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UNIVERSAL RELAXATION BEHAVIOUR OF DISORDERED SOLIDS

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INTRODUCTION

At low temperatures the thermal, ultrasonic and dielectric properties of glasses and various types of cristalline solids strongly depart from current Debye-model.

Usually these glassy anomalies are explained by the phenomenological tunneling-states model (TSM) /1,2/ without any information about the nature of the tunneling entities.

Due to the high sensitivity of the dielectric constant to the low-lying excitations ferroelectric crystals serve as excellent samples for studying the problem and testing any theoretical interpretation of the effect.

The purpose of this paper is to present some typical results obtained by dielectric measurements on ferroelectric crystals at low temperatures and to mention substantial characteristics of a new approach to understanding the glassy behaviour, first proposed in /3/.

THE UNIVERSAL DIELECTRIC RELAXATION

In general, the complex dielectric susceptibility of disodered solids exhibits a broad relaxation peak at temperatures $T \gtrsim 10K$. This is shown, for example, for the relaxation ferroelectric $Sr_{0.5}Ba_{0.5}(Nb_2O_6)$ in Fig.1.

Such peaks in the dielectric losses χ'' do not adhere to Debye--theory and can be considered using the universal response (UDR)-idea, outlined first in Ref./4/.

Owing to the empirical formula

$$1/\chi''(\omega,T) \sim T\left[(\tau\omega)^{1-n} + (\tau\omega)^{-m}\right] \quad (1)$$

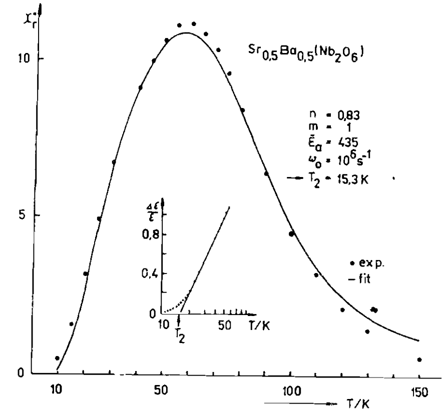

Fig.1. Dielectric loss peak of a SBN (50% Ba)-single crystal, ($\omega = 10^4\ s^{-1}$).

with $0 < n, m < 1$, ω – the angular frequency and a thermally activated relaxation time $\tau = \tau_o \exp(\tilde{E}_a/K_B T)$, a three-level fit of the characteristic exponents n,m and of the attempt frequency $\omega_o = 1/\tau_o$ provides a good confirmation of the experimental data and a deduced activation energy $\tilde{E}_a$. The Kramers-Kronig transformation gives the variation of the real part $\varepsilon(T)$ in accordance with the experimental results, as shown in the inset of Fig.1.

The universality of this Non-Debye relaxation suggests a many-body interpretation where the characteristic exponents indicate the interaction strength of the relaxing units /5/. The value of n found for the above samples emphasizes the high degree of the dipole-dipole interactions in this single crystals. It has been recently suggested /6/, that the obtained $\tilde{E}_a$ is only an apparent activation energy, related to the actual one by:

$$E_a = (1 - n)\,\tilde{E}_a \quad .$$

The obtained activation energy (E_a/K_B = 74 K) agrees well with the main Einstein-mode (Θ_E = 73 K) found in the specific heat C of a SBN (55% Ba) – sample /7/, that causes the C/T^3 – peak just at temperatures near the characteristic plateau in the thermal conductivity /8/.

An **extension** of the UDR-model was proposed to explain both the analogous effects in internal friction and ultrasonic relaxation behaviour of disordered solids and the unifying features of other low-frequency fluctuation, dissipation and relaxation phenomena /6/.

