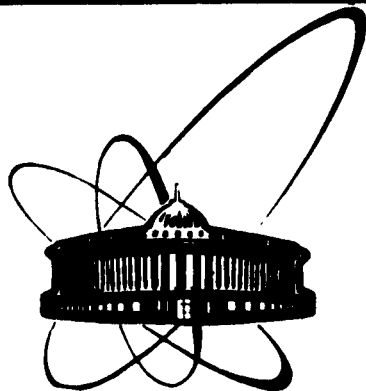


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GLASSY BEHAVIOUR OF LOW TEMPERATURE  
ENERGY RELAXATION IN  $\text{YBa}_2\text{Cu}_3\text{O}_7$   
AND  $\text{YBa}_2\text{Cu}_3\text{O}_6$

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## 1. INTRODUCTION

In all calorimetric experiments with high temperature superconductors (HTS) a linear term of the heat capacity was observed at low temperatures. For  $\text{YBa}_2\text{Cu}_3\text{O}_7$  reported data range from 4 to 80  $\text{mJ/mole}\cdot\text{K}^2$ <sup>1/</sup>. The sample dependent excess contribution above 4  $\text{mJ/mole}\cdot\text{K}^2$  must be attributed to other phases or chemical impurities. However, smaller values could be of intrinsic origin - for example due to the existence of two-level systems (TLS) like in glasses. Indeed, the results of dynamic experiments (thermal) conductivity<sup>2/</sup>, sound attenuation and the relative variation of the sound velocity<sup>3,4/</sup>, are consistent with the results of amorphous solids. Since the broad distribution of the relaxation time  $\tau$  up to very large values ( $10^6 \text{ s} < \tau$ ) seems to be a characteristic feature not only of amorphous solids, but also of structural glasses<sup>5,6/</sup>, a long-time energy relaxation after rapid temperature changes can be expected for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and other HTS. . Measuring the time and temperature dependence of the power  $Q(T_1, T_0, t)$  released (absorbed) after rapid cooling (heating) the sample from the equilibrium temperature  $T_1$  to  $T_0$ , the density of states (DOS) of TLS, and its energy and time dependence can be obtained. The advantage of this method is that only relaxation processes with large relaxation time contribute to the power released or absorbed. While the contribution of normal electrons, magnetical impurities and - at higher temperatures - the phonons complicate essentially the analysis of the heat capacity measurements, there is no such influence on the long-time energy relaxation, and the DOS of TLS with its energy dependence can be determined exactly.

## 2. MODEL

Low temperature long time energy relaxation observed in amorphous solids can be treated within the standard phenomenological tunnelling model<sup>7,8/</sup>. If one assumes a uniform distribution of TLS,

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

where  $\Delta$  is the asymmetry energy; and  $\lambda$ , the tunnelling parameter, the standard tunnelling theory gives a time dependent density of states

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

a time dependent heat capacity

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

and a corresponding power released or absorbed after changing the temperature from the equilibrium temperature  $T_1$  to  $T_0$ .

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

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

### 3. EXPERIMENTAL

Forty-six cylinders of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  ( $T_c = 91\text{K}$ , diameter: 1.2 cm, thickness:  $0.1 \div 0.3$  cm, mass density:  $3,4 \text{ g/cm}^3$ ) were connected by copper foils and a copper holder with a Ge-thermometer, a heater and a copper wire to provide the thermal contact with a mechanical heat switch. The sample (42.845 g  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and 37.615 g Cu) hung in the calorimeter on 12 kapron threads (0.15 mm in diameter, 30 mm long).

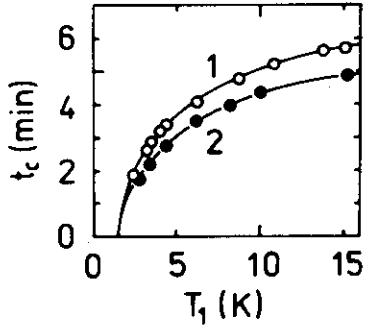
The phase analysis by X-ray diffraction of our sample gives the well-known superconducting ortho-rhombic phase as the main component. The total amount of the other phases is less than 5%.

For the heat release study the resistance drift of the thermometer was measured as a function of the time  $t$  after the rapid cooling of the specimen from equilibrium temperature  $T_1$  (where the specimen remained for at least 10 hr) to  $T_0 = 1.5 \text{ K}$ . The time required for the cooling from  $T_1$  down to  $T_0$  is shown in fig.1.

