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Physica C 282–287 (1997) 186–189

PHYSICA C

Evidence for Two-Component Superconductivity in the Femtosecond Optical and Transient Photoconducting Response of $YBa_2Cu_3O_{7-\delta}$.

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Femtosecond time-resolved spectroscopy and transient photoconductivity measurements have been used to investigate the time-dynamics, frequency dependence and symmetry of electronic excitations associated with the opening of the superconducting gap in $YBa_2Cu_3O_{7-\delta}$. Two clearly identifyable relaxation components are observed operative on nanosecond and picosecond timescales respectively, implying the co-existence of localized states and band-like carriers both in the superconducting and normal state. From the temperature dependence of the two components it is clear that both relaxation phenomena are directly connected with the formation of the superconducting condensate. The spectral response for both excitations closely follows the spectrum observed in thermal modulation spectroscopy and implies that the electron-boson coupling function includes a charge-transfer component at optical excitation energies in the region of 1.5-2 eV.

Although the comparison of the measured normal - to - superconducting state optical conductivity ratio σ_s/σ_n in the infrared region of the optical spectrum when compared with theory has been one of the most decisive factors influencing the acceptance of BCS theory for "normal" superconductors, in high-temperature superconductors the optical response has been found to be much more complicated, not least because of the high anisotropy and complex electronic structure of these materials. Thus the temperature dependence of these spectra did not reveal a simple gap picture, but rather showed large temperature-dependent features which were highly anisotropic and in addition to the expected changes associated with the onset of a superconducting transition, significant changes in the optical spectra were observed also above T_c [1] and at energies well above the gap frequency[2]. Because localized state spectra - this applies to optical spectrosopies as well as ARPES for example - are indistinguishable from those originating from itinerant electron bands, the identification of particular spectral features is very difficult on the basis of its spectrum alone. This problem has inevitably resulted in controversy and whereas the low-frequency component has by now almost universally accepted as the electrodynamic (ED) response of in-plane excitations, the so-called mid-infrared feature in the optical conductivity $\sigma(\omega)$ remained controversial. Of all the possible interpretations, the polaronic one has been the most common[3][4], with the implication that localized states states exist in both the normal and superconducting state.

Recently we have found that measurement of the carrier time-relaxation dynamics at different energies and on different timescales using optical spectroscopies can give qualitatively new information regarding the issue of localized versus itinerant states. In recent ultrafast Raman carrier relaxation measurements we have unambiguously found a signature of localised states in the normal state of optimally doped superconducting YBCO and LaSCO samples[5]. To pin down the origin of the excitations more effectively, we have since performed time-resolved measurements of optical reflectivity, transmission and photocurrent on different $YBa_2Cu_3O_{7-\delta}$ samples as a function of temperature both in the normal and superconducting state - the former also as a function of energy in the mid-infrared region - revealing a significant amount of qualitatively new information regarding the dynamics of the super-carriers in these materials. The time-resolved femtosecond measurements on optimally doped YBCO

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PII S0921-4534(97)00190-1

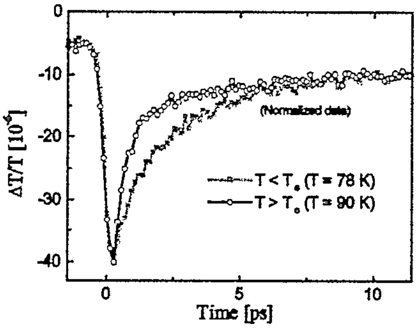

Figure 1. The transmission through an underdoped YBaCuO sample ($T_c = 85$K) as a function of time after photoexcitation by a 1.5 eV laser pulse below and above T_c. (The peak amplitude has been normalized.)

