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METHODS FOR RESEARCHING THE LOCALIZATION AND DELOCALIZATION OF CARRIERS IN YBA2CU3O6+X FILMS

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Abstract

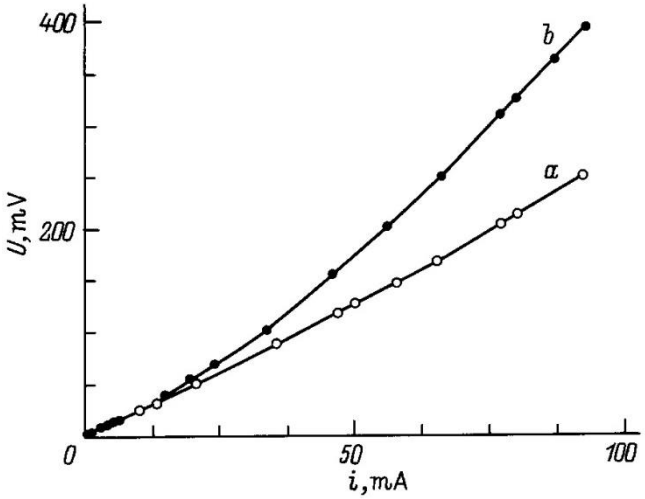
The results of the study of optical absorption of metal films $YBa_2Cu_3O_{6+x}$ when passing through the film direct current to 100 mA are presented. Analysis of the data indicates that the relationship between absorption and current arises due to the possibility of filling the localization areas with carriers in the current mode (or vice versa, due to the emission of localized carriers to the mobility level). Fdor films with substantially nonlinear voltammper characteristic, a strong effect of current on the value of interzone absorption was found. It has been experimentally established that the EO-effect occurs at interzone transitions only in the mode of nonlinear VAC, the amplitude of the effect increases with the growth of current (voltage), but with the linearization of VAC EO-effect disappears. In the field of optical transitions at local levels, this effect was not observed. The analysis of results from the point of view of carrier localization and delocalization processes.

Key words: *volt-ampere characteristics, high-temperature superconductors, electro-optical effect, localization delocalization.*

The competition of the processes of localization and delocalization of carriers largely determines the parameters of high-temperature superconductors (HTSE) in the normal and superconducting phases. At present, optical spectroscopy of absorption and reflection in the visible and infrared spectral ranges is widely used to diagnose these processes in HTSC systems [1–3]. This work presents the results of the detection of the strong electro-optical effect (EO-effect) on $YBa_2Cu_3O_{6+x}$ metal films, which can be considered as a new method of studying the delocalization localization processes. The essence of the effect is that the optical absorption of $YBa_2Cu_3O_{6+x}$ films changes when a direct current is transmitted through the film. An analysis of the data indicates that the relationship between absorption and current arises due to the possibility of carriers filling localization areas in the current mode (or, conversely, due to the release of localized carriers to the mobility level).

Electro-optical measurements were made at 300 K for a series of *c* - oriented metal films $YBa_2Cu_3O_{6+x}$ (x = 0.6−0.9) with a thickness of l= 2500Å ($SrTiO_3$ substrates). In the current range up to i=100mA the films had linear and nonlinear volt-ampere characteristics (VAC), but the most significant experiments were the experiments in which the film with initially linear VAC was switched to the mode with nonlinear VAC and back. The optical absorption of these films depending on the current was measured in the spectral range $\hbar\omega \gtrsim$ 1.4eV. Here, at $\hbar\omega \lesssim$ 2eV, transitions at local levels are concentrated, mainly of d−d character in Cu^{2+} [4], and at 2eV - interzone transitions between the valence zone and the upper (empty) Hubbard zone. These interzone transitions with charge transfer from oxygen to copper belong to the CuO_2 metal plane [1,3]. Note that for films $YBa_2Cu_3O_{6+x}$ films, the currents used by us up to 100 mA (current density $\cong 4 \cdot 10^3$ A/cm^2, fields <5V/cm) do not cause noticeable bolometric effects [5] and do not

