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OPTICAL MICROSCOPIC STUDY OF THE NH_4Cl PHASE TRANSITION WITH OBSERVATIONS OF SLIP BANDS, HETEROPHASE AND DOMAIN STRUCTURE

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Résumé. — Des bandes de glissement parallèles aux plans $\{100\}$ ont été observées presque systématiquement dans des monocristaux de NH_4Cl . Celles-ci sont décrites et caractérisées et on montre qu'elles induisent des contraintes internes inhomogènes qui jouent un rôle important dans le processus de nucléation lors du changement de phase ordre-désordre. La structure hétérophase, pendant la coexistence de phases, est constituée de lames parallèles aux plans $\{111\}$, en accord avec les prédictions théoriques concernant la forme d'inclusions dans une matrice de symétrie cubique. Le changement de phase met en jeu de grandes contraintes internes qui provoquent des déformations plastiques. Des photographies de la structure en domaines dans la phase ordonnée révélée par effet électro-optique, sont présentées pour la première fois :

Elles indiquent que les parois de domaines sont aussi des plans $\{111\}$. Des arguments théoriques sont exposés pour expliquer cette orientation.

Abstract. — Slip bands parallel to $\{100\}$ planes have been observed almost universally in NH_4Cl single crystals. They are described and characterized and it is shown that they induce inhomogeneous internal stresses which play an important role in the nucleation process at the order-disorder phase transformation. The heterophase structure at the phase coexistence consists of slabs parallel to $\{111\}$ planes, in agreement with theoretical predictions concerning the shape of inclusions in a matrix of cubic symmetry. The phase transformation involves large internal stresses which result in plastic deformations. Pictures of the domain structure in the ordered phase using the electro-optical effect are presented for the first time : they show that domain boundaries are also $\{111\}$ planes.

Theoretical arguments are given to explain this orientation.

Introduction. — Recently considerable attention has been given to the order-disorder phase transition of NH_4Cl which, at room pressure, is weakly first order and which becomes second order above a pressure of 1.5 kbar [1, 2]. However the exact nature of this singularity is not known, as the measured value [3] of the critical exponent β is nearer to $1/6$ than to $1/4$, as would be required for a tricritical point [4, 5].

Another point of interest in NH_4Cl is the possibility of a central peak in light scattering. Despite a strong increase in the light intensity at the transition [6, 7, 8], there is no conclusive evidence of a dynamical effect [9]. Recent discussion of the central peak has shown the importance of defects in this phenomenon [10]. As NH_4Cl is reported to be a soft crystal [11], it would seem interesting to investigate experimentally the

influence of plasticity and dislocations on this transition, following some conjectures by Bartis [12, 13].

The NH_4Cl transition is particularly interesting as the transition mechanism is quite simple [1]. Below 183°C , NH_4Cl has the CsCl structure with the NH_4^+ tetrahedra at the centre of a Cl^- simple cubic lattice. The NH_4^+ can have two orientations where NH bonds point towards the neighbouring Cl^- along $\langle 111 \rangle$ directions. Above $T_0 = -30^\circ\text{C}$, the NH_4^+ are disordered and the crystal symmetry is $m\bar{3}m$ (centrosymmetric structure). Below T_0 there is a preferred parallel ordering of the NH_4^+ which produces $43m$ symmetry and the disappearance of the centre of symmetry. Huller [14] has shown that there is a competition between the direct octupole-octupole interaction which favours parallel ordering of the NH_4^+ and an indirect octupole-dipole-octupole interaction via the anion which favours an antiparallel orientation, which actually exists in NH_4Br . As these forces are of short-

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range character, NH_4Cl can be considered as a realisation of a 3-dimensional Ising model on a compressible lattice.

