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Amorphous dielectric behavior of incommensurate ferroelectric $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ at low temperatures

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The complex dielectric susceptibility, $\epsilon = \epsilon' + i\epsilon''$, of the incommensurate ferroelectric $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ has been measured at various frequencies (300 Hz–100 kHz) and excitation voltages (0.15–7.5 V/mm) in the temperature range from 10 to 500 mK. It is found that ϵ' of the crystal studied, similarly to amorphous materials, exhibits a minimum in the millikelvin temperature region. The real part of the dielectric constant at the high-temperature side of this minimum shows an appreciable frequency dependence, and the temperature where the minimum occurs, T_{min} , obeys a power-law dependence on the frequency. This amorphous dielectric behavior of an incommensurately modulated crystal can be described using the two-level system model. We have also observed a strong dependence of ϵ' on the magnitude of the measuring electric field at the low-temperature side of the minimum. We suggest that this nonlinear dielectric behavior is associated with a variation of the height of the potential barriers in the two-level systems as a function of the electric-field strength. [S0163-1829(97)04902-3]

I. INTRODUCTION

It is well known that at low temperatures ($T < 1$ K) the properties of amorphous materials differ considerably from the properties of crystals. In particular, amorphous solids exhibit a linear temperature dependence of the specific heat, a T^2 dependence of the thermal conductivity, and a characteristic temperature and frequency dependence of ultrasonic and dielectric properties, which can all be accounted for by a phenomenological tunneling model.¹ In this model it is assumed that in amorphous materials localized excitations with very low energy exist. These excitations are associated with the spatial disorder inherent for amorphous solids which distinguishes them from crystalline solids. Physically these excitations correspond to the movement of atoms or groups of atoms in a double-well potential. The energy splitting due to tunneling of atoms from one minimum to another needed to explain the properties of amorphous solids below 1 K is very small (less than 10^{-4} eV).¹ In contrast to high temperatures, where the change of the configuration state occurs by thermal activation, at low temperature this occurs through a tunneling process. Although admittedly not amorphous, some ferroelectric crystals have been found to possess thermal and dielectric properties similar to those of amorphous materials.^{2,3} They exhibit a large dielectric response, which makes dielectric measurements on these materials a very sensitive tool for the investigation of amorphous behavior at very low temperatures.

In this paper we report measurements of the dielectric constant of a ferroelectric $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ mixed crystal at temperatures below 1 K. At temperatures below ~ 120 K this compound is in an incommensurate phase. The loss of translational symmetry along the vector of the incommensurate modulation renders the properties of this compound close to those of amorphous materials. Furthermore, the substitution of Sn atoms by Pb atoms causes some structural disorder in the crystal. The high value of the dielectric con-

stant at low temperatures (the main contribution of which comes from the relaxation dynamics of the incommensurate modulation⁴), as well as the two effects mentioned above make this crystal a very attractive object for the study of amorphous behavior of crystalline materials. This is particularly so with respect to the nonlinear dielectric effect (dependence of the dielectric constant on the applied ac electric field) observed in some amorphous materials.⁵ The nature of this effect has not yet been elucidated. It should be mentioned that in amorphous solids this effect is weak, which makes a quantitative and even a qualitative analysis difficult.

The technical importance of the material studied here has been demonstrated⁶ by its use as a sensitive and tunable thermometer at low temperatures, unaffected by large magnetic fields (up to 20 T). However, here we will focus on a comparison between the dielectric properties of $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ and those of amorphous dielectric solids.

In the following we report on measurements of the dielectric constant of a $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal made as a function of temperature, applied electric field, and frequency. We show that the observed dielectric behavior resembles that of amorphous dielectric solids in many respects. We compare the observed frequency dependence of ϵ with a calculation based on the two-level system (TLS) model and give a tentative explanation for the observed electric-field dependence.

II. EXPERIMENT

The $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal was grown using a Bridgmann technique. The crystal (typical size 10 mm in diameter and 25 mm long) was cut and polished to obtain platelets (size $5 \times 5 \times 0.8$) perpendicular to the [100] direction. The samples had sputtered electrodes, which were connected to stainless-steel coaxial wiring with silver paint and a small amount of epoxy.

