Dielectric properties of triglycine selenate ferroelectric near the phase-transition temperature

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The results of nonlinear-susceptibility experimental investigations are given. It has been confirmed that the dielectric data obtained in the ferroelectric phase can be interpreted on the basis of Landau-tricritical-point theory with equation of state $E = A\tau P + CP^5$, where $T_t = (295.73 \pm 0.01)$ K, $A = (1.172 \pm 0.003) \times 10^{10}$ V^2 J^{-1} m, $C = (1.46 \pm 0.03) \times 10^{15}$ V^6 J^{-5} m^9. On the other hand, the paraelectric phase measurements results indicate neither critical nor tricritical Landau behavior. The experimental evidence of such a duality is presented. [S0163-1829(98)02301-7]

I. INTRODUCTION

The triglycine selenate (TGSe) crystals belong to the triglycine sulphate (TGS) family of uniaxial ferroelectrics. Although the continuity of the phase transition in TGS has been confirmed in many experiments (e.g., in Ref. 1), the results of analogous measurements in TGSe have been interpreted ambiguously. It is rather a surprising fact for such a popular ferroelectric crystal. In Ref. 2 the transition in TGSe compound has been treated as that of second order (continuous). On the other hand, in Ref. 3 Gesi suggests that the behavior of TGSe is ''almost critical between the second and the first order ones.'' In Ref. 4 the experimental result $\Delta = 0.33_0$ has been presented, where Δ is a parameter (not a gap exponent here) in the free energy density or in the anomalous part of the heat capacity and $\Delta = 1/3$ for the tricritical point. There are also many other experimental results which suggest the existence of a tricritical point in TGSe.[3,5,6] A small b parameter (in present paper denoted by B) documenting a transition near a tricritical point has been also obtained for the paraelectric phase in Ref. 7. In Ref. 8 evidence of a first order transition very close to the tricritical point has been given. On the other hand, the experimental results presented in Refs. 9 and 10 indicate a continuous transition in TGSe. The influence of deuteration, high-pressure application, and also γ radiation on the phase transition in TGSe has been investigated in Refs. 9 and 11–13.

On the basis of the papers cited above we can conclude that the majority of tricritical behavior evidence has been obtained in TGSe below the phase-transition temperature, e.g., the tricritical type of temperature dependence of the spontaneous polarization $P^4 \sim (T_c - T)/T_c$ in Ref. 8. We have measured the electric susceptibility of TGSe in both phases. The results for the paraelectric phase have been published in our earlier papers, e.g., in Ref. 14. It is interesting that the power exponents ratio Δ/γ obtained by us in the paraelectric region takes neither critical $\Delta/\gamma = 3/2$ nor tricritical $\Delta/\gamma = 5/4$ Landau values (here and below Δ denotes a gap exponent). On the basis of our experiments in the paraelectric region the intervals for the most frequently measured exponents values can be estimated as $1.38 \leqslant \Delta/\gamma \leqslant 1.45$ for TGSe. Not only do power exponents and the form of the susceptibility scaling function differ from those for Landau critical and tricritical ones. Also our experimental ratio $Q = 1.5$ defined in Ref. 15 lies between the critical $Q = 2$ and tricritical $Q = 4/3$ values. The conclusion arises from the above that the interpretation of the experimental results obtained in the paraelectric phase seem to be more complex than that in the ferroelectric one where evidence of a simple Landau tricritical behavior mentioned above can be easily found in the literature. The main purpose of our paper is to point out such an inconsistency.

II. EXPERIMENT

Below we present the results of susceptibility experimental investigations. The measurement method was the same for both phases. Therefore a comparison of results obtained in both regions was possible. The constant electric field E was applied in parallel to the ferroelectric axis and to the measuring field of a TESLA BM595 LCR meter. The amplitude of the measuring field was $E_0 = 47$ V/m and its frequency $f = 1$ kHz. The measurement circuit was the same as in Ref. 16. Two gold electrodes were evaporated on a rectangular crystal plate with an area of $S = 2.047 \times 10^{-5}$ m^2 and thickness $d = 1.07 \times 10^{-3}$ m. The results of measurements (e.g., thermal hysteresis) may depend on sample quality. From samples investigated by us we have chosen the crystal with the smallest—invisible in the scale as in the inset of Fig. 1—susceptibility thermal hysteresis loop (although with not the highest susceptibility value at phase transition temperature) in order to verify its tricritical nature.

