Dynamics of electronic excitations in YBCO at low temperatures

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We present the results of low-temperature optical experiments (absorption and reflection) in visible and middle infrared frequency regions with epitaxial films of copper oxide materials like a $Y_1Ba_2Cu_3O_{6+x}$. Based on our data and reference data, we consider the anomalous spectral effects in HTSC having no analogy with conventional BCS superconductors: (1) the optical response to superconducting transition at T_c; (2) the spectral weight redistribution induced by chemical doping and temperature; (3) the drastic enhancement of low-temperature photodoping; (4) the long spin-structure relaxation via temperature variations seen in the optical spectra. The thorough analysis of the results obtained is fully compatible with the concept of two-component system of light and heavy carriers (holes), being in dynamical coexistence with each other. The dynamical coexistence of the intraband carriers occurs on the background of strongly correlation interrelation of the heavy quasiparticles with the optical interband (charge transfer) excitations.

Introduction

In recent years the optical spectroscopy of HTSC at frequencies much higher than the superconducting gap, $\hbar\omega >> \Delta$, has been found to be informative for understanding the nature of high-T_c superconductivity, in particular, for studying the anomaly of the normal-state properties of HTSC. With regards to room temperature (RT) optical experiments, the following distinct features of the HTSC spectra, which were originally associated with the unusual electron structure, deserve mention: (i) the pronounced optical effect of strongly correlated electrons (holes) under doping, manifesting itself in spectral weight redistribution between the high-energy (interband) region of spectra and the low-energy (intraband) one; (ii) the declination of the optical conductivity from the Drude-like behavior (standard for free carriers) and the appearance of an additional band in the middle infrared region; (iii) the inherent potentiality of photodoping, which results in pumping of carriers under irradiation with light quanta at the frequencies of interband charge transfer transitions above the optical gap $\hbar\omega > \hbar\omega_g \cong 1.6$ eV; (iv) the optical relaxation of the reflection coefficient in the course of aging

and the oxygen ordering of HTSC samples after cooling from high temperature ($T \leq 450$ K) to RT.

The mentioned important results for different HTSC are presented, for example, in [1–6]. All the above features were measured in the optical range from 10^{-2} to 5 eV with different doping regimes of $Y_1Ba_2Cu_3O_{6+x}$: the dielectric phase, $x < 0.3$; the strange metal phase at underdoping, $0.35 < x < 0.8$; the metal at optimal doping, $x \approx 0.9$; the overdoped metal with depressed superconductivity, $x > 0.95$. Because there are two different cuprate structures in the ab geometry of $Y_1Ba_2Cu_3O_{6+x}$–CuO_2 (active plane and CuO_x (chain structure along the b axis), it is essential that the contributions from these planes to absorption and reflection should be separated in experiments with light polarization. This can be most easily done in polarization experiments with $E \parallel a$ and $E \parallel b$ on untwinned crystals. In this respect most intriguing are the data of polarization spectrum measurements on untwinned single crystals of YBaCuO for a and b axes [2,6]. For example, the optical conductivity spectrum of $Y_1Ba_2Cu_3O_{6+x}$ for different doping amounts at RT according [2] is shown in Fig. 1. Below the energy of the charge transfer optical gap, $E \cong 1.6$ eV, separating the
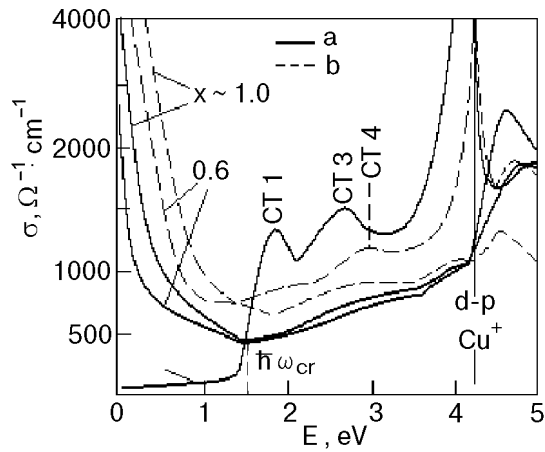
Fig. 1. Optical conductivity of untwinned YBCO crystals for various composition x at RT [2]: along a axis (——); along b axis (– – –).

lower valent and the upper Hubbard bands there are intraband transitions. Above this energy, $E > E_g$, one can observe interband absorption bands with charge transfer from O^{2-} to Cu^{2+} — CT1, CT3, CT4 which are most prominent at low doping. Still higher in energy, at $E = 4.1$ eV, there is a band which belongs to the transition $(d–p)$ in the local center of Cu^+ of CuO_x plane. Figure 1 clearly shows the evolution of optical conductivity spectra with doping from the dielectric to a superconducting state. The correlation effect in the spectra with doping is revealed in the form of integral spectral weight redistribution from the interband charge transfer (CT) transitions in the visible (VIS) region to the intraband transitions in the middle infrared one (MIR). The spectral change indicates that the conduction and valence band states of YBCO insulator are redistributed by doping to construct new states which give rise to low-energy excitations below crossover point $\hbar\omega_{cr}$. As seen from Fig. 1, there is the crossover point ω_{cr} at which $\sigma_1(\omega)$ is constant with doping. It should be mentioned that in general the existence of the doping-independent crossover point ω_{cr} suggests that the optical gap in $Y_1Ba_2Cu_3O_{6+x}$ is not generated by the charge density wave (the spectrum of optical excitations associated with the charge density wave shifts dramatically to a red region as a whole on doping). The optical spectra of YBaCuO in the vicinity of intraband transitions have a Drude peak, centered at $\omega = 0$ with width of $1/T$ and additional spectral weight in the MIR region (with the MIR maximum at 0.6 eV). It is the occurrence of additional absorption in the MIR region that results in the difference of the frequency dependence $\sigma(\omega)$ from the law $\sigma(\omega) \sim \omega^{-2}$ expected for the ordinary Fermi liquid.

