Mechanism of nonequilibrium optical response of high-temperature superconductors

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This paper discusses the mechanism of the nonequilibrium optical response of high-temperature superconductors below T_c. Optical-response studies include pulsed-photoresponse measurements and modulated-reflectivity (transmission) measurements using femtosecond spectroscopy. A model is presented to explain the mechanism of the optical response based on the nonequilibrium dynamic transitions of electrons (quasiparticles and Cooper pairs) and phonons. These nonequilibrium transitions may cause flux motion due to activation by high-energy quasiparticles and phonons in the frame of BCS theory; moreover, these transitions may also change the kinetic inductance due to the reduction in the superconducting-electron density. Relaxation of the high-energy quasiparticles (generated by photons) through the electron-phonon and electron-electron scattering is rather fast: on the order of a picosecond, however, the speed limit of the photoresponse is governed by the phonon escape time. The results of the presented analysis suggest that further femtosecond-spectroscopy measurements may reveal more information about the superconducting anisotropic energy gap and band structure, pairing mechanism, quasiparticle-vortex interactions, and vortex energy structure in high-T_c superconductors. The results also strongly suggest that with proper optimization of device parameters (geometry and thermodynamic properties for fast heat removal, increasing pinning site density, critical current density, etc.), high speed (on the order of a ps response time), and sensitive detectors covering a broad electromagnetic spectrum (e.g., from ultraviolet to far infrared and beyond) can be developed.

I. INTRODUCTION

During the past few years there has been an active study of the optical response of high-temperature (cuprate) superconductors because they have attracted a great deal of attention as a basis for broadband detectors. This has been motivated by the previously reported observation of extremely sensitive and fast optical detection using sputtered films of $BaPb_{0.7}Bi_{0.3}O_3$ (Ba-Pb-Bi-O) (Ref. 1) and niobium tunnel junctions.[2] Furthermore, broadband optical detection with high-T_c superconductors is anticipated because of their low reflectivity (less than 50%) (Ref. 3) and high absorption coefficient (α is on the order of 10^4-10^5 cm^{-1}) (Ref. 4) from ultraviolet to infrared. We can divide the mechanisms responsible for detection of optical radiation by superconductors into two classes: equilibrium (bolometric) and nonequilibrium (non-bolometric) modes. In the equilibrium mode the detector responds to a change in lattice temperature (mostly determined by low-energy acoustic phonons) and acts like a bolometer. The bolometric response is the most sensitive mechanism that has been carefully documented and understood.[5] The nonequilibrium (or nonbolometric) mechanism has a faster response. It is presumably produced by photons breaking Cooper pairs (creating high-energy quasiparticles and, subsequently, high-energy phonons).[11] Both bolometric and nonbolometric mechanisms are present in most devices. Results of the latest developments in bolometric and nonbolometric responses of high-temperature superconductors are reviewed by Frenkel.[6] There are two principal nonbolometric detection mechanisms: the Josephson effect at the grain boundaries, and a nonequilibrium effect involving electrons and phonons. The former mechanisms is mostly determined by the grain boundary properties and it is not discussed in this paper. The latter mechanism is a fundamental phenomenon in superconductors and it is the prime subject of this paper. The results of nonbolometric measurements of high-T_c materials were reported by several groups, including Frenkel *et al.*,[7,8] Shi *et al.*,[9] Johnson,[10] Bluzer,[11] and Semenov *et al.*[12]

Another important study includes measurements of modulated reflectivity (transmission) using femtosecond spectroscopy. A major goal is a real-time study of the relaxation of charge carriers in high-temperature superconductors which can help to provide unique information about the nature of superconductivity in such materials and the role of electrons, phonons, and their interaction. Several groups reported femtosecond-spectroscopy results in cuprates, including Chwalec *et al.*,[13] Brorson *et al.*,[14] Kazeroonian *et al.*,[15] Han *et al.*,[16] Eesley *et al.*,[17] Reitze *et al.*,[18] and Chekalin *et al.*[19]

This paper discusses the mechanism of the nonequilibrium optical response of high-temperature superconductors below T_c by using the measurements of pulsed photoresponse (PR), and modulated reflectivity (transmission) with femtosecond spectroscopy. The model presented explains the mechanism of the optical response based on nonequilibrium dynamic transitions of electrons (quasiparticles and Cooper pairs) and phonons. These nonequilibrium transitions may cause flux motion due to the activation by the generated high-energy quasiparticles and phonons in the frame of BCS theory, and also possi-

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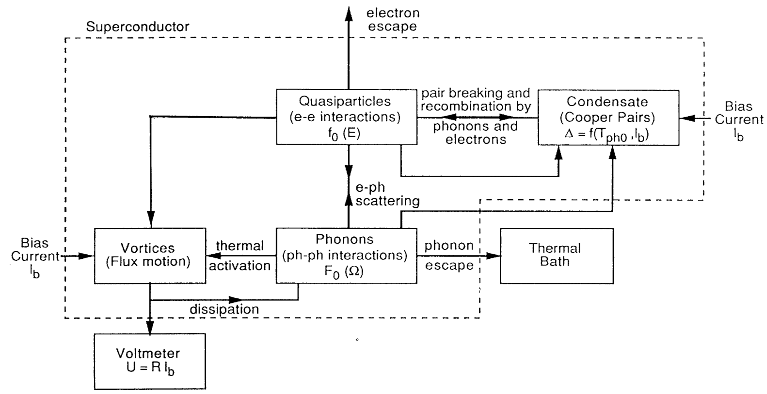