In Fig. 1 the temperature dependences of the reciprocal zero field susceptibility χ_0^{-1} for the investigated TGSe sample are shown. It should be stressed that results of experimental dielectric investigations in the ferroelectric phase depend also on the method of measurement. For example, there are two well-known ways of susceptibility investigations near the phase-transition temperature: one during the cooling or heating of the sample with a sufficiently low rate of temperature changes and another one when the temperature is being altered step by step. The advantage of the second method is the fact that zero-field susceptibility can be obtained after a slow lowering of the electric field E from $E > 0$ to $E = 0$ at a constant temperature (circles in Fig. 1). In such a case the susceptibility values should correspond to stationary (stable) polarized states. Otherwise the susceptibility is measured for a not fully macroscopic polarized crystal

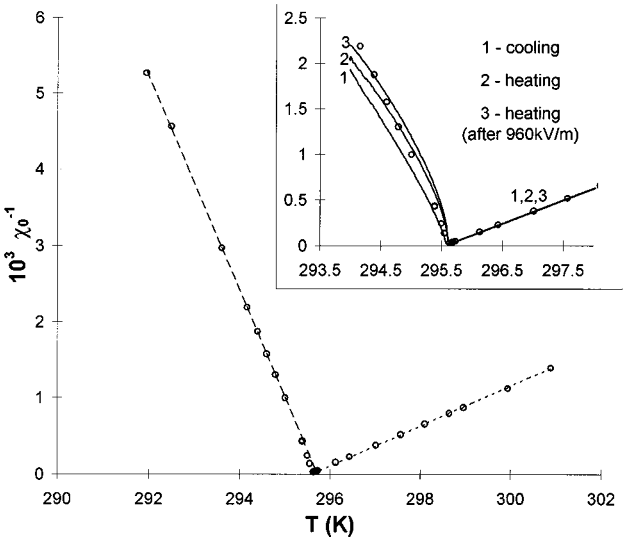

FIG. 1. Reciprocal zero-field susceptibility χ_0^{-1} vs temperature T dependences for TGSe. Circles represent experimental values of the susceptibility for stationary states. The dashed line represents the tricritical model (2) fit for the ferroelectric phase, the dotted line the linear fit for the paraelectric phase. Inset: solid lines represent cooling and heating with constant temperature changing rate 0.004 K/min: 1, cooling; 2, heating; 3, heating also in $E=0$ but after seasoning in $E=960$ kV/m at $T=298$ K for 24 h.

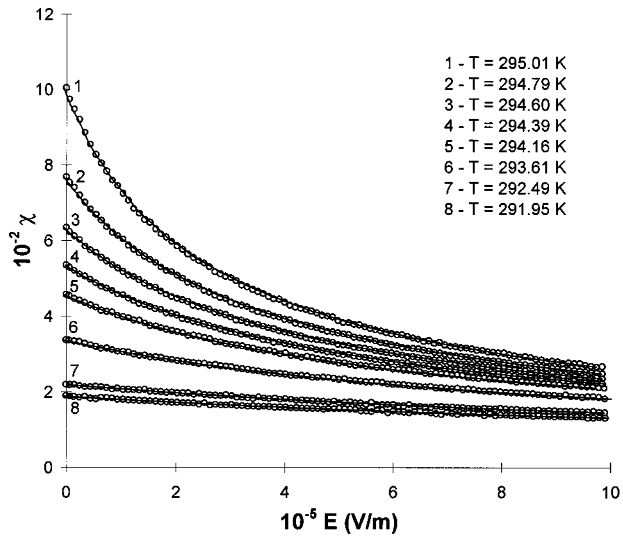

FIG. 2. Comparison of the theoretical (solid lines) and experimental (circles) electric susceptibility isotherms (i.e., electric susceptibility vs electric field dependences) in the ferroelectric phase for eight temperatures in the interval 291.9 K$<T<$295.1 K corresponding to eight circles in Fig. 1.

due to the domain structure. While cooling or heating the sample in $E=0$ (solid lines in the inset of Fig. 1) we usually measure the susceptibility values corresponding to nonequilibrium states. Then the results depend on the temperature changing rate and a comparison with theoretical models is rather very difficult. It should be stressed that in the case of data in the inset of Fig. 1 very low temperature change rate, i.e., 1 K/250 min has been used. Nevertheless, there occur differences which are shown in the inset of Fig. 1. In particular, in the ferroelectric phase, linear dependence has been observed by us only for experimental data obtained after a slow lowering of the electric field at a constant temperature. This fact should been taken into account in further investigations.

