Quasiparticle energy relaxation in the cuprate superconductors

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At high flux-flow velocities in the mixed state of a type-II superconductor the nonequilibrium distribution of the quasiparticles is shifted to higher energies. As shown theoretically by Larkin and Ovchinnikov, a distinct signature of this nonequilibrium effect is an electronic instability, resulting in a sharp kink in the currentvoltage characteristic at a critical vortex velocity v_{φ}^* . From measurements of v_{φ}^* the quasiparticle-energy relaxation rate τ_{ε}^{-1} can be found. We have measured this instability point for epitaxial c axis oriented films of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_{4-x}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. In both cuprates the observed temperature dependence of τ_{ε}^{-1} could be well fitted with the function $\tau_{\varepsilon}^{-1} = a \exp[-2\Delta(T)/k_BT]$, suggesting that the electron-electron recombination process, perhaps in combination with the emission of another excitation, is crucial. As an example of a low- T_c superconductor we have also studied amorphous Mo₃Si films and found that τ_{ε}^{-1} can be fitted by a function similar to that we have used for the cuprates, again indicating the importance of the electron-electron interaction. [S0163-1829(97)07745-X]

I. INTRODUCTION

Recently, we have reported experimental observations of an electronic instability at high vortex velocities in the mixed state of superconducting $YBa_2Cu_3O_{7-\delta}$ films.¹ Based on a theoretical analysis of Larkin and Ovchinnikov (LO),² from our experiments we have extracted the quasiparticle energy relaxation rate τ_{ε}^{-1} as a function of temperature over an extended temperature range below T_c . The relaxation rate τ_{ε}^{-1} showed an exponential decrease with decreasing temperature reaching a value around 10^8 s^{-1} at $T_c/2$. It is an interesting question whether this strongly temperature-dependent relaxation rate is a unique feature of the cuprate superconductors and how it behaves in low- T_c materials. With this question in mind, we have extended our measurements to amorphous Mo₃Si films as an example of a conventional superconductor. Furthermore, we report on results obtained for a second cuprate material, namely, $La_{1.85}Sr_{0.15}CuO_{4-x}$.

We have selected *a*-Mo₃Si since it is an extreme type-II superconductor with a relatively low critical current density. Further, with its critical temperature near 8 K, it presents a convenient temperature range below T_c for experimental investigation. On the other hand, the cuprate $La_{1.85}Sr_{0.15}CuO_{4-x}$ had a critical temperature of about 25 K, again promising an extended temperature range for experiments on the nonlinear effects at high vortex velocities.

II. NONEQUILIBRIUM SUPERCONDUCTIVITY

Nonequilibrium effects in low- T_c superconductors have been studied extensively some time ago and summaries can be found in Refs. 3 and 4. In high- T_c materials, this subject is just beginning to be investigated. The basic ideas for a theoretical understanding of the electronic nonequilibrium effects have been generated by Eliashberg.⁵ Changes in the quasiparticle energy distribution due to external energy input (irradiation with light, microwaves, or phonons; injection of quasiparticles; etc.) affect the superconducting parameters such as critical temperature, critical current, and superconducting energy gap and can result even in an enhancement of these quantities. As an example, the Eliashberg theory yields the following equation for the stimulation of superconductivity by an electromagnetic field:⁵

$$\frac{T_c - T}{T_c} - \frac{7\zeta(3)}{8\pi^2} \left(\frac{\Delta}{k_B T_c}\right)^2 - \frac{\pi\alpha_{\omega}}{2k_B T_c} - 0.17 \frac{\alpha_{\omega}}{\gamma} \left(\frac{\hbar\omega}{k_B T_c}\right)^2 - 2\int_{\Delta}^{+\infty} \frac{f(\varepsilon) - f^0(\varepsilon)}{\sqrt{\varepsilon^2 - \Delta^2}} d\varepsilon = 0.$$
(1)

Here $\zeta(x)$ is the Riemann zeta function, Δ is the superconducting energy gap, ω is the frequency of the electromagnetic field, ε and $f(\varepsilon)$ are the energy and distribution function of the quasiparticles, respectively, γ $\propto g[(k_B T)^3/(\hbar \omega_D)^2]$ with

$$\frac{1}{g} = \int_{\Delta}^{\omega_D} \frac{1 - 2f(\varepsilon)}{\sqrt{\varepsilon^2 - \Delta^2}} d\varepsilon,$$

 ω_D is the Debye frequency, $\alpha_{\omega} = (2e^2D/\hbar c^2)A_{\omega}^2$ is the power of the field, D is the quasiparticle diffusion coefficient, and A_{ω} is the field amplitude.