And that once entailed the need for development of new concepts of HTSC electronic spectrum (for instance, the concept of «marginal» and «nesting» Fermi-liquid [7]). It should be emphasized that in conventional BCS-superconductors no radically new optical effect were observed at low temperatures (LT) as compared to RT in the region $\hbar\omega \gg \Delta$, moreover, no optical response to the emergence of the SC state was detected. In the case of HTSC the LT dynamics of charges displays new effects which would signaled on precursory processes to superconductivity.

This paper presents the experimental results of later years on some nonordinary spectral properties of the normal phase of $Y_1Ba_2Cu_3O_{6+x}$ in VIS and MIR, revealed at different regimes of doping and temperature variations within the LT range below $T = 200$ K down to 20 K. In the main these are the data of investigations under the Program of the National Academy of Sciences of Ukraine. All optical measurements were carried out on thin epitaxial films $l \approx 2000–3000$ Å in thickness oriented parallel to the ab plane. The films were grown in German scientific centers (University of Erlangen; Munich Technical University). The films were tested for their electrical, magnetic and structure characteristics. Some details of the LT optical measurments were described in [8].

At present it can be said with confidence that the hole subsystem dynamics in HTSC materials is largely determined by the Fermi-liquid effects and the strong correlation interaction between intra- and interband transitions. However, up to now the correlation redistribution along spectra has been studied only for the doping effect at RT and has not been considered for other processes of charge dynamics. According to the data [8] and the results presented here, the integral redistribution of spectral weight in HTSC also occurs in experiments with temperature variations, in photoinduced spectra, in experiments with provocation of spin-structure instability and so on. Therefore, the LT spectral data should be analyzed from the general viewpoint. We would like to stress that all LT features in the MIR and VIS spectra can be naturally examined in terms of the Mott-Hubbard model of $2D$ strongly correlated electrons with taking into account the intense spin fluctuations at LT.

Optical response to SC transition at T_c and temperature dependences of spectral functions

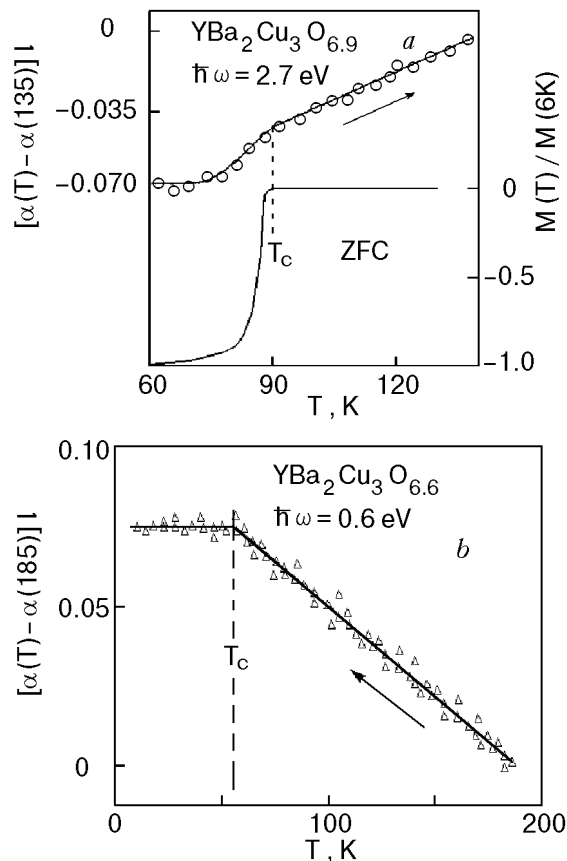One of the unexpected spectroscopic results in copper oxides was the finding of sensitivity to the

Fig. 2. Temperature dependences of the differential absorption coefficient for VIS (*a*) and MIR (*b*) with different *x* (in (*a*) shown are magnetic measurements of the film for ZFC regime also).

onset of superconductivity for the transmitted electromagnetic field of optical frequencies $\hbar\omega \gg \Delta$. The optical response in absorption and reflection to T_c was detected both in VIS and MIR [9–12]. The subsequent experiments confirmed the occurrence of this peculiar effect in YBaCuO and other HTSC in a wide range of doping. It was shown that the response arose under optical transitions belonging to the conducting plane of CuO_2. The *T* dependence of the differential absorption coefficient at fixed frequency $\Delta\alpha_\omega(T)l = [\alpha_\omega(T) - \alpha_\omega(T_0)]l$ (for the CT maxima at 2.6 eV) is shown in Fig. 2,*a* and that for the differential absorption coefficient of the MIR maximum at 0.6 eV is shown in Fig. 2,*b*. As it can be seen, the T_c point coincides closely with the the kink of the functions $\alpha_\omega(T)$. Note that the optical response to the SC transition in HTSC was also detected in luminescence spectra on oxygen *F* centers at frequencies near 2–3 eV [13].

The temperature behavior of the optical spectra in YBaCuO was found to be uncommon. Below T_c one can observe a continuos freezing of the *T* dependences. Particularly striking is the *T* behavior of $\alpha_\omega(T)$ and $R_\omega(T)$ for the normal state at $T > T_c$:

for the regimes of underdoping and optical doping the *T* dependences of the absorption and reflection coefficients have different signs in the VIS and MIR region: both $\alpha_\omega(T)$ and $R_\omega(T)$ are increasing functions of *T* in VIS and they are decreasing ones of *T* in MIR (see Fig. 2,*a,b* and [8,10]). However, in the overdoping regime ($x = 0.95$) the CT transitions in VIS undergo fundamental changes with temperature and the VIS spectral function (*T*) measured decreases with temperature similar to the MIR one [8]. It is like that the correlation spectral weight redistribution is terminated in the overdoping regime where YBaCuO becomes an uncorrelated metal, in which the upper and lower Hubbard bands are merged together.