III. RESULTS AND DISCUSSION

A. Landau tricritical behavior in the ferroelectric phase

Although much evidence of Landau tricritical behavior has been given in the literature (mainly for the ferroelectric phase and $E=0$), no equation of state for TGSe in the ferroelectric phase—as far as known to us—has been published up to now. Below we use nonlinear susceptibility measurements results shown in Fig. 2 in order to check the possibility of the existence of such an equation. The solid lines in Fig. 2 represent a good quality numerical fit of a two-variable function

$$E=E(\tau,\chi)=(1-\varepsilon_0 A\tau\chi)^{1/4}\frac{4\varepsilon_0 A\tau\chi+1}{C^{1/4}(5\varepsilon_0\chi)^{5/4}}, \qquad (1)$$

obtained under assumption that the equation of state is of the Landau tricritical form:

$$E=A\tau P+CP^5, \qquad (2)$$

where ε_0 is a permittivity of vacuum, A,C are constants, P is the polarization, and $\tau=(T-T_t)/T_t$ is the reduced temperature. The equation-of-state parameters obtained numerically in such a way in the ordered region (solid lines in Fig. 2) are $A=(1.172\pm0.003)\times10^{10}$ V^2 J^{-1} m, $C=(1.46\pm0.03)\times10^{15}$ V^6 J^{-5} m^9, and $T_t=(295.73\pm0.01)$ K. Only the isotherms for 291.9 K$<T<$295.1 K shown in Fig. 2 have been used during the fitting procedure (see Fig. 2). For low temperatures errors in experiments were too large. On the other hand, the data obtained closer to the temperature of the susceptibility maximum have been excluded because of their deviations from the tricritical Landau scaling function χ/χ_0 versus $E\chi_0^{\Delta/\gamma}$ for $\Delta/\gamma=5/4$ in Fig. 3(a). The relations χ/χ_0 versus $E\chi_0^{\Delta/\gamma}$ have been obtained using experimental points from the dependences χ versus E measured in various temperatures, where $\Delta/\gamma=\delta/(\delta-1)$, Δ is a gap exponent, γ and δ determine the relations $\chi_0\sim\tau^{-\gamma}$ for $\tau\neq0$ and $E\sim P^{\delta}$ for $\tau=0$. For isotherms in the interval 291.9 K$<T<$295.1 K the scaling holds good for the Landau tricritical ratio $\Delta/\gamma=5/4$ in Fig. 3(b), although a better one appears for $\Delta/\gamma=1.30$ in Fig. 3(c). In our opinion, such a difference between $\Delta/\gamma=1.25$ and $\Delta/\gamma=1.30$ is too small to be conclusive evidence of deviations from Landau tricritical behavior. In Fig. 3(d) the dependences χ/χ_0 versus $E\chi_0^{\Delta/\gamma}$ for the Landau critical value $\Delta/\gamma=3/2$ have been also presented for comparison.

It should be stressed that the numerically obtained T_t temperature of the tricritical point is slightly higher (by about 0.1 K) than the Curie-Weiss temperature T_{CW} extrapolated from the paraelectric phase. It can be due to defects and inhomogeneities in the sample. The small electric field hysteresis in the paraelectric phase of TGS and TGSe ferroelectrics measured in our experiments may be evidence of such imperfections and local fields in real crystals (cf. Ref. 17 for γ-damaged TGS). On the other hand, one can state that the experimental fact $T_t-T_{CW}\approx0.1$ K may be evidence of a