THE UNIVERSAL RELAXATION CONCEPT AND THE GLASSY BEHAVIOUR

A further generalization of the universal relaxation conception was shown to give a physical relevant interpretation of the correlating glassy anomalies in thermal, ultrasonic and dielectric properties /3/. According to that, the thermal conductivity λ can be obtained using the mean free-path $l(\omega)$ of the phonon scattering determined by the generalized susceptibility corresponding to

$$l(\omega) = [\exp(\hbar\omega/K_B T) - 1]/\chi''(\omega,T). \qquad (2)$$

In general χ'' is a more comprehensive function of ω and T than given by eq. (1) /9/, but in the particular case using this empirical formula eq. (2) will provide a simple explanation of the well-known plateau in λ (T) in terms of the dominant phonons ω_{Dom} :

$$\hbar\omega_{Dom}(T_p) \approx E_a \qquad (3)$$

with T_p – the mean temperature of the plateau.

Summing up, at $T > 1$ K the glassy anomalies can be explained by means of the universal Non-Debye relaxation mechanism due to many-body interactions. Therefore any extrapolation of the TSM beyond the low-temperature region using various forms of an energy dependent density of states P(E) /10-12/ is not required and even contradicts a growing number of experimental data /3,13/.

THE UNIVERSAL RELAXATION BELOW 1 K

At $T \leq 1$ K the characteristic properties of disordered solids seem to be in accordance with the prediction of the TSM given in Ref. /1,2/, suggesting a constant density of states P_o. This model explains, for example, the minimum in the real part of the dielectric constant $\varepsilon(T)$ at T_o as a result of the competitive influence of resonant and relaxation scattering of the electromagnetic waves on tunneling states.

At $T > T_o$ the relaxation process dominates and a logarithmic increase of $\Delta\varepsilon/\varepsilon = [\varepsilon(T)/\varepsilon(T_o)]-1$ is observed. However, the measurements often reveal a lineary dependence of $\varepsilon(T)$ /3,14/. The most distinct representation of this was found for a $PbTiO_3$ - ceramic sample, exhibiting the lineary behaviour for about two decades in temperature (Fig.2).

Analogous results are not compatible with the above TSM, even by considering any modified form of P(E).

To investigate this problem, $\varepsilon(T)$ and the thermal conductivity were measured on two ceramic $SrTiO_3$-samples, prepared by different burning processes.

A qualitative different behaviour of $\varepsilon(T)$ was found for the two samples (Fig.3). Instead of the logarithmic dependence **(full** line), obtained for sample 2, a well defined linear variation **dashed line) in connection with a distinct lower thermal conductivity is observed for the first sample.**

Taking into account the strong correlation between the dielectric and thermal anomalies, a proof analysis of the thermal conductivity and dielectric data obtained for the two samples and for a large range of other disordered solids leads to the following conclusions /3/:

At $T \leq 1$ K the characteristic scattering process, responsible for the approximate T^2 - behaviour of λ, in dielectric (or ultra-

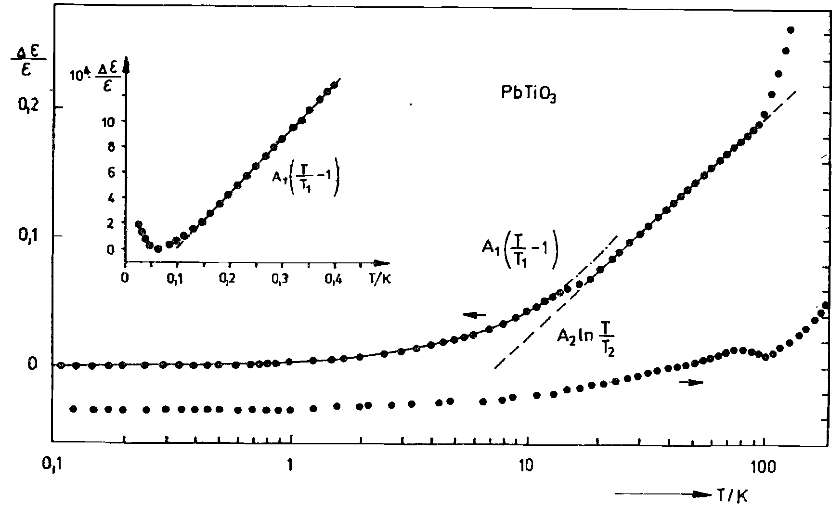

Fig.2. Dielectric behaviour of $PbTiO_3$ ($\omega = 10^4\ s^{-1}$).