FIG. 1. Block diagram of a superconductor for an equilibrium condition before the optical pulse.

ble change in the kinetic inductance due to the reduction in the superconducting electron density. Discussions also include phonon escape, dissipation mechanism, and dynamic behavior near T_c. Based on the model presented and subsequent discussions, conclusions are made about the importance of further femtosecond spectroscopy measurements for studies of superconducting properties, and about the potential for broadband, fast and sensitive photodetectors using high-T_c materials.

II. NONEQUILIBRIUM DYNAMICS OF OPTICAL EXCITATION

Using the results of the optical-response studies including nonbolometric pulsed PR and modulated femtosecond reflectivity and transmission measurements, the nonequilibrium mechanism of optical response in high-temperature (type-II) superconductors based on BCS theory is described. The general block diagram of a nonequilibrium condition created by the optical pulse is presented in Sec. II A, and the quasiparticle and phonon dynamic behavior is discussed in Sec. II B.

A. General block diagram

A general block diagram describing the development of optical dynamic effects in a superconductor is shown in Figs. 1 and 2. We consider a general case of a superconductor below T_c which is in a mixed state created by a magnetic field or, equivalently, by a bias current. Figure 1 presents the equilibrium situation in a superconductor biased with a dc current, I_b, before the optical pulse is applied. It is assumed that non-Ohmic dissipation below T_c is predominantly caused by the flux motion through thermal activation.[20] We use the BCS description of a superconductor in this model. The condensate (Cooper pairs) exchanges energy with quasiparticles (unpaired electrons above energy gap) through pair breaking and recombination via phonons (electron-phonon scattering) and via quasiparticles (electron-electron scattering). The quasiparticle energy spectrum (or spectral density), $f_0(E)$, is in equilibrium which is maintained (in addition to the exchange with condensate) by the electron-phonon scattering (exchange with phonons), by the electron-electron scattering, by the diffusion escape, and possibly by the interactions with vortices, thus, causing flux motion dissipation (the terms "vortices," and "flux" are used interchangeably in the text). Phonons participate in electron-phonon and phonon-phonon interactions [the phonon equilibrium energy spectrum $F_0(\Omega)$], and they may also thermally activate vortices, causing flux motion dissipation. Phonon equilibrium is also maintained by their escape to the thermal bath.

A short optical pulse creates a nonequilibrium condition in the superconductor shown in Fig. 2. As a result

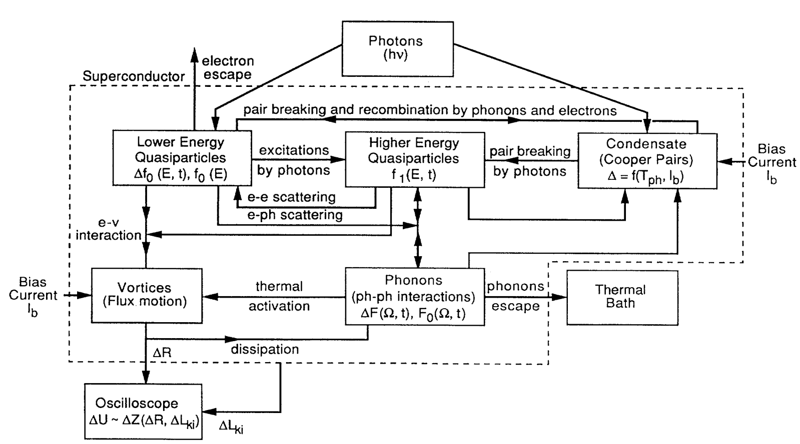

FIG. 2. Block diagram of a superconductor for a nonequilibrium condition created by the optical pulse.

of optical excitation, the spectrum of quasiparticles is significantly changed. There are two types of quasiparticles: lower-energy quasiparticles which initially correspond to the equilibrium energy spectrum $f_0(E)$, and higher-energy quasiparticles which are instantaneously created by interactions of photons with electrons (breaking Cooper pairs and exciting lower-energy quasiparticles). The quasiparticle time-dependent energy spectrum, $f(E,t)$ (E is the quasiparticle energy and t is the time), is determined by a rather complex exchange of electrons and phonons shown in Fig. 2, governed by the electron-phonon and electron-electron scattering (quasiparticle interactions with themselves, condensate, phonons, and vortices). The quasiparticle spectrum, $f(E,t)$, and its nonequilibrium change, $\Delta f(E,t)$, are given by