Take note that the dependences $\alpha_\omega(T)$ and $R_\omega(T)$ have no selective character in frequency and the *T* changes touch on the whole wide region in the VIS and MIR spectra. The differential absorption spectra $\Delta\alpha(\omega)l = (\alpha^f(\omega) - \alpha^i(\omega))l$ (where *f* and *i* mark the final and initial conditions of the measurements) are shown in Fig. 3,*a* for doping and in Fig. 3,*b* for cooling from 250 to 80 K (for comparison). In both cases one can observe a decrease in the integral spectral weight of the CT absorption in
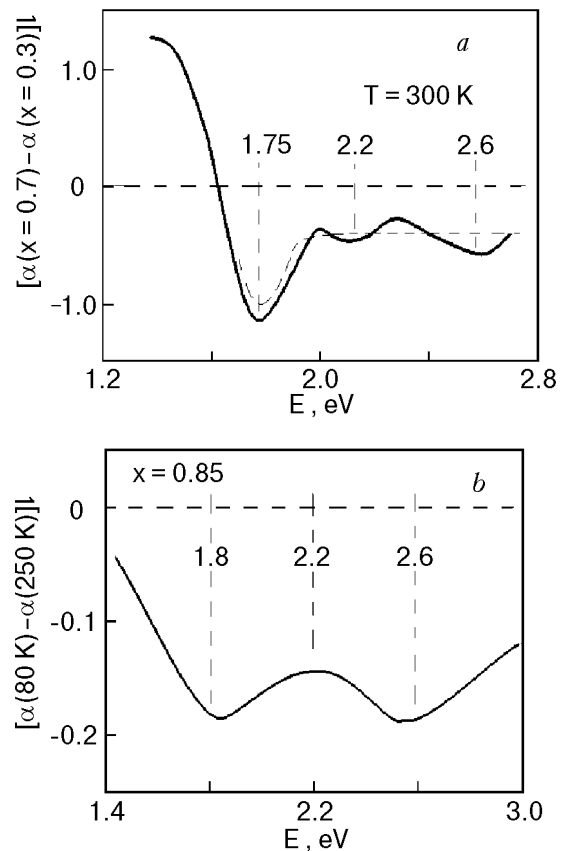

Fig. 3. The differential absorption spectra in YBCO in the CT interband region of frequencies: on doping from $x = 0.3$ to 0.7 at RT (*a*); on cooling from $T = 250$ K to 80 K for $x = 0.85$ (*b*).
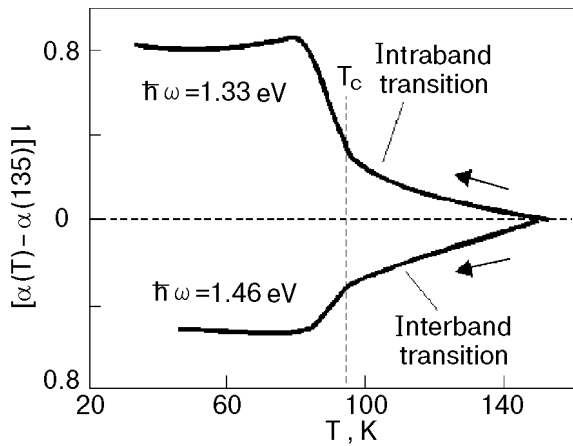
Fig. 4. Temperature dependences of the differential absorption coefficient for two frequences in the vicinity of the crossover point (the upper curve for $\hbar\omega < \hbar\omega_{cr}$, the lower curve for $\hbar\omega > \hbar\omega_{cr}$).

favour of the MIR absorption under cooling as well as under doping (but the scale of the T effect is much smaller than that of the doping one). Another remarkable common feature of the spectral evolution under doping and T variations is the existence of the crossover point ω_{cr} positioning near E_g and dividing the VIS interband and the MIR intraband transitions: $\omega_{cr} = 1.55$ eV at doping and $\omega_{cr} = 1.4$ eV at T variations (Fig. 4). It is seen that the overall integral spectral weight redistribution from VIS to MIR occurs on around the crossover point at which $\Delta\alpha_{\omega} = 0$.

The above data on sensitivity of spectral functions to T_c and on their different signs of the temperature dependences above and below the crossover point are in agreement with the results in [14]. In this work the ratio of reflection coefficients for the superconducting and the normal phases near T_c was measured by applying the temperature modulation of HTSC samples. The data of the measurements are shown in Fig. 5 for a BiCaCuO
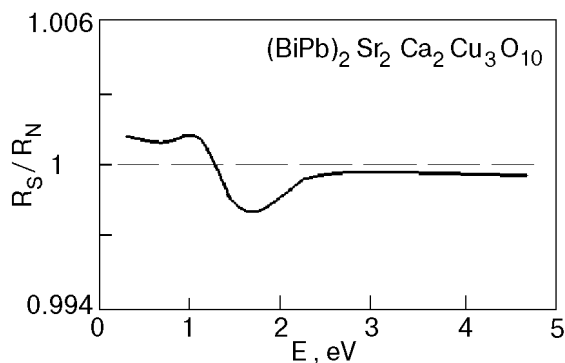

Fig. 5. The superconducting to normal-state reflectance ratio, R_s/R_n of Bi-contained HTSC at $T = 90$ K (R_s/R_n deviates significantly from unity at energies 0.5–2.5 eV) [14].

film. As is evident, the response of the reflection spectra to the SC transition is prominent for the 1.0–2.5 eV region, within which there is an crossover point ($\hbar\omega \cong 1.4$ eV). Since the temperature modulation range nearby T_c was quite narrow ($\pm$ 5 K), it is believed that the peculiarity nearby T_c is jumplike in character. It should be noted that by the data on reflection shown in Fig. 5 the jump scale is $\Delta R/R \cong 0.3\%$ while by our data on absorption (Fig. 4) it is equal to $\Delta\alpha/\alpha \cong 1–2\%$.

