Scaling in ferroelectrics with critical points induced by an electric field

B. Westwański, B. Fugiel, A. Ogaza, and M. Pawlik
Institute of Physics, Silesian University, Uniwersytecka 4, 40-007 Katowice, Poland

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A method of constructing a susceptibility scaling function and extracting critical exponents from experimental susceptibility data for physical systems with discontinuous phase transitions and/or unsymmetrical critical points induced by a nonzero field is proposed. The coordinates of these points do not appear in the derived form of the scaling function. The transformation of the field E conjugate to the order parameter to a new field $h=E-E_0(T)$ where $E_0(T)$ represents the critical isochore and/or two-phase equilibrium line, is shown to reduce the scaling function for unsymmetrical systems to that for symmetrical ones. Transforming original isothermal experimental data for deuterated triglycine selenate with a deuterium content of $x\simeq 100\%$, the asymmetrical scaling function and values of the nonzero-field critical invariants δ, $a=\Delta/\gamma$, and Q (equal to 1.5, 3, and 1.2, respectively) were obtained.

I. INTRODUCTION

Most of the ferromagnets are well known to undergo continuous phase transitions, while most solid and liquid ferroelectrics[1–5] exhibit discontinuous phase transitions, similar to the liquid-gas transition. Systems with continuous phase transitions have symmetrical critical points: $P_c=0$, $E_c=0$, $T_c\neq 0$, whereas systems with discontinuous ones show an unsymmetrical critical point: $P_c\neq 0$, $E_c\neq 0$, $T_c\neq 0$, where E is the (electric) field conjugate to the order parameter P (polarization). The two-phase equilibrium line $E=E_0(T)$ and its continuation above T_c, the critical isopolare ($P=P_c$), are both dependent on temperature [$E=E_0(T)\neq 0$] for unsymmetrical systems in contrast with symmetrical ones, where $E=E_0(T)=0$. The above name, isopolare, is introduced by analogy to isochore for the liquid-gas transition. We refer the reader to the couple of schematic diagrams in other papers,[6,7,1] where more information about symmetrical and unsymmetrical points (Fig. 1 in Ref. 6, Fig. 1.1 in Ref. 7) as well as the phase diagram (Fig. 11.11 in Ref. 1) for ferroelectrics with discontinuous phase transitions are given.

In the prevailing investigations[6,7] of scaling the coordinates (P_c,E_c,T_c) of the critical point were required. These studies are much easier for a symmetrical critical point, where only one parameter, T_c, remains to be determined. We have proposed[8–16] another approach which makes the knowledge of the critical temperature T_c unnecessary. The only thing required is the certitude that measurements are carried out in a definite phase (mainly above T_c).

Our method is based on the relation[9]

$$\chi(T,E)/\chi(T,0)=f\{E[\chi(T,0)]^{a}\}\ ,\qquad a=\delta/(\delta-1)=\Delta/\gamma\ , \tag{1}$$

where $\chi(T,E)$ is the electric susceptibility at temperature T and electric field E, $\chi(T,0)$ is the zero-field susceptibility. Parameter δ is a critical exponent and represents the power-law relation, $E\sim P^{\delta}$, between external electric field E and the polarization P on the critical isotherm ($T=T_c$). The exponent γ describes the power-law form, $\chi(T,0)\sim(T/T_c-1)^{-\gamma}$, of the initial susceptibility $\chi(T,0)$. Δ is the gap exponent, which appears in Eq. (2c) in Ref. 9 [referred to hereafter as Eq. (2c.9)], $E\sim[T_m(E)/T_c-1]^{\Delta}$, where $T=T_m(E)$ is the temperature of maximum of susceptibility $\chi(T,E)$ for a given value of the electric field E. From Eq. (1) the existence of the nonzero-field critical invariant Q was deduced:[9]

$$Q=\chi[T_m(E),0]/\chi[T_m(E),E]\ . \tag{2}$$

Relation (1) defines the susceptibility scaling function and was derived[9] from the scaling hypothesis. It is true for all physical systems satisfying the scaling hypothesis and belonging to different universality classes, which have a symmetrical critical point.