$$f(E,t)=f_0(E)+\Delta f_0(E,t)+f_1(E,t), \quad (1)$$

$$\Delta f(E,t)=\Delta f_0(E,t)+f_1(E,t),$$

where $\Delta f_0(E,t)$ is a change in the spectrum of lower-energy quasiparticles, and $f_1(E,t)$ is the spectrum of higher-energy quasiparticles created by the optical excitation (after one electron-phonon or electron-electron interaction, the higher-energy quasiparticles become lower-energy quasiparticles by our convention). As a result of electron-phonon scattering, extra phonons of high energy (optical phonons) with the spectrum $\Delta F(\Omega,t)$ are generated. These nonequilibrium transitions may cause additional flux motion (resulting in a resistance change ΔR) due to the activation by the generated high-energy quasiparticles and phonons in the frame of BCS theory, and possible change in the kinetic inductance, ΔL_{ki}, due to the reduction in superconducting electron density. This results in an impedance change, ΔZ, measured by the oscilloscope signal (kinetic inductance measurements are described by Bluzer[11]) as a function of time. The rate of phonon and electron escape to the thermal bath determines in many respects the nonequilibrium dynamics of PR and modulated reflectivity, since these extra phonons and electrons continue to cause nonequilibrium transitions. These transitions will be analyzed and discussed below.

B. Quasiparticle and phonon dynamics

After applications of an optical pulse, electron and phonon temperature may vary differently. Let us define electron and phonon temperature, $T_e=T_e(t)$ and $T_{ph}=T_{ph}(t)$, respectively, as follows:

$$T_c=T_{e0}+\Delta T_e(t), \quad \Delta T_e(t)=\int_\infty \Delta f(E,t)dE \ , \quad (2)$$

$$T_{ph}=T_{ph0}+\Delta T_{ph}(t), \quad \Delta T_{ph}(t)=\int_\infty \Delta F(\Omega,t)d\Omega \ , \quad (3)$$

where T_{e0} and T_{ph0} are the equilibrium electron and phonon temperatures before the optical pulse is applied. Let us assume that the electron and phonon temperatures are in a local equilibrium with themselves, and, therefore, the superconductor can be described as a coupled, two-temperature system. When a short optical pulse is applied to a superconductor, the space and time evolution of T_e and T_{ph} is governed by a pair of coupled nonlinear differential equations:[21]

$$C_e\delta T_e/\delta t=+K_e\nabla^2T_e-G_{e\text{-ph}}(T_e-T_{ph}) -G_{e\text{-}v}\Delta T_e(t)+P(r,t) \ , \quad (4)$$

$$C_{ph}\delta T_{ph}/\delta t=+K_{ph}\nabla^2T_{ph}+G_{e\text{-ph}}(T_e-T_{ph}) -G_{ph\text{-}v}\Delta T_{ph}(t)+P_d(t) \ , \quad (5)$$

where C_e and C_{ph} are the electron and phonon heat capacities, K_e and K_{ph} are the electron and phonon thermal conductivity, $G_{e\text{-ph}}$ is the electron-phonon coupling constant, $G_{e\text{-}v}$ is the electron-vortex interaction constant, $G_{ph\text{-}v}$ is the phonon-vortex interaction constant, $P(r,t)$ is the space- and time-dependent absorbed optical power density, and $P_d(t)$ is the dissipated power density due to the flux motion. The first term in these equations reflects the diffusion process, the second term reflects the electron-phonon interaction, the third term reflects the interaction of extra high-energy electrons [Eq. (4)] and phonons [Eq. (5)] with vortices. The optical power source term appears only in Eq. (4) since optical pulse interacts directly with electrons. The dissipation power term due to the vortex motion appears in Eq. (5).

Let us consider two extreme cases which correspond to two types of measurements discussed here (modulated reflectivity and PR). First, the optical pulse width, τ, is shorter than or on the same time scale as electron-phonon scattering time, $\tau_{e\text{-ph}}$, and also shorter than the electron-electron scattering time, $\tau_{e\text{-}e}$, (preliminary calculations show that these times could be on the order of femtoseconds[22]); at the same time $\tau_{e\text{-ph}}$ and $\tau_{e\text{-}e}$ are much shorter than electron and phonon escape (diffusion) times, τ_{ee} and τ_{phe}, respectively. For this case we can simplify Eqs. (4) and (5) to

$$C_e\delta T_e/\delta t=-G_{e\text{-ph}}(T_e-T_{ph})-G_{e\text{-}v}\Delta T_e(t)+P(r,t) \quad \text{for } t<\tau_{e=ph} \ , \quad (6)$$

$$C_{ph}\delta T_{ph}/\delta t=+G_{e\text{-ph}}(T_e-T_{ph})-G_{ph\text{-}v}\Delta T_{ph}(t)+P_d(t) \quad \text{for } t<\tau_{e=ph} \ , \quad (7)$$

$$C\delta T/\delta t=-K\nabla^2T \quad \text{for } t>\tau_{e=ph} \ , \quad (8)$$

where $\tau_{e=ph}$ is the time when T_e arbitrarily approaches T_{ph}, T is an equilibrium temperature between T_e and T_{ph}, $C=C_e+C_{ph}$, and $K=K_e+K_{ph}$.