Experimental studies of the effects due to high-frequency irradiation are usually performed at zero applied magnetic field. Nonequilibrium effects in the mixed state of a type-II superconductor can become important for the process of current-induced vortex motion. In particular, near the transition temperature strong changes in the electric resistivity and in the superconducting order parameter Δ can occur, even when the quasiparticle distribution function shows only little deviation from equilibrium. This situation has been analyzed

by LO,² yielding the following equation for the order parameter Δ and its variation with the distance *r* from the center of the vortex for $T \approx T_c$:

$$\frac{T_c - T}{T_c} - \frac{7\zeta(3)}{8\pi^2} \left(\frac{\Delta}{k_B T_c}\right)^2 + \frac{\pi}{8} \frac{\hbar D}{k_B T_c} \left(\frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial}{\partial r} - \frac{1}{r^2}\right) + \int_{\Delta}^{+\infty} \frac{1}{\sqrt{\varepsilon^2 - \Delta^2}} \left(f(\varepsilon) - \tanh\frac{\varepsilon}{2k_B T}\right) d\varepsilon = 0.$$
(2)

We note the similarity of Eqs. (1) and (2). A change in the quasiparticle distribution function, due to the electric field induced by the vortex motion, mainly causes a change in the $\Delta(r)$ dependence near the vortex core. In the vortex core, the quasiparticles are shifted upward in energy compared to the equilibrium distribution. The quasiparticles diffuse out of the vortex core, the core shrinks, and, as a consequence, the viscous damping coefficient decreases with increasing vortex velocity. According to LO,² the damping force passes through a maximum at the critical vortex velocity v_{φ}^* given by

$$v_{\varphi}^{*2} = \frac{D[14\zeta(3)]^{1/2}(1 - T/T_c)^{1/2}}{\pi\tau_{\varepsilon}}$$
(3)

 $(D = \frac{1}{3}v_F l, v_F)$ being the Fermi velocity and *l* the electron mean free path). Reaching the maximum of the damping force manifests itself as an abrupt change of the slope of the current-voltage characteristic (IVC). The nonequilibrium quasiparticle distribution and the critical vortex velocity v_{φ}^* are fixed by the quasiparticle energy relaxation rate τ_{ε}^{-1} . The electric field E^* corresponding to v_{φ}^* is given by

$$\mathbf{E}^* = -(\mathbf{v}^*_{\boldsymbol{\omega}} \times \mathbf{B}). \tag{4}$$

The relaxation rate τ_{ε}^{-1} can be found from measurements of the nonlinear IVC.¹ We emphasize that τ_{ε}^{-1} is extracted from intrinsic bulk phenomena of the material. In the derivation of Eq. (3), spatial homogeneity of the nonequilibrium quasiparticle distribution represents an important assumption. This means that the inelastic quasiparticle diffusion length l_{ε} must be at least equal to the intervortex distance *a*. A more detailed discussion of this point has been given elsewhere.⁶

III. INELASTIC ELECTRON SCATTERING

Since the inelastic electron scattering processes play a central role in limiting the deviation of the quasiparticle distribution from equilibrium due to the injection of energy into the quasiparticle system, in the following we briefly discuss some aspects relevant for our experiments. In the classical superconductors *inelastic electron-phonon scattering* is highly important. These processes perform the transfer of excess quasiparticle energy to the crystal lattice and eventually to the heat bath. The energy relaxation can take place either by means of a scattering process with a phonon, changing the quasiparticle excitation energy, or by a recombination process reducing the number of quasiparticles by 2 and transferring the excess quasiparticle energy to an emitted phonon. The temperature dependence is distinctly different for both processes. The inelastic electron-phonon scattering

