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дубна

E8-88-638

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GLASSY BEHAVIOUR OF $YBa_2Cu_3O_7$

Submitted to "International Journal
of Modern Physics B"

1988

1. INTRODUCTION

For the understanding of high temperature superconductivity it is necessary to analyse such features in which the new materials distinguish themselves from usual superconductors. One feature is the heat capacity at low temperatures, where a term linear in temperature was observed for high temperature superconductors[1-4]. As was shown recently[3,4] this anomaly is not only related to an impurity effect. In measuring the heat capacity of superconducting $YBa_2Cu_3O_7$ and dielectric $YBa_2Cu_3O_6$, prepared by oxygen reduction of the former, Collocott et al.[3] observed a decrease of the linear term to (1.16 ÷ 1.91) mJ/mole·K^2. Similar results were obtained by Kumagai et al.[4] for the $La_{2-x}Ba_xCuO_4$ compounds. Here the appearance of the superconductivity for $x>0.02$ is connected with an increase of c/T of about 5 mJ/mole·K^2. The authors[4] explain this behaviour within the resonating valence-bond model[5] where the T-term is a consequence of the highly correlated ground state. An alternative explanation is given in ref.[3] assuming in consistency with the acoustic measurements of Golding et al.[6] and the bipolaronic theory of de Jongh[7] that the excess heat capacity is caused by two-level systems (tls) like in glasses.

A very sensitive method for the test of the last version are the measurements of the long time energy relaxation. As was shown for various glasses (see e.g. ref.[8]) the wide relaxation time spectrum of tls leads to a long time relaxation of energy after a rapid temperature change. The first results presented in this paper demonstrate the glass-like behaviour of energy relaxation in $YBa_2Cu_3O_7$.

2. MODEL

At low temperatures the long time relaxation observed in amorphous solids can be treated within the standard phenomenological tunneling model[9,10]. If one assumes a uniform distribution of tls,

$$P(\Delta,\lambda) = \bar{P} = \text{const} \quad , \tag{1}$$

where Δ is the asymmetry energy and λ is the tunneling parameter, the standard tunneling theory gives a time dependent density of states

$$n(E,t) = n_o(t) = (\bar{P}/2)\ln(4t/\tau_{min}) \quad , \tag{2}$$

a time dependent heat capacity

$$c_t = (\pi^2 k_B^2/12)\bar{P}T\ln(4t/\tau_{min}) \quad , \tag{3}$$

and a corresponding power release after cooling the system from equilibrium temperature T_1 down to T_o

$$\dot{Q}(T_1,T_o,t) = V(\pi^2 k_B^2/24)\bar{P}(T_1^2 - T_o^2)/t \quad , \tag{4}$$

where E is the energy difference between two levels, τ_{min} is the shortest relaxation time and V the volume of the specimen.

Indeed, an accurate t^{-1} dependence of power release was observed for various glasses. However, the temperature dependence holds for low enough temperatures ($T_1,T_o \leq 2$ K) only. For higher temperatures T_1 one achieves better agreement with the experimental data using the energy dependent distribution function[8)]

$$n(E,t) = n_o(t)/(1 + \exp(E - E_f)/k_B T_b) \quad , \tag{5}$$

where $E_f = k_B T_f$ and T_b are constant. With (5), eqs.(3) and (4) are transformed into

$$c_t = (\pi^2 k_B^2/12)\bar{P} I_1(T/T_f,T_b) T\ln(4t/\tau_{min}) \tag{6}$$

and

$$\dot{Q}(T_1,T_o,t) = V(\pi^2 k_B^2/24)\bar{P}(I_2(T_1/T_f,T_b)T_1^2 - I_2(T_o/T_f,T_b)T_o^2)/t \quad , \tag{7}$$

where

$$I_1(T/T_f,T_b) = (6/\pi^2)\int_0^\infty \frac{x^2\exp(-x)\,dx}{(1+\exp(-x))^2(1+\exp((xT/T_f - 1)/(T_b/T_f)))} \quad , \tag{8}$$

$$I_2(T/T_f,T_b) = 2(T_f/T)^2 \int_0^{T/T_f} (T/T_f) I_1(T/T_f,T_b)\, d(T_b/T_f) \quad . \tag{9}$$

Similar values of T_f were obtained for all amorphous solids (13 K $\leq T_f \leq$ 24 K)[8)]. Equation (7) holds only for $T_o < T_f/10$, because the tunneling parameter λ becomes temperature dependent at higher temperatures[8,11)].