The measurements of the temperature dependence of the dielectric constant at various excitation voltages (from 0.01 to 50 V/cm) at frequency 1 kHz were performed using an

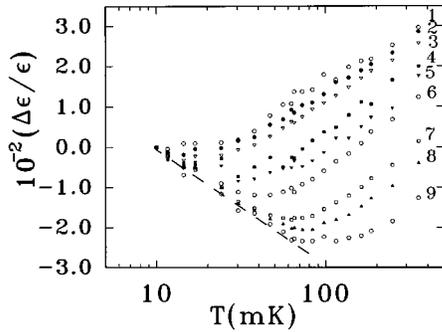


FIG. 1. Temperature dependence of $\Delta\epsilon'/\epsilon'(T_0)$ for $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal at various frequencies: 1–300 Hz, 2–500 Hz, 3–1 kHz, 4–3 kHz, 5–5 kHz, 6–10 kHz, 7–30 kHz, 8–50 kHz, and 9–100 kHz at excitation voltage of 0.2 V/cm; $\epsilon'(T_0)$ is defined here as ϵ' at $T=10$ mK.

Andeen-Hagerling model 2500 automatic capacitance bridge. For the measurements at different frequencies in the range from 300 Hz to 100 kHz, we have used a General-Radio type 1615 A capacitance bridge.

The samples were mounted in the plastic mixing chamber of an adapted SHE dilution refrigerator. Temperatures down to 10 mK were measured with two types of thermometers in the mixing chamber: calibrated Speer 100 Ω carbon resistors and a CMN thermometer.

III. RESULTS

To demonstrate the similarity between amorphous dielectrics and the present compound, the temperature dependence of ϵ' and ϵ'' at various frequencies are shown in Figs. 1 and 2, respectively. To compare experimental results with theory, the variations of the ϵ' with temperature, frequency, or electric field are presented in reduced form: $\Delta\epsilon'/\epsilon' = [\epsilon'(T, \omega, U) - \epsilon'(T_0, \omega_0, U_0)] / \epsilon'(T_0, \omega_0, U_0)$. At high temperatures, where ϵ' increases with temperature, we find a logarithmic temperature dependence for $\Delta\epsilon'/\epsilon'(T_0)$. For almost all frequencies a pronounced minimum in $\Delta\epsilon'/\epsilon'(T_0)(T)$ is observed at $T=T_{\min}$. Only at the lowest frequency used (300 Hz), the minimum in the $\Delta\epsilon'/\epsilon'(T_0)$ dependence is unobservable in the temperature region studied. With an increase in the frequency the minimum becomes observable and gradually shifts to higher temperatures. As is clear from Fig. 1, dielectric dispersion (frequency dependence of ϵ) occurs in the whole temperature region studied. However the depth of the dispersion at temperatures below T_{\min} is smaller than above T_{\min} . Moreover, below T_{\min} the dispersion in question comes to a close: its edge lies at frequencies around 50 kHz. At the same time $\epsilon'(T)$ above T_{\min} decreases monotonically as a function of frequency in the whole frequency range studied.

Concomitant with the minimum in ϵ' , we observe a maximum in ϵ'' . The maximum in $\epsilon''(T)$ gradually disappears as the frequency increases (Fig. 2): at $f \geq 10$ kHz the $\epsilon''(T)$ curves flatten out. As the minimum of $\Delta\epsilon'/\epsilon'(T_0)$ is shifted with an increase in frequency, the part of the $\epsilon''(T)$ curve corresponding to the rapid decrease in ϵ' is shifted to higher temperatures as well. At $T > T_{\min}$ the slope of $\Delta\epsilon'/\epsilon'(T_0)$ curve is nearly constant for all frequencies.

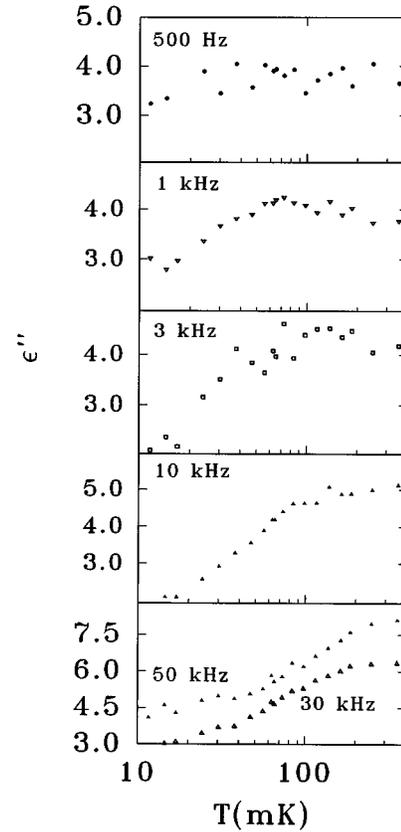


FIG. 2. $\epsilon''(T)$ at various frequencies.