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

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

Fig. 1. The time  $t_c$  required for the cooling the specimen from  $T_1$  down to  $T_0 = 1.5$  K. 1 -  $\text{YBa}_2\text{Cu}_3\text{O}_7$  2 -  $\text{YBa}_2\text{Cu}_3\text{O}_6$ .



where

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

$$\dot{Q}_A = A(T_K - T), \quad (7)$$

C is the heat capacity of the specimen,  $\delta R/\delta T$  is the sensitivity of the thermometer, 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 ( $10^{-4}$  K). For  $T(t) - T_0 = 10^{-2}$  K the temperature was set to  $T_0 = T_K$  by help of the heat switch. The values of  $A = 0.14$  nW/mK and  $Q_B = 0.5$  nW were determined experimentally. The accuracy of the measurement is limited by the time fluctuations of  $\dot{Q}_B$ , approximately 0.1 nW.

To estimate the heat release in the holder due to possibly hydrogen bubbles in copper<sup>9,10/</sup>, the specimen was kept at 4.2 K for one week before beginning the heat release measurements. As was shown, the power release in copper is negligible for  $T_1 < 20$  K<sup>20/</sup>. At last, the heat capacity was measured.

To convert  $\text{YBa}_2\text{Cu}_3\text{O}_7$  into the nonsuperconducting tetragonal  $\text{YBa}_2\text{Cu}_3\text{O}_6$  the cylinders were kept 7 hr at 800°C in vacuo, 5 hr at 800°C in a  $\text{N}_2$ -atmosphere and quenched. Then the cylinders were mounted to the copper holder and the power release and the heat capacity were measured again.

"Short heating" experiments were performed with both samples: after complete power relaxation at  $T_0 = 1.5$  K the sample was in two minutes heated to  $T_H = 15.1$  K and after the time interval  $t_H = 1$  hr rapidly cooled down to  $T_0$  again, then the power release  $Q(T_H, T_0, t_H, t)$  was measured. In these experiments the same results were obtained as for  $t_H = 20$  hr.

## 4. RESULTS AND DISCUSSION

### 4.1. Heat Capacity

Results of the heat capacity measurement between 1.2 and 15 K for our  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{YBa}_2\text{Cu}_3\text{O}_6$  are shown in fig.2 as

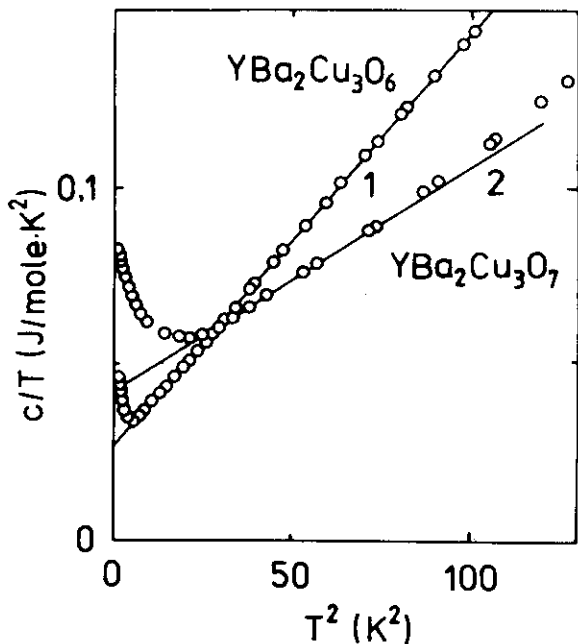


Fig. 2. The heat capacity as a plot  $c/T$  against  $T^2$ . 1 - eq.(8) with  $a=27$  mJ/mole $\cdot$ K $^2$  and  $b=1.18$  mJ/mole $\cdot$ K $^4$ . 2 - eq.(8) with  $a=42$  mJ/mole $\cdot$ K $^2$  and  $b=0.64$  mJ/mole $\cdot$ K $^4$ .

a plot of  $c/T$  against  $T^2$ . The upturn in  $c/T$  which arises from the presence of paramagnetic transition metal and rare earth ion impurities has the character of the high-temperature tail of a Schottky anomaly (proportional to  $T^{-2}$ ), with a Schottky peak

resolvable by applying a high magnetic field<sup>/11/</sup>. Assuming

$$c/T = a + bT^2, \quad (8)$$

in the temperature range with a linear dependence of  $c/T$ , one obtains  $a$  by extrapolation to  $T = 0$  K. The obtained values of the linear term  $a$ , the Debye-Temperature  $\theta_D$  (calculated from  $b$ ) and the transition temperature  $T_C$  are given in table 1 together with some results from ref. 12 and 13.