3. EXPERIMENTAL

46 cylinders of $YBa_2Cu_3O_7$ (T_c = 91 K, diameter : 1.0 cm, thickness : 0.1 ÷ 0.3 cm) were connected by copper foils and a copper holder with a Ge-thermometer, a heater and a copper wire to provide contact with a mechanical heat switch. The sample (42.854 g $YBa_2Cu_3O_7$ and 37.615 g Cu) hung in the calorimeter on 12 kapron threads (0.15 mm in diameter, 30 mm long).

For the heat release study, the resistance drift $\dot{R}$ of the thermometer was measured as a function of time t after the rapid cooling of the specimen from equilibrium temperature T_1 (2.35 K $\leq T_1 \leq$ 15.11 K) (where the specimen remained for at least 10 hr) to T_o = 1.5 K. The time required for the cooling from T_1 down to T_o = 1.5 K is shown in fig. 1.

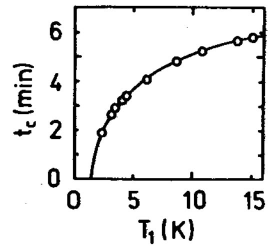

Fig. 1. The time t_c required for cooling the specimen from T_1 down to T_o = 1.5 K

The corresponding power release was then determined from the directly measured power release $\dot{Q}_m$, the heat leak $\dot{Q}_A$ and the background $\dot{Q}_B$ as

$$\dot{Q}(T_1,T_o,t) = \dot{Q}_m - \dot{Q}_A - \dot{Q}_B \quad , \tag{10}$$

where

$$\dot{Q}_m = \dot{R}C/(\partial R/\partial T) \quad , \tag{11}$$

$$\dot{Q}_A = A(T_K - T) \quad . \tag{12}$$

C is the heat capacity of the specimen, $\partial R/\partial T$ is the sensitivity of the thermometer, and A is the coupling constant between the specimen and the body of the calorimeter. The sample temperature T was chosen close to the calorimeter temperature, which was kept constant (to 10^{-4} K). For $T(t) - T_o = 10^{-2}$ K the temperature was referred to $T_o = T_K$ by the heat switch. The parameters A = 0.14 nW/mK and $\dot{Q}_B$ = 0.5 nW were determined experimentally.

Table. The distribution parameter $\bar{P}_m$ of tls in $YBa_2Cu_3O_7$ and vitreous silica obtained by heat capacity, acoustic and energy relaxation measurements

$\bar{P}_m (10^{38}/Jg)$

	heat capacity	acoustic meas.	energy relaxation
am. SiO_2	2.4 ÷ 3.4 ref.[16]	1.0 ÷ 1.4 ref.[17,18]	0.45 ÷ 1.5 ref.[14,19]
$YBa_2Cu_3O_7$	3.7 ÷ 6.0 ref.[3]	4.5 ref.[6]	2.4 this work

