PHYSICAL REVIEW B VOLUME 37, NUMBER 10 1 APRIL 1988

Dilatometric study of critical behavior in $CsPbCl_3$: Bicritical point at 41 °C

T. H. Johansen, J. Feder, and T. Jøssang
Department of Physics, University of Oslo, P.O. Box 1048 Blindern, 0316 Oslo 3, Norway

(Received 22 June 1987)

High-quality crystals of $CsPbCl_3$ were grown by the Czochralski technique with automatic diameter control by the weighing method. A sample was prepared for high-resolution dilatometric measurements of the thermal expansion along a cubic [100] direction. The linear strain, $\Delta L/L$, was measured in the temperature interval 28–55 °C, and anomalies associated with three phase transitions were observed. Emphasis was put on the critical behavior in the close vicinity of the second-order II-III phase transition at 41 °C. The temperature variation of the anomalous thermal expansivity follows a power law $\sim |t|^{-0.18}$ in the ordered phase. This is in excellent agreement with the expected behavior of the spontaneous strain in cubic perovskites at an $n=2$ bicritical point. Also the [100]-stress–temperature phase diagram was measured for the II-III transition. The phase boundary line for compressional stress σ is well described by $T_c(\sigma)-T_c(0)\propto\sigma^{1/\phi}$ with $\phi=1.175$, consistent with the unstressed transition being an $n=2$ bicritical point. This is the first observation of such bicriticality in a structural system.

I. INTRODUCTION

Cesium lead chloride, $CsPbCl_3$, crystallizes from the melt in the perovskite structure. On cooling, it undergoes a succession of structural phase transitions at 46, 41, and 36 °C. From the high-temperature cubic phase (phase I, O_h^1 or $Pm3m$) it transforms to phase II, tetragonal (D_{4h}^5 or $P4/mbm$), then to phase III, orthorhombic (D_{2h}^{17} or $Cmcm$), and finally to phase IV, orthorhombic (D_{2h}^{16} or $Panm$).[1–3] It has been shown that these phase transitions can be interpreted in terms of condensations of soft phonon modes at the Brillouin-zone boundary of the cubic perovskite lattice.[2] The structural changes are associated with a rotation scheme of the $PbCl_6$ octahedra around the pseudocubic crystal axes; at 46 °C an M_3 mode at the M point condenses, while R_{25} modes at the R point are responsible for the transitions at 41 and 36 °C. The II-III transition is believed to be of second order, whereas the other two are reported to be first-order transitions.

For some time the crystals of the cubic perovskite family that undergo displacive phase transitions have served as important model systems for the studies of critical phenomena. Renormalization-group[4] analyses have predicted that a variety of multicritical points can occur if symmetry-breaking terms are introduced in the Hamiltonian.[5,6] In structural transitions the application of uniaxial stress can break the symmetry, as was first investigated by Aharony and Bruce.[7] Their analysis of the antiferrodistortive transition in strontium titanate, $SrTiO_3$, brought up the idea that the stress-free transition at 105 K should be a bicritical point, i.e., a point in the stress-temperature phase diagram where two critical lines meet. The presence of a uniaxial stress along a [100] axis not only shifts the transition temperature in a nontrivial way, but also changes the critical behavior. In $SrTiO_3$ a tension leads to an Ising-like ($n=1$) behavior, a compression to an XY-like ($n=2$) behavior, and for the stress-free state a Heisenberg-like ($n=3$) behavior is observed.[8,9]

In the case of $CsPbCl_3$ a large number of investigations have concentrated on the first-order transition at 46 °C,[10–12] while less attention as been paid to the critical variation of thermodynamic quantities near the continuous II-III transition. Moreover, the effects of symmetry-breaking stress on this substance have not been explored previously.

In this work we have applied dilatometry to measure the thermal expansion of $CsPbCl_3$ in a [100] direction. A high resolution has enabled us to extract more information concerning the critical behavior than before. This includes a determination of a critical exponent for the diverging thermal expansivity. In addition, the dilatometer permits application of a uniaxial stress, thus we were able to locate the II-III phase boundary in the [100]-stress–temperature plane. We describe here these measurements, and show that the results are fully consistent with a picture where the unstressed II-III transition is a bicritical point characterized by $n=2$. To our knowledge, this is the first observation of such bicriticality in a structural system.

Several crystals of $CsPbCl_3$ were grown by the Czochralski technique. Since this method has not been applied to $CsPbCl_3$ earlier, we give first a short description of our crystal production procedures.