The Schottky anomaly and a large linear term  $a = 42$  mJ/mole $\cdot$ K $^2$  (about 10 times larger than the observed minimum value) indicate a high concentration of chemical and phase impurities. After the oxygen reduction both the Schottky anomaly and the linear term ( $a = 27$  mJ/mole $\cdot$ K $^2$ ) decreases noticeable: the annealing procedure not only changes the concentration of the labile oxygen but also influences the impurities and defects. In particular, the heat treatment changes the low temperature heat capacity anomaly of  $BaCu_2O_{2+x}$ <sup>/13/</sup>. As was shown recently, even a small proportion (1%) of  $BaCu_2O_{2+x}$  in  $YBa_2Cu_3O_7$  could contribute significantly to the linear term and the Schottky anomaly<sup>/13,14/</sup>. Therefore only an upper limit of the DOS can be obtained from the heat capacity measurements.

Table 1

Transition temperature  $T_C$ , linear term of the specific heat  $a$ , Debye temperature  $\theta_D$  for  $YBa_2Cu_3O_{7-x}$

compound	$T_C$ , K	$a$ , mJ/mole $\cdot$ K <sup>2</sup>	$\theta_D$ , K	reference
$YBa_2Cu_3O_7$	91	42	343	this work
$YBa_2Cu_3O_6$	-	27	280	this work
$YBa_2Cu_3O_7$	95	7.5	426	/12/
$YBa_2Cu_3O_6$	-	5.6	330	/12/
$YBa_2Cu_3O_7$	90.5	3.9	392	/13/

#### 4.2. Heat Release

The specific power release  $\dot{q} = \dot{Q}/m$  ( $m = 42.854$  g) after cooling the  $YBa_2Cu_3O_7$  sample from various  $T_1$  to  $T_0 = 1.5$  K as a function of time ( $t = 0$  at the beginning of cooling down) is given in fig. 3. The power release shows a time dependence typical of glasses:  $\dot{Q}$  is exactly proportional to  $t^{-1}$  for all  $T_1$  and  $t > 3t_c$ , without any systematic deviation even after 30 hr.

For  $YBa_2Cu_3O_6$  the same time dependence of  $\dot{q}$  was observed with 20% larger values of  $\dot{q}$  for the same  $T_1$ ,  $T_0$  and  $t$ .

Two sources of an anomalous long time heat relaxation at low temperatures are only known up to now: the ortho-para conversion of molecular hydrogen, which is present in some materials in form of bubbles<sup>/9/</sup>, and the existence of TLS with a broad distribution of relaxation times up to very large values in glasses. The highest equilibrium temperature  $T_1$  was about 15 K in our experiments. The power release in hydrogen bubbles is proportional to the square of the ortho hydrogen concentration<sup>/10/</sup>, which is about  $5 \cdot 10^7$  times larger at room temperature than at 15 K. Since the sample was kept at helium temperature for one week before beginning the heat release measurements, a possible contribution would be negligible. Such conclusion can be made from the exact  $t^{-1}$  dependence of the power release, since a  $t^{-2}$  dependence would be observed at large  $t$  for the ortho-para conversion. Thus the appearance of the low temperature long time energy relaxation