The second extreme case is when τ is longer than $\tau_{e\text{-ph}}$, $\tau_{e\text{-}e}$, and also the electron-vortex interaction time, $\tau_{e\text{-}v}$. In this case electrons and phonons will remain in thermal equilibrium and the temperature of the system, T, can be described by simplifying Eqs. (4) and (5) as

$$C\delta T/\delta t=+K\nabla^2T+P(r,t)+P_d(t) \ . \quad (9)$$

Figure 3 shows the projected time evolution of T_e and T_{ph} for these two cases. Equations (6)–(8) were solved numerically for low-temperature superconductors.[22] Their solutions are applicable to the model described here. Figure 3(a) presents a time evolution based on these equations for the first extreme case corresponding to a very short optical excitation pulse (femtoseconds), which is confirmed qualitatively by the experimental results.[13–19] Typically, T_e reaches its maximum at the end of the optical pulse (τ_{em}, in Fig. 3) which corresponds to the maximum energy of quasiparticles excited by photons. However, the time corresponding to the maximum (or minimum if reflectivity change is negative) reflectivity (τ_{r-m} in Fig. 3) may be delayed relative to τ_{em} because it corresponds to the maximum number of quasiparticles, N_{qpm}, due to the continuing breaking of Cooper pairs after the end of the optical excitation. This phenomenon was noticed, for example, by Han *et al.*[16] τ_{r-m} can be found by solving a couple of Rothwarf and Taylor equations:[23]

$$dN_{\rm qp}/dt = I_{\rm qp} - RN_{\rm qp}^2 + 2\beta_1 N_{\rm ph} + 2\beta_2 N_{\rm qp} - \gamma_1 N_{\rm qp}, \qquad (10)$$

$$dN_{\rm ph}/dt = I_{\rm ph} + \frac{RN_{\rm qp}^2}{2} + SN_{\rm qp}^2 - \beta_1 N_{\rm ph} - \gamma_2 N_{\rm ph}\ , \qquad (11)$$

where $N_{\rm qp}$ and $N_{\rm ph}$ are, respectively, the number of quasiparticles and high-energy phonons with energy exceeding energy gap ($\Omega > 2\Delta$), $I_{\rm qp}$ and $I_{\rm ph}$ are, respectively, the quasiparticle and phonon generation rates due to the external excitation, R is the quasiparticle recombination rate constant, β_1 and β_2 are, respectively, the probabilities of breaking Cooper pairs by phonons and high-energy quasiparticles, γ_1 and γ_2 are, respectively, the probability rates of quasiparticle and phonon thermal escape, S is the probability rate of electron-phonon scattering. When T_e arbitrarily approaches $T_{\rm ph}$ [conventionally shown as $\tau_{e=\rm ph}$ time in Fig. 3(a)], then the further relaxation of T_e follows relaxation of $T_{\rm ph}$ and is primarily determined by the phonon escape time [Eq. (8)] which will be discussed in the next section. This slow component due to slow phonon escape was relatively insignificant in femtosecond reflectivity measurements[13–19] because the maximum electron temperature change, ΔT_{em}, is presumably much larger than the maximum phonon temperature change, $\Delta T_{{\rm ph}m}$. Modeling of the photoresponse is not straightforward because experimental data is limited by the resolution of the oscilloscope (typically, about 5–10 ps). However, we can make some observations and predictions based on the available data.[6–12] We assume that PR rise time would be the same as for T_e, and PR reaches its maximum at the end of the optical pulse because of the limited instrument time resolution of several picosecond in the PR data. A fast component in the PR relaxation[7–10] suggests the presence of an electron-vortex interaction because $T_e \gg T_{\rm ph}$ at τ_{em} (otherwise T_e should follow $T_{\rm ph}$). In general, relaxation of PR could be somewhat slower than relaxation of T_e if the vortex dissipation time τ_d is longer than $\tau_{e=\rm ph}$ (τ_d is discussed below in the text). However, in principle, PR could be only limited by the phonon escape time; thus PR could be as fast as $\tau_{e=\rm ph}$ or on the order of a ps.[16,17] In case of PR governed by a change in kinetic inductance,[11] the kinetic inductance is inversely proportional to the number of superconducting (Cooper) electrons [refer to Eq. 21 later in the text] and the number of superconducting electrons is determined by the relaxation of quasiparticles which follows from Rothwarf and Taylor equations [Eqs. (10) and (11)].

Figure 3(b) presents another extreme case described by Eq. (9). Here T_e, $T_{\rm ph}$, and PR are presented by the same curve. This curve reaches its maximum at the end of the optical pulse which follows from Eq. (9). The speed of PR is determined by the phonon escape time which will be discussed below.