II. CRYSTAL GROWTH AND SAMPLE PREPARATION

The preparation of $CsPbCl_3$ started by mixing powdered CsCl and $PbCl_2$ (Merck 99.5% purity) in stoichiometric proportions. The mixture was mechanically homogenized by thorough shaking, and then sealed with vacuum in a fused quartz ampoule for horizontal zone refining. A molten zone, maintained by radiofrequency (rf) heating, traversed the ampoule 10–15 times until the pure side of the ingot showed the characteristic pale yellow color of $CsPbCl_3$. The best half of the refined material was selected for crystal pulling in a Czochralski apparatus, shown in Fig. 1.

The charge material, which melts at 620 °C, was contained in a fused quartz crucible of diameter 30 mm. The heating element in the furnace consists of a resistance wire (Kanthal 3.65 Ω/m) wound on the inside of a fused quartz cylinder, and attached to the glass by ceramic glue (Cerastil *C*10). The power supply for the element has a thyristor-based digital control unit, which was operated remotely by a MINC 23 laboratory computer. The thermal growth conditions were found to be best represented by the temperature measured immediately below the crucible, in a cavity well protected from convective fluctuations. The temperature can be stabilized at $T = T^{\mathrm{ref}} \pm 0.05$ K by applying proportional and integrating (PI) corrective feedback action using the computer.

Seed crystals of $CsPbCl_3$ were not available, thence solidification was initiated on the air-cooled quartz tip of a pulling rod. During growth the rod was lifted at a constant speed of $V = 3.8$ mm/h and rotated by 20 rpm. To monitor and control the crystal diameter we adopted the melt weighing method.[13,14] For this purpose the crucible was mounted on a pedestal connected directly to a 0.01-g resolution Mettler PT1200 digital balance. The strategy for the computerized diameter regulation is outlined in the Appendix.