As can be seen from Fig. 1, the $\Delta\epsilon'/\epsilon'(T_0)(T)$ dependences are typical for amorphous dielectric behavior, which is characterized by a minimum. The temperature of the minimum, T_{\min} , depends on the excitation frequency: T_{\min} is shifted upwards with increasing frequency. This behavior is well described by a power law: T_{\min} varies proportional to $\omega^{0.3}$.

To show the dependence of $\epsilon'(T)$ on the strength of the applied ac electric field, the temperature dependence of the real part of the complex dielectric constant, ϵ' at various excitation voltages at 1 kHz, is presented in Fig. 3. We present in Fig. 4 the $\epsilon''(T)$ dependence only for the highest measuring voltages, since the low measurement accuracy of

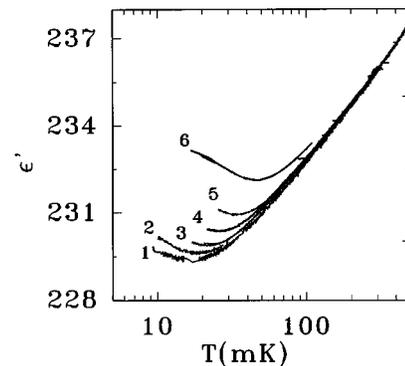


FIG. 3. Temperature dependence of ϵ' for $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal at different excitation voltages 1–0.15; 2–0.30; 3–0.50; 4–1; 5–3.75; 6–7.5 V/cm.

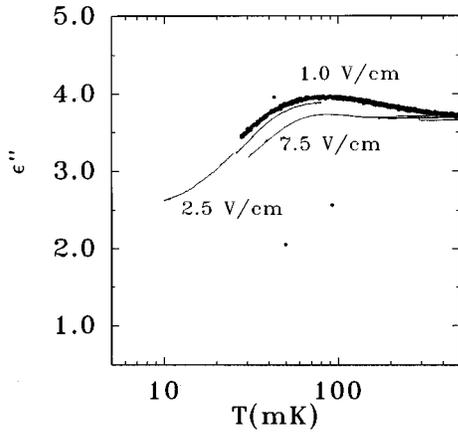


FIG. 4. Temperature dependence of ϵ'' at various excitation voltages.

ϵ'' , especially at low excitation voltages, results in large scatter in these data.

An unexpected peculiarity in the behavior of ϵ' is apparent from Fig. 3, where ϵ' is shown to exhibit a strong dependence on the amplitude of the measuring field near the minimum and in temperature region below T_{\min} . The nonlinear effect observed becomes negligibly small at higher temperatures, at least at the voltages used.

The imaginary part of ϵ , ϵ'' , grows with increasing temperature and after passing the maximum becomes nearly temperature independent. At the highest excitation voltages the maximum becomes less pronounced: ϵ'' as a function of temperature shows a plateau. At higher temperatures ϵ'' tends to the same value for various measuring fields.

IV. DISCUSSION

We first discuss the temperature dependence of the dielectric constant at fixed voltage but with varying frequency. As can be seen from the frequency-dependent data, the dielectric properties of the $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal exhibits amorphous behavior at low temperatures: the occurrence of a minimum in ϵ' that shifts with frequency and a logarithmic temperature dependence on either side of the minimum. This fact has also been observed in some ferroelectric single crystals as well as in polycrystalline ferroelectrics,^{2,3} and has been associated with the presence of ferroelectric domains. However, we have verified that amorphous behavior at low temperatures occurs also in single-domain ferroelectric samples. In particular, the characteristic minimum in ϵ' is observed in both single-domain and poly-domain crystal $\text{Sn}_2\text{P}_2\text{Se}_6$ which crystalizes at low temperatures in the ferroelectric phase. Thus, the microscopic nature of the amorphous behavior of ferroelectrics at low temperatures is not yet entirely known.

The crystal we are dealing with is in the incommensurate phase at low temperatures. The large dielectric response in the incommensurate phase is caused by a high polarizability of the incommensurate structure. Upon lowering the temperature below 50 K the dielectric susceptibility of $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ sharply decreases. It has been suggested that this effect is caused by freezing of the relaxation dynamics of the incommensurate modulation pinned by defects.⁴ A

smooth decrease in the dielectric constant beginning at ~ 50 K continues until very low temperatures. This suggests that the amorphous part of the dielectric response of the crystal at very low temperatures is due to the incommensurate modulation.