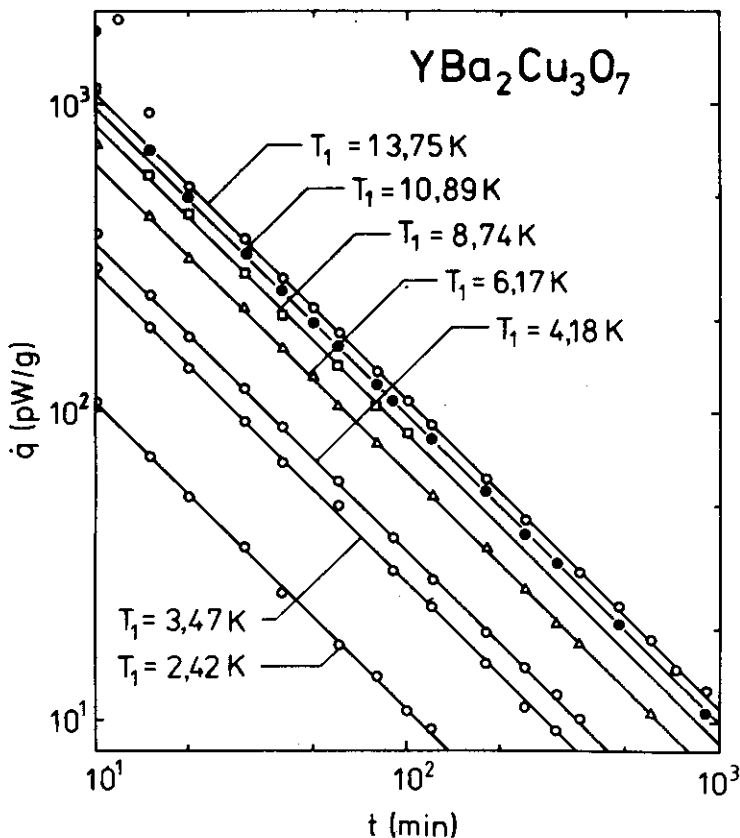


Fig. 3. The specific power  $\dot{q} = \dot{Q}/m$  released in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  after rapid cooling from various equilibrium temperature  $T_1$  to  $T_0 = 1.5$  K as a function of time  $t$ . Straight lines:  $\dot{q}$  proportional to  $t^{-1}$ .

is a clear evidence that TLS exist in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{YBa}_2\text{Cu}_3\text{O}_6$  like in amorphous solids. The  $t^{-1}$  dependence of the power release shows the validity of the tunnelling theory distribution of the form (1) even for very large values of the tunnelling parameter  $\lambda$ .

For  $T_1 < 3.5$  K the temperature dependence of  $\dot{q}$  predicted by the tunnelling theory (see eq. (4) and fig. 3) was found and the distribution parameter  $\bar{P}$  can be determined. Table 2 shows the parameter  $\bar{P}_m = \bar{P}/\rho$  ( $\rho$  is the mass density) obtained by acoustic, heat capacity and heat release measurements for  $\text{YBa}_2\text{Cu}_3\text{O}_7$ ,  $\text{YBa}_2\text{Cu}_3\text{O}_6$  and vitreous silica. Reported minimum

values of the linear term of the heat capacity  $a = 3.9 \text{ mJ/mole}\cdot\text{K}^2$ <sup>/13/</sup> and  $a = 5.6 \text{ mJ/mole}\cdot\text{K}^2$ <sup>/12/</sup> were used to estimate the upper limit of  $\bar{P}_m$  for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{YBa}_2\text{Cu}_3\text{O}_8$  respectively. For  $\bar{P}_m$ -calculations from the value  $\bar{P}_m^2$  obtained in acoustic<sup>/3/</sup> and vibrating reed experiments<sup>/4/</sup> the  $\gamma$ -values of Golding et al.<sup>/3/</sup> ( $\gamma_1 = 0.03 \text{ eV}$  and  $\gamma_t = 0.02 \text{ eV}$ ) were used. In comparison to vitreous silica ( $\gamma_1 = 1.6 \text{ eV}$  and  $\gamma_t = 1.1 \text{ eV}$ )<sup>/15/</sup> the coupling constants of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  are significantly smaller.

Table 2

The distribution parameter  $\bar{P}_m = \bar{P}/\rho$  developed from heat capacity, acoustic and heat release measurements  
 $\bar{P}_m, 10^{38}/\text{Jg}$

material	acoustic	heat release	heat capacity
vitr. silica	$1.0 \div 1.4$ <sup>/16,17/</sup>	$0.45 \div 1.5$ <sup>/18-20/</sup>	$2.4 \div 3.4$ <sup>/21/</sup>
$\text{YBa}_2\text{Cu}_3\text{O}_7$	$0.9 \div 4.5$ <sup>/3,4/</sup>	2.4 this work	$< 13.7$ <sup>/12/</sup>
$\text{YBa}_2\text{Cu}_3\text{O}_8$	-	2.9 this work	$< 16.8$ <sup>/13/</sup>

According to the data of table 2 three important conclusions can be made:

1. The DOS of TLS in the polycrystalline rare earth compounds are even larger than in vitreous silica. Therefore these materials are of interest for detailed study of the glassy behaviour of crystalline solids independently of the question whether the TLS are of intrinsic or extrinsic origin.