The crystals produced with automatic diameter control

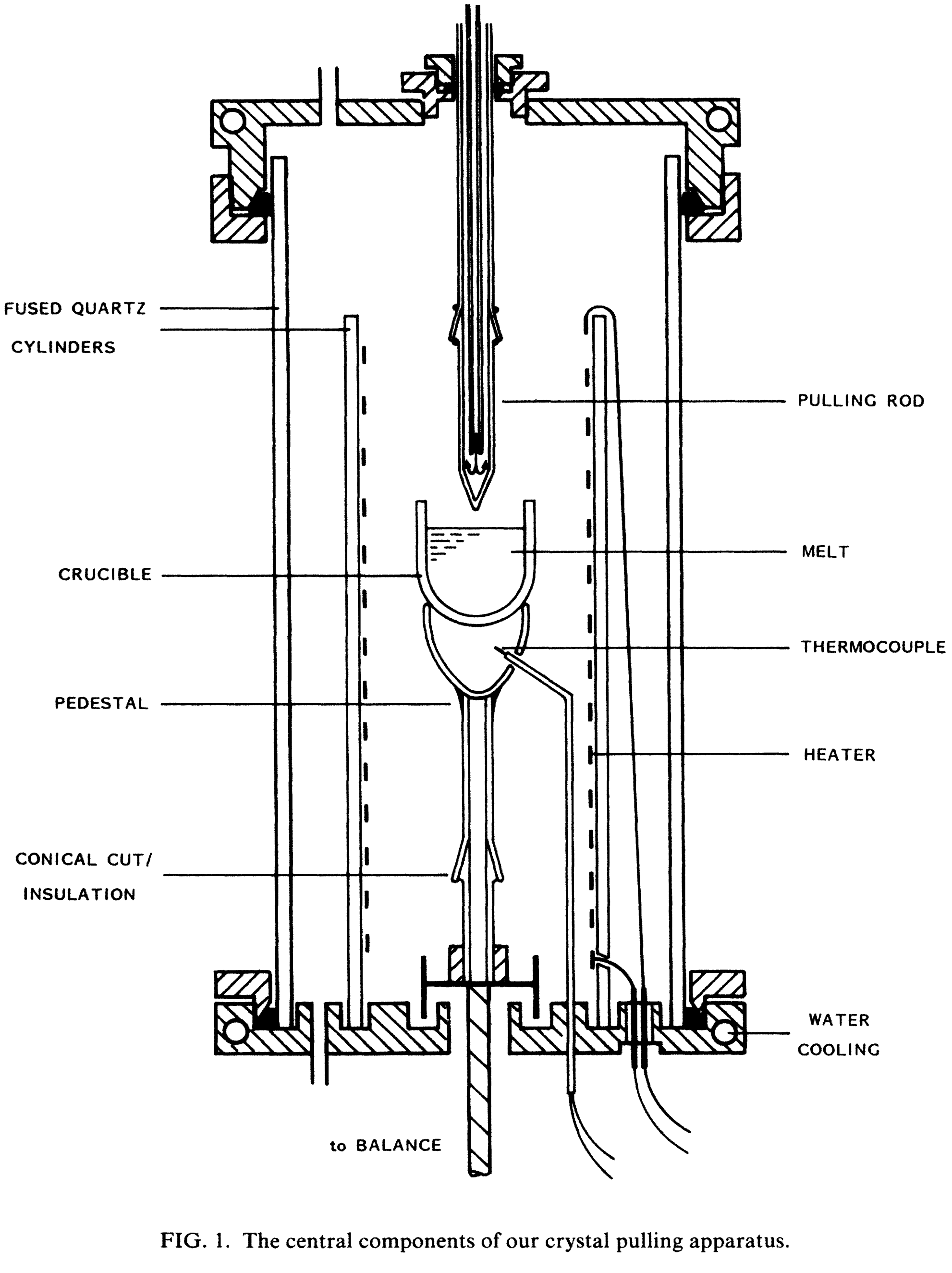

FIG. 1. The central components of our crystal pulling apparatus.

were grown with a thin 1 cm long neck in the initial stage, thus permitting grain boundaries to stop propagating. The bulk part is a single crystal pulled as a right cylinder of diameter near 10 mm. The surface is smooth, having a diameter ripple of only 1–3 %. The $CsPbCl_3$ crystals grown by this method show good structural quality—mosaic spreads of 0.3–0.45 deg were determined by neutron diffraction experiments.

To prepare an oriented sample it was convenient to use the fact that $CsPbCl_3$ cleaves along planes parallel to cubic (100). Since the mother crystal was pulled with an arbitrary growth direction, x-ray backscattering was used to find the cleavage planes. The sample was shaped as a right prism with plane parallel end surfaces normal to a cubic [100] axis. At room temperature the prism height was 3.310 mm. The cross-sectional area of the endfaces was 13.5 mm^2.

Prior to dilatometric measurements the prepared sample was annealed at 450 °C for 30 h.

III. THERMAL EXPANSION

The linear thermal expansion was measured using a parallel-plate capacitance dilatometer and Peltier oven system. The apparatus, which has a resolution of 0.1 Å in dilatation and 0.1 mK in temperature, has been described in detail earlier.[15,16] The dilatometer responded to variations in the distance L between the endfaces of the specimen. A small mounting load of 100 g was acting on the crystal as a uniaxial compressional stress of 74 kPa parallel to L. The measurements were made as the temperature was slowly decreasing at a rate of 0.6 K/h. Figure 2 displays the observed temperature dependence of the linear strain $\Delta L/L$ over the interval 28–55 °C.

The successive phase transitions are seen as three distinct anomalies in the dilatation. At 46 °C a sudden change in ΔL occurs, corresponding to a strain discontinuity of $\delta = 1.75\times 10^{-3}$. If the I-II transition caused the tetragonal c axis to orient perpendicular to the measurement direction throughout the sample, the jump δ will be equal to the relative change in the lattice constant a. X-ray investigations have given $\Delta a/a$ close to 1.8×10^{-3}.[1] The good agreement between δ and $\Delta a/a$ shows that in our sample the c axis preferred an orientation perpendicular to L. This choice was induced by the compressional mounting stress. On removing the stress we could observe nonreproducible behaviors near the I-II transition. The value for δ is in good agreement with the previous dilatometric results of Hirotsu,[17] and Hirotsu and Suzuki,[18] who also applied external stress for sample detwinning.

The anomaly associated with the II-III transition near 41 °C appears in Fig. 2 as an abrupt change in the slope of the thermal expansion curve. In that region no discontinuity in strain was observed, which is consistent with the second-order nature of this transition. We find that the spontaneous strain in phase III is of the *opposite* sign when compared to the results shown in Ref. 18. There, a dilation of the type indicated in the plot by a broken line was reported. However, in the earlier work[17] Hirotsu presented a thermal expansion curve very similar to ours. The two behaviors in the spontaneous strain along cubic [100] can occur if the direction of L coincides with each of two orthogonal directions normal to the c axis. Indeed, such results were obtained for $CsSrCl_3$,[19] which is isomorphous to $CsPbCl_3$, and undergoes the same sequence of phase transitions. Since the mounting of our sample always tends to align the elongated axis perpendicular to L, we assume that $\Delta L/L$ represents the dilatation of the shortest lattice constant of the orthorhombic phase.