lead to the oxygen electro-migration along the lattice, which begins at the threshold level $\cong 10^6$ A/cm^2 [6]. It has been experimentally established that the EO-effect occurs at interzone transitions only in the nonlinear VAC mode, the amplitude of the effect increases with the increase of current (voltage), but with the linearization of VAC the EO-effect disappears. Fig. 1 shows VAC for the film, which originally had a weakly nonlinear VAC (curve a in Fig. 1), and then by heat treatment in a vacuum was switched to the mode with a strong nonlinear addition to VAC (curve b in Fig. 1), corresponding to the growth of dynamic resistance of the film $R_d = dU/di$. The curves *a* and *b* of Fig. 2 demonstrate for VAC (see curves a and b in Fig. 1) the changes in the absorption coefficient α at the frequency of interzonal transitions ħω = 2.6eV when the current is switched on and off at moments t_1 and t_2, respectively. The absolute value of the absorption coefficient at this frequency was $\alpha l = 2.6$, where l is the film thickness. It can be seen that at t = t1 immediately after switching on the current $i = 80$mA there is a sharp increase in absorption with an output for saturation in a few minutes, when the current is switched off at the moment $t = t_2$, the absorption immediately reduced. For weakly nonlinear VAC the amplitude of the effect is very small: $\Delta(\alpha l) \cong 0.01$ ($\Delta\alpha/\alpha \cong 0.4\%$), but for strongly nonlinear VAC a significant increase in absorption is observed: $\Delta(\alpha l) \cong 0.22$ ($\Delta\alpha/\alpha \cong 9\%$), and after the current is switched off the absorption does not return to the original level at $t = t_1$.



1.

Fig.1. VAC with weak (a) and strong (b) nonlinearity for two electron-structural states of $YBa_2Cu_3O_{6+x}$($x \cong 0.9$) film at 300 K.

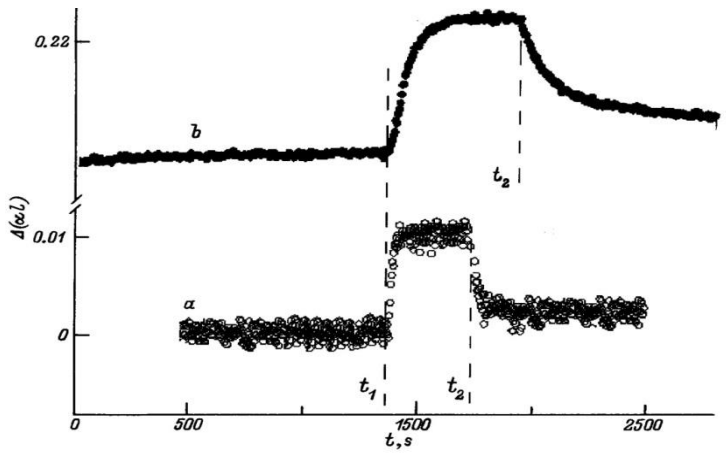


Fig.2. Change of optical absorption at the frequency ħω= 2.6eV for VAC *a* and *b* (see Fig. 1) when switching on and off the current of 80 mA at the moments of time t_1 and t_2 respectively.

Special experiments were carried out on non-equilibrium films, which were slowly, for tens of hours, relaxed to their equilibrium state for 300 K with linear VAC. In the process of such electron-structural relaxation, when there is a change in the mobility of carriers [5], a decrease in the nonlinear additive to the VAC was accompanied by a decrease in the amplitude of the EO-effect. In the area of frequencies of $d-d$ transitions near ħω≅1.5 eV, the considered EO-effect was not found.

