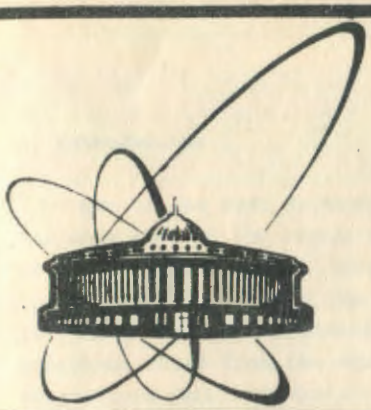

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LONG-TIME RELAXATION OF LOW ENERGY EXCITATIONS IN $Bi_2CaSr_2O_8$

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1. Introduction

One of the most interesting features of amorphous solids is the existence of low energy excitations with a broad distribution of their energies and relaxation times. The relaxation time spectrum extends from a few Nanoseconds up to very large values (at least 10^5 sec). Therefore, after a rapid cooling of an amorphous solid from the equilibrium temperature T_1 to T_0 the excitations with relaxation times larger than the time t_c necessary for cooling remain in the excited state. Their further relaxation leads to a time dependent power $\dot{Q}(T_1,T_0,t)$ released in the solid, which can be measured in calorimetric experiments. For all investigated amorphous solids roughly a t^{-1} dependence of the long-time power release was observed at low temperatures T_1 and T_0 .[1]

Recently this behaviour and other low temperature anomalies typical of glasses have been found for crystalline ceramics PLZT[2-4], a ferroelectric material with a diffuse phase transition, and $YBa_2Cu_3O_{6+x}$ in the superconducting ($x = 1$)[5,6] and the nonsuperconducting phase ($x = 0$)[7]. In this work the low temperature heat capacity and the long-time thermal relaxation of polycrystalline ceramic $Bi_2CaSr_2Cu_2O_8$ - an other high temperature superconductor (HTS) - were investigated. The behaviour typical of amorphous solids was observed again.

2. Model

At low temperatures, the long-time relaxation in amorphous solids can be treaded within the standard phenomenological tunnelling model[8,9]. If one assumes a uniform distribution of two-level systems (TLS)

$$P(\Delta,\lambda) = P = \text{constant} , \qquad (1)$$

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

$$n(E,t) = n_0(t) = (P/2) \cdot \ln(4t/\tau_{min}) , \qquad (2)$$

a time dependent heat capacity

$$c_t = (\pi^2 k^2/12)\cdot P\cdot T\cdot \ln(4t/\tau_{min}) \ , \qquad (3)$$

and a corresponding power release after cooling the solid from the equilibrium temperature T_1 down to T_0

$$\dot{Q}(T_1,T_0,t) = (\pi^2 k^2/24)\cdot V\cdot P\cdot (T_1^2 - T_1^2)\ t^{-1} \ , \qquad (4)$$

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

3. Experimental

Six $Bi_2CaSr_2Cu_2O_8$ ceramics (total mass: 31,7 g, mass density: 5.0 g/cm^3 and T_c = 80K) were connected by copper foils and a copper holder with a Ge-thermometer, a heater and a copper wire to provide the contact with a mechanical heat switch. The sample (31.7 g Bi-ceramics and 23.6 g Copper) hung in the calorimeter on 8 kapron threads.

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 sample from various equilibrium temperatures T_1 (where the specimen remained for at least 10 hours) to T_0 = 1.5K. 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

$$\dot{Q}(T_1,T_0,t) = \dot{Q}_m - \dot{Q}_A - \dot{Q}_B \ , \qquad (5)$$

where

$$\dot{Q}_m = \dot{R}C/(\delta R/\delta T) \ , \qquad (6)$$

$$\dot{Q}_A = A(T_K - T) \ . \qquad (7)$$

C is the heat capacity of the specimen, $\delta R/\delta T$ is the sensitivity of the thermometer, and A is the thermal resistance of the kapron threads and of the electrical wires connected to the heater and the thermometer. The sample temperature T was chosen close to the calorimeter temperature T_K, which was kept constant (to 10^{-4}K). For $T(t) - T_0 = 10^{-2}$K the temperature was

return to $T_0 = T_K$ by the heat switch. The parameter $A = 0.15$ nW/mK and $\dot{Q}_B = 0.5$ nW were determined experimentally. The accuracy of the measurement is limited by the time fluctuations of $\dot{Q}_B$, approximately to 0.1 nW.