We proceed to analyze the II-III transition in more detail. The thermal expansion coefficient

$$\beta_1 = \frac{1}{L}\frac{d\Delta L}{dT}\ , \tag{1}$$

in the neighborhood of $T_c^{\text{II-III}}$ is displayed in Fig. 3. β_1 was obtained by sliding two-point derivation of the

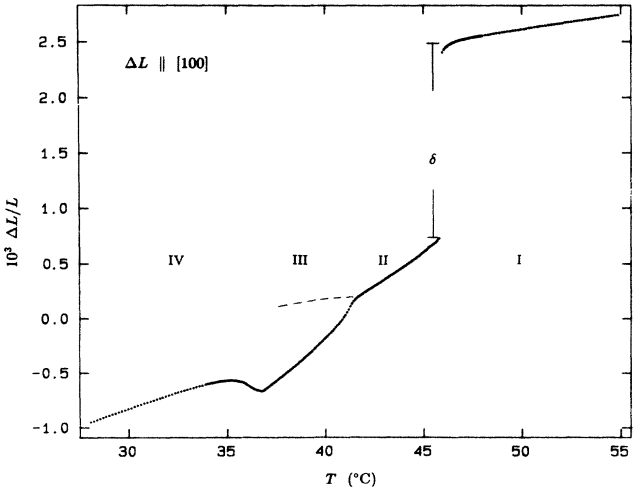

FIG. 2. Linear thermal expansion of $CsPbCl_3$ measured along a cubic [100] direction. Only a small fraction of the data points is shown.

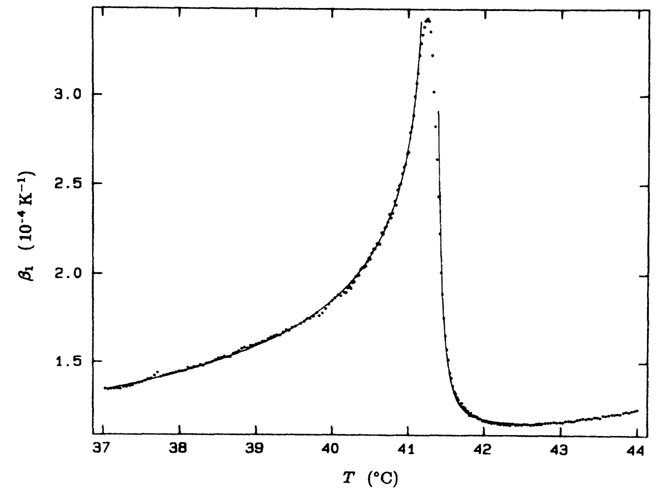

FIG. 3. Coefficient of thermal expansion along [100] as function of temperature. The fully drawn curves represent fitted theoretical functions (see text).

thermal expansion data. The curve thus computed is free from statistical correlation between the points. The expansivity goes through a sharp maximum at 41.25 °C. The peak has a strong nonsymmetric profile, rising very steeply on the high-temperature side.

Much of the interest in critical phenomena has been directed towards the exponents of the power-law equations which describe the asymptotic variation of thermodynamic quantities near the transition temperatures. To search for power-law behavior near $T_c^{\text{II-III}}$, hereafter abbreviated T_c, attempts were made to fit β_1 by a function of the form

$$\beta^{\text{fit}} = A \frac{|t|^{-a} - 1}{a} + B \ . \tag{2}$$

Here $t = (T - T_c)/T_c$ with $T_c = 41.25\,^\circ\text{C}$, and a, A, and B are constants to be determined from the fits. This was done by plotting β_1 versus $(|t|^{-a} - 1)/a$ for different values of a, and looking for the best approximation to a straight line. Because of the nonlinear temperature axis a number of points were removed far from the transition in order to give a uniform distribution of data. As shown in Fig. 4, a highly linear relationship can be produced for the data below T_c. Systematic deviation due to rounding sets in first at 0.08 K from T_c, i.e., for $|t| = 2.5 \times 10^{-4}$. The inset gives the rms deviation from a least-squares fitted line as function of the exponent a, which takes values from 0 to 0.40. The best fit occurs when $a = 0.18$. The corresponding line is characterized by $A = 1.728$ and $B = 2.164$ with units 10^{-5} K^{-1}, and is drawn as full curves in both Figs. 3 and 4.

Above T_c it was not possible to produce a linear relationship of the form Eq. (2) without increasing the rms deviation by about 2 orders of magnitude.

