Some Thermal and Electronic Transport Properties of YBa₂Cu₃O_{7-x} Obtained by the Photoacoustic Method

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The possibility of simultaneous determination of some thermal and transport properties of polycrystalline $YBa_2Cu_3O_{7-x}$, using a photoacoustic (PA) method was investigated. Both the amplitude and phase PA signals were measured as a function of the modulation frequency of the laser beam. Thermal diffusivity, the coefficients of the carrier diffusion, the excess-carrier lifetime and optical absorption coefficient were calculated by fitting experimental spectra and theoretical photoacousic signals.

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The discovery¹⁾ of oxides that remain superconducting above the boiling point of nitrogen 13 years ago caused a great interest. Various methods have been used in search for cooperative phenomena to high temperature superconductivity in $YBa_2Cu_3O_{7-x}$, which may shed light on the mechanism of superconductivity of superconducting ceramics²⁾. Among other things, the possibility of feroelectric-like instabilities related to superconductivity has been considered³⁾. Far-infrared reflectivity difference of $YBa_2Cu_3O_{7-x}$ were measured⁴⁾ and results were used to calculate the high frequency dielectric permittivity $\epsilon_{\infty}{=}3.8$. The dielectric response of $YBa_2Cu_3O_{7-x}$ was also remeasured, and the real and imaginary parts of the dielectric permittivity in the range from 20 kHz⁵⁾ to 20 GHz⁶⁾ have been determined.

Following Kamimura and Ushio^{7,8)} and using the results of the cluster calculations by Kamimura and Sano⁹⁾, Nomura and Kamimura calculated the many – electron energy bands the Fermi surfaces and density of states of YBa₂Cu₃O_{7-x}¹⁰⁾. Many authors^{11,12,13,14)} investigated ceramic and thin films of Y-Ba-Cu-O, using the photoacoustic and photothermal technique.

In this paper, we present the results of a photoacoustic investigation of thermal and transport properties of freshly made and 13 years old $YBa_2Cu_3O_{7-x}$ samples. The thermal diffusivity, the coefficient of carrier diffusion, the optical absorption coefficient and the excess carrier life time were determined..

Experimental

Polycrystalline YBa₂Cu₃O_{7-x} samples were synthesized from superconducting powders as explained elsewhere¹⁵⁾ and the critical temperature was about 94 K. The samples were of a disc shape with a diameter of about 10 mm. PA phase and amplitude spectra were measured using a PA cell which was protected against the surrounding influence and that was previously described elsewhere¹⁶⁾. The sample was mounted directly on the electret microphone that had a 3 mm diameter window.

The PA signals were measured using an experimental set-up, with a He-Cd laser (25 mW) as an optical source. The laser beam was modulated with a mechanical chopper. A large laser spot, about 3 mm in diameter, irradiated the sample, in order to decrease the effects of lateral diffusion.

In Fig. 1, the phase (a) and amplitude (b) PA diagrams versus the modulation frequency for three different freshly made samples with thicknesses (480, 410 and 345 μ m) are given.

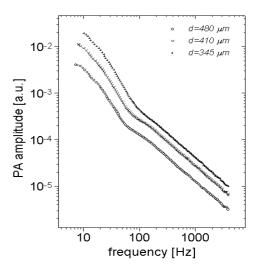


Fig. 1a. Amplitude PA spectra versus the modulation frequency for three various thicknesses of $YBa_2Cu_3O_{7-x}$; (o) $480\mu m$, (x) $410 \mu m$ and (+) $345 \mu m$.

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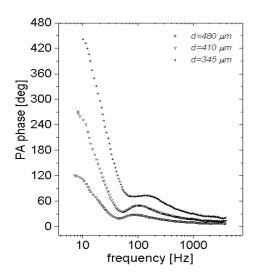


Fig. 1b. Phase PA spectra versus the modulation frequency for three various thicknesses of $YBa_2Cu_3O_{7-x}$; (o) $480\mu m$, (x) $410\mu m$ and (+) $345\mu m$

Discussion

The PA signal is a consequence of heat processes in the observed sample. It depends on periodic temperature variation on the rear sample surface, Φ (-l, ω) which is in contact with the electret microphone (Fig.2).

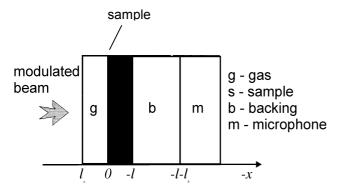


Fig. 2. The gas – sample – backing – microphone detection configuration.

The PA signal can be expressed by the following relation:

$$S(\omega) = \frac{\gamma P_o}{T_o k_b(\omega) l_b} \Phi(-l, \omega)$$
 (1)

where l is the sample thickness; ω is the frequency modulation of the excitation light beam; γ is the adiabatic constant and P_0 and T_0 are ambient pressure and temperature, respectively; $k_b(\omega) = (1+j)/\mu_b(\omega)$, where $\mu_b(\omega)$ is the thermal diffusion length of the backing (gas); l_b is the distance between the sample and the microphone membrane and $\Phi(-l,\omega)$ is the temperature variation of the sample surface which is in contact with the electret microphone.