were recently described[6]. The data show a ubiquitous presence of two distinct carrier relaxation processes which occur on very different timescales. The faster relaxation component occurs on picosecond timescales typical of band-carrier relaxation, whereas the slow component displays non-exponential relaxation dynamics on timescales extending from nanoseconds to hundreds of microseconds and beyond[7], characteristic of localized or polaronic states. In Fig. 1 we show the relaxation of the transmission through an YBCO sample above and below T_c. Apart from the almost 10 fold longer lifetime of the fast band-like carrier relaxation in the superconducting state, the amplitude of this signal increases dramatically below T_c and in the optimally doped sample is described quite well by a BCS curve for $T < T_c$ as seen in the bottom panel of Figure 2. Similar measurements on underdoped and entirely undoped samples (see Fig. 2) show a dramatic difference in temperature dependence, and the amplitude can no longer be described by anything resembling a BCS curve. In fact the signal amplitude shows a much slower drop with increasing temperature and eventually drops to zero at near $T = 150$ K. We note that in this sample ($T_c = 85$ K) this temperature $T = 150$ K corresponds closely to the pseudogap temperature T^* found by other measurement techniques. Measurements on different samples show that this T^* is very sensitive to the O concentration. For this reason a large number of samples have been measured with different O concentrations and on different substrates. Although it is not possible to discuss these results in detail in this paper, the measurements confirm qualitatively the behaviour outlined above.

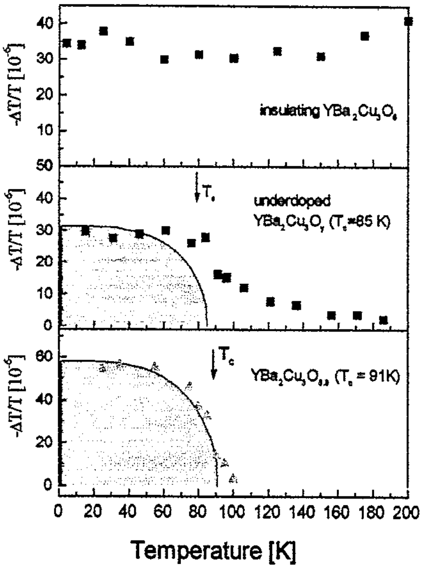

Figure 2. The temperature dependence of the amplitude of the fast carrier relaxation process for undoped (top), underdoped (middle) and an optimally doped sample (bottom).

The temperature dependence of the *slow* signal is qualitatively different than of the fast signal, and in the optimally doped sample the temperature dependence below T_c can be described in terms of an single thermally - activated temperature dependence *below* T_c and a rapid drop in amplitude *above* T_c to a small constant, i.e. temperature-independent value. Consideration of possible processes which can give rise to such behavior has lead us to the conclusion that the long-lifetime states which we are probing in these experiments are thermally activated quasiparticle states, not least because the activation energy in the optimally doped samples $E_a = 2\Delta = 4.0 \pm 0.5 k_B T_c$. For the underdoped sample (middle panel in Figure 3) the peak in the amplitude at T_c is augmented by an additional temperature-independent signal. In the undoped $YBa_2Cu_3O_6$ sample the temperature dependence of the slow signal is essentially monotonic, as expected (see top panel of Figure 3). In this case also, just as in the case of the fast signal discussed above, the measurements on a variety of samples give essentially the same results.

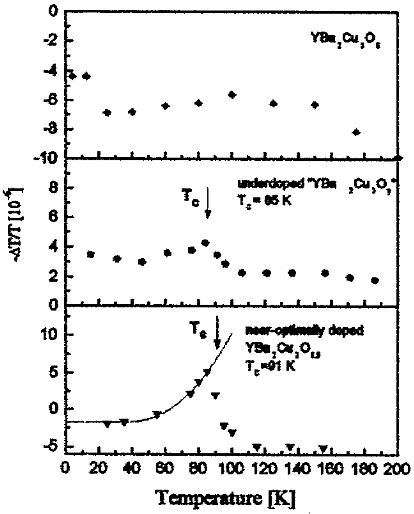

Figure 3. The temperature dependence of the amplitude of the *slow* carrier relaxation process for undoped (top), underdoped (middle) and near-optimally doped samples (bottom). The amplitude of the signal follows thermally activated behaviour below T_c with $E_a = 4 \pm 0.5 kT_c$.

