THERMOPHYSICAL PROPERTIES AND SUPERCONDUCTIVITY OF THE METAL OXIDE Y–Sm–Ba–Cu–O

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We have studied the specific heat capacity and the coefficient of thermal conductivity for the superconductive ceramic $Y_{0.8}Sm_{0.2}Ba_2Cu_3O_X$ in the temperature range 4.2-300 K.

The problem of seeking out new metal-oxide compounds with a temperature of transition to the superconductive state (T_c > 100 K) resulted in the appearance of numerous publications (for example, [1-3]) dealing with the anomalous relationships between temperature and the electrical resistance ρ. We have carried out studies of the ceramic system Y–Sm–Ba–Cu–O [4], and these have demonstrated that when 20% of the yttrium is replaced with samarium, the function ρ(T) in the region 100-170 K exhibits anomalies in the form of an exponential (by 2-3 orders) reduction in the resistance, subsequent to which, with T ~ 100 K, the complete superconductive transition takes place. Analysis of our data, as well as studies of the structural state and phase composition, have demonstrated that the unique features involved in the behavior of the ceramic systems are associated with the fact that they are not of a single phase. A majority of researchers have noted a characteristic property of the above-indicated anomalies, and namely: after 3-7 repetitions of the heat-treatment cycle in a temperature range of 77-300 K the jumps in ρ diminished in magnitude and disappeared entirely. The function ρ(T) assumed the linear nature that is usual for such materials all the way to T_c. At the present time we are unable to isolate and identify the phase responsible for these anomalies, nor to come up with an acceptable physical interpretation (whether or not this is truly superconductivity).

In order to observe and study the temperature features of the thermophysical characteristics, in the present study we have undertaken measurements of the heat capacity c_p and of the thermal conductivity λ for the compound $Y_{0.8}Sm_{0.2}Ba_2Cu_3O_X$ in the temperature interval 4.2-300 K, conducting our measurements every 0.5 K. The specimens of the superconductive ceramic were fabricated by high-temperature solid-phase synthesis from oxides of yttrium, samarium, copper, and barium carbonate, in stoichiometric proportions. The synthesized material was subjected to triple annealing at 950°C for 6 h, and the specimens were ground and pressed at the conclusion of each stage. This made it possible (on the basis of the data from the measurement of electrical resistance) to achieve adequate homogeneity for the specimens. They exhibited resistive transition to the superconductive state on attainment of zero resistance at a temperature of 94 K and a transition depth of 2.5 K. The thermophysical characteristics were studied by means of an installation which made it possible to achieve a quasisteady method for the specimens in the shape of plates, and moreover, allowed us to determine the specific heat capacity c_p and the coefficient of thermal conductivity λ from a single experiment. With this purpose in mind, the specimen being studied was cooled to a specified temperature and then subjected to heating from a direct-current power source under adiabatic conditions. The adiabatic shell has been formed out of a copper-foil parallelepiped, onto which a constantan-wire heater has been wound in bifilar fashion. Thermocouples were positioned on the inside surface of the shell, and by means of these temperature was automatically controlled. The measurements were conducted on two identical rectangular plates exhibiting dimensions of 35 × 35 × 3.6 mm, between which a plane heater was attached with glue. Measurements of the adiabatic conditions (the temperature difference across the shell and the specimen must equal zero) was monitored by a double-layered

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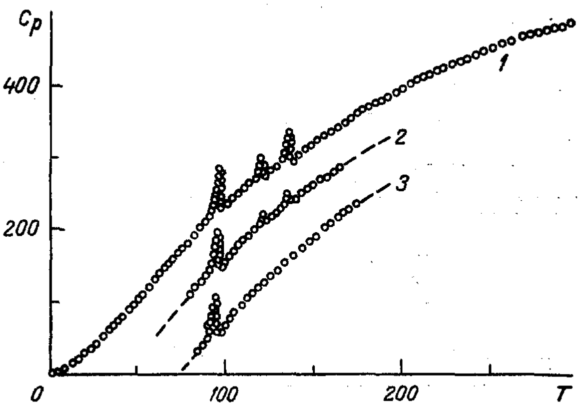

Fig. 1

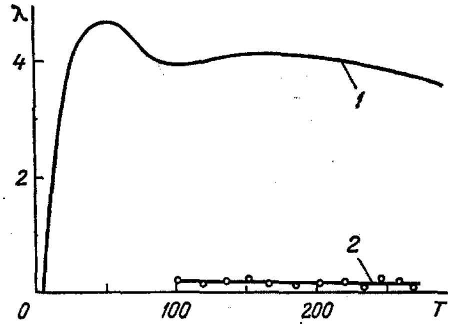

Fig. 2

Fig. 1. Specific heat capacity c_p for the ceramic $Y_{0.8}Sm_{0.2}Ba_2Cu_3O_x$ as a function of temperature: 1, 2, 3) respectively, the one-, three-, and five-pass heat-treatment cycle. The curves have been displaced along the vertical. c_p, J/(kg·K); T, K.