IV. DISCUSSION

The critical variation of the thermal expansivity is most frequently discussed in terms of the Pippard relations.[20,21] From these one expects that for ordinary critical points (on a λ line) the singularities in expansivity and in specific heat, as well as the components of the elastic compliance, are described by the same critical exponents. However, the renormalization-group calculations for $n = 1-3$ do not give specific-heat exponents α close to our fitted value of 0.18 for the expansivity exponent. Moreover, the fact that the power law holds only below T_c strongly suggests that the II-III transition in CsPbCl_3 is a special critical point.

In analogy to the transition in SrTiO_3 we expect that

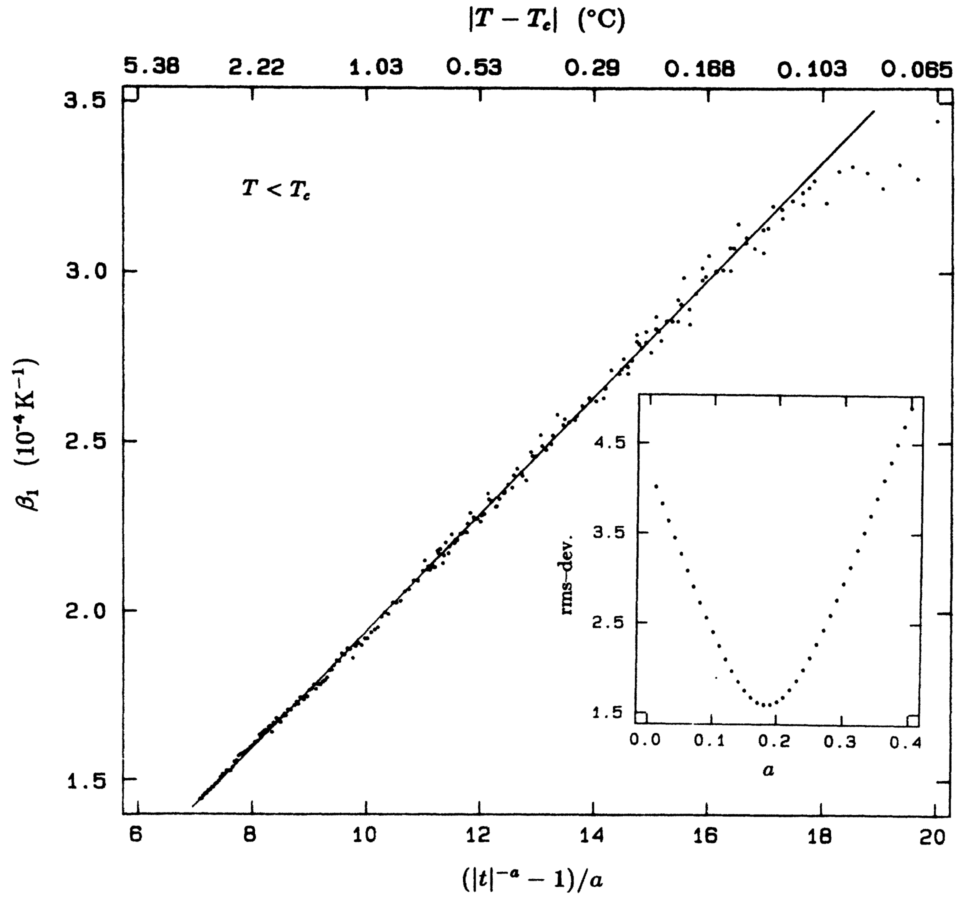

FIG. 4. Expansivity as function of $(|t|^{-a} - 1)/a$ with $a = 0.18$ for a temperature interval of 4 K below the II-III transition. Inset: Root-mean-square deviation in units of 10^{-6} K^{-1} between data and fitted straight line for various a.

[100]-stressed $CsPbCl_3$ near the II-III transition has a topology similar to a uniaxial antiferromagnet near the spin-flop bicritical point. Whereas $SrTiO_3$ is cubic in the high-temperature phase, the symmetry of $CsPbCl_3$ in phase II is tetragonal because the $PbCl_6$ octahedra have already rotated about one axis. Thus, in the unstressed II-III transition there are only two equivalent rotation axes for the octahedra. A bicritical point in $CsPbCl_3$ should therefore be characterized by an $n=2$ component order parameter, instead of $n=3$ as in the case of $SrTiO_3$.