In this paper we generalize relations (1) and (2) and all the results given in Ref. 9 for symmetrical systems to unsymmetrical ones. In Sec. II, considering a simple model of the discontinuous phase transition, we show the existence of a general structure of scaling function for an unsymmetrical critical point. This scaling law is independent of coordinates of this point. Finally, in Sec. III, the experimental isothermal data for the susceptibility of DTGSe ferroelectric are transformed into the scaling function due to the results of Sec. II.

II. DISCONTINUOUS PHASE TRANSITION AND THE SCALING

As the structure of the susceptibility scaling function (1) is common for all experimental and theoretical symmetrical systems obeying the scaling law, we can expect a similar result for the unsymmetrical critical point. To get the generalization of Eq. (1) it is sufficient to consider the simplest theoretical model which is subject to the scaling law and provides the symmetrical critical point as a limit of an unsymmetrical one. The best candidate for that purpose is the Landau-DeGennes model. It satisfies all the mentioned requirements and does not contain any corrections to the scaling.

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The free energy (F) for this model[17] is given by

$$F=\tfrac{1}{2}C_2\tau P^2+\tfrac{1}{3}C_3P^3+\tfrac{1}{4}C_4P^4 ,$$
$$\tau=T/T_0-1 , \quad C_2>0 , \quad C_3<0 , \quad C_4>0 , \tag{3}$$

where P is the polarization, C_n's are constant coefficients, and T_0 is the lowest temperature at which the metastable paraelectric phase may exist. The free energy (3) and, of course, the thermodynamic potential $G=F-EP$ are not invariant under the transformation: $P\to -P$, $E\to -E$. Therefore, a discontinuous phase transition at zero electric field and unsymmetrical critical point induced by nonzero electric field E do exist.

The equation of state, $E=\partial F/\partial P$, and the susceptibility, $\chi=\partial P/\partial E$, are needed to get the coordinates of the unsymmetrical critical point

$$P_c=-C_3/(3C_4) , \quad E_c=(-C_3)^3/(27C_4^2) ,$$
$$T_c=T_0[1+C_3^2/(3C_2C_4)] . \tag{4}$$

Inserting $P=P_c$ into the equation of state we obtain the linear relation ($E_0-E_c\sim\tau'$) of the two-phase equilibrium curve ($T<T_c$) and the critical isopolare ($T>T_c$)

$$E\equiv E_0(T)=E_c+P_cC_2'\tau' , \quad \tau'=T/T_c-1 ,$$
$$C_2'=C_2T_c/T_0=C_2+C_3^2/(3C_4) . \tag{5}$$

Substituting P_c for P in the corresponding equation for χ we get the susceptibility $\chi[T,E_0(T)]$ on the critical isopolare (5)

$$1/\chi[T,E_0(T)]=C_2'\tau'=1/\chi(T,0)-C_3^2/(3C_4) \tag{6}$$

and its relation to the zero-field susceptibility $\chi(T,0)=(C_2\tau)^{-1}$. Moreover, it is seen from Eq. (6) that the difference of reciprocal susceptibility at zero field ($E=0$) and on the critical isopolare $[E=E_0(T)]$ is constant for all temperatures $T>T_c$. Of course, $\chi[T,E_0(T)]$ is the largest value of $\chi(T,E)$, which can be gained for a constant temperature when E is changing. From this fact and Eqs. (5) and (6) the following behavior of isothermal susceptibility can be deduced. When E is increased from zero to $E_0(T)$ ($0\le P\le P_c$), $\chi(T,E)$ increases from $\chi(T,0)$ to the maximum value $\chi[T,E_0(T)]$. Subsequent increase of E in the region $E\ge E_0(T)$ ($P>P_c$) causes decreasing of $\chi(T,E)$. If T is increased, $E_0(T)$ increases too, but $\chi[T,E_0(T)]$ decreases. Besides, it should be mentioned that if $E>E_0(T)$ the points (E,T) are situated above the critical isopolare (5) on the phase diagram and correspond to the polarization $P>P_c$, while for $E<E_0(T)$ they lie below the isopolare and are connected with polarization $P<P_c$. This behavior is in a qualitative but not quantitative agreement with the experimental data, given in Fig. 1(a).