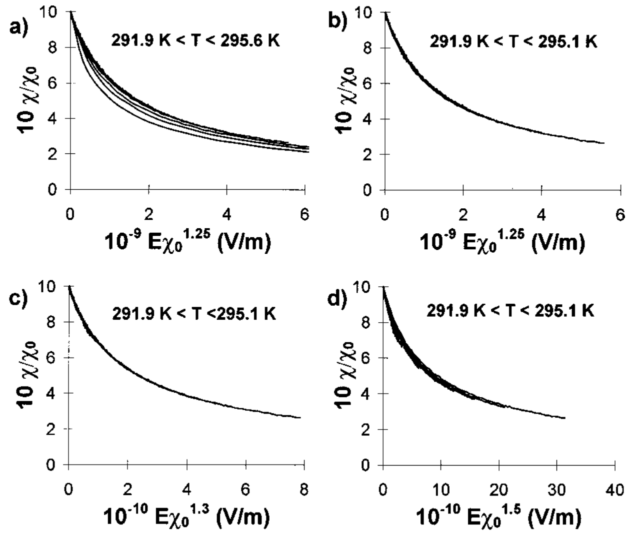

FIG. 3. The scaling relations χ/χ_0 vs $E\chi_0^{\Delta/\gamma}$ in the ferroelectric phase for a tricritical Landau value $\Delta/\gamma=5/4$ obtained for 11 (a) and 8 (b) isotherms (cf. Fig. 1); the best fit for $\Delta/\gamma=1.3$ (c) and deviation from scaling for the Landau critical value $\Delta/\gamma=3/2$ (d). In the cases of (c) and (d) eight isotherms have been used.

first-order transition (cf. Ref. 8). However, on the basis of a numerical analysis of the more general equation of state (cf. Ref. 18),

$$E=A\tau P+BP^3+CP^5, \tag{3}$$

we have concluded that at most a small (comparable with experimental error) positive B parameter might be permissible in the case of our experimental data. Contrary to Ref. 8, if not tricritical, we propose rather a continuous phase transition with B much smaller than for TGS (cf. Ref. 18). In Fig. 4 the isotherm for $T=294.6$ K as well as theoretical curves obtained from the model (3) for A, C fitted and presented above for various positive and negative B parameters are shown. From this figure we conclude that an experimental error for the B value can be estimated as -6.6×10^8 V^4 J^{-3} m$^5<B<6.6\times10^8$ V^4 J^{-3} m^5. In Fig. 5 the values of three terms $f_2=\frac{1}{2}A\tau P_s^2$, $f_4=\frac{1}{4}BP_s^4$, and $f_6=\frac{1}{6}CP_s^6$ in the free energy density expansion for $E=0$ and $T=294.6$ K$<T_t$ are shown in a log-log scale, where P_s is a spontaneous polarization calculated from Eq. (3). It is evident that for a B value of order of 10^9 V^4 J^{-3} m^5 the term with P_s^4 is less than 1% of those with P_s^2 and P_s^6.

In Fig. 6(a) the scaling form of the equation of state,

$$e=Ap+Cp^5, \tag{4}$$

is also verified, where $e=E/(-\tau)^{\Delta}$, $p=P/(-\tau)^{\beta}$, $P=P(E)=P_s+\varepsilon_0\int_0^E\chi(E')dE'$, and $\tau=(T-T_t)/T_t$. The temperature dependence of the spontaneous polarization [Fig. 6(b)],

$$P_s=\left(-\frac{A\tau}{C}\right)^{1/4}, \tag{5}$$

has been here calculated on the basis of Landau tricritical point model (2). The values of $P(E)$ have been obtained by numerical integration of isotherms in Fig. 2. The solid line has been drawn as a result of a calculation for the Landau tricritical equation of state with numerically fitted A and C given above. In Fig. 6(c) the temperature dependence of coercive field is also presented. These values concern investigations for which an electric field was slowly lowered step by step, contrary to standard polarization hysteresis loop measurements at frequency of 50 Hz.