A scaling theory of a bicritical point may be formulated[22] by including a modifying field σ with a crossover exponent ϕ in a scaling form of the free energy. In the absence of an ordering field the singular part of the free energy takes the asymptotic form

$$G_s(T,\sigma) \sim |t|^{2-\alpha} f(\sigma |t|^{-\phi}) \ . \tag{3}$$

Following Aharony and Bruce,[7] the field σ can represent a symmetry-breaking [100] stress. The thermodynamic conjugate, the spontaneous strain $\Delta\epsilon$, is therefore given by

$$\Delta\epsilon \sim |t|^{2-\alpha-\phi} f'(\sigma |t|^{-\phi}) \ , \tag{4}$$

where $f'(x)=df(x)/dx$. In zero stress the temperature dependence of the thermal expansivity $\Delta\beta = d\Delta\epsilon/dT$ then follows the power law

$$\Delta\beta \sim |t|^{1-\alpha-\phi}, \quad t<0 \ . \tag{5}$$

This behavior is not contained in the Pippard relations.

For $n=2$ in three dimensions one has $\alpha=-0.008$ and $\phi=1.175$,[23,24] giving $\Delta\beta \sim |t|^{-0.167}$. There is excellent agreement between this theoretical result and our observed behavior of the anomalous expansivity. Note here that the present experiments determined β_1, while $\Delta\beta=\beta_2-\beta_1$ is the difference in expansivity along [010] and [100]. The leading singular behavior should, however, be the same in both cases.

The strain measurements were not performed under total stress-free conditions, but the sample experienced a small [100] stress. This implies that possible corrections dictated by the function $f'(x)$ should be considered. For $t<0$ and $\sigma |t|^{-\phi} << 1$, i.e., not too close to the transition temperature, one may write $f'(x) \simeq f'(0)+f''(0)x$. This gives an additional contribution to $\Delta\beta$ in Eq. (5) proportional to $|t|^{-1.34}$. The effect of such a term could not be resolved from the data. For $t>0$ the stress-free transition will by symmetry show no spontaneous strain; thus the presence of a small [100] stress should become more evident. Indeed, from Fig. 3 one sees that above T_c there is a clear contribution to the singular part of the expansivity. When we use that $f'(0)$ vanishes for the branch $t>0$ the scaling function becomes

$$f'(x)=f''(0)x+\tfrac{1}{2}f'''(0)x^2+\cdots \ ,$$

for small $\sigma |t|^{-\phi}$. To leading order this gives $\Delta\beta \propto t^{-1.34}$ with a correction term proportional to $t^{-2.52}$. As illustrated by a full curve in Fig. 3, such behavior represents the excess expansivity reasonably well over an intermediate temperature range above T_c.

V. PHASE DIAGRAM FOR THE II-III TRANSITION

To substantiate the suggestion of a bicritical point further, experiments were carried out in order to locate the II-III phase boundary in the [100]-stress–temperature plane. The theory predicts[25,7] that the shape of the boundary lines close to a bicritical point is described in terms of the same crossover exponent ϕ introduced in Eq. (3). The transition temperature T_c as function of symmetry-breaking stress σ is given by

$$T_c(\sigma)=T_c(0)+W\sigma^{1/\phi}+(\text{regular terms}) \ . \tag{6}$$

Previous measurements of the $SrTiO_3$ phase diagram have confirmed the theory in the case of structural transitions.[26]

Compressional [100] stress was accomplished by filling the bellows of the dilatometer[15] with excess gas pressure. The pressure was measured by a pressure transducer (Gulton Ind., Inc. GS613-092EE2U) and a voltmeter. The force on the sample was determined from the transducer voltage by first having the bellows arrangement calibrated against an electronic balance.

The stress dependence of the II-III transition temperature was found by performing five new thermal expansion runs. In all the measurement series the strain varied continuously with temperature, and any indication of a change in the order of the transition was not observed. We assume that T_c can be well represented by the temperature at which the expansivity reaches a maximum. A slight broadening of the expansivity peak at the higher stresses increased the uncertainty in our choice of T_c. At the maximum stress of $\sigma=2.9$ MPa the peak temperature could be determined within ±25 mK. The resulting phase diagram is shown in Fig. 5.

The II-III transition temperature shifted towards higher values as the applied stress increased. One observes that the stress dependence of T_c is not well approximated by a linear function. Instead, a straight line was fitted to $\sigma^{1/\phi}$ versus T_c setting $\phi=1.175$. The best fit

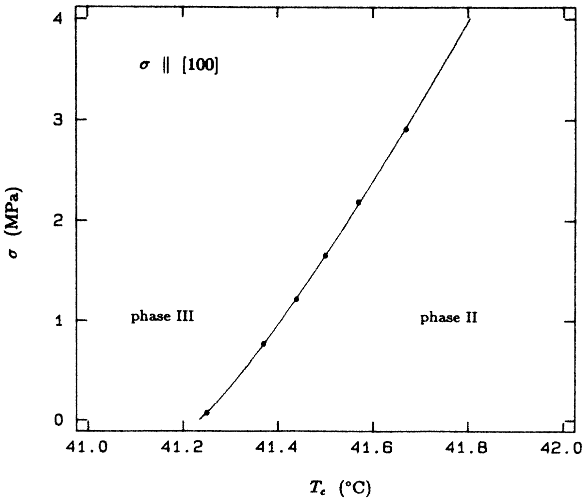

FIG. 5. Phase diagram of $CsPbCl_3$ with uniaxial stress along [100] showing the transition temperature vs stress.