By elimination of polarization P, equations of state and susceptibility may be reduced to one relation

$$\{\chi[T,E_0(T)]/\chi(T,E)-1\}^{1/2}\{\chi[T,E_0(T)]/\chi(T,E)+2\}$$
$$=3(3C_4)^{1/2}|E-E_0(T)|\{\chi[T,E_0(T)]\}^a \tag{7}$$

between susceptibilities $\chi(T,E)$ and electric fields E on

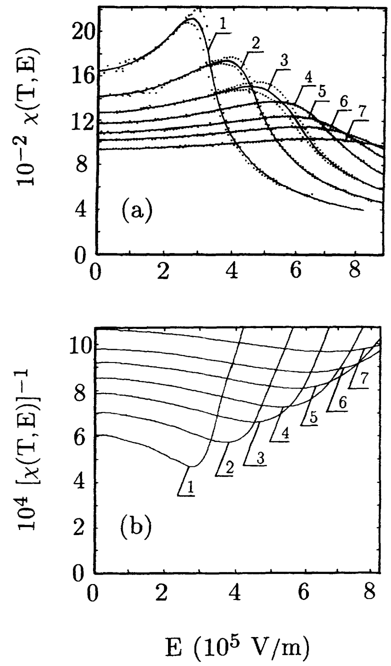

FIG. 1. Functions: (a) $\chi(T,E)$ and (b) $[\chi(T,E)]^{-1}$ vs electric field E for seven temperatures ($\tau=T/T_c-1<0.006$, $T_c=309.9$ K); T: 1-309.95, 2-310.30, 3-310.60, 4-310.87, 5-311.13, 6-311.37, 7-311.70 K. The experimental data for the DTGSe ferroelectric crystal with deuterium contents $x=100\%$ are displayed by dots: about 200 values of E for each temperature. The solid lines represent polynomial fits using the least-squares method.

$[E=E_0(T)]$ and beyond $[E\neq E_0(T)]$ the critical isopolare, where the values of critical exponents, $a=\delta/(\delta-1)=\Delta/\gamma=3/2$, $\delta=3$, $\gamma=1$, $\Delta=3/2$, are the same as for Landau theory of symmetrical critical point, i.e., for $C_3=0$ in Eq. (3). The presence of the absolute value of the difference $E-E_0(T)$ in Eq. (7) causes the ratio $Y=\chi(T,E)/\chi[T,E_0(T)]$ to be an even function (f) of the argument

$$X=[E-E_0(T)]\{\chi[T,E_0(T)]\}^a .$$

It is the sought susceptibility scaling function

$$\chi(T,E)/\chi[T,E_0(T)]=f([E-E_0(T)]\{\chi[T,E_0(T)]\}^a) \tag{8}$$

for an unsymmetrical critical point of Landau-DeGennes model. This function is thus symmetrical with respect to Y axis. This is a consequence of symmetry of each isothermal susceptibility $\chi(T,E)$ with respect to a direct line perpendicular to the electric-field axis at the point $E=E_0(T)$ and coming through the maximum value, $\chi[T,E_0(T)]$, of susceptibility. There are two branches of function (8). One branch ($X>0, 0\le Y<1$) originates from the points (E,T) of phase diagram situated above $[E>E_0(T)$ or $P>P_c]$ and the other ($X<0, 0\le Y<1$) is determined by the points lying below $[E<E_0(T)$ or $P<P_c]$ the critical isopolare $[E=E_0(T)$ or $P=P_c\neq 0]$.

The whole isopolare is reduced to one point $X=0$, $Y=1$ of the scaling function (8).

In the limit $C_3 \to 0$ due to Eqs. (4)–(6) the Landau-DeGennes model (3) goes over the Landau model of a symmetrical point with $P_c=0$, $E_c=0$, $T_c=T_0$, $E_0(T)=0$, $C_2'=C_2$ and Eq. (7) is reduced to Eq. (3a) in Ref. 10 [referred to hereafter as Eq. (3a.10)] for the susceptibility scaling function (1) for Landau symmetrical point.