Analogous to amorphous materials, the changes in the dielectric constant with temperature and frequency in some crystalline ferroelectrics² at very low temperatures were explained using the tunneling two-level states model.

Let us check the applicability of the tunneling model to the crystal studied here. The two-level states model suggests an energy-dependent density of states, $P(E)$ and a broad spectrum of thermal relaxation times, τ for each two-level state energy E . The energy E is a function of the double-well asymmetry energy Δ and tunneling splitting Δ_0 : $E = \pm(\Delta^2 + \Delta_0^2)^{1/2}$. The thermal relaxation time for a given state is given by¹

$$\tau^{-1}(E) = \sum_{\alpha} \frac{\gamma_{\alpha}^2}{\vartheta_{\alpha}^5} \frac{E \Delta_0^2}{2\pi\rho h^4} \coth\left(\frac{E}{2k_b T}\right), \quad (1)$$

where γ_{α} is the coupling constant characterized by the coupling of the two-level states to phonons, ϑ_{α} is the acoustic velocity for the three phonon polarizations α , and ρ is the bulk density. Thus, the relaxation time has a certain distribution which is defined by the variation of E . The smallest relaxation time τ_{\min} occurs for $\Delta_0 = E$, i.e., for a symmetric potential well. The temperature variation of the dielectric constant in the two-level system model is associated with the resonant and the relaxation mechanism of scattering of electromagnetic waves by two-level systems and it is given by the following relations:¹

$$\left(\frac{\Delta\epsilon'}{\epsilon'}\right)_{\text{res}} = -B p_0^2 P \ln(T/T_0), \quad (2)$$

$$\left(\frac{\Delta\epsilon'}{\epsilon'}\right)_{\text{rel}} \sim \ln(T/T_0), \quad (3)$$

where T_0 is an arbitrary reference temperature, p_0 is the average microscopic dipole moment, P is the density of states, and $B = (-2\pi/27)p_0^2(\epsilon' + 2)^2/\epsilon'$.

Equation (2) is only valid when $h\omega \ll kT$, a condition that is always fulfilled in our case. Thus the resonant contribution to ϵ shows a logarithmic decrease with temperature.

At $\omega\tau_{\min} \gg 1$ the relaxation contribution is suppressed and the behavior of the dielectric constant is governed by the resonant process. But for $\omega\tau_{\min} \ll 1$ in the higher temperature region the contribution of the relaxation mechanism of polarization becomes predominant and $\Delta\epsilon'/\epsilon'$ increases obeying Eq. (3). In the temperature region where this condition is satisfied, the frequency dependence of ϵ' follows the logarithmic law⁵

$$\epsilon' \sim \ln(T/\omega). \quad (4)$$

Thus, the minimum on the temperature dependence of the dielectric constant corresponds to the balance of two contributions resulted from the resonant and relaxation mechanism of scattering in the two-level systems model.

The temperature dependences of $\Delta\epsilon'/\epsilon'(T_0)$ for the crystal studied at different frequencies (Fig. 1) show that a logarithmic

mic dependence of $\Delta\epsilon'/\epsilon'(T_0)$ on temperature occurs above T_{\min} . At low frequencies the relaxation contribution remains large until very low temperatures. Because of this, the minimum in $\Delta\epsilon'/\epsilon'(T_0)$, in particular at 300 Hz, is not observable. As the frequency increases the relaxation contribution is gradually suppressed. At frequencies above 50 kHz the dielectric dispersion, at least below 50 mK, is not observed anymore. This implies that the relaxation contribution at these frequencies is completely suppressed and consequently the dielectric response measured resulted only from the resonant scattering mechanism. Indeed, as can be seen in Fig. 1, (dashed line) the temperature dependence of $\Delta\epsilon'/\epsilon'(T_0)$ is in this case logarithmic, which is in good agreement with the theory outlined above.

Thus, the deviation of the temperature behavior of $\Delta\epsilon'/\epsilon'(T_0)$ from logarithmic dependence with an approach to T_{\min} from above results from the fact that τ_{\min} for certain TLS's becomes comparable to ω^{-1} and subsequently those TLS's do not take part in the relaxation process. However, τ_{\min} cannot be considered as a characteristic quantity of the system, since it has a certain distribution defined by the distribution of energies E . A rough estimate of some average τ_{\min} can be made, assuming that the conditions $\omega\tau_{\min}\approx 1$ and $E=\Delta_0=kT$ are satisfied at T_{\min} .¹ Using the temperature dependence of τ_{\min} we have estimated the coupling constant γ from Eq. (1) to be approximately equal to 4.7×10^{-19} J. This value is of the same order of magnitude as for disordered ferroelectrics² and amorphous solids.⁷ (The sound velocity ϑ_α was taken from Ref. 8.)