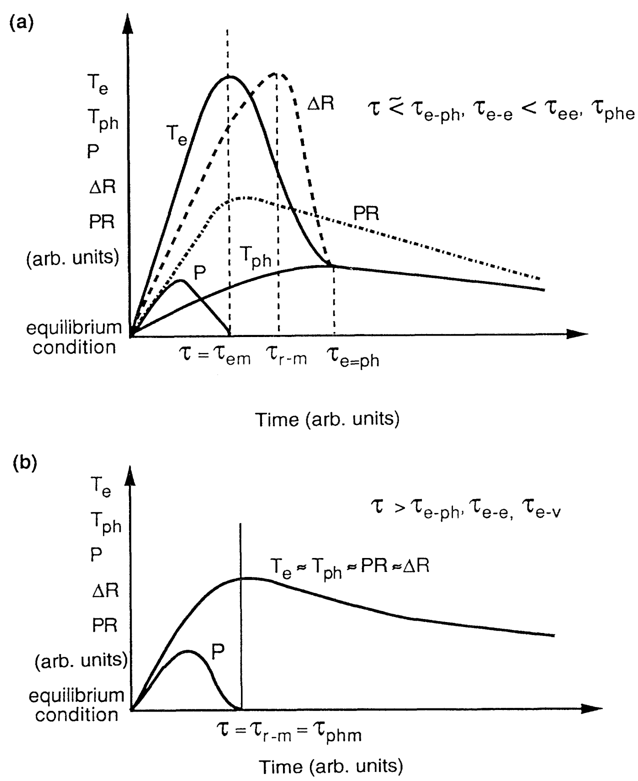

FIG. 3. Nonequilibrium dynamic behavior in a superconductor for two extreme cases: (a) $\tau < \tau_{e\text{-ph}}$, $\tau_{e\text{-}e} < \tau_{ee}$, $\tau_{{\rm ph}e}$; (b) $\tau > \tau_{e\text{-ph}}$, $\tau_{e\text{-}e}$, $\tau_{e\text{-}v}$; T_e is the electron temperature, $T_{\rm ph}$ is the phonon temperature, P is the optical pulse, PR is the photoresponse, ΔR is the reflectivity response.

III. DISCUSSIONS

A. Phonon escape

Frenkel *et al.* pointed out earlier[7,8] that phonon escape becomes a bottleneck for the PR relaxation which is shown in Fig. 3. Therefore, minimization of this escape time determines the speed of the PR. A simplified model for calculating escape time was presented by Frenkel *et al.*[7] In this model the film temperature rise due to the incident radiation can be estimated from a heat transfer model,[24] wherein the optical pulse of duration τ supplies heat (absorbed optical energy) at a constant rate F per unit time per unit area to the very thin superconducting bridge lying on a semi-infinite substrate. Thermal boundary resistance between the film and the substrate is neglected in this model assuming that it is compensated by lateral heat transfer in the substrate and in the film. Then the temperature rise ΔT as a function of time and coordinate x (in the direction perpendicular to the film) is given by[24]

$$\Delta T = 2F\left[\frac{t}{Kc\rho}\right]^{1/2} i\,\mathrm{erfc}\frac{x}{2(kt)^{1/2}} \quad \text{for } 0<t<\tau\,, \tag{12}$$

$$\Delta T = 2F\left[\frac{t}{Kc\rho}\right]^{1/2}\left[i\,\mathrm{erfc}\frac{x}{2(kt)^{1/2}} - \left[1-\frac{\tau}{t}\right]^{1/2} i\,\mathrm{erfc}\frac{x}{2[k(t-\tau)]^{1/2}}\right] \quad \text{for } t>\tau\,, \tag{13}$$

where K, c, and ρ are the thermal conductivity, specific heat, and density of the substrate, respectively, and $k=K/(c\rho)$. Figure 4 shows the dependence of the normalized temperature rise $\Delta T/\Delta T_\tau$ as a function of time (also normalized to τ). As expected, the temperature rise reaches its maximum at the end of the optical pulse ($t=\tau$) and then slowly decays, reaching about 10% at $t=20\tau$. Thus, for example, for a 150-ps optical pulse the thermodynamic model predicts the temperature relaxation time on the order of a nanosecond which is consistent with the reported results.[7,8]

However, boundary resistance, R_{bd} (defined as the ratio of the temperature difference across the interface to the power per unit area flowing across it), can play an important role. It is very difficult to model boundary resistance in high-temperature superconductors because it depends on variable growth conditions, though attempts were made to do that (e.g., Flick, Phelan, and Tien[25]). Boundary resistance was studied experimentally by Nahum, Verhese, and Richards[26] where they also observed a nanosecond PR time. It was pointed out and demonstrated[12] that phonon escape time can be significantly reduced to the picosecond range by using a narrow stripe geometry (the width of a stripe could be on the order of a micron or less) of a photodetector due to the significant reduction of a back phonon flow from the substrate to the superconductor.

We want to make one step further and propose a configuration which incorporates this narrow stripe geometry and also eliminates the boundary resistance problem (Fig. 5). In this configuration a relatively thick layer of deoxygenated superconducting material (nonsuperconducting) is deposited on the substrate first (or we can even use a bulk sample directly attached to the cold stage). Then, a thin layer of superconducting material (oxygenated) is deposited and patterned as narrow superconducting stripes using conventional lithography. In addition, nonsuperconducting areas may be covered by a high reflectance material to minimize heating. This structure minimizes the negative effect of a boundary resistance on the phonon escape. Thus in principle, PR could be on the order of a ps (as fast as $\tau_{e=\mathrm{ph}}$) for the device with the optimized phonon escape time.