The isothermal susceptibility $\chi(T,E)$ behaves quite differently in symmetrical and unsymmetrical cases. The electric field, $E=E_0(T)$, of maximum of $\chi(T,E)$ is equal to zero for all temperatures $T>T_c$ in the first case, whereas in the second one it increases with a rise of T due to the formula (5). Therefore, the isothermal maxima $\chi[T,E_0(T)]$ of $\chi(T,E)$ decrease, widen and shift to the higher fields E with increase of T for the unsymmetrical case and do not move otherwise; they are just standing at $E=0$ and decreasing with rise of T.

The dependence of $\chi(T,E)$ on T for constant values of E is completely different in continuous (1), (3a.10) and discontinuous (8), (7) cases. The relations (2) and (2c.9), which hold for the continuous transition, are no longer true for the discontinuous one. The susceptibility calculated from Eq. (7) as a function of T at constant values of E exhibits maxima which move with a change of E. However, the relation (2c.9) is not satisfied and the ratio (2) is not constant and depends on temperature.

After all the relations (2) and (2c.9) are also true for unsymmetrical case (7), however, not at constant field E but at a constant new field h:

$$h=E-E_0(T)\ , \tag{9}$$

which is a difference of the electric fields E and $E_0(T)$ (of maximum of isothermal susceptibility) on the critical isopolare (5). This statement is a consequence of the structure of Eqs. (3a.10) and (7) taking into account relations (4)–(6). The calculations or experimental measurements at constant values of new field h are equivalent to those run on a line

$$E=h+E_0(T)=h+E_c+P_cC_2'(T/T_c-1)\ ,$$

parallel to the critical isopolare (5), where h is an usual translation of (or a constant distance from) the critical isopolare. According as the translation is positive or negative the system is above $[E>E_0(T), P>P_c]$ or below $[E<E_0(T), P<P_c]$ the critical isopolare on the phase diagram E versus T. The calculations on the line $E=E_0(T)+h$ due to Eq. (7) give us (a) the invariant $Q=2$; (b) the formula (2c.9) in the form

$$[T_m(h)/T_c-1)]^{\Delta}=(27C_4/16C_2'^3)^{1/2}|h|\ ,$$

where $\Delta=\frac{3}{2}$, T_c and C_2' are given in Eqs. (4) and (5), respectively. The symmetrical limit of these results is reported in Eq. (13) of Ref. 10.

Let us notice that in the limit $C_3 \to 0$, Eq. (8) coincides with the general form (1) for the susceptibility scaling function for all different universality classes and symmetrical critical points. Therefore, we can set up a hypothesis that relation (8) also represents a general structure of the scaling function for all different universality classes with unsymmetrical critical points (or discontinuous phase transitions). Formula (8) is very important for the experimental investigations of scaling in unsymmetrical systems, because, in contrast with the traditional approach,[6,7] makes the knowledge of the coordinates of the critical point redundant. The only information needed is that for which phase the measurements are made.

To get Eq. (8) more similar to Eq. (1) let us make a change of variables due to Eq. (9). Then the scaling function (8) takes the form

$$\chi[T,E_0(T)+h]/\chi[T,E_0(T)]=f(h\{\chi[T,E_0(T)]\}^{a})\ ,$$
$$a=\delta/(\delta-1)=\Delta/\gamma\ . \tag{10}$$

In the limit $C_3 \to 0$, $E_0(T)=0$, $h=E$, and Eq. (10) assumes form (1) for the symmetrical case. Furthermore, the unsymmetrical form (10) can exactly be reduced to the symmetrical one (1);

$$\chi'(T,h)/\chi'(T,0)=f\{h[\chi'(T,0)]^{a}\}\ , \tag{11}$$

by the transformation $\chi'(T,h)=\chi[T,E_0(T)+h]$. It is now completely clear that all the statements derived in Ref. 9 for the symmetrical point are also true for the unsymmetrical critical case, when we go over from the field E to the new field h (9). Of course, the linearity of field $E_0(T)$ is a consequence of relation (5) for Landau-DeGennes model. In general, $E_0(T)$ may be nonlinear or even singular. It may happen when

$$E_0(T)=E_c+P_cC_2'(T/T_c-1)^{b}$$

with noninteger exponent b.