The data shown suggest an idea of the T-induced correlation redistribution between VIS and MIR spectra and the strong coupling of oscillators at CT and MIR transitions. Our data are interesting because they were obtained at LT under the conditions where the charge transfer between CuO_2 and CuO_x planes is hindered. It is known that with decrease in temperature down to the pseudogap state, the interaction between the CuO_2 and the CuO_x planes is sharply diminished (there occurs a strengthening of the two-dimensionality of the CuO_2 plane), as evidenced by the considerable reduction in the optical conductivity along the c axis at frequencies of the scale of the pseudogap value $\hbar\omega < 40$ meV [6]. It is obvious that this process reflects the freezing of charge transfer between the different planes of the YBCO cell.

So, there exists an optical response of all the spectral functions at frequencies hundreds times higher than the superconducting gap which is opened under the transition of HTSC systems to a superconducting state. It may be assumed that this nonconventional effect in HTSC is responsible for by a high ratio $(\Delta/\varepsilon_F)^2 \cong 10^{-2}$ which determines the effect scale and is hundreds times more than that in conventional BCS materials. The optical response mechanism was considered theoretically from different viewpoints in Refs. 15, 16 and still remains debatable. Moreover, the sensitivity of spectral functions to T_c occurs against the background of temperature-induced correlation mixture low- and high-frequency excitations. Therefore, the optical response suggests that either (i) superconditivity stimulates an extra additional correlation redistribution of excitations or (ii) high-frequency excitations of 1 eV are directly involved in the mechanism of high-T_c superconductivity.

Photodoping at low temperatures

One of the important questions of the optical spectroscopy of HTSC is the question of photoirradiation critical dose, D_0, and the threshold spectral energy, $\hbar\omega_{th}$, which determine permissible doses and energies of photoirradiation without persistent
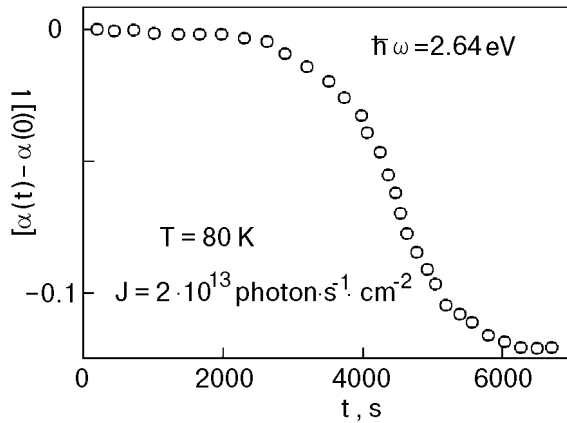
Fig. 6. Dose dependence of the absorption in YBCO with $x = 0.35$ under irradiation and at measurements with $\hbar\omega = 2.64$ eV. At doses $D = Jt > 10^{17}$ photon·cm^{-2} the sharp decrease of VIS absorption occurs due to photoinduced insulator-metal phase transition.
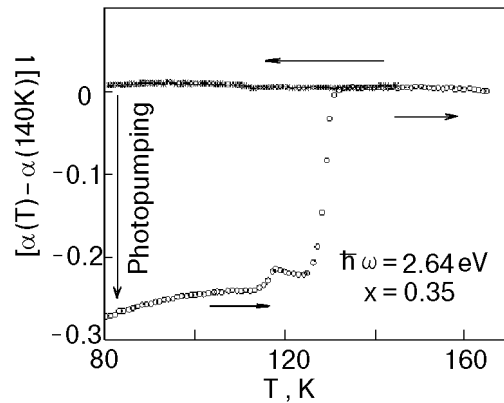

Fig. 7. Temperature dependences of the differential absorption coefficient in YBCO with $x = 0.35$ before (the upper curve) and after (the lower curve) LT photopumping with $\hbar\omega = 2.64$ eV, $D = 10^{17}$ photon·cm^{-2}.

photoinduced effects in the lattice structure and in the current characteristics. It is known that a HTSC material is sensitive to exposure dose in VIS with the threshold spectral energy $\hbar\omega_{th} > \hbar\omega_g$ (in the region of the CT transitions) and with the doses $D > D_0 \cong 10^{19-20}$ photon/cm^2. That is, using these conditions of photoirradiation at RT it becomes possible to observe an enhancement of T_c in HTSC underdoped samples after photopumping [5,17,18]. The accepted explanation of the photoinduced conductivity is based on the assumption that the photo-excitation at RT causes the ordering of oxygen vacancies in the CuO$_x$ chains that in turn leads to a subsequent transfer of electrons between the CuO$_2$ and the CuO$_x$ planes and the emergence of excess holes in CuO$_2$ [17].