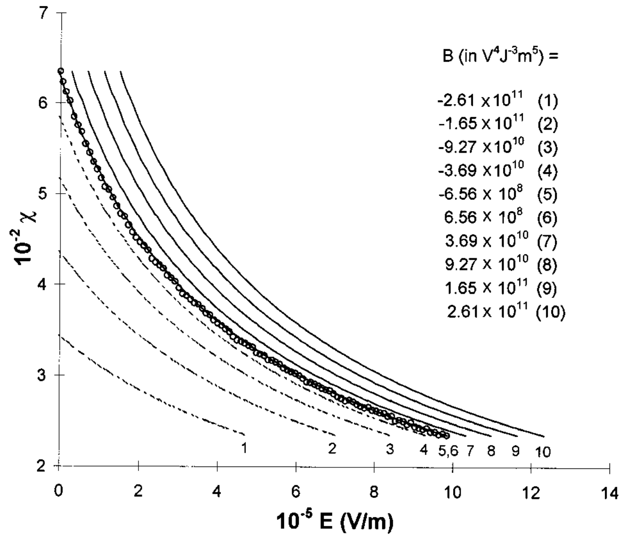

FIG. 4. The susceptibility isotherm for $T=294.6$ K (circles) and theoretical curves obtained from the model (3) for $A=1.172\times10^{10}$ V^2 J^{-1} m and $C=1.46\times10^{15}$ V^6 J^{-5} m^9 and for various positive (solid 1–5 curves) and negative (dotted 6–10 curves) B parameters.

B. Non-Landau behavior in the paraelectric phase

As has been shown above (Fig. 2) the dielectric properties of TGSe in the ferroelectric region for 291.9 K$<T$

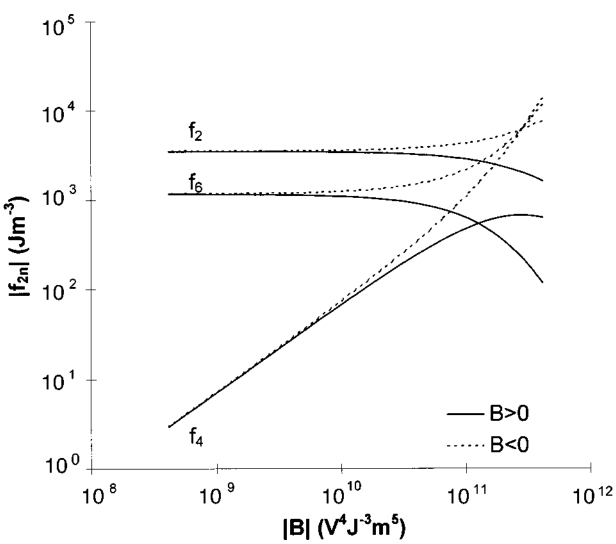

FIG. 5. The terms $f_2=\frac{1}{2}A\tau P_s^2$, $f_4=\frac{1}{4}BP_s^4$, and $f_6=\frac{1}{6}CP_s^6$ of the free energy density expansion vs the B parameter theoretical dependences [on the basis of Eq. (3)] in a log-log scale for $T=294.6$ K, $E=0$, $A=1.172\times10^{10}$ V^2 J^{-1} m, $C=1.46\times10^{15}$ V^6 J^{-5} m^9 for various positive (solid lines) and negative (dotted lines) B parameter values.

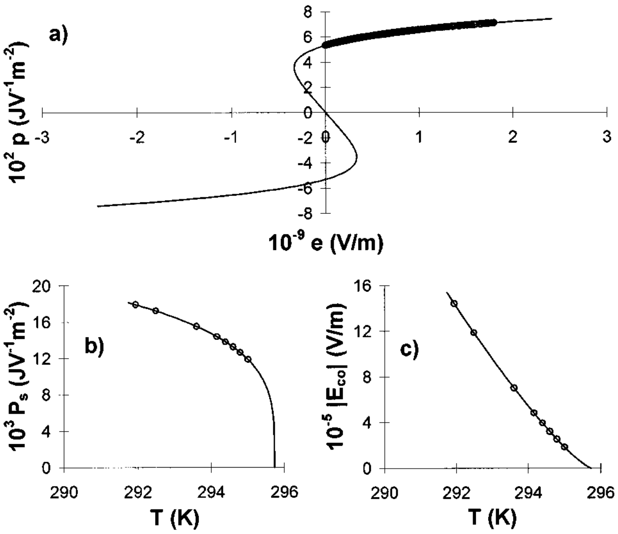

FIG. 6. Equation of state p vs e (circles correspond to experimental data) in the scaling form (a), spontaneous polarization P_s vs T (b), and coercive field E_{co} vs T (c) dependences. In (b) and (c) circles correspond to temperatures of the isotherms in Fig. 2. Solid lines represent the theoretical calculations on the basis of Eq. (2) for $A=1.172\times 10^{10}$ $V^2 J^{-1}$ m and $C=1.46\times 10^{15}$ $V^6 J^{-5} m^9$.