As we have already stated, the scaling function (8) for the Landau-DeGennes model is even. The same is true for function (10). However, for real physical systems, we can expect several departures: (a) exponents δ, Δ, and γ may be different from their classical counterparts 3, $\frac{3}{2}$, and 1, respectively; (b) scaling function (8) or (10) does not have to be even; (c) the relation $E=E_0(T)$ describing the critical isopolare does not have to be linear, it may be singular.

The ferroelectric system investigated experimentally in this paper (DTGSe, about 100% of deuterium, determined as in the paper by GeSi[3]) shows all the mentioned deviations and confirms our hypothesis that relation (8) or (10) as derived for Landau-DeGennes model is indeed a general structure of susceptibility scaling function for systems with discontinuous phase transitions. Of course it is impossible to carry out the measurements at constant distance h from the critical isopolare $E=E_0(T)$ because we do not know it before.

The experiments can be realized at constant temperatures T or at constant fields E. Both these methods are applicable for symmetrical systems (1). The isothermal experiments are preferred for unsymmetrical systems because maxima of the isothermal susceptibility determine the critical isopolare, which is not true for the experiments at constant electric field E, where the maxima have nothing in common with critical isopolare. We have shown elsewhere that the critical isopolare is the envelope of a family of curves $\chi(T,E)$ versus T for constant values of E and is situated to the right of the maxima of $\chi(T,E)$. The point of tangency of this envelope with a given curve is approaching its maximum when E ($>E_c$) goes to the critical value E_c. So it is clear that the determination of the critical isopolare from the measurements of $\chi(T,E)$ at constant electric fields E is more difficult than from analogous measurements at constant temperature T.

III. SUSCEPTIBILITY SCALING FUNCTION FOR DTGSe FERROELECTRIC

The experimental results have been achieved from the measurements of ferroelectric sample DTGSe with deuterium contents $x \simeq 1$. The gold evaporated electrodes completely covered the surfaces of area about 4 mm^2 of crystal plate with crystal thickness equal about 1.1 mm. The susceptibility was calculated from the formula $\chi = C/C_g - 1 \simeq C/C_g$ for $\chi \gg 1$, where C is the measured capacity of the capacitor with the sample and C_g is the geometric capacity. The measurements of electric capacity were carried out by means of TESLA BM 595 RLCG meter. The frequencies of measuring the electric field and its amplitude were 1 kHz and 44 V/m, respectively. The measuring electric field and the constant electric field E were applied along the ferroelectric axis. The measurements were made at the constant temperatures $T > T_c = 309.9$ K and the electric field was increased and decreased step by step.

In Fig. 1(a) the experimental data of susceptibility $\chi(T,E)$ versus the electric field E are given by seven isotherms of about 200 points each (~ 100 points for increasing and decreasing field E, respectively). The temperatures of these isotherms fulfill the inequalities: $T > T_c$, $\tau = T/T_c - 1 < 0.006$, where $T_c = 309.9$ K. The critical value of T_c was estimated due to different behavior of field hysteresis above and below T_c. It is worthwhile to mention that the electric-field hysteresis loops can be measured also above T_c. However, they are of different type from the classical ones observed below T_c. Our experiments showed that they were smaller when the change of electric field was slower. But slowing down the change did not always allow us to eliminate these loops completely. It seems that other factors must be involved, for example, history of the sample, its inhomogeneity, etc. The critical temperature was also estimated[18] from similar behavior of the temperature hysteresis of $\chi(T,E)$ for measurements at constant electric fields. Let us point out that Landau-DeGennes and other models do not provide hysteresis above T_c. The solid lines in Fig. 1(a) are the mean values of $\chi(T,E)$ for increasing and decreasing field E and are represented by polynomials generated using the least-squares method. Their reciprocals are shown in Fig. 1(b).

