowered; at each temperature, the exponent increases as the photon energy decreases. Figure 4 shows the numerical simulation of the laser power dependence of the FOR signal at different temperatures using parameters described earlier. The simulated curves are also fitted to a simple power law and the results are included in Table I.

It can be seen from Table I that according to the kinetic inductance model and the Rothwarf-Taylor theory, the FOR signal is a nonlinear function of the laser power, at least at relatively high temperatures such as 80 K and 60 K. However, the simulated exponents are consistently and significantly smaller than the experimental results at all temperatures. For example, the simulated exponents are ~ 1.44 , \sim 1.32, and \sim 0.92 for T=80 K, 60 K, and 20 K, respectively, compared to ~ 2.13 , ~ 3.31 , and ~ 5.51 observed experimentally for the photon energy of 1.50 eV. Secondly, the exponent of the simulated laser power dependence is smaller at lower temperatures, opposite to what was observed experimentally where the nonlinearity is greatly enhanced. Finally and most importantly, the Rothwarf-Taylor theory does not have significant photon energy dependence at all. As a result, it cannot explain the observed systematic change in the exponents as the photon energy is tuned. Therefore, unlike the case of temperature dependence of the FOR signal, these systematic variations cannot be accounted for by the kinetic inductance model and the Rothwarf-Taylor theory alone. Additional nonlinear effect(s), which is (are) not contained in this simple numerical simulation and is (are) stronger as the temperature and the photon energy are lowered, must be taken into consideration. Here, it is important to point out that the number of Cooper pairs broken by incoming photons in this experiment is only a very small fraction (less than 1%) of the total Cooper pair density as estimated earlier, and the superconducting system is not very far away from equilibrium conditions. Therefore, the interband transitions and associated nonlinear optical effects due to the saturation of free carrier absorption observed in many metallic systems under very high-power laser irradiation²⁹⁻³¹ do not seem to play any role in our experiments.

The nonlinear power dependence of the FOR signal was also observed by Sobolewski *et al.* in optimally doped YBCO thin film using the EO sampling pump-probe technique with subpicosecond resolution.³² There, quasiquadratic laser power dependence was obtained at 77 K at relatively low laser power when only the nonequilibrium kinetic inductive optical response was observed, and it was seen to change into a linear dependence for relatively high laser power when the bolometric optical response was also observed at the



FIG. 3. Laser power dependence of the fast optical response for optimally doped YBCO thin film in the superconducting state at photon energies (a) E = 1.62 eV, (b) E = 1.54 eV, and (c) E = 1.50 eV. The photon energy dependence of the fast optical response for optimally doped YBCO at T = 80 K and P = 1 mW is redrawn in (d) for comparison (Ref. 8).

same time. Such a nonlinear behavior is not surprising at all considering the complexity of the nonequilibrium dynamics, especially in the case of high-temperature superconductivity. The Rothwarf-Taylor theory, developed in the 1960s for non-equilibrium superconductivity in conventional superconductors, may not contain all the information necessary to describe the nonequilibrium processes in high-temperature superconductors. For example, the symmetry of the order parameter in high-temperature superconductors has been shown to be *d* wave instead of *s* wave^{33,34} and this has been shown to demand significant modifications to the kinetic inductance model itself.³⁵ In addition, the characteristic time

TABLE I. Power law fitting of the laser power dependence of the fast optical response for optimally doped YBCO thin film in the superconducting state and numerical simulations.

| | E (eV) | | | Simulation |
|----------|---------------------------------|---------------------------------|-------------------|---------------------------------|
| T (K) | 1.50 | 1.54 | 1.62 | |
| 80 | $\sim P^{2.13}$ | $\sim P^{1.86}$ | $\sim P^{1.58}$ | $\sim P^{1.44}$ |
| 60 20 | $\sim P^{3.31}$ $\sim P^{5.51}$ | $\sim P^{3.18}$ $\sim P^{5.23}$ | $\sim P^{3.01}$? | $\sim P^{1.32}$ $\sim P^{0.92}$ |

scales in the nonequilibrium dynamics such as the Cooper pair-breaking time by phonons and the quasiparticle recombination time have been found to depend on substrate temperature²⁵ as well as laser power³⁶ rather strongly, contrary to the assumption of the Rothwarf-Taylor theory in this