Finaly, the heat capacity was measured.

4. Results and Discussion

The Results of our heat capacity measurements together with data of other Bi-HTS[10,11] are given in fig. 1. On the assumption that

$$c = \alpha T^{-2} + \gamma T + \beta T^3 + \delta T^5 \quad , \tag{8}$$

the best fit of our heat capacity data ($1.5K \leq T \leq 6K$) with $\alpha = 40\ \mu J\ K/g$, $\gamma = 1\ \mu J/gK^2$, $\beta = 1.83\ \mu J/gK^4$ and $\delta = 0.005\ \mu J/gK^6$ was obtained. Because of the Schottky like contribution to the heat capacity at low temperatures and the large δ we cannot distinguish between the cases $\gamma = 0$ and $\gamma = 1\ \mu J/gK^2$ (see fig. 1, curve 2). A similar situation exists for other investigated Bi-HTS. In reference 8 the upper limit $\gamma = 1\ \mu J/gK^2$ was given (see fig. 1, curve 3). The corresponding upper limit of the distribution parameter is $P = 12 \cdot 10^{44}/Jm^3$, where $t = 100$ sec, $\tau_{min} = 10^{-9}$sec and $\rho = 5.0\ g/cm^3$ were used for the calculation. From the heat capacity data we cannot answer the question: whether low energy excitations exist in the Bi-HTS as in glasses or not. The answer to this question can be undoubtedly obtained from the power release measurements, where the long-time power release typical of amorphous solids was observed in our Bi-HTS.

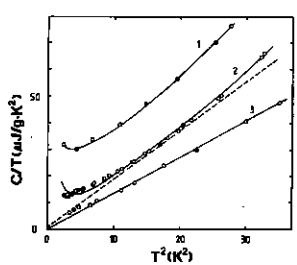

Fig. 1: The heat capacity of Bi-high temperature superconductors:
curve 1: $Bi_{2.15}Ca_{1.17}Sr_{1.68}Cu_2O_{\ 8}$, (R.A. Fisher et al.)[10]
curve 2: $Bi_2CaSr_2Cu_2O_8$ (this work)
curve 3: $Bi_4(Sr_{0.6}Ca_{0.4})Cu_4O_{16}$, (M. Sera et al.)[11]

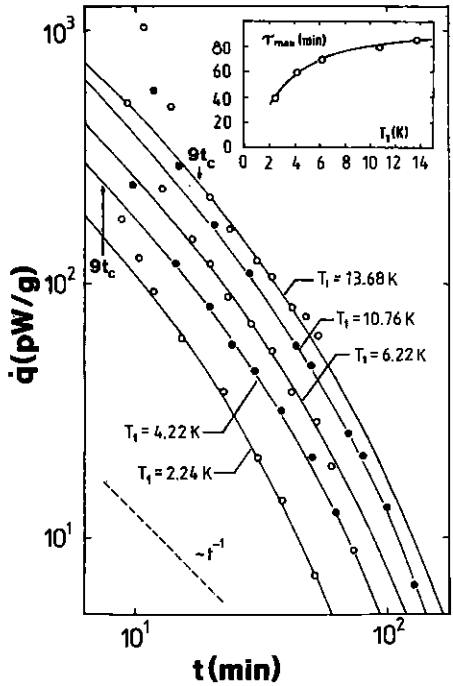

Fig. 2: The specific power $\dot{q} = \dot{Q}/m$ released in $Bi_2CaSr_2Cu_2O_8$ after rapid cooling from various equilibrium temperature T_1 to $T_0 = 1.5K$ as a function of time t. t_c is the time necessary for cooling.
Curves: the best fits of our experimental data with eq. (9). The fit parameters τ_{max} and q_0 as a function of time are given in the inset and in fig. 3.

Fig. 2 shows the power released after cooling the sample from various equilibrium temperatures T_1 to $T_0 = 1.5K$. The sample was cooled down from T_1 to T_0 in 1-2 minutes. However, in the first time ($t_c < t < 9t_c$) an additional power with a roughly exponential time dependence was observed. This additional power release is probably caused by the large thermal resistance between the Bi-ceramics and the copper foil.