The data from Fig. 1(a) are plotted in Fig. 2 due to Eq. (1) for two values of δ ($=1.5$, 3). The first value is the best one estimated in Fig. 5 according to Eq. (10) whereas the second one arises from the Landau theory. We investigated that Eq. (1) gives no concentration of points around any curve for any value of δ. This indicates that Eq. (1) for the symmetrical critical point cannot be used

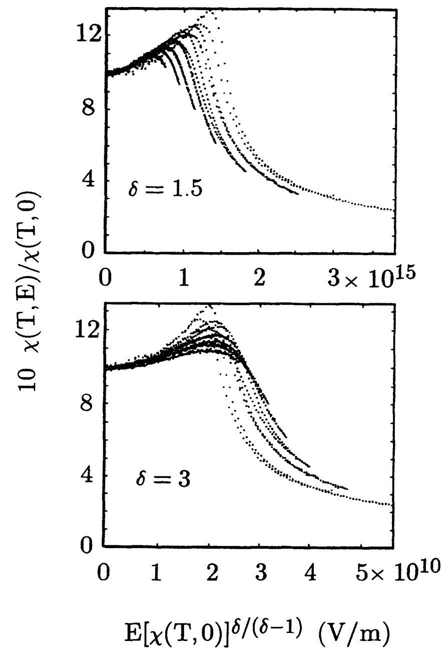

FIG. 2. Symmetrical critical point function (1) for the experimental data [the dots in Fig. 1(a)] and two values of δ; the lack of scaling for any value of δ.

with the data for the unsymmetrical critical point and there is a necessity to discover a new relation, which is already suggested in this paper, cf. Eq. (10).

The results in Fig. 1(b) are utilized in Fig. 3 to plot the reciprocals of susceptibility $\chi(T,E)$ versus temperature T at $E=0$ and on the critical isopolare $E_0(T)$. These curves are not parallel in contrast with the curves given by Eq. (6) for the Landau-DeGennes model. The inset in Fig. 3 shows the behavior of the critical isopolare, $E_0(T)$, i.e., the temperature dependence of electric field E at maximum of the isothermal susceptibility $\chi(T,E)$. This relation is nonlinear. It does not agree with Eq. (5) for the Landau-DeGennes model, where the critical isopolare is linear in temperature.

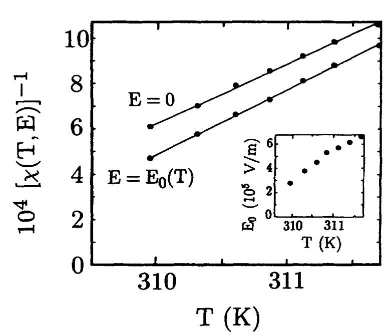

FIG. 3. Function $1/\chi(T,E)$ from Fig. 1(b) vs T for $E=0$ and on the critical isopolare $E=E_0(T)$ shown in the inset. $E=E_0(T)$ is the electric field of minimum in Fig. 1(b) for a given value of T.

To obtain the results discussed below we were obliged to run computer simulations on polynomial fits of experimental data from Fig. 1(a) to form new data, $\chi'(T_k,h)$, "measured" at a constant distance h from the critical isopolare $E_0(T)$.

In Fig. 4 we show the transformed susceptibility

$$\chi'(T_k,h)=\chi[T_k,E_0(T_k)+h]$$

versus T_k ($k=1,\ldots,7$). The solid lines join the dots of the same translation h of the electric field E. The highest curves in Figs. 4(a) and 4(b) are just the maximum values, $\chi[T,E_0(T)]=\chi'(T,0)$, of $\chi(T,E)$, i.e., the values on the critical isopolare, $P=P_c$, $E=E_0(T)+h$; $h=0$, shown in the inset of Fig. 3. The successive five curves, $\chi[T,E_0(T)+h_i]=\chi'(T,h_i)$, in Figs. 4(a) and 4(b) from the top to the bottom are obtained for the sequence of translations, $h_i=ih_\pm$ ($i=1,\ldots,5$); $E_i=E_0(T)+h_i$, made to the right [Fig. 4(a), $h_+=3\times10^4$ V/m, $P>P_c$] or to the left [Fig. 4(b), $h_-=-5.5\times10^4$ V/m, $P<P_c$] of the maxima for lines in Fig. 1(a).