At the same time in some works it is found that the threshold dose of photoinduced effects become less by about two orders of magnitude if photopumping is carried out at low temperatures, $D_0 \approx 10^{17}$ photon/cm^2 [9,19]. The progressive increase of photodoping and the LT photoinduced phase transition of Y$_1$Ba$_2$Cu$_3$O$_{6.35}$ from the dielectric to a metallic state can be traced by the dose dependence of the absorption coefficient. Figure 6 shows the changes in the absorption coefficient as compared to the initial moment of irradiation with a light of $J = 2 \cdot 10^{13}$ photon·s^{-1}·cm^{-2} at 80 K of Y$_1$Ba$_2$Cu$_3$O$_{6+x}$ ($x = 0.35$). As is evident, there occurs a sharp passage of the dependence $\Delta\alpha_\omega(t)$ to saturation at $D > Jt \approx 10^{17}$ photon·cm^{-2}, suggesting the insulator—metal phase transition. It should be mentioned that the phase transition under LT photopumping follows the pattern of the second-kind phase transition. In Fig. 7 the process manifestation

of YBaCuO metallization under LT photopumping is seen from the temperature dependence of the absorption coefficient. The upper curve (on cooling the sample) corresponds to the dielectric state of YBaCuO with $x = 0.35$ prior photopumping. As is evident, in this case the behavior of $\alpha(T)$ is temperature independent, which is typical of the insulator phase. After exposure of the sample to the light flux of $D = 10^{17}$ photon·cm^{-2} at 80 K and with subsequent increase in temperature, one can observe a temperature dependence of absorption typical of HTSC metals (compare with Fig. 2,b). At $T = 135$ K a sharp reduction in the photopumped holes occurs due to their recombination with electrons so that the sample reverts to the original initial insulator state. The temperature of the reduction in the photopumped carriers (the threshold temperature) is dependent on doping of samples. The threshold temperature $T_{th} = 135$ K was also observed in [4] by the decrease weakening of metallization on the curves $\alpha(T)$.
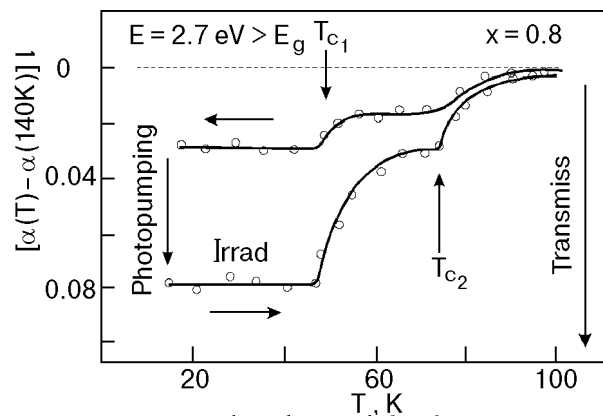

Fig. 8. Temperature dependences of the absorption in YBCO with $x = 0.8$ before (the upper curve) and after (the lower curve) LT photopumping into SC phase with $\hbar\omega = 2.7$ eV, $D \geq 10^{17}$ photon·cm^{-2}.

The effect of photodoping is also observed at immediate pumping into the superconducting phase. The experimental results for a two-phase sample with SC transitions at $T_{c1} = 50$ K and $T_{c2} = 75$ K are illustrated in Fig. 8. After the LT photopumping the sample is seen to become more transparent and with subsequent heating it returns to the initial state. The sharp decrease in the nonequilibrium concentration of photopumped holes is occurring at superconducting transition temperatures. We assume that the superconducting state is favorable to the retention of photopumped holes. As is seen from Fig. 9, the photopumping results in the correlation spectral weight redistribution between VIS and MIR, i.e., it acts like chemical doping. We can see a decrease in the integral absorption coefficient in $Y_1Ba_2Cu_3O_{6.8}$ after LT photopumping, i.e., due to the generation of nonequilibrium extra holes.

Our understanding of the process of persistent photodoping at LT is distinguished from the accepted ones at RT and is as follows. We think that the photoinduced holes appear in the valence band as a result of the charge-transfer excitations in the CuO_2 plane with $\hbar\omega > E_g = 1.6$ eV ($Cu^{2+}O^{2+} \overset{\hbar\omega}{\rightarrow} \overset{\hbar\omega}{\rightarrow} Cu^+O^-$). Thus photopumping will be persistent if some obstacles to a reversible process exist. We suggest that a new process arises at LT photopumping that hampers the reversible recombination. This new process is considered in [20]. It is based on the concept of appropriate generation of low-temperature heavy quasi-particles—fluctuons. The physical concept of fluctuons was first introduced in the physics of solids by I. Lifshitz [21], and their phenomenological model was developed in [22]. In HTSC systems a hole fluctuon in the active CuO_2 plane is easily generated through hole trapping by spin fluctuations inherent in the pseudogap state at $T < T^*$. In the $2D$ case particularly deep fluctuon wells are generated with reduction in temperature, the well
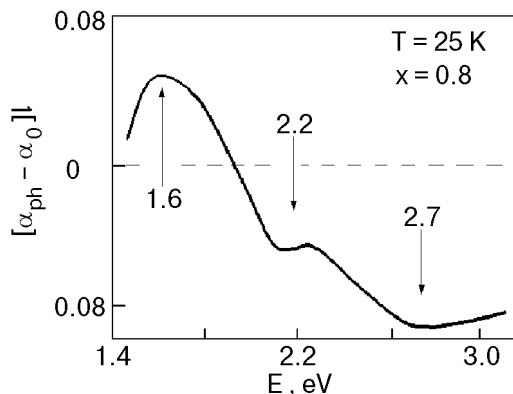
depth increasing as T^{-2} [20]. It is the hole trapping by these spin fluctuons that results in the retention and accumulation of photoinduced holes. As the number of photoinduced holes increases, the fluctuon-fluctuon interaction determines a nonlinear process of photopumping (Fig. 6) and gives rise to an infinite metallic cluster following the pattern of the second-order phase transition.

So, the LT photopumping of HTSC at $T < T^* \approx 150–200$ K (in the pseudogap state) with reduced threshold doses may be attributed to the effective hole trapping by spin fluctuations followed by generation of heavy fluctuons. It is likely that it is heavy fluctuons that determines a temperature- and photo-dependent addition to the spectral weight of the MIR region at LT, resulting in the correlation redistribution in the CuO_2 plane.