Let's discuss some features of the relationship of VAC and optical absorption interrelation with the processes of delocalization localization. Deviations of VAC from the Ohm's law can be of two types: 1) mode with excess current, when VAC is described by $U^n \sim i$ (n>1) and dynamic resistance R_d decreases with current growth; 2) the mode with excess voltage $U \sim i^n$(n>1), when R_d increases. The first case corresponds to the release of localized carriers from the traps and the appearance of injection current in the sample. It is observed, for example, for amorphous films $YBa_2Cu_3O_{6+x}$, and for monoenergetic traps VAC has the form $U^2 \sim i$[7]. In the second case, on the contrary, the capture of mobile carriers to local levels and the VAC follows the law $U \sim (lni)^n (n>1)$ or a simpler one $U \sim i^2$ depending on the temperature and energy behavior of the density of local states [8]. In particular, the strongly nonlinear VAC in Fig. 1, close to the law $U \sim i^2$ in the area of high currents, indicates the existence in the films of spatial areas of localization, where in the current mode are effectively thrown mobile carriers.

For revealing of interrelation of optical absorption of HTSE-systems with processes of localization delocalization it is necessary to take into account the "rule of sums" connecting optical conductivity $\sigma(\omega)$ with kinetic energy of system $\langle T\rangle$. For a square grid with N nodes, this relationship is as follows [1]:

$$\int_0^\infty Re\sigma(\omega)d\omega = (e/\hbar)^2\langle T\rangle/N \qquad (1)$$

A similar integral relation can be written for the absorption coefficient α, since $Re\sigma(\omega) = (c/4\pi)\alpha(\omega)n(\omega)$, where $n(\omega)$ is the refractive index. For a dielectric with a completely filled valence zone, the conductivity (absorption) is entirely concentrated in the area of interzone transitions $\hbar\omega \geq E_g$, where $E_g \cong 1.7$eV is the optical gap for $YBa_2Cu_3O_6$. The feature of the Hubbard system is that during the metallization of the dielectric the energy $\langle T\rangle$ changes slightly if $u/t \overset{\smile}{<} 10-12$, where u is the Hubbard repulsion energy and t is the interstitial transfer integral [1]. For $YBa_2Cu_3O_{6+x}$, the value $u/t \cong 10$, so at metallization of the integral (1) is preserved, but there is a spectral redistribution of conductivity (absorption) over different areas of the frequency range. In the low-frequency area, where the Drude component appears, the conductivity (absorption) will be proportional to the kinetic energy of free oxygen holes $\langle T\rangle^D$. Then for metal in the high-frequency area above some boundary frequency $\omega_c$we have

$$(c/4\pi)n\int_{\omega_c}^\infty \alpha(\omega) \sim const - \langle T\rangle^D \qquad (2)$$

where it is taken into account that in the high-frequency area the refractive index n does not practically depend on the frequency [9]. Experimental data on the redistribution of the spectrum in $YBa_2Cu_3O_{6+x}$ at oxygen doping show that $\hbar\omega_c \cong 1.2$eV and this boundary energy at metallization practically do not shift [1]. It follows from (2) that absorption in the high-frequency area depends on the behavior of $\langle T\rangle^D$,. When the hole carriers are captured in the localization area, when the kinetic energy of the local holes increases and accordingly decreases $\langle T\rangle^D$, the

absorption at the inter-zone transitions should grow. This case corresponds to the above mentioned EO-effect for the film, in which there are spatial localization areas that determine the nonlinearity of VAC in Fig. 1. As the current increases, more and more carriers are thrown into the localization area, and the amplitude of the EO-effect increases. At the same time, the "asymmetric" behavior of absorption in Fig. 2 means that after the current is switched off, some carriers remain in the localization areas. For $d - d$ transitions, when absorption is determined by highly localized copper holes, the considered EO-effect should be absent, which, as already noted, and is observed in the experiment. At the same time, we should expect a strong EO-effect in the area of Drude absorption ($\hbar\omega < 1$eV), when the decrease of $\langle T \rangle^D$ when capturing oxygen carriers in the area of localization will lead to a significant reduction of absorption in current mode. Similarly, based on the above, we can consider the electro-optical effect for HTSC films with "injection" type VAC (for example, for small-crystalline or amorphous films), when the carriers are released to the mobility level.

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