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process is governed by the thermal or nonequilibrium phonon population. On the other hand, the recombination process represents a binary reaction and the recombination rate decreases exponentially according to $\exp[-\Delta(T)/k_BT]$, reflecting the exponential temperature dependence of the quasiparticle population.⁷

The direct *inelastic electron-electron scattering* can also affect the quasiparticle lifetime. In combination with the emission and absorption of phonons these processes can also accomplish the inelastic energy transfer. As pointed out already by Eliashberg,⁸ these processes can play an important role in nonequilibrium superconductivity, as we encounter, for example, at high vortex velocities and, in particular, near the instability point, we are investigating in this paper. Here two processes must be distinguished.9 Two quasiparticles are scattered into two new quantum states, with the total energy and quasiparticle number remaining constant. Phase breaking occurs because of the inelastic nature of this process. In the second process, three quasiparticles interact with each other, two recombine to a Cooper pair and one quasiparticle with energy $\varepsilon \ge 3\Delta$ is generated (instead of three quasiparticles with energy $\varepsilon \approx \Delta$). In the inverse process a Cooper pair is broken by impact ionization. The recombination rate due to electron-electron interaction scales with $\exp[-2\Delta(T)/k_{B}T]$ since three quasiparticles are involved.⁷ This process gains importance at higher temperatures. In order to transfer energy out of the quasiparticle system, it must be combined with the emission of another excitation such as a phonon or spin fluctuation.

It appears that usually for the classical superconductors the electron-phonon interaction is dominant compared to the electron-electron interaction.⁹ However, there are also cases where the electron-electron interaction is more important as, for example, in materials with a high Debye temperature. On the other hand, there are strong indications that electronelectron scattering plays an important role in the cuprate superconductors. Hence, in the discussion of the strong nonequilibrium effects observed in our experiments the electronelectron recombination process may have to be taken into account. We return to this point in Sec. VI.

IV. EXPERIMENTS WITH a-Mo₃Si

For comparing a low- T_c superconductor with the cuprates we chose thin films of amorphous Mo₃Si. This material is an extreme type-II superconductor in the dirty limit with a Ginzburg-Landau parameter $\kappa \approx 60^{10}$ Its transition temperature of 8 K yields a convenient temperature range below T_c for experimental studies. Furthermore, flux pinning in this material is very low, which represents an important advantage for experiments dealing with vortex dynamics. A typical value of the critical current density at 4.2 K and in the magnetic field range 1–4 T is $j_c = 500 \text{ A/cm}^{2.11}$ As has been discussed in more detail elsewhere,¹ the minimization of Joule heating of the sample is an important requirement for our measurements. Hence single-crystalline MgO has been used as substrate material because of its high thermal conductivity. Furthermore, the IVC's were recorded using single triangular current pulses (typical pulse width equal to 1 ms), as in our previous experiments.¹

The a-Mo₃Si films were deposited by means of rf sput-



FIG. 1. Applied current and resistive voltage versus time for a single triangular current pulse for amorphous Mo_3Si at T=6.7 K and B=0.5 T. The inset shows the resistive transition to superconductivity.

tering. Details of sample fabrication and characterization are given elsewhere.¹² Microfabrication of our four-point sample geometry was performed by standard photolithography. For attaching the current and voltage leads, silver contact pads with an area as large as $2 \times 2 \text{ mm}^2$ were placed on top of the Mo₃Si film. The contact pads for the current were at least 2 mm away from the sample gauge length between the voltage leads. The geometrical sample parameters were typically as follows: thickness, 100 nm; length (between voltage leads), 200 μ m; and width, 20 μ m. The MgO substrate was in good thermal contact with a temperature-regulated copper block. allowing a temperature variation between 4.2 and 30 K. The samples had a sharp superconducting transition of 50 mK width. The zero-resistance critical temperature was 7.67 K. Just above T_c the resistivity was 150 $\mu\Omega$ cm. A magnetic field was applied perpendicular to the film plane. We investigated three samples, all yielding nearly the same results.