2. The reasonable agreement between the  $\bar{P}$ -values obtained from heat capacity, acoustic and heat release measurements show that the DOS is only weakly sample dependent. Higher DOS in our sample with a large linear term of the heat capacity ( $c/T = 42 \text{ mJ/mole}\cdot\text{K}^2$ ) would be otherways expected. With  $\bar{P}_m = 2.4 \cdot 10^{38}/\text{Jg}$  and  $\ln(4t/\tau_{\text{min}}) = 30$ , eq.(3) yields  $c(T, 100 \text{ s})/T = 0.8 \cdot \text{mJ/mole}\cdot\text{K}^2$ , i.e. only 2% of the linear term observed for our  $\text{YBa}_2\text{Cu}_3\text{O}_7$  specimen are due to TLS. Thus, it is unprobably that the TLS are caused by chemical or phase impurities. If we take into account a typical glassy behaviour of the sound velocity and the internal friction of all investigated HTS<sup>/2/</sup>, we can expect a long time energy relaxation at least for all high-temperature superconducting ceramics. Systematical investigations of the long time energy relaxation in HTS seem to be reasonable.



3. The conversion of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  to  $\text{YBa}_2\text{Cu}_3\text{O}_6$  does not change the DOS essentially. If the TLS are caused by the labile oxygen a decrease of the DOS after the oxygen reduction is expected. However, the dependence of the oxygen concentration can be more complicate. If we assume that the tunnelling rate depends on the vacancy and oxygen concentration, a maximum value of the DOS appears for an intermediate oxygen concentration. Indeed, an increase of the internal friction plateau was observed even after a small reduction of the oxygen concentration<sup>/9/</sup>, which could be due to an increase of the DOS.

At temperatures  $T_1 > 3.5$  K, the temperature dependence of the power release disagrees with eq. (4) (see fig. 4). Similar deviations were observed for all investigated glasses<sup>/5/</sup>.

The simplest explanation of this behaviour would be that the DOS of the TLS does not extend beyond energies above a cut-off energy  $E_f$ . Indeed, a surprisingly good agreement with the experimental data of all investigated glasses was obtained using the energy dependent distribution function<sup>/5/</sup>

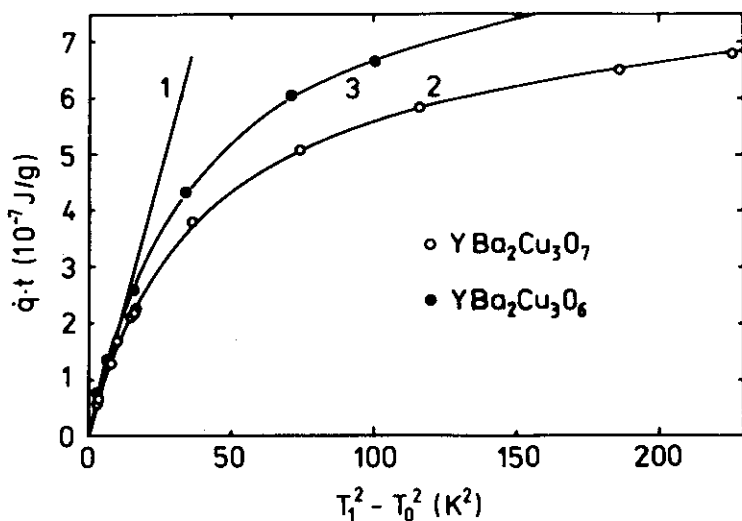


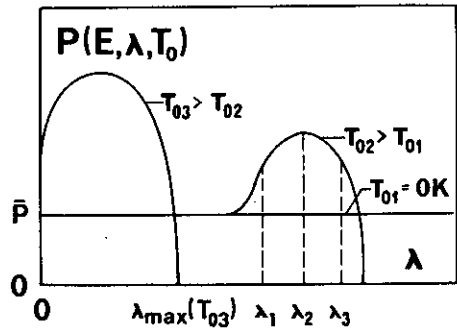
Fig. 4. The value  $\dot{q} \cdot t$  as a function of  $T_1^2 - T_0^2$  ( $T_0 = 1.5$  K). 1 - standard tunnelling theory<sup>/7,8/</sup> (eq.(4) with  $\bar{P}_m = 2.4 \cdot 10^{38}$ /Jg. 2,3 - modified tunnelling theory<sup>/5/</sup> based on the energy dependent DOS according to eq. (9) with  $E_f/k = 13$  K,  $T_b = 0$  K,  $\bar{P}_m = 2.4 \cdot 10^{38}$ /Jg (curve 2) and  $\bar{P}_m = 2.9 \cdot 10^{38}$ /Jg (curve 3).