was obtained with $T_c(0)=41.23\,^\circ\mathrm{C}$ and $W=0.176$ $\mathrm{K/(MPa)}^{1/\phi}$, and the corresponding curve has been included in the plot. It is seen to give a very good description of the phase diagram.

An independent fit of the exponent ϕ from the phase diagram was not attempted because of insufficient precision in the phase diagram data.

VI. CONCLUSIONS

From measurements of thermal expansion in $CsPbCl_3$ along [100] we arrive at the following conclusions:

(i) Critical behavior in the linear [100] strain is observed over a temperature range of about 4 K below the continuous II-III transition at 41.2 °C.

(ii) The anomalous thermal expansivity diverges below T_c according to a power law $\sim |t|^{-a}$ with $a=0.18$ giving best fit.

(iii) The II-III phase boundary in the [100] stress–temperature plane has a nonclassical shape when T_c is chosen as the temperature of maximum expansivity. The shift in T_c is well described by $\Delta T_c \propto \sigma^{1/\phi}$ with $\phi=1.175$.

(iv) The above observations provide evidence that the unstressed II-III transition in $CsPbCl_3$ is an $n=2$ bicritical point, analogous to the transition in $SrTiO_3$ at 105 K.

Finally, we have in this work shown that good quality crystals of $CsPbCl_3$ can be grown by the Czochralski technique when the weighing method of diameter control is applied.

ACKNOWLEDGMENTS

The authors are deeply indebted to Professor A. Aharony for stimulating discussions. They also wish to express gratitude to Dr. O. Steinsvoll for performing the neutron scattering rocking curve experiments. The financial support of NAVF (Norwegian Research Council for Science and Humanities) is gratefully acknowledged.

APPENDIX: DIAMETER CONTROL STRATEGY

The weighing method of diameter control in Czochralski growth, pioneered by Bardsley *et al.*,[13] presumes that the growth interface area can be detected by observing the force F experienced by a weighing cell holding the crucible (or the crystal). A simple analysis of the mass transfer in the pulling process yields that the reduced crystal diameter d—crystal diameter in units of crucible diameter $2R_C$—can be related to the rate of change in the force $dF/dt=-G$ according to[14]

$$d=\left[\frac{G}{\rho G+\pi R_C^2\rho_s V}\right]^{1/2} . \tag{A1}$$

Here ρ_s is the mass density of the crystal, and $\rho=\rho_s/\rho_l$ is the ratio of the mass densities in the solid and liquid phases.

To derive the growth rate G numerically from force data F_n the MINC 23 computer repeatedly performs in real time a least-squares line fitting to batches of N data collected at equidistant point in time t_n. The fitting criterion,

$$\sum_n w_n[F_n-F(t_n)]^2=\text{minimum} ,$$

with

$$w_n=\begin{cases}1-n/N \text{ for } n=0 \text{ (latest sample)},1,\ldots,N-1 ,\\ 0, \text{ otherwise}\end{cases} \tag{A2}$$

has been chosen to ensure that latest recorded data enter the computation with a higher weight. The fitting function is linear, $F(t)=at+b$, and gives

$$G=-a=-\frac{1}{\Delta t}\sum_{n=0}^{N-1}\frac{(N-n)(N-1-3n)F_n}{N(N+2)(N^2-1)/12} . \tag{A3}$$

Useful values for the sampling interval Δt and the averaging parameter N are 30 s and 20–30, respectively.

As indicated in Sec. II, the temperature at a point under the crucible is regarded as the primary control variable in the regulation of the growth. Thus, the crystal diameter is controlled by making adjustments in the set-point temperature

$$T_n^{\text{ref}}=T_{n-1}^{\text{ref}}+\Delta_n . \tag{A4}$$

The increment Δ_n is determined on the basis of the observed diameter d^{obs}, derived from Eqs. (A1) and (A3),