In Fig. 1 we show a typical plot of current and voltage versus time for a single triangular current pulse. At the voltage V=30 mV (marked by the arrow), the curve abruptly switches to a branch with nearly vertical slope. We interpret this voltage level as the value associated with the maximum vortex velocity v_{φ}^* by means of the relation $\mathbf{V}^* = -(\mathbf{v}_{\varphi}^*)$ \times **B**)*L*, where *L* is the sample gauge length. A typical resistive transition curve for our a-Mo₃Si films is shown in the inset of Fig. 1. The critical vortex velocity v_{ω}^{*} obtained from the kink in the IVC and calculated using Eq. (4) is plotted in Fig. 2 versus magnetic field for four temperatures. The magnetic-field independence of v_{φ}^* expected from LO's theory and Eq. (3) can be seen, except for the low-field end of the data. As we have discussed in detail elsewhere,^{6,13} there exists a crossover magnetic field, below which the field independence of v_{α}^{*} turns into the proportionality v_{α}^{*} $\sim B^{-1/2}$. This latter behavior comes from the requirement that the quasiparticle nonequilibrium distribution is spatially homogeneous. As has been demonstrated elsewhere,¹⁴ Joule heating of the sample leads to a distinct magnetic-field dependence of v_{α}^{*} . From Fig. 2 we see that such effects appear to be negligible. Calculating the quasiparticle energy relaxation rate τ_{ε}^{-1} from our measured values of v_{φ}^{*} using Eq. (3), we obtain Fig. 3, where τ_{ε}^{-1} is plotted logarithmically versus



FIG. 2. Critical vortex velocity v_{φ}^* versus magnetic field for four temperatures in *a*-Mo₃Si.

the normalized temperature T/T_c . Here we have used the values $v_F = 1.5 \times 10^6$ m/s and l = 0.3 nm.¹¹ For comparison, we show in Fig. 3 also our previous data for YBa₂Cu₃O_{7- δ}.¹

As can be seen from Fig. 3, in *a*-Mo₃Si the decrease of τ_{ε}^{-1} with decreasing temperature is less steep than for YBa₂Cu₃O_{7- δ} However, *a*-Mo₃Si also shows the exponential temperature dependence of τ_{ε}^{-1} , which one can see for YBa₂Cu₃O_{7- δ} Our data for *a*-Mo₃Si clearly reach normalized temperatures T/T_c closer to 1 than for YBa₂Cu₃O_{7- δ}. This upper limit of the temperature range, where the distinct kink in the IVC can be observed, appears to be related to the magnitude of the quasiparticle energy relaxation rate τ_{ε}^{-1} and extends closer to T_c for smaller values of $\tau_{\varepsilon}^{-1.2}$ A more detailed discussion of this point is given elsewhere.¹³

The solid lines in Fig. 3 represent fits to the function $a \exp[-2\Delta(T)/k_BT]$. For the function $\Delta(T)$ we have taken the BCS temperature dependence. For a-Mo₃Si we have taken $\Delta(0) = 1.76k_BT_c$, whereas for YBa₂Cu₃O_{7- δ} we used $\Delta(0) = 3.5k_BT_c$. The fitting values of a are listed in Fig. 3. As we can see, a good fit of our data is obtained both for a-Mo₃Si and YBa₂Cu₃O_{7- δ}.

V. EXPERIMENTS WITH La_{1.85}Sr_{0.15}CuO_{4-x}

Our comparison in Fig. 3 of the temperature-dependent quasiparticle energy relaxation rate τ_{ε}^{-1} showed a similar



FIG. 3. Quasiparticle energy relaxation rate τ_{ε}^{-1} plotted logarithmically versus the normalized temperature T/T_{c} for *a*-Mo₃Si and YBa₂Cu₃O_{7- δ}. The solid lines represent the function $a \exp[-2\Delta(T)/k_{B}T]$ fitted to the data.



FIG. 4. Applied current and resistive voltage versus time for a single triangular current pulse for $La_{1.85}Sr_{0.15}CuO_{4-x}$ at T = 10.6 K and B = 0.16 T. The resistive transition to superconductivity is shown in the inset.

behavior for the cuprate (YBa₂Cu₃O_{7- δ}) and the classical (*a*-Mo₃Si) superconductor. We have also studied La_{1.85}Sr_{0.15}CuO_{4-x} as another cuprate material. The *c*-axis-oriented epitaxial La_{1.85}Sr_{0.15}CuO_{4-x} films were deposited on single-crystalline SrTiO₃ substrates by laser ablation. The typical sample dimensions were thickness, 100 nm; length, 100–200 μ m; and width, 10–20 μ m. Microfabrication and the measurements were performed similarly as for *a*-Mo₃Si. The critical temperature was $T_c = 25$ K. The magnetic field was applied parallel to the *c* axis. A typical curve of resistivity versus temperature is shown in the inset of Fig. 4.