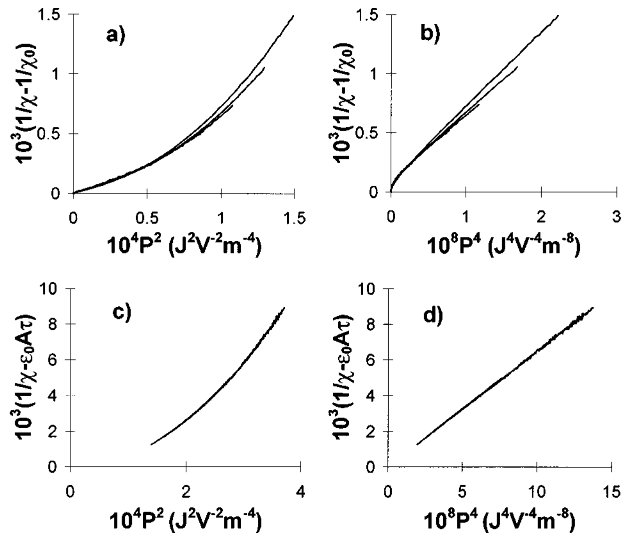

FIG. 7. The experimental relations $1/\chi-1/\chi_0$ vs P^2 (a), $1/\chi-1/\chi_0$ vs P^4 (b) in the paraelectric phase for 297 K$<T<$300 K and the experimental relations $1/\chi-\varepsilon_0 A\tau$ vs P^2 (c), $1/\chi-\varepsilon_0 A\tau$ vs P^4 (d) in the ferroelectric phase for eight temperatures in the interval 291.9 K$<T<$295.1 K (cf. Fig. 1). Only segments connecting the experimental points are shown.

$<$295.1 K and $0<E<935$ kV/m can be described with the help of a simple Landau tricritical model. Such a result confirms the data in the literature cited earlier in our present paper. On the other hand, in the paraelectric phase neither a critical nor tricritical Landau model can be accepted. Below we compare experimental results obtained in both phases by the same experimental method. According to the equation of state (3) the relation for susceptibility can be obtained:

$$1/\chi-\varepsilon_0 A\tau=3\varepsilon_0 BP^2+5\varepsilon_0 CP^4. \qquad (6)$$

Then for the critical point ($B>0$) the linear experimental dependence $1/\chi-\varepsilon_0 A\tau$ versus P^2 should be observed for small P. On the other hand, in the case of the tricritical point ($B=0$) the linearity of $1/\chi-\varepsilon_0 A\tau$ versus P^4 function is expected (for $P\rightarrow 0$, due to higher-order terms). In Figs. 7(a) and 7(b) the experimental relations $1/\chi-1/\chi_0$ versus P^2 and P^4, respectively, obtained in the paraelectric phase ($P_s=0$, $\varepsilon_0 A\tau=1/\chi_0$) for six temperatures in the interval 297 K$<T<$300 K (cf. Fig. 1) are shown. It is visible that neither critical nor tricritical Landau behavior is observed in the disordered region. Such a non-Landau behavior above the transition temperature has been investigated by us in Refs. 14 and 19–21. The model following from works of Domb and Hunter[22] and Patashinskii and Pokrovskii[23] with the equation of state,

$$E=a\tau^{\gamma}P+b\tau^{3\gamma-2\Delta}P^3+c\tau^{5\gamma-4\Delta}P^5, \qquad (7)$$

with constants a, b, c and not too small τ, has been assumed. On the other hand, the linearity of $1/\chi-\varepsilon_0 A\tau$ versus P^4 in Fig. 7(d) in the ferroelectric region for eight temperatures in interval 291.9 K$<T<$295.1 K (cf. Fig. 1) is evidence of Landau-tricritical-point behavior. For comparison, the relation $1/\chi-\varepsilon_0 A\tau$ versus P^2 in the ferroelectric phase is shown in Fig. 7(c). It is easy to notice that due to nonzero spontaneous polarization, values of P in the ordered phase [Figs. 7(c) and 7(d)] are higher than those in the paraelectric region [Figs. 7(a) and 7(b)], although the intervals for E ($0<E<935$ kV/m) remain the same in two phases. In Fig. 8 we combine the results from two sides of the transition point. At first sight the values of P seem to form one straight line. However, as follows from a comparison of Figs. 7(b) and 7(d), the data from the paraelectric phase cannot be treated as an extension of the tricritical linear dependence obtained below T_t.