For $t > t_c$ the power release $\dot{Q}$ is given by:

$$\dot{Q} = m \cdot q_0 \cdot t^{-1} \cdot \exp(-t/\tau_{max}) \quad , \qquad (9)$$

where the fit parameters q_0 and τ_{max} depend on T_1 (see fig. 3 and the inset of fig. 2).

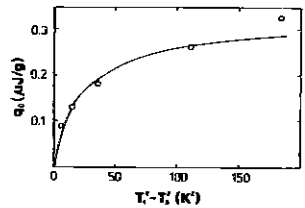

Fig. 3: The fit parameter q_0 of eq. (9) used for calculation of the power release data in fig. 2 as a function of $T_1^2 - T_0^2$ ($T_0 = 1.5K$).
Curve: calculation of q_0 based on the energy dependent distribution function (10) with $P = 12.5 \cdot 10^{44}/Jm^3$, $E_f/k = 8K$ and $T_b = 0$.

The following interpretation of the observed time dependence is possible. In the short-time range ($t \ll \tau_{max}$) the power release is proportional to t^{-1} in agreement with eq. (4). In the long-time range ($t > \tau_{max}$) the time became longer than the upper limit of the relaxation time spectrum and the time dependence changes from t^{-1} to $\exp(-t/\tau_{max})$. In our experiment only the long-time range was investigated.

From q_0 the distribution parameter P can be estimated. Fig. 3 shows q_0 as a function of $T_1^2 - T_0^2$ (T_0=1.5K). The temperature dependence of q_0 disagrees with eq. (4). Similar deviations were observed for all investigated amorphous solids, where the assumption of a energy dependent density of states of TLS

$$n(E,t) = (P/2) \cdot \ln(4t/\tau_{min}) \cdot (1 + \exp((E - E_f)/kT_b) \qquad (10)$$

leads to a good fit of the experimental data (E_f and T_b are constants)[11]. The results of this calculation agree with eq. (4) only at low T_1 ($T_1/T_f < 0.2$).

For the Bi-HTS the calculation of q_0 with function (10) also yields a good fit ($P = 12.5 \cdot 10^{44}/Jm^3$, $E_f/k = 8K$ and $T_b = 0$). Because of the low $E_f/k = 8K$ one expects that q_0 is proportional to $T_1^2 - T_0^2$ only at $T_1 < 1.6K$ and was therefore not observed in our experiment ($T_1 \geq 2.24K$).

Table 1 shows the distribution parameter P obtained by acoustic, heat release and heat capacity measurements for vitreous silica, PLZT and various HTS.

Table 1

The distribution parameter calculated from acoustic, heat release and heat capacity data of vitreous silica, PLZT and various HTS.

distribution parameter P (10^{44}/Jm^3)

material	acoustic	Ref.	h. release	Ref.	heat cap.	Ref.
vitr. silica	2.2-3.1	/12,13/	1-3.3	/14,15/	5.3-7.5	/16/
PLZT			12.0	/2-3/		
$YBa_2Cu_3O_7$	4-28	/17,18/	4.3	/5/	<75	/19/
$YBa_2Cu_3O_{6.85}$			8.2	/6/		
$YBa_2Cu_3O_6$			9.9	/7/	<90	/20/
$Bi_2CaSr_2Cu_2O_8$			12.5	this work	<12	this work

The distribution parameter of the Bi-HTS is not far from the P-values of PLZT and the Y-compounds. Thus the low temperature glassy behaviour is at least a common feature of high temperature superconducting ceramics. It is remarkable, that in polycrystalline ceramics the density of states of TLS is even larger than in vitreous silica. Therefore, these ceramics are convenient for a more detailed study of the long-time power release.

The observed T_1 dependence of the power release in the Bi-HTS and in other glasses can be explained by the cut-off energy E_f according to function (10) or by the existence of a temperature dependent $\tau_{max}(T_0)$ - the upper limit of the relaxation time spectrum. In the first case there are no excitations with energies above E_f, in the second case the excitations with larger energies relax during the cooling. In both cases τ_{max} is in agreement with our experimental results (see fig. 2) independent of T_1 at $T_1 > E_f/k$.