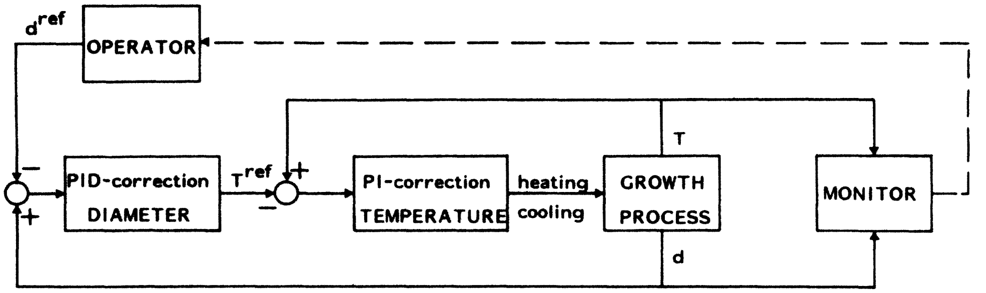

FIG. 6. Block diagram of control strategy for computerized diameter regulation. The dashed line indicates that an operator is free to choose the time variation of the diameter set-point d^{ref} as the growth develops.

and its deviation from a desired value d^{ref}. A proportional-integrating-differentiating (PID) correction

$$\Delta_n = C_P E_n + C_I I_n + C_D D_n \tag{A5}$$

is updated by the computer at each timestep. Here $E_n = d_n^{\text{obs}} - d_n^{\text{ref}}$ is the current error, I_n is the integral (sum) of previous errors, D_n is the two-point differential error, and the coefficients C_P, C_I, and C_D are corresponding gain factors. A schematic picture of the process control is shown in Fig. 6.

---

[1] C. K. Møller, K. Dan Vidensk. Selsk. Mat. Fys. Medd. **32**, No. 2 (1959).

[2] Y. Fuji, S. Hoshino, Y. Yamada, and G. Shirane, Phys. Rev. B **9**, 4549 (1974).

[3] S. Plesko, R. Kind, and J. Roos, J. Phys. Soc. Jpn. **45**, 553 (1978).

[4] A. D. Bruce, Ferroelectrics **12**, 21 (1976).

[5] A. Aharony, Ferroelectrics **24**, 313 (1980).

[6] A. Aharony and D. Blankschtein, Phys. Scr. **T1**, 53 (1982).

[7] A. Aharony and A. D. Bruce, Phys. Rev. Lett. **33**, 427 (1974); A. D. Bruce and A. Aharony, Phys. Rev. B **11**, 478 (1975).

[8] K. A. Müller and W. Berlinger, Phys. Rev. Lett. **35**, 1547 (1975).

[9] W. Rehwald, Solid State Commun. **21**, 667 (1977).

[10] S. Hirotsu and Y. Kunii, J. Phys. Soc. Jpn. **50**, 1249 (1981).

[11] K. Hamano and S. Hirotsu, J. Phys. Soc. Jpn. **48**, 168 (1980).

[12] S. Hirotsu and T. Suzuki, Ferroelectrics **26**, 179 (1978).

[13] W. Bardsley, G. W. Green, C. H. Holliday, and D. T. J. Hurle, J. Cryst. Growth **16**, 277 (1972).

[14] T. H. Johansen, J. Cryst. Growth **80**, 343 (1987).

[15] T. H. Johansen, J. Feder, and T. Jøssang, Rev. Sci. Instrum. **57**, 1168 (1986).

[16] T. H. Johansen, J. Appl. Phys. **60**, 2754 (1986).

[17] S. Hirotsu, J. Phys. Soc. Jpn. **31**, 552 (1971).

[18] S. Hirotsu and T. Suzuki, J. Phys. Soc. Jpn. **44**, 1604 (1978).

[19] M. Midorikawa, Y. Ishibashi, and Y. Tagaki, J. Phys. Soc. Jpn. **41**, 2001 (1976).

[20] V. Janovec, J. Chem. Phys. **45**, 1874 (1966).

[21] T. H. Johansen, J. Feder, and T. Jøssang, Z. Phys. B **56**, 41 (1984).

[22] M. E. Fisher and D. R. Nelson, Phys. Rev. Lett. **32**, 1350 (1974).

[23] J. C. LeGuillou and J. Zinn-Justin, Phys. Rev. Lett. **39**, 95 (1977).

[24] M. E. Fisher, *Magnetism and Magnetic Materials—1974 (San Franciso)*, Proceedings of the 20th Annual Conference on Magnetism and Magnetic Materials, AIP Conf. Proc. No. 24, edited by C. D. Graham, G. H. Lander, and J. J. Rhyne (AIP, New York, 1975).

[25] P. Pfeuty, D. Jasnow, and M. E. Fisher, Phys. Rev. B **10**, 2088 (1974).

[26] S. Stokka and K. Fossheim, Phys. Rev. B **25**, 4896 (1982).