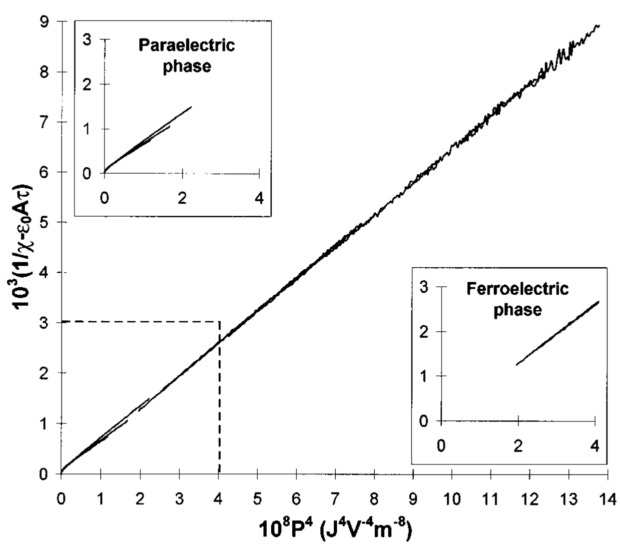

FIG. 8. The experimental relation $1/\chi-\varepsilon_0 A\tau$ vs P^4 combined from diagrams from Figs. 7(b) and 7(d) for both phases (only segments connecting the experimental points are shown). In the insets two corresponding dependences are presented separately.

C. Data obtained at the transition point

The investigations in the immediate neighborhood (closer than, e.g., 0.5 K) of the transition point may lead to incorrect conclusions because in experiments the susceptibility never tends to infinity and spontaneous polarization does not disappear exactly at the transition temperature.[24] Such a behavior is caused, among others, by surface layers effects,[25] inhomogeneities of the sample as well as local electric fields present sometimes even in the paraelectric phase. Apart from the finite susceptibility maximum, additional effects are observed, e.g., rather nonphysical intersections of various isotherms $P(E)$ obtained by integration of $\chi(E)$ dependences from temperatures very close to the transition point (cf. Ref. 26). On the other hand, the question arises as to what behavior can be expected for TGSe below the T_t temperature and closer than 0.5 K to it, i.e., for sufficiently small P_s. It should be stressed here that dielectric measurements were carried out very close to the transition point and, what is more, the tricritical isotherm was investigated. Unfortunately, the results of such experiments are not accurate enough and should be interpreted with criticism for the sake of the arguments mentioned above. Up to now, in our experiments the exponent δ has been only calculated from the scaling relation $\delta/(\delta-1)=\Delta/\gamma$, for Δ/γ determined as a scaling parameter fitted below or above the transition temperature. As can be concluded from the discussion above, different δ values follow from investigations carried out in both phases. Hence, it was also a very interesting question for us as to what was the experimental value of the critical exponent δ obtained directly from susceptibility measurements at the transition point. Although, in our opinion, any fitting procedures cannot be carried out with satisfactory accuracy for $T_t-T<0.5$ K, the values of the scaling parameters obtained further from the transition temperature (for isotherms as in Fig. 2) may be verified (compared with experiment) very close to T_t or even on the critical (tricritical) isotherm. For this reason we show the results of our electric susceptibility measurements at $T=T_t$. Since on the critical (tricritical) isotherm the relation $P^{\delta}\sim E$ occurs, we have also $\chi^{-\Delta/\gamma}\sim E$, where $\Delta/\gamma=\delta/(\delta-1)$. In Fig. 9 the dependence of $1/\chi$ versus E for $T=T_t=295.73$ K on a log-log scale has been presented. For small E we can see a nonlinearity caused, among others, by the fact that for $E\rightarrow 0$ and $\tau\rightarrow 0$ the experimental dependence differs from the theoretical one for which infinite susceptibility is expected for $\tau=0$ [cf. Fig. 3(a)]. Since $1/\chi\sim E^{(\delta-1)/\delta}$, the slope of the linear part of the dependence in Fig. 9 should be equal to $(\delta-1)/\delta$. For comparison, we have drawn the Landau tricritical dependence $1/\chi$ $(=5\varepsilon_0 C^{1/5}E^{4/5})$ versus E (solid line) for $C=1.46\times 10^{15}$ $V^6\,J^{-5}\,m^9$. It is shown that, within the limits of experimental errors, tricritical behavior also occurs for $T=T_t$ for the same C as that determined for the ferroelectric region (cf. $1/\delta=0.23\pm 0.03$ in Ref. 8). Such a result suggests that not only for temperatures as in Fig. 2 but also closer to the transition temperature in the ordered phase the tricritical Landau model may be valid.