$$n(E, t) = n_0(t) / (1 + \exp((E - E_f) / kT_b)), \quad (9)$$

where  $E_f$  and  $T_b$  are constant and  $n_0(t)$  is determined by eq.(2). Moreover, the small variation of  $E_f$  for various glasses ( $7.5 \text{ K} \leq E_f/k \leq 24 \text{ K}$ ) indicates an universal behaviour of the energy dependence. Assumption (9) with  $E_f/k = 13 \text{ K}$  and  $T_b = 0 \text{ K}$  yields a good fit for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{YBa}_2\text{Cu}_3\text{O}_6$  (see fig. 4, curves 2 and 3), too.

However, measurements of the  $T_0$  dependence of  $\dot{q}$  for epoxy resin<sup>/22/</sup>, and short heating experiments with  $\text{YBa}_2\text{Cu}_3\text{O}_7$ ,  $\text{YBa}_2\text{Cu}_3\text{O}_6$  and various glasses<sup>/5,23/</sup> show that assumption (9) is uncorrect. In particular, it was found for epoxy resin that the value of the power  $\dot{Q}(T_1, T_0, t)$  released disagrees with the absolute value of the power  $\dot{Q}(T_0, T_1, t)$  absorbed for the same  $T_1, T_0$  and  $t$  if  $T_0 > 2 \text{ K}$ . For explanation of these results we must assume that the relaxation time or the tunneling parameter became temperature dependent at higher temperatures, i.e. another temperature dependent relaxation process exist. The temperature dependence of the distribution function  $P(\Delta, \lambda)$  necessary for explanation of the long-time energy relaxation experiments is shown in fig.5 qualitatively. For low temperatures  $T_0$  the temperature dependent relaxation process influences only the distribution for large  $\lambda$ , while for shorter  $\lambda$  the distribution function believes unchanged. With increasing  $T_0$  the maximum of the distribution function shifts to lower  $\lambda$ . From fig. 5, we can predict some unusual behaviour of the time and temperature dependence of the power release.

Fig. 5. A qualitative plot of the distribution function  $P(E, \lambda)$  as a function of  $\lambda$  for various  $T_0$ . The parameters  $\lambda_1, \lambda_2, \lambda_3$  and  $\lambda_{\min}$  see text.



1. A maximum value  $\lambda_{\max}$  and a corresponding  $\tau_{\max}$  exist in  $T_0$  dependence. For high temperatures ( $T > T_f$ ), the sample cooling is not fast enough and all excitations relax during the cooling procedure. This excitation with energies  $E > E_f$  does not contribute to the power release after cooling.

2. The main part of the power released or absorbed is due to TLS with  $\tau \approx t$  and the corresponding effective tunnelling parameter  $\lambda_{\text{eff}} = \lambda(\tau) = \lambda(t)$ . With increase of  $t$  the effective  $\lambda_{\text{eff}}$  also increases. If  $\lambda_{\text{eff}}$  reaches the distribution anomaly, the time dependence changes, the power relaxes first slower ( $T_0 = T_{02}$ ,  $\lambda_{\text{eff}} = \lambda_1$ ) and then faster than  $t^{-1}$  ( $T_0 = T_{02}$ ,  $\lambda_{\text{eff}} = \lambda_3$ ).

3. Measuring the power release after cooling from high  $T_1$  ( $T_0 \ll T_1$ ), an increase of the power release can be observed after an increase of  $T_0$ , changing the distribution anomaly in the region of  $\lambda_{\text{eff}}$  ( $\lambda_{\text{eff}} = \lambda_2$ ,  $T_{01} \rightarrow T_{02}$ ). A similar deformation explains that the absolute value of the power release  $\dot{Q}(T_1, T_0, t)$  can be larger than the power absorption  $\dot{Q}(T_0, T_1, t)$  for the same  $T_1$ ,  $T_0$  and  $t$  ( $T_{01} = T_0$ ,  $T_{02} = T_1$ ,  $\lambda_{\text{eff}} = \lambda_2$ ).