The experimental amplitude and phase PA signals should be corrected in the modulation frequency range between 10 and 100 Hz because the electret microphone sensitivity decrease in the mentioned frequency range.

In this paper we have used a simple method of normalization, using PA experimental results for the same sample but with a different thickness. In this way, assuming that the conditions of measurement were the same in both cases, the normalized amplitude and phase spectra are obtained, described by the following equation¹⁷⁾:

$$\frac{S_1(\omega)}{S_2(\omega)} = A_n \exp(i\varphi_n) \tag{2}$$

where S_1 and S_2 are the PA signals for two different thicknesses of the same sample, A_n is the amplitude ratio, φ_n is the phase difference of those two PA signals. The normalized experimental PA amplitude and phase diagrams were then fitted with theoretically calculated PA diagrams for our superconducting sample. The fitting program was developed in the program language FORTRAN MS V5 explained elsewhere 17). This program enables the user to choose the values of the parameters in the mathematical model. The user selects the magnitude of change for each parameter, and also, the desired fitting criteria. In this way, the experimental curves can be compared with the theoretical ones which are obtained from the theoretical model explained previously, and whose values can be determined during the fitting procedure. The change of the phase (a) and amplitude (b) diagrams for the YBa₂Cu₃O_{7-x} sample, where the sample 480 µm thick was normalized with the sample 410 µm thick, are given in Fig. 3.

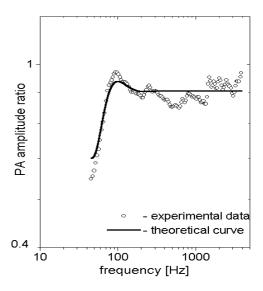


Fig. 3a. PA amplitude ratio normalized fitted diagrams for two samples of $YBa_2Cu_3O_{7-x}$ (480µm / 410 µm ratio of thicknesses); (o) experimental data and () theoretical curve;

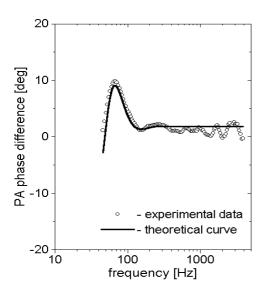


Fig. 3b. PA phase difference normalized fitted diagrams for two samples of $YBa_2Cu_3O_{7-x}$ (480µm / 410 µm ratio of thicknesses); (o) experimental data and () theoretical curve;

The values of five parameters were fitted: thermal diffusivity coefficient (D_T) , optical absorption coefficient (α) , excess carrier lifetime (τ) , and thermal conductivity (K). These values are given in Table 1.

Table 1. The values of thermal and electronic transport parameters of YBa₂Cu₃O_{7-x} superconducting ceramics, obtained by the fitting procedure

D _T [m ² /s]	α	τ	D	K
	[m ⁻¹]	[μs]	[m ² /s]	[W/mK]
0.16x10 ⁻⁵	8062	113	$0.2x10^{-5}$	1.0

The obtained value of the parameters can not be compared with literature data, because as far as we know they almost do not exist for room temperature. Dyer *et al.*¹⁴⁾ only estimated, on the basis of a photoacoustic technique, the values of the specific heat $J \approx 1$ J/gK, thermal conductivity $K \approx 1$ W/mK, and the thermal diffusivity $D_T \approx 2 \times 10^{-7}$ m²/s, for molten YBaCuO samples. Thermal diffusion coefficient length (μ) was calculated using the phase diagrams from Fig.1b and relation:

$$\mu = \sqrt{\frac{D_T}{\pi f_c}} \tag{3}$$

where f_c is the critical frequency when the phase diagram has a minimum. For the sample 480 μm thick μ was determined to be about 100 μm – almost five times smaller than the thickness of the used sample.

The extinction coefficient was calculated $k=0.515x10^{-3}$ using the obtained value for the optical absorption coefficient. The room temperature extinction coefficient can be calculated using Rey *et al.*⁵⁾ results. But, they measured room temperature

real (ϵ) and imaginary (ϵ) part of dielectric permittivity between $2x10^8$ Hz and $2x10^{10}$ Hz. They showed that both values decrease for almost two orders of magnitude in that range. If we suppose the same decreasing method of ϵ to the frequency of our laser beam ($f=6.7x10^{14}$ Hz), then we can get the same order of magnitude for the extinction coefficient.

The hot probe method was used to determine the majority free carriers of our $YBa_2Cu_3O_{7\text{-}x}$. It was clearly confirmed that all of our $YBa_2Cu_3O_{7\text{-}x}$ samples were of the p - type. The obtained value for the carrier diffusivity coefficient, $D=0.2x10^{-5}$ m^2/s , was used to calculate the free carrier mobility $\mu_n\approx 1~m^2/Vs$. This was confirmed using the Van der Pauw method. In this way, it was found that the hole concentration was about $10^{18}~cm^{-3}$. Rey $et~al.^{5)}$ suggested that the charge density was $>10^{19}~cm^{-3}$.

For the sample 13 years old we reported the same procedure of measurements and numerical analysis for the samples of two various thicknesses and we obtained that the value of thermal diffusivity slightly decreased to about 0.1×10^{-5} m²/s

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