We have at least two facts, which show that the second version is more realistic. First, the existence of an upper limit was found for the Bi-HTS (E_f/k = 8K) and PLZT (E_f/k = 7.5K)/2,3/, e.g. for the materials with small E_f. Secondly, "short heating" experiments demonstrate that at higher temperatures ($T_0 \geq E_f/k$) τ_{max} is sufficiently shorter than at 1.5K for all investigated glasses/1/. Moreover, a strong temperature dependence of τ_{max} follows from the standard tunnelling theory with a fixed maximum value λ_{max} of the tunnelling parameters: τ_{max} is proportional to T^{-3} (one phonon process)/21/ and at higher temperatures to T^{-7} (the first order Raman process)/22/ with $E = 2KT$.

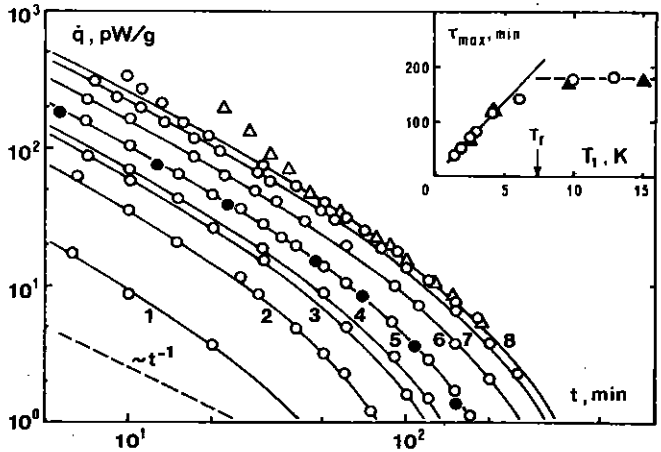

Fig. 4: The time dependence of the specific power released in PLZT after cooling from various T_1 ($1.49K \leq T_1 \leq 13K$) to 1.3K. Curves: the best fits of the experimental data with eq. (9). Inset: the maximum relaxation time used for the fit as a function of T_1. For more details see references 2 and 3.

However, some experimental facts cannot be explained by this way, in particular the T_1 dependence of τ_{max} observed for the Bi-HTS and PLZT. It is seen from fig. 2 and 3 that τ_{max} increases with increasing T_1 at $T_1 < E_f/k$. After cooling the sample from T_1 to T_0 the main contribution to the power release yields the TLS with the energy $E = 2kT_1$. Thus, in contradiction with the tunnelling theory τ_{max} increases with the energy, e.g the strong temperature dependence of τ_{max} is not caused by strong energy dependence of τ_{max} but by the thermal activation of a probably unknown relaxation process. For example, the tunnelling between the ground state and the first excited level of the double well potential could dominate at large λ and E. In this case τ_{max} is roughly proportional to $\exp(E_{TA}/kT_0)$, where E_{TA} is the energy of the first excited level[23].

For a better understanding of the problem it is necessary to measure the exact T_0 dependence of τ_{max}. Since τ_{max} of the Bi-HTS and PLZT is relatively short at 1.5K, a variation of T_0 below 1K is only reasonable, which is difficult to do with our apparatus. We also attempt to check τ_{max} for a $YBa_2CU_3O_7$ - sample (E_f/k = 17K)[23]. Here an analogous change of the time

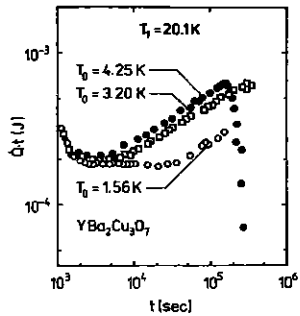

Fig. 5: The Power released in $YBa_2Cu_3O_7$ after cooling from the equilibrium temperature T_1 = 20.1K to various T_0 as a function of time.

dependence of the power release was observed after cooling the sample from T_1 = 20.1K to 4.2K for $t \geq 2 \cdot 10^5$ sec.(s. fig. 5) At lower T_0 τ_{max} shifts to very large values (above 10^6 sec at 3.2K), while at higher T_0 (T_0 > 4.2K) the sensitivity of the power release measurement decreases rapidly because of the higher heat capacity of the sample and the lower sensitivity of the Ge-thermometer. Thus, the exact T_0 dependence of τ_{max} is still unknown.

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