4. If we heat the sample rapidly from  $T_{01}$  to  $T_H = T_{03}$  and keep the sample at  $T_{03}$  for the time interval  $t_H$ , the power released after the cooling to  $T_{01}$  is independent of  $t_H$  for  $t_H > \tau_{\text{max}}$ , where  $\tau_{\text{max}}$  is determined by  $\lambda_{\text{max}}(T_{03})$ . Thus the "short heating" experiments can be explained.

The behaviour according to 1. and 4. was found for  $\text{YBa}_2\text{Cu}_3\text{O}_7$ ,  $\text{YBa}_2\text{Cu}_3\text{O}_8$  and all other investigated glasses<sup>/5/</sup>. Until now, the features 2 and 3 were observed for epoxy resin only. For other glasses the  $T_0$  dependence was not investigated.

If we assume that the temperature dependent transformation of the distribution function according to fig. 5 is correct, experimental parameters depending on shorter  $\tau$  or  $\lambda$  will also show a different behaviour than predicted by the tunnelling theory. However, these anomalies we can expect to find at higher temperatures than the temperature, where the long-time power release deviate from the tunnelling theory. Indeed, at higher temperature a plateau of the thermal conductivity and an "unknown" relaxation peak of the internal friction was observed for the most investigated glasses. Thus these anomalies seem to be connected. In particular, the same  $T_1$  dependence of the power release ( $T_f = 13$  K,  $T_b = 0$  K) was observed for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and vitreous silica<sup>/11,5/</sup>, and an increase of the internal friction above 5 K with a relaxation peak at 36 K<sup>/4/</sup> and 31 K<sup>/24/</sup> respectively were found for both materials. Moreover, in accordance to fig. 5, the internal friction relaxation peak of vitreous silica<sup>/24/</sup> and  $\text{YBa}_2\text{Cu}_3\text{O}_7$ <sup>/25/</sup> moves to higher temperatures increasing the measuring frequency.

The temperature dependent relaxation process can be the consequence of, at least, two reasons: the Raman process

(interaction between the TLS and two or more phonons) and the thermal activation process.

At low temperatures only the first order Raman process is of interest (two phonons), yielding a  $T^7$  dependence<sup>/26/</sup> of the relaxation rate. However, both relaxation rates of TLS interacting with one and two phonons are proportional to  $(\Delta_0/E)^{2/28/}$ , where

$$\Delta_0 = \hbar\Omega \exp(-\lambda), \quad (10)$$

and  $\hbar\Omega/2$  is the ground state energy.

That means that for a given energy and temperature the deformation of the distribution function is the same for small and large  $\lambda$ , assuming the coupling constants to be  $\lambda$  independent. Moreover, it was shown in the framework of the "soft configuration" theory<sup>/27/</sup> - yielding identical with the tunnelling theory results for low temperatures - that the two and more phonon interaction can be neglected below 100 K<sup>/28/</sup>. Thus, the temperature dependent relaxation process is not or only partially caused by the Raman process.

If we assume that the temperature dependence is due to the thermal activation process, an unusual distribution of the potential barrier heights will be obtained. Since the thermal activation sets in at low temperatures only for large  $\lambda$ -values, the potential barrier height  $V_b$  must decrease with increasing  $\lambda$ . However,  $\lambda$  is determined by

$$\lambda = d(2mV_b)^{1/2}/\hbar. \quad (11)$$

Thus the decrease of  $V_b$  must be compensated by a stronger increase of the distance  $d$  between the potential minima or the mass  $m$  of the tunnelling particles. This result is difficult to be understood in the "one particle" picture of the tunnelling model. Since so far the nature of the tunnelling entity is not known neither in general nor in specific cases, this result indicates ones more that the "one particle" picture can be too simple for explanation of the low temperature anomalies of glasses in detail.

For various HTS a long-time relaxation of the magnetization was observed for temperatures near  $T_c$ <sup>/29/</sup> and for low temperatures ( $T \ll T_c$ )<sup>/30/</sup>, too. For example, if a small field  $H_1 < H_{c1}$  is applied to a  $\text{Sr}_{0.2}\text{La}_{1.8}\text{CuO}_4$  or  $\text{Ba}_{0.15}\text{La}_{1.85}\text{CuO}_4$  sample at  $T = 0.8$  K, the isothermal dc magnetization  $M$  is unstable in time, following the law

$$M - M_0 \sim TH_1^n \ln(t/t_0) \quad (11')$$

with  $n = 3$  during a  $10^5$  sec observation<sup>/30/</sup>.