Fig. 2. Coefficient of thermal conductivity as a function of temperature: 1) phonon term; 2) electron term; λ, λ_e, W/m·K; T, K.

differential thermocouple Cu + 0.1% Fe + 0.024% Li, i.e., Chromel. Identical thermocouples were mounted on each plate to record the changes in the specimen temperatures and in the temperature drops across the thickness of the specimen. As a result, the error in the measurement of the temperature for the absolute thermocouple amounted to ≤0.25 K, while for the differential thermocouple it amounted to ±0.05 K in the range 4.2-35 K and ±0.01 K in the range 35-300 K. The quasisteady experiment was conducted at a heating rate of $dT/d\tau \leq$ 3 K/min and the maximum relative error for the method amounted to 7-8%.

Figure 1 shows the heat capacities measured on identical specimens subsequent to synthesis (curve 1) and after the heat-treatment cycle within the limits of the temperature measurement interval (curves 2 and 3) as functions of temperature. In the first experiment, curve 1 clearly shows three jumps in the heat capacity, with maxima at 98, 120, and 137 K. The initial 98-deg peak in Δc_p corresponds to the temperature for the onset of the superconductive transition T_c on the ρ(T) curve for the given system. Taking into consideration that such anomalies in c_p are characteristic for the superconductive transition, and that they have also been observed in the T_c region for Y—Ba—Cu—O and La—Sr—Cu—O superconductors [5, 6], the relationship between Δc_p and the onset of the superconductive state in the metalloceramic electron system becomes obvious.

The physical interpretation of two other high-temperature peaks in Δc_p is of considerable interest, since no stable superconductive phases at T > 125 K have been observed to the present time. Comparison of the functions ρ(T) [4] with existing results points to the possibility of a single nature for these anomalies in the behavior of the kinetic and thermodynamic coefficients. We should point out that the jumps in electrical resistance and the peaks in heat capacity at T > 100 K are analogous from the qualitative standpoint, and close in the quantitative standpoint, to the corresponding features encountered in the region of the superconductive transition. Thus, the conclusions in [4] as to the possibility for the existence of superconductivity in metal-oxide compounds of the "1-2-3" type at T > 100 K are confirmed by the heat-capacity data. We also have confirmation of the viewpoints to the effect that these states exhibit limited stability. Curves 2 and 3 in Fig. 1 have been plotted after three- and five-pass heat-treatment cycles, respectively, in the temperature range 4.2-300 K. We can see that the jumps in heat capacity for T = 120 and 137 K become noticeably smoother after three cycles and disappear entirely after five; the peak in Δc_p at 98 K undergoes no changes. In this regard, we also note complete correlation with the functions ρ(T) [4], transformed after completion of the heat-treatment cycles into one that is ordinary and linear all the way to the start of the superconductivity transition.

The plates used as the specimens were of inadequate thickness to determine the coefficient of thermal conductivity with required accuracy at low temperatures. Therefore, in the temperature region 4.2-25 K, λ was measured by a steady-state method in which gaseous helium served as the heat carrier. The onset of the steady state was determined from constancy of the temperatures on either side of the plate.

Figure 2 shows the results from the measurements of thermal conductivity in the temperature range 4.2-300 K. The nature of the derived function is in good agreement with the data from a number of studies involving a Y—Ba—Cu—O ceramic [7, 8] and demonstrates the predominant role of phonons in the process of heat transfer. In the region 100-300 K the coefficient λ changes little with temperature, and its magnitude (by 2-3 orders smaller than in metal conductors) was determined by a strong electron-phonon interaction. The observed inflection on the $\lambda(T)$ curve in the region of 95 K is associated with the suppression of the electron-phonon scattering due to the transition of the electron subsystem to the superconductive state. As a result, we find a pronounced increase in the phonon contribution to λ, reaching a maximum at 50-60 K. A further reduction in temperature leads to the "freezing" of the phonons and to a reduction in λ.

Using the Lorentz relationship $Lo = \lambda_e \rho T^{-1}$, we determined the electron component of the thermal conductivity λ_e. Since for the case in which $T \geq 100$ K inelastic scattering of electrons against long-wave phonons is virtually absent, the Lorentz number $Lo = 2.45 \cdot 10^{-8}$ $W \cdot \Omega \cdot K^{-1}$. The magnitude of the electrical resistance prior to the superconductive transition $\rho \approx 10^{-5}$ $\Omega \cdot m$. Thus, the quantity λ_e for T = 100 K is equal to 0.25 $W \cdot m^{-1} \cdot K_1$, which amounts to 5-7% of the phonon thermal conductivity, and is virtually independent of temperature (Fig. 2).

Unlike c_p, the absence of anomalies in the function $\lambda(T)$ in the temperature interval 100-150 K is possibly associated with the fact that the contribution of λ_e to the total thermal conductivity is quite insignificant. Moreover, from all of the kinetic parameters the coefficient λ_e is least sensitive both to the retuning of the electron spectrum and to the type of scattering mechanisms.

NOTATION

c_p, specific heat capacity; λ, thermal conductivity; λ_e, electron thermal conductivity; T, temperature; ρ, resistance; τ, time; Lo, Lorentz number.

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