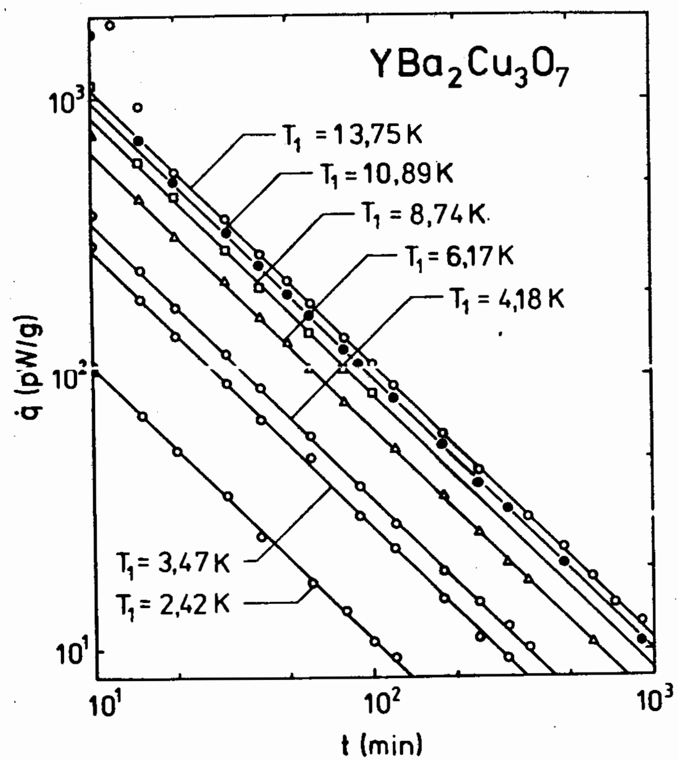

Fig. 2. The specific power released in $YBa_2Cu_3O_7$ after cooling from various T_1 to $T_o = 1.5$ K as a function of time. Straight lines : $\dot{q} \sim t^{-1}$

The accuracy of the measurement is limited by the time fluctuations of $\dot{Q}_B$, approximately to 0.1 nW.

For excluding the heat release in the holder due to hydrogen bubbles in copper[12,13], the specimen was kept at 4.2 K for a weak before beginning the heat measurements. As was shown[13] the power release in copper is negligible for $T_1 \leq 20$ K.

At last the heat capacity was measured.

4. RESULTS AND DISCUSSION

The specific power release $\dot{q} = \dot{Q}/m$ (m = 42.854 g) after cooling from various T_1 to $T_o = 1.5$ K as a function of time (t = 0 at the beginning of the cooling down) is shown in fig. 2. Exact t^{-1}-dependence is observed for all T_1 and $t > 2t_c$, without any systematic deviation even after 30 hr ($T_1 = 15.11$ K). We have also got for the T_1-dependence of the power release a typical glassy behaviour. Figure 3 shows the values $\dot{q} \cdot t$ as a function of $T_1^2 - T_o^2$, ($T_o = 1.5$ K). With $\bar{P}_m = 2.4 \cdot 10^{38}/Jg$, $T_f = 13$ K, $T_b = 0$ K eq. (7) (curve 2) agrees with the experimental results very well, while eq. (4) (curve 1) holds for $T_1 \leq 3$ K only. The same T_1-dependence ($T_f = 13$ K, $T_b = 0$ K) and a similar value of $\bar{P}_m$ (see the table) were obtained for amorphous silica[14,15].

In agreement with acoustic measurements of Golding et al.[6] the long-time heat relaxation study shows that tls exist in $YBa_2Cu_3O_7$. Now, we must answer the question whether the existence of tls is an intrinsic feature or it is caused by impurities or defects.

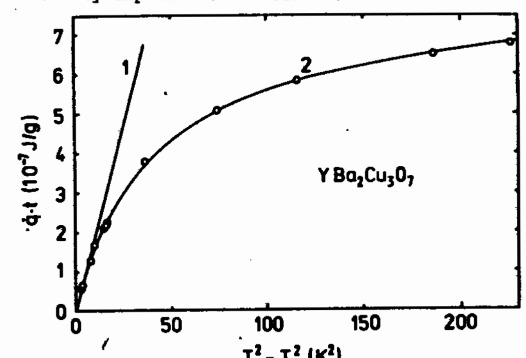

Fig. 3. The value $\dot{q} \cdot t$ as a function of $T_1^2 - T_o^2$ ($T_o = 1.5$ K),
1 - standard tunneling theory (eq. (4) with $\bar{P}_m = 2.4 \cdot 10^{38}/Jg$).
2 - modified tunneling theory (eq. (7) with $\bar{P}_m = 2.4 \cdot 10^{38}/Jg$), $T_f = 13$ K and $T_b = 0$ K)

From the difference between measured heat capacities for superconducting $YBa_2Cu_3O_7$ and dielectric $YBa_2Cu_3O_6$ ($c/T = 1.16 \div 1.91$ mJ/mole·K^2)[3] eq.(3) gives $\bar{P}_m = (3.7 \div 6.0)\cdot 10^{38}$/Jg using $\ln(4t/\tau_{min}) = 30$, ($\tau_{min} = 10^{-10}$ sec, t = 300 sec), not far from $\bar{P}_m = 4.5\cdot 10^{38}$/Jg from acoustic measurements[6], and our value $\bar{P}_m = 2.4\cdot 10^{38}$/Jg. The differences between the $\bar{P}_m$-values are not larger than between the corresponding values for amorphous silica (see the table). Therefore, it is probable that in all measurements the anomalies are of the same origin, i.e. it is an intrinsic feature of $YBa_2Cu_3O_7$.