B. Dynamic behavior in the transition region and near T_c

PR measurements[7-12] and most femtosecond reflectivity measurements[14-19] show similar dynamic behavior near T_c in the transition region: slowing down at higher temperatures and diverging near T_c. In the case of reflectivity this divergence directly relates to the change in the order parameter. We can compare the results of the dynamic optical response with the dynamical effects in low-temperature nonequilibrium superconductors.[27] In the frame of nonequilibrium superconductivity, the short optical pulse is a transient energy-mode pertur-

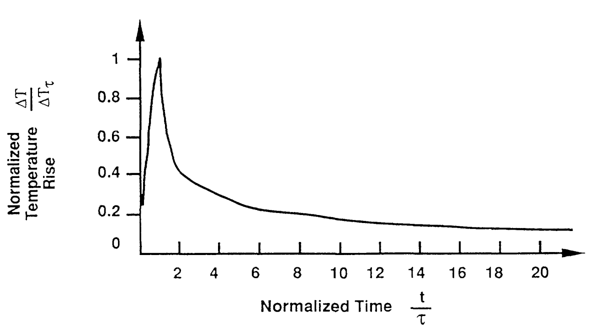

FIG. 4. The dependence of the normalized temperature rise $\Delta T/\Delta T_\tau$ as a function of the normalized time normalized to the laser pulse duration τ [from Frenkel *et al.* (Ref. 7)].

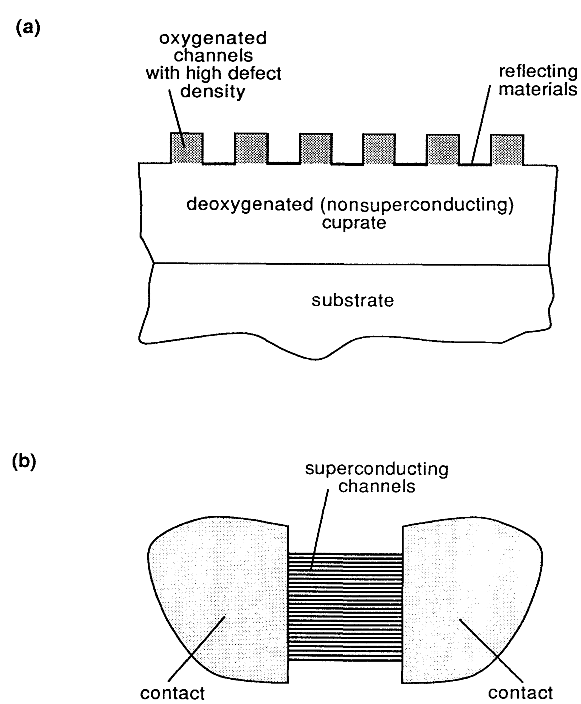

FIG. 5. Proposed superconductor detector configuration: (a) cross section, (b) general view.

bation which causes a nonequilibrium redistribution of quasiparticles [change in quasiparticle distribution f (E)]. The dynamic equation based on BCS theory suggests that the energy-mode transient relaxation time of quasiparticles, τ_Δ, can be expressed as follows:[27,28]

$$\tau_\Delta = 3.7\tau_{e\text{-ph}}\frac{k_B T}{\Delta} = 1.15\tau_{e\text{-ph}}\frac{T}{T_c}\left[1-\frac{T}{T_c}\right]^{-1/2}, \tag{14}$$

where the superconductor order (gap) parameter, Δ, is given by

$$\Delta = 3.1 k_B T_c \left[1-\frac{T}{T_c}\right]^{1/2}, \tag{15}$$

where $\tau_{e\text{-ph}}$ is the characteristic electron-phonon scattering time. The quasiparticles relax to the "local" equilibrium defined by the instantaneous value of the energy gap. Thus, relaxation of the energy gap becomes a bottleneck of the quasiparticle relaxation. Decrease in the superconducting energy gap parameter causes additional phonons with less energy participating in exchange between quasiparticles and condensate which may be equivalent to a change in the electron-phonon coupling constant $G_{e\text{-ph}}$ or it may be equivalent to a larger number of transitions involved in the quasiparticle relaxation (i.e., it takes more time for T_e to relax to T_{ph}). In the transition region Δ diverges to 0 which significantly slows the relaxation of quasiparticles. Thus, the nonequilibrium BCS description and our model of nonequilibrium optical excitation presented in Sec. II are in a good agreement with experimental data.[16,17]

PR effects are similar to our description above: with temperature increase additional phonons with less energy participate in the exchange between quasiparticles and condensate which leads to the additional interaction with vortices leading to increasing dissipation due to the vortex motion. In addition to the divergence of the energy gap to zero in the transition region, we may have a significant Ohmic dissipation in the transition region near T_c which slows the relaxation further due to the additional phonons in the superconductor. Thus, Eqs. (5), (7) and (9) should take the Joule heating term into consideration in the transition region.