Studying the maxima, $\chi'[T_m(h),h]$, of four curves, $\chi'(T,h_i)$, in Fig. 4(a) we have estimated the value 1.2 for the invariant

$$Q=\chi'[T_m(h),0]/\chi'[T_m(h),h]\ .$$

Δ may be found due to the relation $h\sim[T_m(h)/T_c-1]^\Delta$, however, more isotherms are required for that purpose. We do not see any maxima on Fig. 4(b). It does not mean that there are no maxima at all below critical isopolare $E_0(T)$ ($P<P_c,h_i<0$). They still may exist very close to T_c. But in this region it is very difficult to approximate the curves $\chi(T,E)$ with very sharp peaks by polynomials in a smooth way. For the Landau-DeGennes model the invariant $Q=2$ both above and below the critical isopolare (5), because the scaling function, $Y=f(X)$, arising from Eq. (7) is symmetrical with respect to the Y axis.

To test the dependence of the ratio $Y=\chi'(T_k,h)/\chi'(T_k,0)$ versus $X=h[\chi'(T_k,0))]^{\delta/(\delta-1)}$ [relations (10) and (11)] on different values of δ, we have previously formed some curves, similar to those in Fig. 4, for 29 values of the translation h: $h=0$, $h_i=ih_\pm$ ($i=1,\ldots,14$), $h_+=1.5\times10^4$ V/m, $h_-=-2\times10^4$ V/m. The positive translations, $h_i=ih_+$, produced the right branches, while the negative, $h_i=ih_-$, produced the left branches of the plots shown in Fig. 5. The best concentration of points is found for $\delta=1.5$ and their location shows the sought scaling function. In contrast with the Landau-DeGennes model, it is asymmetrical with respect to the Y axis. For $\delta=1.5$, Eq. (10) provides the relation $\Delta=3\gamma$.

It is worth mentioning that the same scaling function can be derived not only from the solid curves in Fig. 1(a), but also by separate consideration of the data for increasing and decreasing field E, respectively. It seems therefore that, within the limits of experimental error, the invariants δ, Q, Δ/γ, and the scaling function do not depend on the field hysteresis above T_c. This is the reason why in Fig. 1(a) we have included the experimental data with field hysteresis.

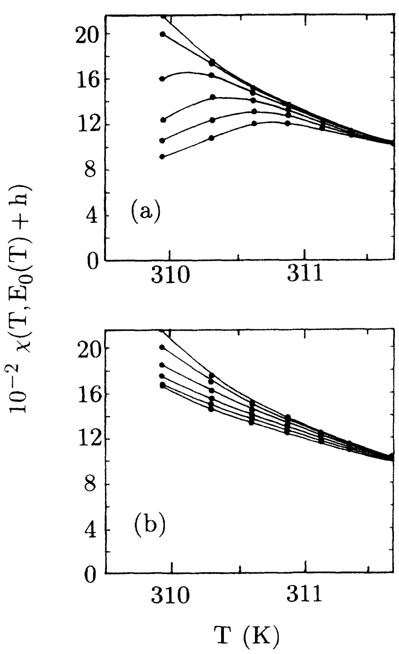

FIG. 4. Function $\chi'(T,h)=\chi[T,E_0(T)+h]$ vs T at constant distances (h) from the critical isopolare $E_0(T)$ (see inset in Fig. 3) as the result of computer simulations due to Eqs. (10) and (11) on the solid curves in Fig. 1(a) for 11 distances h. In both (a) and (b), the value $h=0$ corresponds to the highest curves; translations $h_i=ih_\pm$ ($i=1,\ldots,5$) numerate the next curves from the top to the bottom; (a) $h_+=3\times10^4$ V/m; (b) $h_-=-5.5\times10^4$ V/m.