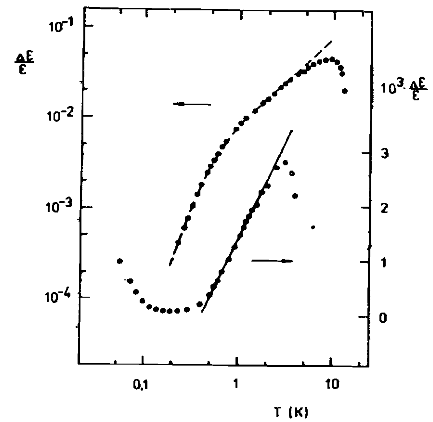

Fig.3. The different behaviour of $\Delta\varepsilon/\varepsilon$ (T) for two $SrTiO_3$--samples, ($\omega = 10^4\ s^{-1}$; full circles - sample 1, open circles - sample 2).

sonic) measurements is not represented by the logarithmic variation with temperature of $\Delta\varepsilon/\varepsilon$ but by the lineary dependence. Then the frequency dependence of this process can be deduced obeying

$$\chi''(\omega,T) \sim \omega^{n-1}\cdot T. \tag{4}$$

Eq. (4) shows that the universal relaxation concept can be extended to explain the glassy anomalies also at $T \lesssim 1$ K. Thus, for instance, the T^2-variation of the thermal conductivity can be understood by considering eqs. (2) and (4) with $n \approx 1$.

CONCLUSIONS

The obtained linear dependence on temperature of the generalized susceptibility is an important new feature of the universal mechanism. This is obviously the result of a cross-over from classical to quantum behaviour of the system, when the temperature is lowered.

These results have confirmed the hypothesis, first put forward in Ref. /3/, that the characteristic glassy anomalies, observed at low temperatures, and the common relaxation behaviour (frequency-, time-, temperature-dependence and others) found in various properties (electric, magnetic, ultrasonic, thermal, ...) of disordered solids are the consequences of one general relaxation mechanism caused by a universal type of many-body interaction. This suggests the possibility of creating a unitary microscopic theory for the mentioned effects probably concerning some fundamental principles of solid state physics.

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Фишер Э. E8-86-241
Универсальное релаксационное поведение дефектных веществ

При низких температурах тепловые, ультразвуковые и диэлектрические свойства стекол и самых разных типов кристаллических твердых тел проявляют сильное отклонение от теории Дебая. Обычно эти аномалии рассматривают на основе феноменологической модели, постулирующей наличие некоего спектра низкоэнергетических туннельных возбуждений. Можно однако показать, что данную проблему следует рассматривать на основе универсального механизма многочастичного взаимодействия, отвечающего в частности и за проявление, так называемой, универсальной диэлектрической релаксации твердых веществ и других эффектов низкочастотных флуктуаций и диссипации. В данной работе представлены некоторые типичные результаты диэлектрических измерений на кристаллических сегнетоэлектриках при низких температурах. Даются главные характеристики нового подхода к данной проблематике.

Работа выполнена в Лаборатории высоких энергий ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1986

Fischer E. E8-86-241
Universal Relaxation Behaviour of Disordered Solids

The low temperature properties of disordered solids known as the glassy behaviour are discussed within a universal relaxation approach. On this basis the characteristic anomalies in thermal and dielectric properties and their strong correlation are shown to be only one aspect of a general relaxation mechanism caused by manybody interaction. At the lowest temperatures the generalized susceptibility of the process is found to obey a linear dependence on temperature, presumed due to quantum effects.

The investigation has been performed at the Laboratory of High Energies, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1986