FIG. 4. Numerical simulation of the laser power dependence of the fast optical response in optimally doped YBCO thin film in the superconducting state.

simple form that these characteristic time scales are constants. More interestingly, based on the proposed assignments of the sharp resonant triplet fine structures (a charge transfer exciton at $\sim 1.62 \text{ eV}$, a charge transfer exciton coupled with an optical phonon³⁷ at ~ 1.54 eV, and a charge transfer exciton coupled with both an optical phonon and the 41 meV magnetic excitation in YBCO systems³⁸), it was suggested that, within the electronic phase separation scenario of high-temperature superconductivity, the nonlinear laser power dependence could be understood, at least qualitatively, based on the strong coupling of charge carriers to the lattice and spin degrees of freedom, and the systematic variation of the nonlinearity as a function of photon energies (higher nonlinearity at low photon energies) could simply be the result of increasing complexity in underlying coupling mechanisms.⁸ According to the stripe phase theory of hightemperature superconductivity,¹⁸ mobile holes in the Cu-O plane are confined in conducting stripes separated by holefree antiferromagnetic insulating regions. Pairing, in this model, originates in the insulating regions, not in the conducting stripes, and it is simply transferred to the mobile holes in the conducting stripes by coherent transverse stripe fluctuations. A charge transfer exciton excited by the incoming photons in the insulating regions can itself break Cooper pairs by generating a local ferromagnetic fluctuation, or it can act as an instantaneous pinning center, thus increasing the effective mass density of the conducting stripes which was shown to be inversely proportional to T_c in the stripe phase model.³⁹ Therefore, a nonlinear laser power dependence of the Cooper pair-breaking rate by incoming photons is expected to be due to multiple exciton generation by the intense laser pulse. In addition, the notion of self-assembled array of conducting charge stripes separated by hole-free antiferromagnetic insulating domains in the Cu-O plane of high-temperature superconductors introduces intermediate length, time, and temperature scales in the physics of the problem.^{16-18,40} In fact, the very concept of kinetic inductance could have an entirely new microscopic interpretation in the stripe phase model or another model of electronic inhomogeneity. The corresponding description of nonequilibrium superconductivity is expected to be much more complex than the simple Rothwarf-Taylor theory due to the strong coupling among spin, charge, and lattice degrees of freedom. Further investigations are needed to make a quantitative connection of this theory to the nonlinear laser power dependence of the FOR signal discussed in this article.

We should like to point out that Huggard *et al.*⁴¹ have also reported a power law behavior of the photoresponse of high- T_c superconductors in the far infrared range over many orders of magnitude of laser intensity. In this regime, however, pair breaking is not expected, hence the corresponding physics is also different. Moreover, their laser pulse widths were \sim 50 ns as against 100 fs in our experiment. In their case, the power dependence was found to be less than linear. At low photon energy, the exponent was seen to be around 0.5. With increase of the photon energy, the exponent was seen to increase, approaching 1. In our case, the exponents for the three peaks are much higher than 1 and decrease with the increase of the photon energy. This again shows that the mechanisms in the two cases are indeed very different, implying a strong correlation effect in our case. Granular films such as those examined by Huggerd et al. basically have a random array of Josephson junctions. For photon energy less than the gap, the properties of each such Josephson junction and also of their complex network would be influenced. As mentioned in their paper, for example, a photon with energy less than the gap could affect the critical current across the junction. Thus, their observed nonlinear behavior may not be related to the intrinsic properties of high- T_c material, in contrast to our study, wherein high-quality epitaxial crystalline films are examined.

In conclusion, the temperature and the laser power dependence of the femtosecond optical pair breaking rate for an optimally doped YBCO thin film were measured in the superconducting state. While the temperature dependence measurements can be described rather successfully by the kinetic inductance model and the Rothwarf-Taylor theory, the strong nonlinearity and its significant temperature and photon energy dependence in the laser power dependence measurements cannot be accounted for by this simple model alone. These data demonstrate the complexity in the state of nonequilibrium superconductivity in high temperature superconductors and the opportunity provided by the femtosecond optical pair breaking technique.

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