C. Flux motion

We consider here superconducting materials without grain boundaries. There is a strong evidence that dissipation properties of such materials (e.g. epitaxial $Y_1Ba_2Cu_3O_{7-x}$ thin films) are most likely described by the flux-creep and flux-flow model which was pointed out elsewhere.[29,30] Therefore, we suggest that the pulsed optical response is most likely caused by the photoactivated flux creep and flux flow. In equilibrium, fluxons are present in the film and are moved by thermal activation.[29] The optical photons create additional flux creep and flux flow (through generated extra electrons and phonons) which apparently cause additional dissipation (equivalent to the increase in the resistance) and which is attributed to the nonbolometric component in the optical response.[7] The activation energy U_0 (which should be overcome to allow flux motion) for low magnetic field is given by[20]

$$U_0 = H_c^2\xi^3/8\pi, \tag{16}$$

where H_c is the thermodynamic critical field, and ξ is the coherence length. Using Eq. (16) for the activation energy with the clean limit Ginsburg-Landau formulas for the thermodynamic critical field, H_c, and the coherence length, ξ:

$$H_c = 1.73H_{c0}(1-T/T_c), \tag{17}$$

$$\xi = 0.74\xi_0(1-T/T_c)^{-1/2}, \tag{18}$$

we get the following expression for the activation energy:

$$U_0 = \frac{1.21H_{c0}^2\xi_0^3}{8\pi}\left[1-\frac{T}{T_c}\right]^{1/2}, \tag{19}$$

where H_{c0} is the thermodynamic critical field at 0 K, ξ_0 is the coherence length at 0 K, T is the superconductor temperature, and T_c is the critical temperature. The energy of photons at the wavelength of 1.06 μm is about 1.2 eV which is typically larger or on the same order of magnitude as U_0 calculated from Eq. (19) (Ref. 31) and measured experimentally.[29,32,33] It has been shown[30] that thin-film devices have a wide gradual nonlinear resistive transition in the $I-V$ curves (induced by the bias current). This indicates that U_0 is not homogeneous through the sample and is widespread. Obviously, with the temperature decrease, the activation energy (i.e., average value of its distribution) increases which should decrease the amplitude of the optical response. This explains the gradual decrease of the amplitude of the nonbolometric component in the optical pulse response (e.g., Fig. 10 in Ref. 7) with the temperature decrease below the transition region. Thus, PR results for epitaxial films are consistent with the photoactivated flux motion model.

If flux motion is the main contributor to non-Ohmic dissipation, the other important aspects are the nature of this dissipation mechanism and its time constant, τ_d. These issues were discussed for low-temperature superconductors.[34] The nature and the time constant of dissipation due to the flux-line motion is an important phenomenon which requires more attention and investigation for highT_c materials. Optimization of PR measurements may provide information about the flux motion dissipation mechanism and its dynamics. If phonon escape time is minimized, then τ_d may become a bottleneck for photodetection relaxation process and can be measured directly.

D. Kinetic inductance

Change in the kinetic inductance of a superconducting thin film caused by the optical excitation may result in a significant change of the impedance of a superconducting thin film which was pointed out in Sec. II. If significant, this kinetic inductance change can be used to monitor the dynamics of PR,[11] as well as to serve as a PR signal in nonequilibrium kinetic inductance detectors.[35] Kinetic inductance as a function of the penetration depth λ is given by[36]

$$L_{\mathrm{ki}}=\mu_0\frac{1}{w}\frac{\lambda^2}{d} \quad \text{for } d \ll \lambda \ , \tag{20}$$

where 1, w, and d are length, width, and thickness of a superconducting film, respectively, and μ_0 is the permeability of free space. Penetration depth in the dirty limit is given by[37]

$$\lambda^2=\frac{mc^2}{4\pi e^2 n_s}\frac{\xi_0}{1_s} \quad \text{for } \xi_0 \gg 1_s,\ \xi_0 \ll \lambda \ , \tag{21}$$

where 1_s is the electronic mean free path, ξ_0 is the coherence length at 0 K, n_s is the density of superconducting electrons, m and e are the electron's mass and charge, respectively, and c is the speed of light. From Eqs. (21) it follows that the kinetic inductance is inversely proportional to the number of superconducting electrons and that number is determined by the relaxation of quasiparticles which follows from Rothwarf and Taylor equations [Eqs. (10) and (11)]. So far, kinetic inductance nonequilibrium detectors were implemented using low-temperature superconductors.[35] Analysis of Eqs. (20) and (21) shows that high-T_c materials have a potential as kinetic inductance detectors because they have larger penetration depth than low-temperature materials and also because the electronic mean free path can be significantly reduced in these materials by increasing defect and pinning site density as discussed below.