Figure 5(b) shows the results of a numerical integration of a more elaborate expression from which Eq. (3) was obtained.⁷ The presented curves have been calculated with $\tau_{\max}/\tau_{\min}=10^{10}$ and $E_{\max}/k=10$ K. For comparison, in Fig. 5(a) the temperature dependences of the relaxation contribution of $\Delta\epsilon'/\epsilon'(T_0)$ derived from our experimental data are shown. The experimental curves were obtained by subtraction of the resonant contribution to ϵ' from the observed ϵ' . The linear part of the $\epsilon'(\ln T)$ curve at a frequency of 100 kHz below T_{\min} and the extrapolation of this dependence to higher temperatures was taken as the resonant contribution. It is evident from these results that the temperature-frequency dependences of the relaxation contribution to ϵ' is adequately described [correct to the $(B\cdot P)^{-1}$ coefficient] by the TLS model.

Furthermore, in Fig. 6 we present a plot of $\Delta\epsilon'/\epsilon'$ versus $\ln(\omega)$. The observed linear dependence in a rather broad temperature region is consistent with Eq. (4). The deviation from logarithmic behavior occurring at temperatures close to T_{\min} (Fig. 6, curve 2) is due to $1/\omega$ becoming comparable to τ_{\min} .

Turning now to the imaginary part of the dielectric constant, a plateau in the temperature dependence of ϵ'' (Fig. 2) is expected in the two-level systems model. There it results from the fact that the number of two-level systems ignoring the condition $\Delta_0=kT$ and $\omega\tau=1$ (TLS for which τ_{\min} is smaller than ω^{-1}) remains constant. In reality, ϵ'' of the $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal in this temperature region shows a slight temperature dependence. This discrepancy between the TLS model and experiment with respect to the temperature behavior of ϵ'' is very likely caused by a not completely flat density of states distribution. The plateau in the $\epsilon''(T)$ dependence at high frequencies (≥ 10 kHz) and the slight

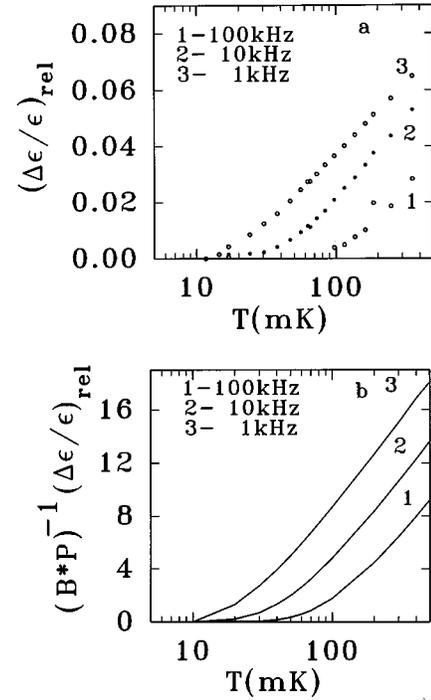


FIG. 5. (a) Relaxation contribution of $\Delta\epsilon'/\epsilon'$ at various frequencies derived from experimental data by subtraction of the resonant contribution from total ϵ' . (b). Results of numerical integration of Eq. (6) in Ref. 7 for a contribution of the relaxation process to the dielectric constant with $\tau_{\max}/\tau_{\min}=10^{10}$ and $E_{\max}/k=10$ K. The calculations have been normalized to $\Delta\epsilon'/\epsilon'=0$ at 10 mK.

temperature dependence of ϵ'' in this temperature region at lower frequencies indicate that the density distribution is not completely constant due to the states with relatively long relaxation times [$\tau > \omega^{-1} = (2\pi \times 10 \text{ kHz})^{-1}$]. It should be mentioned that the temperature of the minimum in the temperature dependence of $\Delta\epsilon'/\epsilon'(T_0)$ correlates with the temperature of the onset of abrupt decrease of ϵ'' . Less pictorially this correlation can be seen at low frequencies when the minimum in $\epsilon'(T)$ is not clearly defined and $\epsilon''(T)$ shows a maximum instead of a plateau.