The LT relaxation in YBCO samples observed in optical spectroscopy

Yet another low-temperature peculiarity in the behavior of spectral functions of HTSC at $T < T^*$ was observed with varying direction and velocity of T scanning of the samples, namely, hysteresis effects in absorption and reflection. The hysteresis loop under cooling and heating of YBaCuO was mentioned in our former work for VIS absorption (Fig. 10) [12] and then subsequently the same effect was observed in measurements of MIR absorption (Fig. 2) and reflection (Fig. 11). As is evident from Figs. 10 and 11, as the samples are cooled, there occurs an accumulation of nonequilibrium that may be discarded (completely or partially) if the sample is held at LT for Δt. The set of our data suggests that the upper temperature boundary of instability is a temperature in the vicinity of the phase transition from the pseudogap
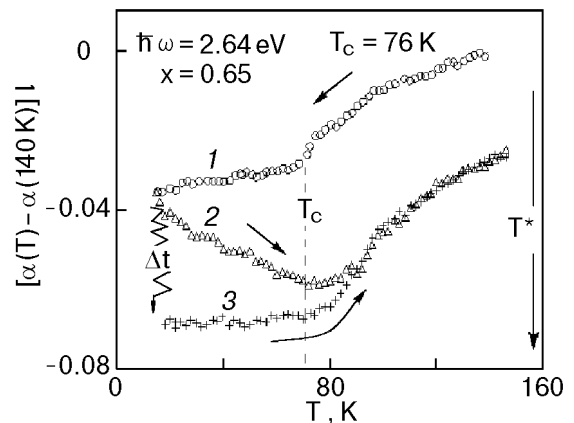

Fig. 9. The differential absorption spectra of YBCO with $x = 0.8$ in the CT interband region of frequencies after LT photopumping.

Fig. 10. Nonreversible effect in VIS absorption upon cooling and heating of the underdoped YBCO. Curves *1* and *2* were measured with uninterrupted cooling and heating. Curve *3* was measured after a long pause as 20 K ($\Delta t \approx 40$ min before heating).
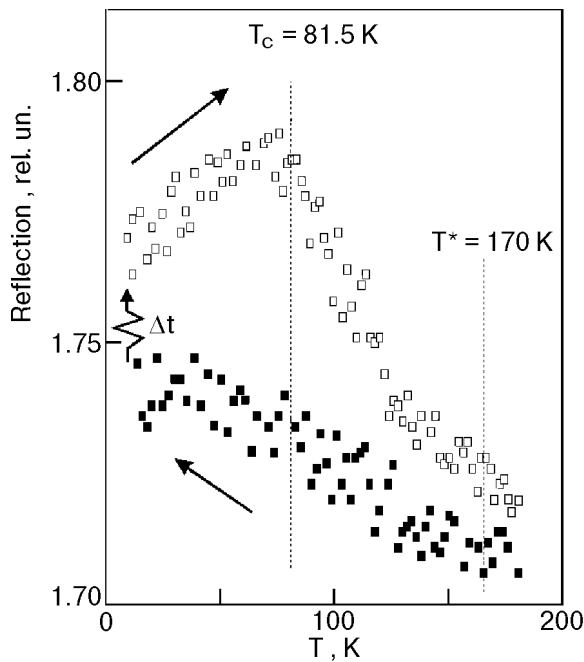
Fig. 11. The reflection spectra of YBCO in MIR region at $\hbar\omega = 0.6$ eV upon cooling and heating.

state to states with disordered spins, T^*. The most pronounced hysteresis (electron-structure instability) is displayed by underdoped samples. With increasing doping, the loop area in the spectral function-temperature coordinates reduces, and the loop itself shifts towards lower temperatures. For samples of optimal doping, the hysteresis effects are minimum and may be observed with rather fast temperature cycling between the SC and N phases solely within the $T < T_c$ range. Generally speaking, the occurrence of hysteresis effects is associated with the existence of relaxation processes within a hysteresis loop temperature range. It is important to stress that the hysteresis loop in the MIR spectra is temperature inverse to that of the VIS spectral measurements. This means that the absorption (re-
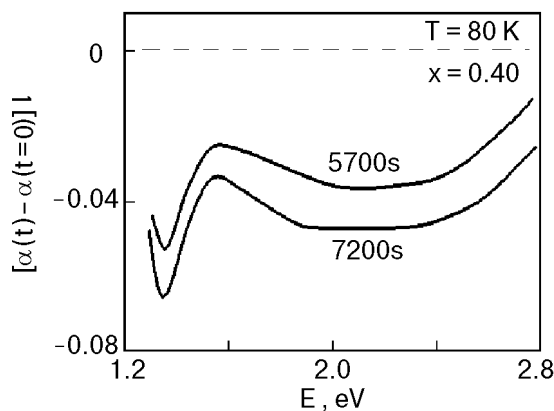
flection) coefficient undergoes changes in different directions in VIS and MIR during relaxation (Figs. 10, 11). All the experiments with instabilities of spectra were performed with low light doses ($D < D_0$). We also carried out a special test-measurement with a dark window during relaxation and therefore we are convinced that the instabilities observed are not associated with the photoinduced effect. Note that the resistive measurements in different HTSC films exhibited no hysteresis effects with a loop span of several percents.