D. Amplitude ratio Γ/Γ'

At the end of this paper we want to point out one more non-Landau result. According to the theory of phase transitions the zero-electric-field susceptibility depends on temperature as a power function:

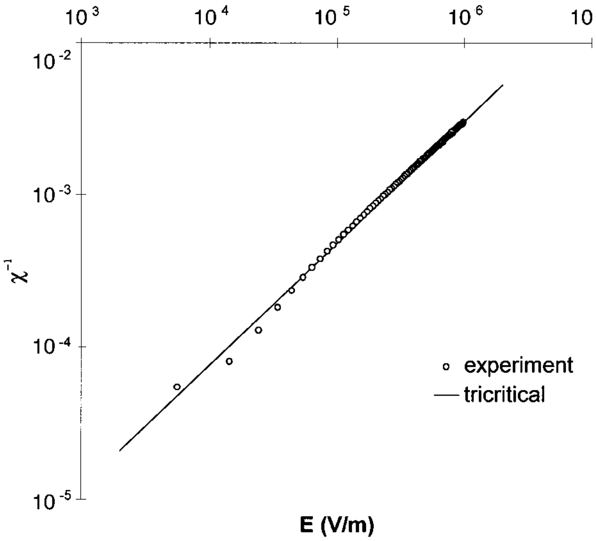

FIG. 9. Reciprocal susceptibility χ^{-1} vs electric field E obtained experimentally (circles) and calculated for the tricritical Landau model for $A=1.172\times 10^{10}$ $V^2\,J^{-1}\,m$ and $C=1.46\times 10^{15}$ $V^6\,J^{-5}\,m^9$ (solid line) for $T=T_t=295.73$ in a log-log scale.

$$\chi_0=\Gamma'\tau^{-\gamma'} \quad \text{for} \quad \tau<0 \quad \text{and} \quad \chi_0=\Gamma\tau^{-\gamma} \quad \text{for} \quad \tau>0. \tag{8}$$

For the Landau-critical-point model [$C=0$ in Eq. (3)] we have $\Gamma=1/A$, $\gamma=1$ and $\Gamma'=1/(2A)$, $\gamma'=1$. In the case of the Landau tricritical behavior [$B=0$ in Eq. (3)] we obtain $\Gamma=1/A$, $\gamma=1$ and $\Gamma'=1/(4A)$, $\gamma'=1$. It is an interesting fact that the amplitude ratio Γ/Γ' measured for TGSe is higher than 4, i.e., than Landau-tricritical-point value and, what is more, the relation $\Gamma/\Gamma'=(4/3)\times 4$ can be accepted (circles in Fig. 1). The 4/3 factor has been earlier obtained experimentally by us for TGS (Ref. 18) where $\Gamma/\Gamma'=(4/3)\times 2$ has been measured in the case of the critical point ($\Gamma/\Gamma'=2$ for the Landau critical point).

IV. CONCLUSIONS

Summing up the results of our work we can state that non-Landau behavior can be easily observed for TGSe crystals in the paraelectric region. In the ferroelectric phase no deviations from the Landau tricritical model have been detected. It is an interesting result because until now the same behavior as well as a similar width of the critical regions has been usually assumed for both phases. The application of two models for TGSe crystals, the non-Landau critical and Landau tricritical one, cannot be excluded. It means that two different equations of state (7) and (2) may exist for both sides of the transition temperature.

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