We have studied two La_{1.85}Sr_{0.15}CuO_{4-x} samples, both showing nearly the same behavior. The La_{1.85}Sr_{0.15}CuO_{4-x} films also displayed a distinct kink in the IVC, indicating the electronic instability at high vortex velocities. A typical case is shown in Fig. 4. Again, we have found that v_{φ}^{*} is independent of magnetic field, as shown in Fig. 5 for different temperatures. The deviation from the field independence at the low-field end of the data is due to the same reason we have discussed in Sec. IV. Calculating τ_{ε}^{-1} from our measurements of v_{φ}^{*} , using Eq. (3), we obtained the plot of τ_{ε}^{-1} versus the normalized temperature T/T_c shown in Fig. 6. Here we have used the values $v_F = 1 \times 10^7$ cm/s and l



FIG. 5. Critical vortex velocity v_{φ}^* versus magnetic field at different temperatures for La_{1.85}Sr_{0.15}CuO_{4-x}.



FIG. 6. Quasiparticle energy relaxation rate τ_{ε}^{-1} plotted logarithmically versus the normalized temperature T/T_c for La_{1.85}Sr_{0.15}CuO_{4-x} and YBa₂Cu₃O_{7- δ}. The solid line represents the function $a \exp[-2\Delta(T)/k_BT]$ fitted to the data.

=5 nm. For comparison, in Fig. 6 we also present our previous data for YBa₂Cu₃O_{7- δ}⁻¹ Similar to YBa₂Cu₃O_{7- δ}, the energy relaxation rate τ_{ε}^{-1} of La_{1.85}Sr_{0.15}CuO_{4-x} can also be well fitted by the function $\tau_{\varepsilon}^{-1} = a \exp[-2\Delta(T)/k_BT]$ shown by the solid line in Fig. 6. Again we have taken $\Delta(0)$ = $3.5k_BT_c$ and the BCS temperature dependence $\Delta(T)$. In this fit the value $a = 3 \times 10^{16} \text{ s}^{-1}$ was used. In both materials a good fit is obtained using $\Delta(0) = 3.5k_BT_c$ and the BCS $\Delta(T)$ dependence. From Fig. 6 we note that in La_{1.85}Sr_{0.15}CuO_{4-x} the instability in the IVC could be detected down to temperatures $T > 0.4T_c$.

VI. DISCUSSION

As the main result of our experiments we found that the two cuprates $YBa_2Cu_3O_{7-\delta}$ and $La_{1.85}Sr_{0.15}CuO_{4-x}$ show a very similar temperature dependence of the quasiparticle energy relaxation rate τ_{ε}^{-1} , which can be well fitted with the function $\tau_{\varepsilon}^{-1} = a \exp[-2\Delta(T)/k_BT]$. In both cases we have performed the fit using $\Delta(0) = 3.5k_BT_c$, i.e., twice the value of the standard weak-coupling BCS behavior. For $YBa_2Cu_3O_{7-\delta}$ the values of $\Delta(0)$ reported in the literature cover the range $\Delta(0) \approx 3-4k_BT_c$, ¹⁵ whereas for $La_{1.85}Sr_{0.15}CuO_{4-x}$ the range is $\Delta(0) \approx 2-4.5k_BT_c$. ¹⁶ Apparently, taking the average $\Delta(0) \approx 3.5k_BT_c$ provides a good fit to our data.