IV. CONCLUSIONS

It is worth it to finally give the list of the most important results. The DTGSe crystal with about 100% of deuterium exhibits an unusual critical behavior. The phase transition is discontinuous at zero external electric field. There is the critical point ($P_c\neq0,E_c\neq0,T_c\neq0$) induced by an external electric field. For this crystal it is impossible to construct the susceptibility scaling function due to Eq. (1) for a symmetrical critical point for any value of δ. The general structure of the unsymmetrical scaling function (10) is crucial for investigation of the scaling in the real unsymmetrical systems. The reciprocals, $1/\chi(T,0)$ and $1/\chi[T,E_0(T)]$, of zero-field susceptibility and the isothermal susceptibility $\chi(T,E)$ at its maximum are not parallel when plotted versus T. The critical invariant Q characterizing the maxima of susceptibility $\chi'(T,h)=\chi[T,E_0(T)+h]$ at constant distances h from the critical isopolare $E_0(T)$ is estimated to be $Q=1.2$. The critical exponent representing the relation $E-E_c\sim(P-P_c)^\delta$ at T_c is, to our surprise, $\delta=1.5$. The gap exponent $\Delta=3\gamma$. Making use of the scaling relations[6,7] between the critical exponents, $\alpha=\alpha'$, $\gamma=\gamma'$, $\beta\delta=\Delta=\gamma+\beta$, $\alpha'+2\beta+\gamma'=2$, one can speculate that critical exponent $\beta=2\gamma$. This relation ($\beta>\gamma$) has never accompanied any experimental or theoretical system so far. Having independently measured Δ or γ, we are able to determine values of all exponents. These results may change, when taking into account surface layers,[1] which

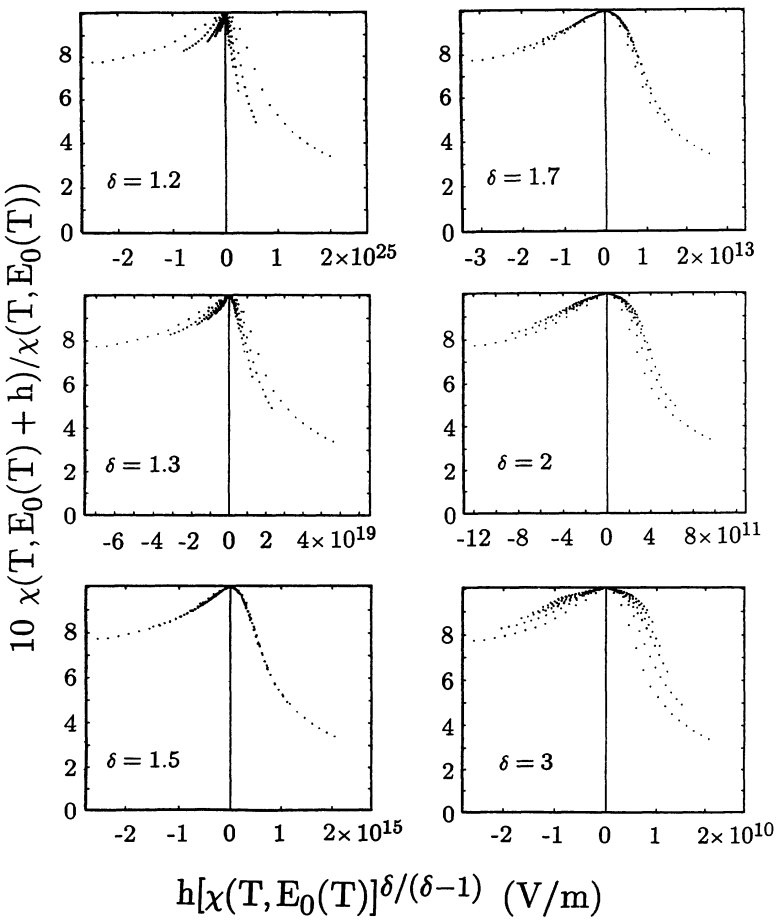

FIG. 5. The test of scaling for transformed experimental data "measured" parallel to the critical isopolare (see the next to last paragraph of Sec. III). The best concentration of points for $\delta=1.5$ qualifies the corresponding curve as the scaling function.

are, unfortunately, difficult to implement, but seem now to be the only way out. The critical isopolare is nonlinear, probably singular,[18] $E_0(T)-E_c \sim (T/T_c-1)^b$ $(0<b<1)$, contrary to the result (5) for the Landau-DeGennes model, where the critical exponent b is equal to the exponent $\gamma=1$ of zero-field $(h=0)$ transformed susceptibility $\chi'(T,0)=1/[C_2'(T/T_c-1)]$. However, the problem of eventual singularity of the critical isopolare $E_0(T)$ should be investigated in more detail.

Finally, let us emphasize that the scaling law (10) is also valid for the liquid-gas system by replacing P by density and E by pressure.

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