The differential spectra of VIS absorption of a sample with $x = 0.4$ (in the vicinity of dielectric — metal transition) measured for two relaxation times ($t_1 = 5700$ s and $t_2 = 7200$ s) after cooling from 220 to 88 K are plotted in Fig. 12. It can be seen that the absorption decreases through the whole VIS region, i.e., the integral bleaching of the CT transitions occurs. The characteristic time of LT-relaxation could be estimated as $\tau \cong (1-2)\cdot 10^3$ s. Time modifications in the absorption and reflection spectra are also observed in the MIR region after rapid cooling down to a certain temperature. These modifications occur with increasing both the integral MIR absorption and the MIR reflection, i.e., during relaxation the MIR and VIS optical characteristics undergo changes in different directions just as it occurs under chemical or photodoping. At the same time, in the near UV region for $\hbar\omega = 4.1$ eV, in which there are optical transitions ($3d$–$4p$) on local centers of Cu^+, one cannot observe relaxation effects in absorption at LT, i.e., the concentration of monovalent Cu^+ ions in the chain structure of CuO_x remains unchanged after temperature variations in the $T < T^*$ region (Fig. 13).

Figure 14 presents relaxation curves for the underdoped metal film of $Y_1Ba_2Cu_3O_{6+x}$ with $x = 0.7$ at $T_c = 70$ K. The data for different temperatures

Fig. 12. The differential absorption spectra of YBCO with $x = 0.4$ in CT interband region of frequencies at different time of relaxation after fast cooling.
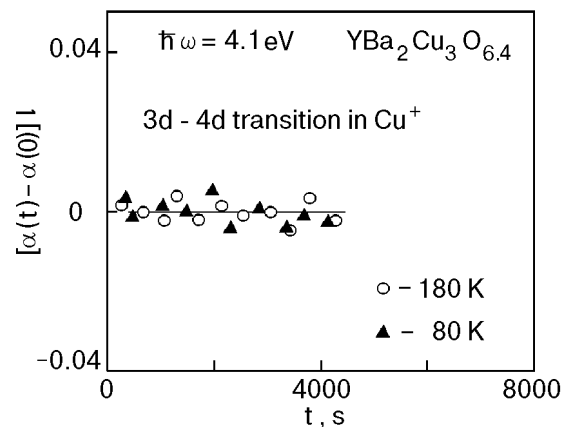

Fig. 13. Time dependence of the differential absorption coefficient at $\hbar\omega = 4.1$ eV after fast cooling of YBCO from 220 K to 180 K (O) and to 80 K (▲).
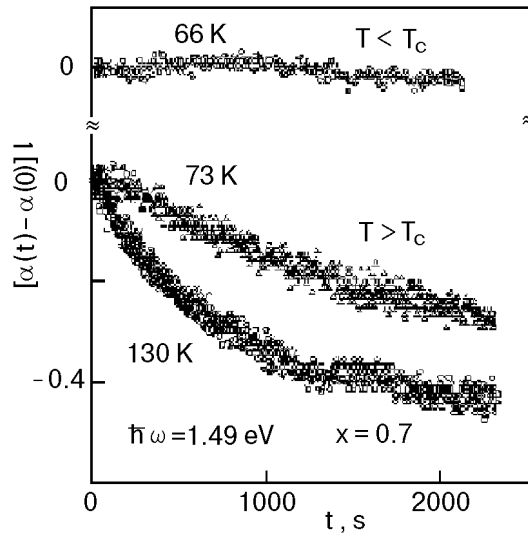
Fig. 14. Time dependences of the differential absorption coefficient at $\hbar\omega = 1.49$ eV in process of relaxation after cooling to different temperatures $T > T_c = 70$ K and $T < T_c$.

were obtained in sequential measurements upon cooling from 250 K. Differential absorption $\Delta\alpha l = = (\alpha^f - \alpha^i)$ vs. T is plotted relative to the initial moment of temperature stabilization (T_i) of α_i $(t = 0)$. After each previous case where the curve (t) reached saturation at a certain temperature, the temperature was quickly decreased to a next level and relaxation was measured once again and so on. On this slow cooling, upon reaching the equilibrium state (saturation) one can see that relaxation is absent below $T < T_c$.

It seems to be likely that the T-induced time instability revealed in VIS optical absorption of CT transitions, which belong to the CuO_2 plane, reflects the inherent electron (spin)-structure relaxation in the active CuO_2 plane at LT variations. In the course of this relaxation the spectral weight redistribution from the VIS to the MIR region occurs. If we accept this assumption, two questions arise immediately: 1) what is the nature of the electron-structure relaxation at low temperatures? 2) what is the reason why the LT relaxation is not so clearly observed in experiments on the dc resistivity?

Let us remember that the T-induced structure relaxation resulting in ortho-ordering and variations of spectral functions of HTSC samples after fast cooling from high temperature (400 °C) down to RT are described in a series of papers [3,23]. The commonly accepted model of RT relaxation of an order parameter is based on the mechanism of diffusion rearrangement of oxygen vacancies making the copper-oxygen chain (Cu–O–Cu) along the b-axis

in CuO_x longer and following the charge transfer between the CuO_x and CuO_2 planes. One of the arguments in favor of this model was the evidence for the variations of the Cu^+ concentration (the variations observed in the optical reflection of the $(3d-4d)$ transitions of the Cu^+ ions at 4.1 eV vs. time [3]).

In contrast to the RT relaxation of the spectral functions, the LT relaxation of the CT interband and MIR intraband spectral weights is not accompanied by variations of the Cu^+ concentration (Fig. 13). This result is against the electron-structure rearrangement in CuO_x, where the Cu^+ ions reside, and the evidences that the LT relaxation occurs exactly in the CuO_2 plane without any charge transfer between the planes (the interplane charge transfer is likely to be hindered at LT). We assume that the most reliable explanation of the LT relaxation of the spectral functions in VIS and MIR is the temperature-induced spin structure rearrangement in the Cu^+ plane due to the formation of hole-spin fluctuons at LT. Strong influence of the system prehistory and its cooling rate on the LT state and long relaxation times in underdoped HTSC suggest that these are similar to irreversible effects in nonergodic systems with competing interactions (for example, to structural or spin glasses [24]). From this viewpoint, the transition to the pseudogap state at T^* may be considered as a transition to a nonergodic system where ordering regions alternate with disordering ones. In the fluctuon model the ordering occurs within a region round a hole trapped by spins. Note also that the concept of holes trapped in a spin bag [25] as well as the concept of holes trapped in the «string» potential created by AF background [26], or the model of stripes, that is the subject of wide speculation [27], are not dramatically different from our pattern of hole-spin fluctuons.