Nevertheless, it is not excluded that the tls are caused by impurities or inhomogeneities. In fig. 4 the heat capacity at low temperatures (1.2 K≤T≤15 K) is shown. With

$$c = AT^{-2} + aT + bT^3 \qquad (13)$$

we get for 3 K<T<9 K : A = 400 mJ·K/mole, a = 42 mJ/mole·K^2, b = 0.64 mJ/mole·K^4 and $\Theta_D = 343$ K. The Schottky-anomaly AT^{-2} and a large linear term together with a low Debye temperature, indicate a high impurity concentration in our $YBa_2Cu_3O_7$. According to $\bar{P}_m = 2.4\cdot 10^{38}$/Jg the linear term due to tls is about 0.8 mJ/mole·K^2, i.e. only 5% of the measured value a = 42 mJ/mole K^2.

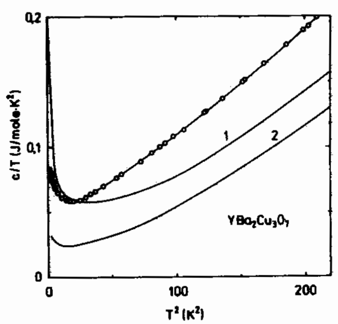

Fig. 4. The specific heat capacity plotted as c/T vs T^2 for $YBa_2Cu_3O_7$. Points : this work, 1 - Phillips et al.[1], 2 - Junod et al.[2]

The measurements of long time energy relaxation in $YBa_2Cu_3O_{7-x}$ ($0\leq x\leq 1$) and $La_{2-x}Ba_xCuO_4$ ($0\leq x\leq 0.15$) could be the answer to the question of the true origin of tls in high temperature superconductors.

5. CONCLUSIONS

The low temperature long time power release after the rapid cooling of $YBa_2Cu_3O_7$ show the behaviour typical of amorphous solids. The power is strongly proportional to t^{-1} (0.2 hr<t<30 hr). For $T_o = 1.5$ K and $T_1 \leq 3$ K the results agree with the tunneling theory. It enables one to determine the distribution parameter of tls $\bar{P}_m = 2.4\cdot 10^{38}$/Jg, which is not far from the $\bar{P}_m$-values obtained by heat capacity and acoustic measurements. Therefore, the assumption seems to be reasonable of the existence of tls being an intrinsic feature of the high temperature superconductors.

For $T_1 \geq 3$ K ($T_o = 1.5$ K) the T_1-dependence of the power release can be understood within the modified tunneling model assuming the existence of a maximum energy $E_f/k_B = 13$ K in the distribution function of the density of states. This parameter is typical for amorphous solids too (13 K≤T_f≤24 K).

ACKNOWLEDGEMENTS

The authors are much obliged to Prof. Oppermann, Drs. Reichelt and Schreiber (Technical University Dresden) for providing them with the $YBa_2Cu_3O_7$ and valuable discussions and to Dr. Mertig (JINR) for useful remarks in the course of the manuscript preparation.

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Received by Publishing Department
on August 24, 1988.

Салинг С. E8-88-638

Стеклоподобное поведение $YBa_2Cu_3O_7$

Измерялись мощность тепловыделения после быстрого охлаждения от равновесной температуры $T_1 (2,35K \leq T_1 \leq 15,11K)$ до T_0 = 1,5K и теплоемкость $(1,2K \leq T \leq 100K)$. Полученные временная и температурная зависимости тепловыделения такие же как для аморфных твердых тел. Результирующая плотность состояния двухуровневых систем близка к величинам, полученным из теплоемкости и акустических измерений.

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1988

Sahling S. E8-88-638

Glassy Behaviour of $YBa_2Cu_3O_7$

Long time power released in $YBa_2Cu_3O_7$ after rapid cooling from the equilibrium temperature T_1 $(2.35K \leq T_1 \leq 15.11K)$ to T_0 = 1.5K and the heat capacity $(1.2K \leq T \leq 100K)$ were measured. The observed power release is similar to those of amorphous solids. The resulting density of states of two-level systems is close to the values obtained from heat capacity and acoustic measurements.

The investigation has been performed at the Laboratory of Nuclear Problems, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1988