As we can see from Fig. 6, the data for La_{1.85}Sr_{0.15}CuO_{4-x} are shifted to lower reduced temperatures T/T_c compared to YBa₂Cu₃O_{7- δ}. This can be explained by examining the validity of the theory we have applied for analyzing our results. LO developed their theory² in the dirty limit, whereas the cuprates are in the moderately clean limit. If inelastic quasiparticle scattering takes place on the background of many elastic processes, as treated by LO, the factor D/τ_{ε} in Eq. (3) for v_{φ}^* may change. On the other hand, at the relatively high temperatures of our experiments the establishment of the strictly clean limit will be difficult. An argument in favor of our values of the energy relaxation rate τ_{ε}^{-1} is based on the fact that the crossover condition

$$k_B(T_c - T) = h \tau_{\varepsilon}^{-1} \tag{5}$$

derived by LO (Ref. 17) for the appearance of the electronic instability is well satisfied by our results. If $h\tau_e^{-1}$ becomes larger than $k_B(T_c - T)$, the quasiparticles are closely coupled to the lattice and the deviation of their distribution from equilibrium becomes too small for the development of the electronic instability. A more detailed discussion is given in Ref. 18. The shift of our data for La_{1.85}Sr_{0.15}CuO_{4-x} to lower reduced temperatures compared to the data for YBa₂Cu₃O_{7-δ} appears to be due to the fact that in both materials the crossover condition (5) is reached at about the same *absolute* distance of the temperature from T_c . Hence, in La_{1.85}Sr_{0.15}CuO_{4-x} (with the lower T_c) the crossover takes place at a lower value of the *reduced* temperature T/T_c compared to YBa₂Cu₃O_{7-δ}.

 $\tau_{\rm e}^{-1}$ temperature The observed dependence $= a \exp[-2\Delta(T)/k_BT]$ of the quasiparticle energy relaxation rate for the two cuprates suggests that the electron-electron recombination process we have discussed in Sec. III may play a central role, perhaps in combination with the emission of another excitation. However, more theoretical and experimental work is clearly necessary for reaching a satisfactory understanding. For instance, the possible influence of phonon trapping on the quasiparticle energy relaxation rate must be studied. However, because of the small thickness of our samples, we do not expect significant changes from phonon trapping. It is important to note that the quasiparticle energy relaxation rate observed in our measurements may not appear in other experiments studying dissipation due to elastic and/or inelastic quasiparticle scattering and where the deviation of the quasiparticle distribution from equilibrium remains relatively small. As an example, in microwave absorption experiments one often measures mostly the elastic and/or inelastic scattering of quasiparticles and not primarily their energy relaxation. Indeed, the quasiparticle scattering rate below T_c determined from the microwave surface impedance of $YBa_2Cu_3O_{7-\delta}$ films¹⁹ and single crystals²⁰ also decreases with decreasing temperature. However, this decrease is much less rapid than our results for τ_{ε}^{-1} shown in Fig. 6 for the cuprates. A direct comparison with the microwave data is presented in Fig. 4 of Ref. 1.

The fact that the a-Mo₃Si films also show the temperature dependence $\tau_{\varepsilon}^{-1} = a \exp[-2\Delta(T)/k_BT]$ of the quasiparticle energy relaxation rate τ_{ε}^{-1} appears surprising on first sight since in the low- T_c superconductors the electron-phonon interaction is expected to dominate compared to electronelectron scattering. However, the Debye temperature θ_D in amorphous Mo₃Si is relatively high, as shown by the measurements of Mirmel'shteyn et al.²¹ These authors measured the heat capacity of Mo₃Si crystals before and after irradiation with fast neutrons (flux equal to 1×10^{20} n/cm²). Before irradiation, the low-temperature limit of the Debye temperature was found to be $\theta_D = 560$ K. Following the irradiation, this value decreased to $\theta_D = 330$ K. Due to the irradiation, the superconducting transition temperature increased from $T_c = 1.56$ to 6.00 K, i.e., to a value similar to that of our a-Mo₃Si films. Hence, for our a-Mo₃Si films we also expect a value of θ_D near 300 K and it seems possible that the role of electron-phonon scattering is strongly reduced at low temperatures. At any rate, it appears that under the particular nonequilibrium situation of our experiments the electronelectron interaction becomes significant. Our present results on a-Mo₃Si films also appear consistent with earlier experiments using oxygen-doped Al films²² where electronelectron scattering^{23,24} dominates the quasiparticle energy relaxation rate.

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