Thus, we think it quite reasonable to associate the LT instability of optical spectra and the effective photodoping at low temperatures with two processes — the hole trapping by spin fluctuations at low temperature and the induced spectral weight redistribution from VIS to MIR (due to strong correlation).

In parallel with the fluctuon domains where the trapped heavy holes reside, there exists a current network for light holes. Therefore, the light and heavy holes are in dynamic equilibrium resulting from the competition between the superexchange energy lost near a trapped hole and its kinetic energy. The spectral weight of intraband transitions is distributed between the contributions from light

and heavy carriers. Note that in the MIR region the contribution from heavy holes is dominant while in the far low-frequency region ($\omega \to 0$) the contribution from light holes is prevalent. The measurements of HTSC sample resistivity ($\omega \to 0$) reveal mainly the contribution of light carriers not related to the spin-structure relaxation. d.c. conductivity $\sigma(0)$ is defined by plasma frequency $\omega_p^2 \sim n_L$ (T, t) and damping $\Gamma(T, t)$ (where n_L is the number of light carriers): $\sigma(0) \sim n/\Gamma$. The relative changes $\Delta\sigma/\sigma$ in the experiments with varying T (or due to relaxation) are $\Delta\sigma/\sigma = \Delta n/n - \Delta\Gamma/\Gamma$. As is evident from the optical measurements, $\Delta n/n \sim \Delta\alpha/\alpha \simeq 5\%$. At the same time $\Delta\Gamma/\Gamma \sim \Delta T/T$ and amounts to 50%. Therefore, unlike the optical measurements, the resistive ones do not sense relaxation and temperature variations associated with the number of carriers and show mainly the T variations in damping of light carriers.

So, the optical data reveal the dramatic effects of nonequilibrium inherent in HTSC systems after their fast cooling to the pseudogap state region below T^*. The effects are most pronounced for underdoped samples and much weaker for the optimal doping regime. Generally speaking, the high nonequilibrium resulted from the competitive interactions occurring in the HTSC system which bring two types of carriers (holes) with different spin surrounding into dynamic coexistence: light (coherent) holes and heavy holes trapped in a spin bag. As a result, the optical conductivity is determined by two contributions — a coherent part at the expense of Drude-like light carriers and a noncoherent one at the expense of heavy hole fluctuons. The optical relaxation observed arises from the self-consistent hole trapping by spin fluctuations and the subsequent spin ordering inside a fluctuon. In this case the ordering time is supposed to be $\tau \simeq \exp(a \sqrt{N})$, where $N >> 1$, the number of spins in a fluctuon, may be rather high.

Conclusion

After the discovery of high-T_c superconductivity a great number of works were concentrated on the problem of adequate description of the behavior of electrons in anisotropic cuprates. For the analysis of the above optical results we used the consequences of the theoretical concepts developed in a series of theoretical works as a basis. These works are directly or indirectly connected with a broader Hubbard-like model of strongly correlated $2D$ electrons submerged in spin media [26, 28–33]. One of the important general results of these theories is that the behavior of holes in the AF background is

accounted for by the competition between the superexchange energy lost near a hole and its kinetic energy. Thus, the Hubbard-like model evidences that there is a dynamical coexistence of mobile (coherent) holes and heavy holes trapped in a spin bag. The impressive calculations of the temperature evolution of DOS in terms of the Hubbard model are carried out in [33]. Away from half-filling of a lower Hubbard band a sharp resonance peak appears near the chemical potential. As the doping is increased the resonance width also increases and it starts to merge with the lower Hubbard band. For the overdoping regime, both low-energy peaks are indistinguishable, implying that the system has become an uncorrelated ordinary metal. The narrow peak in DOS evolves like the MIR band in the optical spectra. The narrow peak appears to be strongly temperature dependent, thus it is pronounced only in the region of low temperatures and within a regime that extends from underdoping to optical doping.

Recent low-temperature experimental and theoretical works have made it clear that in HTSC materials there appears a state below $T < T^*$ that is precursory to superconductivity. The temperature T^* coincides with the temperature of the pseudogap opening with spin singlet ordering. This LT precursory region turns out to be optically active in the absorption and reflection spectra. Using the basic consequences of the theory of correlated electrons and the optical experimental findings, we propose the following principal ansatze:

1. The consistent concept of strongly correlated hole excitations interacting with spin fluctuations in the CuO_2 plane is attractive for understanding and interpreting the anomalous optical effects in HTSC at LT having no analogue with conventional BCS superconductors (the sensitivity to T_c, the T-induced spectral weight redistribution, the LT relaxation of optical functions, the LT-enhanced photodoping).

2. A nontrivial consequence of the correlation model in the intermediate regime is the coexistence of light (Drude-like) and heavy (hole-spin-fluctuons) holes, the latter being generated in the LT region below the temperature of spin ordering (opening spin gap). Thus, the high-T_c superconductive scenario is realized in dynamical two-component systems of carriers.

3. Subsystems of light and heavy carriers are in dynamical equilibrium upon doping or T variations. If the doping of HTSC is increased the localization effects are weakened, while if the temperature is decreased, the localization effects become stronger

and the density of heavy trapped holes increases. Thus, the temperature serves as fine tuning for high-T_c superconductivity.

Acknowledgments

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