The optical data on quasiparticle relaxation above are complemented by measurements of the photoconducting transient signal recorded as the voltage across a YBCO microbridge after excitation by a short laser pulse. In this case the laser pulse drives the superconductor normal which results in the appearance of a voltage transient across the superconductor. The time-relaxation dynamics are essentially determined by the quasiparticle relaxation dynamics. The most surprising feature of the observed photoresponse signal shown in Fig. 4 is that the signal voltage shows first a fast transient and subsequently drops to a negative value and then relaxes on an unusually long timescale. Detailed analysis and careful modelling shows that this behaviour cannot be described in terms of any kind of bolometric signal. As shown in Figure 4, both the calculated bolometric signal arising from the kinetic inductance and the direct bolometric signal due to change in resistivity of the sample with temperature show qualitatively different time-dependences than the observed signal and have been consequently excluded as possible mechanisms for explaining the observed photoconducting transient response. The long lifetime kinetics are thus apparently independent evidence of long quasiparticle lifetimes already observed in the optical relaxation measurements. We should also mention here that although the dynamics of resistive vortex flow on these timescales is still not understood in detail, we estimate that this contribution to the signal is too small and of the wrong sign to explain the "inductive" signal observed in the photoconductivity.

The general conclusion from these experiments is that there exist two different types of quasiparticle relaxation processes in these materials. The existence of two components in the relaxation do not necessarily mean that we are probing two independent carrier bands, but rather, the data give a clear indication that the supercarriers have a dual character, judging especially from the fact that both the fast and the slow signal have a similar spectral response [6]. This inference of the existence of a dual or composite quasi particle is entirely consistent with the hypothesis discussed

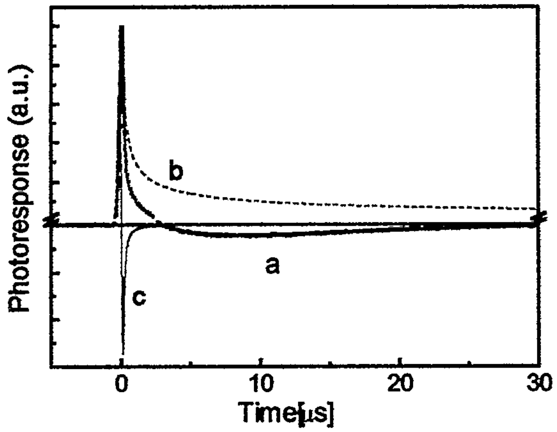

Figure 4. a) The photoconducting response following a laser pulse incident on a microstrip of YBCO. b) is the calculated bolometric response and c) is the calculated kinetic inductance. Clearly the experiment (a) cannot be reconciled with either (b) or (c).

in the introductory paragraph regarding the polaronic and itinerant carrier components in the optical conductivity $\sigma(\omega)$. Moreover the two different relaxation experiments (optical and transport) compare rather well regarding quasiparticle lifetimes, in spite of the fact that the details of the relaxation processes in the two cases are necessarily different.

An important point regarding the spectral dependence of the optical signal is that it matches rather closely the spectral shape of the thermal differential reflectance (TDR) peak at 1.6 eV measured on the same material[2]. In both the time-resolved and TDR measurements the feature at 1.6 eV appears only in the superconducting phase (for optimally doped samples) and is thus naturally assigned to the electrodynamic response of the pairs or the condensate. The corollary is that the non-zero signal in the underdoped samples at temperature above T_c is naturally interpreted as a sign of pairs - possibly bipolarons - in the underdoped materials at intermediate temperatures as suggested by some theories[9].

We wish to acknowledge J.E.Evetts and V.I.Dediu for kindly providing YBCO samples for these experiments.

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