There is a surprising similarity between the relaxation behaviour of HTS and a certain class of perovskite ferroelectrics like  $\text{Pb}_{0.915}\text{La}_{0.085}(\text{Zr}_{0.65}\text{Ti}_{0.35})\text{O}_3$  (PLZT), exhibiting a diffuse phase transition. In these materials properties typical of glasses were observed at low temperatures<sup>/31/</sup>. In particular, experiments with PLZT show not only a logarithmic time dependence of the electric polarization after switching on or off an electric field for high ( $T \approx T_c$ ) and low temperatures ( $T \ll T_c$ ), but also a  $t^{-1}$  dependence of the power released after rapid cooling or after switching on or off an electric field<sup>/6/</sup>. This indicates that in both materials the glassy behaviour is due to structural instabilities. It seems to be remarkable, that these low temperature anomalies were found not only for ferroelectrical polycrystalline ceramics, but also for monocrystals as, for example,  $\text{Pb}(\text{Mg}_{0.33}\text{Nb}_{0.67})\text{O}_3$ <sup>/31/</sup>. From this point of view glass like behaviour of high temperature superconducting monocrystals can be expected, i.e. this behaviour is of intrinsic origin.

At last, a connection between the low temperature anomalies and the high  $T_c$  could exist. An attempt to explain the high  $T_c$  and the glassy behaviour as a consequence of structural instabilities is given in Ref. 32.

## 5. CONCLUSIONS

A typical of amorphous solids and structural glasses time and temperature dependence of the power  $Q(T_1, T_0, t)$  released after rapidly cooling a  $\text{YBa}_2\text{Cu}_3\text{O}_7$  sample from various equilibrium temperatures  $T_1$  ( $2.35 \text{ K} \leq T_1 \leq 15.1 \text{ K}$ ) to  $T_0 = 1.5 \text{ K}$  was observed.

For all  $T_1$  and  $T_0 = 1.5 \text{ K}$  an exact  $t^{-1}$  dependence was found for  $3t_c < t < 30 \text{ hr}$ . At low temperatures ( $T_1 < 3.5 \text{ K}$ ) the results agree with the tunnelling theory well and the distribution parameter  $\bar{P}_m$  can be calculated. The  $\bar{P}_m$  value of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  agrees with values obtained in acoustic experiments. The TLS give a contribution to the linear term of the heat capacity ( $c(T, t)/T = 0.8 \text{ mJ/mole}\cdot\text{K}^2$ ), which is significantly smaller than the observed linear term of our sample ( $42 \text{ mJ/mole}\cdot\text{K}^2$ ), caused mainly by phase and chemical impurities.

While the linear term of the heat capacity decreases noticeable, the power released and the DOS of TLS increases of about 20% after the conversion of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  to  $\text{YBa}_2\text{Cu}_3\text{O}_6$ . This result indicates that the TLS are probably not caused by chemical or phase impurities.

For  $T_1 > 3.5$  K the temperature dependence of the power release disagrees with the tunnelling theory. Similar deviations were observed for all investigated amorphous solids and structural glasses.

The assumption that the DOS of TLS does not extend beyond energies above a cut-off energy  $E_f$  yields a good fit for the observed  $T_1$  dependence. However, "short heating" experiments indicate that an unknown temperature dependent relaxation process exists, probably due to the thermal activation. From our experiments a qualitative plot of the distribution function  $P(E, \lambda)$  as a function of  $T_0$  can be obtained. At low  $T_0$  the unknown relaxation process produces an anomaly of the distribution function only for large  $\lambda$ -values. With increasing  $T_0$  this anomaly changes to lower  $\lambda$ . This relaxation process probably causes also the "unknown" relaxation peak of the internal friction and the plateau of the thermal conductivity observed for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and the most of investigated amorphous solids and structural glasses.

Astonishing similarities exist between HTS and perovskite-ferroelectrics with diffuse phase transitions in the sense of low temperature anomalies and relaxation phenomena. This indicates that in both materials the glass like behaviour is due to structural instabilities which probably exist also in monocrystals.

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