E. Detector sensitivity issues

Optimization of device geometry (thickness, device pattern, maximization of the absorbed power) and operation at higher bias currents play an important role in increasing of the detector responsivity.[7,8] Also, increase of the boundary resistance between the thin film and the substrate (which will slow the device speed) can improve the responsivity (the tradeoffs between responsivity of bolometers and their speed are discussed in detail by Frenkel[6]). However, it is desirable to find a way for increasing responsivity of the device withoug sacrificing its speed. Therefore, we still want to keep the boundary resistance between the film and the substrate to a minimum, and maximize the additional flux dissipation and kinetic inductance change caused by the optical pulse. One solution is to increase the density of defects in type-II superconductors. It was demonstrated[38] that it is possible to significantly increase the number of pinning sites (defects) and increase critical current density (by increasing U_0) at the same time, using heavy ion irradiation. This should significantly increase the additional nonequilibrium flux dissipation (somewhat proportionally to the pinning site density and critical current density) and also increase the kinetic inductance impedance signal (proportionally to the critical current density and inversely proportionally to the decrease in the electronic free path), thus, increasing the responsivity according to our model. Therefore, the ideal configuration for a high speed sensitive detector is the one shown in Fig. 5 with superconducting channels of high defect density.

F. Femtosecond spectroscopy

Based on the above discussion we can identify major directions for femtosecond-spectroscopy measurements of high-temperature superconductors. The major controversy in the reported data[13–19] is the different sign of the reflectance change. It becomes clear that the sign of the changes depends on the material itself and its energy band structure. Varying wavelengths of both pumping and probing beams can be a way to resolve the controversy. The attempts to use the white source as a probe,[19,30] or to change doping[15,18,40] were made, but it is not enough. The measurements should include spectral dependence of the pump and probe pulses as an important goal which may reveal more information on the superconducting properties. One such goal would be to study superconducting anisotropic properties of a Fermi level (applying generalized Hubbard model[14,15]) and superconducting energy gap by detecting the value of the probe beam frequency at which the change of the sign and slope in the modulated reflectance or transmission occurs. Anisotropic studies can be performed by changing the angle of incidence of the pump and probe beams and their optical polarization. These studies can contribute to the resolution of the argument about possible anisotropic energy gap (which implies a different angular momentum of the electron pair, e.g., s wave, d wave, or others) in a copper oxides and reveal more information about superconducting energy band structure. Another important aspect which can be addressed using the pump-probe technique is to study the contribution of electron-phonon scattering and electron-electron scattering to the quasiparticle relaxation. For example, an electron-phonon coupling constant could be found by applying Allen's theory[41] for different anisotropies in the superconducting material. Then it can be compared with the value expected from the BCS theory which should tell us if the quasiparticle relaxation is mostly due to the electron-phonon scattering or electron-electron scattering. Thus, based on these findings the coupling mechanism (strong vs weak coupling and mediated by phonons vs electrons) of the paired superconducting electrons can be evaluated. Another important superconducting mechanism which can be studied using femtosecond spectroscopy is flux dynamics which is especially important because the coherence length in copper oxides is much smaller than in metallic type-II superconductors which may imply a different mechanism of the flux pinning (e.g., collective effects of the vortex lines[42] should be considered), dissipation, and vortex core energy structure in copper oxides. Therefore, it is important to study interactions of high-energy quasiparticles and phonons with vortices and to understand energy transfer mechanism from the electrons and phonons to vortices, and to determine vortex core excitation energy levels by applying the magnetic field (or, equivalently, bias current). By varying the magnetic field we should be able to see the change in the fast-Fourier-transformation spectra of the time-resolved reflectivity[43] which should give us information about the energy transitions related to vortices and their core excitation energies and compare these results with the results reported for magneto-optical studies in the far infrared.[44] Careful attention in all pump-probe studies should be paid to the pulse width, because due to the uncertainty principle, spectral bandwidth even in a transform limited case can be significant enough (several nm) to obscure the results. Therefore, the use of transform limited longer pulses (perhaps, on the order of a picosecond) with a corresponding reduced spectral bandwidth could be considered.

IV. SUMMARY

The mechanism of the nonequilibrium optical response of high-temperature superconductors below T_c is discussed in this paper. Nonequilibrium dynamics of electrons (quasiparticles and Cooper pairs) and phonons is used to explain the mechanism of the optical response. The nonequilibrium transitions of electrons and phonons may cause flux motion due to the activation by the high-energy quasiparticles and phonons generated by the pulse and also possibly change in the kinetic inductance due to the reduction in the superconducting electron density. Relaxation of the high-energy quasiparticles (generated by photons) through the electron-phonon and electron-electron scattering is rather fast: on the order of a picosecond, however, the speed limit of the PR is governed by the phonon escape time. The results of the presented analysis suggest that further femtosecond spectral dependent measurements should reveal more information about superconducting anisotropic energy gap and band structure, pairing mechanism, quasiparticle-vortex interactions, and vortex energy structure in high-T_c superconductors by varying pump and probe wavelengths and applying external perturbations (magnetic field or bias current). The results strongly suggest that with a proper optimization of device parameters (geometry and thermodynamic properties for a fast heat removal, increasing pinning site density, critical current density, etc.) high speed (on the order of a ps response time) and sensitive detectors covering a broad electromagnetic spectrum (e.g., from γ rays to millimeter waves) can be developed. A superconducting photodetector configuration is proposed.

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