Finally we comment on the observed strong nonlinear behavior of the dielectric constant in our measurements. Actually, a nonlinear dielectric effect is not foreseen in the TLS model. However, our set of experimental data allows us to draw some conclusions to the nature of the effect. The anal-

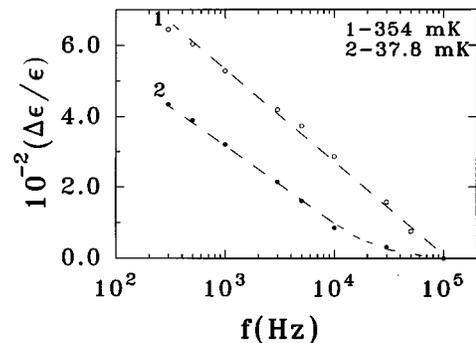


FIG. 6. $\Delta\epsilon'/\epsilon'$ versus $\ln(\omega)$ at different temperatures.

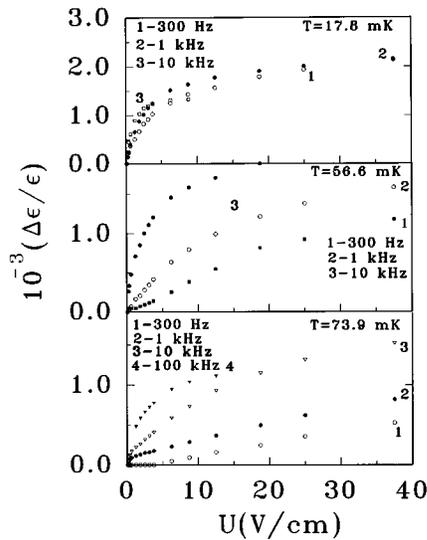


FIG. 7. Variation of dielectric constant vs measuring voltage for $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal.

ogy in the dependence of T_{\min} on frequency and excitation voltage evidences that a change in the measuring field may result in a renormalization of the minimum relaxation time. In particular, the larger the excitation voltage is, the higher the temperature will be, below which the quickest two-level systems exceeds the condition $\omega\tau=1$. A strong argument in support of this interpretation is the result of a study of the voltage dependence of $\Delta\epsilon'/\epsilon'$ at various frequencies and temperatures shown in Fig. 7. At temperatures below T_{\min} and at the lowest frequency used, the dielectric nonlinearity is the greatest and almost frequency independent. That means that the number of two-level systems being involved in the relaxation dynamics with an increase in the measuring voltage is frequency independent if $\tau_{\min}(U\rightarrow 0) > \omega^{-1}$. At higher temperatures, when τ_{\min} is getting comparable to or smaller than ω^{-1} the nonlinear effect becomes weaker. And finally, at temperatures where $\tau_{\min} \ll \omega^{-1}$ and therefore the number of two-level systems in the “frequency window” remains unaltered at various voltages, the dielectric nonlinear effect becomes negligible.

A decrease in the imaginary part of the dielectric constant below T_{\min} in the TLS model is attributed to the fact that a

certain number of TLS go beyond the condition $\omega\tau=1$. Therefore, it may be safely suggested that with an increase in the amplitude of the measuring field, those two-level systems in the relaxation dynamics which are practically eliminated from the process at lower field because of high barriers, will start to play a role again. In the TLS model it can be considered as a reduction of the effective height of the potential barriers. The minimum relaxation time for such two-level systems subjected to a weak external electric field. As a result the minimum in the $\epsilon'(T)$ and kink in the $\epsilon''(T)$ dependences shift to higher temperatures. Thus, the dielectric nonlinearity can be attributed to the renormalization of the height of the barriers.

A similar conclusion with respect to the nature of the observed nonlinear dielectric behavior was reached for another amorphous system at low temperature in Ref. 5. The nonlinear behavior of the polarizability of this system was attributed to a voltage dependence of the number of contributing two-level systems. The power dependence of polarizability of the form $\alpha \sim \epsilon \sim U^{1/2}$ was calculated in Ref. 5. We repeat here that curiously the voltage dependence of T_{\min} in $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ is adequately described by a power-law dependence as well. We have not yet formed a quantitative theory on this point.

V. CONCLUSION

We have shown that the crystalline material $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ possesses dielectric properties which resemble greatly those of amorphous dielectric solids. We have pointed out that the strong nonlinear behavior of the dielectric constant in these materials can possibly, at least qualitatively, be incorporated in the TLS model. A quantitative basis is still lacking.

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