Since the first observation of this transition by Simon [15] (1922) a great number of experiments have been performed. However some features remain puzzling, such as a *tail* of the order parameter which is systematically observed in the high temperature phase [3]. In a second harmonic generation experiment I. Freund *et al.* [16] have observed the persistence of long-range order several degrees above T_0 . Some authors have mentioned the existence of *domains* around or below the transition temperature, but this word seems to have been used with some ambiguity. Sometimes [7, 9] it means the mixture of high and low temperature phases in the coexistence region, already observed with X-rays by Dinichert [17], which, following Roidburd [18], we will call *heterophase structure*. This mixed state leads to an intense light scattering at the phase transition, as also observed in quartz [19]. At other times the expression *domain structure* is used with its usual meaning and refers to a structure where the NH_4^+ (or the order parameter) have the same orientation in a given region but different orientations from one region to the neighbouring one. These domains produce the non-linear diffraction observed by Freund in the low temperature phase [20]. Heterophase and domain structures are both reported to be lamellae parallel to $\{111\}$ planes [7, 20]. Surprisingly no direct optical microscopic observation of these phenomena seems to have been reported.

We present here the first results of our observations. In part 1 we describe the room temperature observations of NH_4Cl crystal in polarized light, which show internal stresses in slip bands. This type of defect is almost always observed due to the rather low limit of elasticity of the crystal. Observations of the heterophase structure of ordered and disordered regions are presented in part 2, while the direct observation of the domain structure by the electro-optic effect is described in part 3.

1. Slip band observations. — For our experiments NH_4Cl single crystals were grown in our laboratory from aqueous solutions with urea as habit modifier [7] following two methods :

A. Slow cooling ($0.1^\circ\text{C}/\text{day}$) around 45°C .

B. Slow evaporation at stabilized temperature ($\sim 25^\circ\text{C}$).

Generally, cubic single crystals $7 \times 7 \times 7 \text{ mm}^3$ in size or larger with $\{100\}$ faces were obtained with method A and smaller crystals with method B. Immediately after growth, the crystals were put in silicon oil to preserve a good surface quality. Optical observations of the grown crystals were made with a polarizing microscope. The picture of figure 1, typical of type A crystals observed with crossed polarizers at 45° from $\langle 100 \rangle$ directions, shows birefringent bands produced by internal stresses. These bands are parallel to crystal

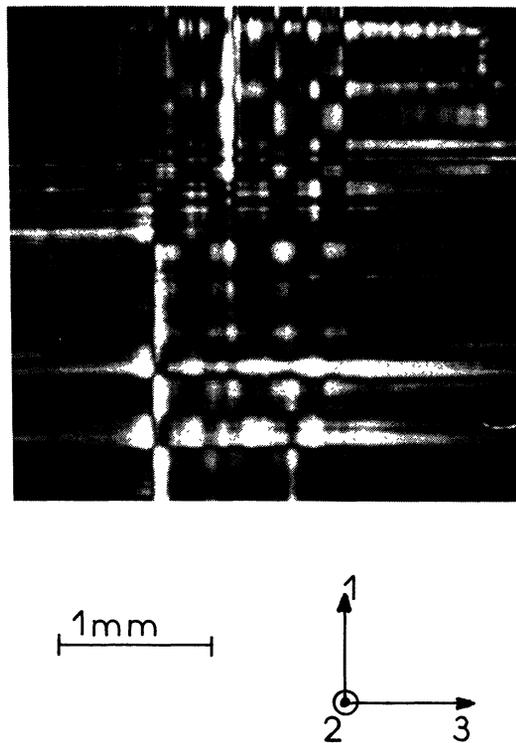


FIG. 1. — Picture of a $3 \times 3 \times 1,5 \text{ mm}$ NH_4Cl single crystal (type A) observed between crossed polarizers at 45° of $\langle 100 \rangle$, showing an inhomogeneous internal stress field.

edges, i.e. perpendicular to $\langle 100 \rangle$ directions as verified by X-rays. Type B samples grown at room temperature and handled with great care to prevent thermal or mechanical shocks, present a very small number of birefringent bands. However these defects appear under a rapid temperature variation (evaporation of an acetone drop for example) or a small mechanical stress. Thus it seems that the difference observed between type A and type B crystals is probably due to handling rather than to the growing method. These observations indicate that the elastic limit of NH_4Cl is very low, probably smaller than the 50 kg/cm^2 reported by Narasimhamurty [11] and nearer to that observed in CsI [21] (5 kg/cm^2) or other ionic crystals. A simple birefringence pattern was observed in a crystal grown as a $0.5 \times 4 \times 8 \text{ mm}^3$ plate, where only one kind of parallel band crossing the whole crystal thickness is present (Fig. 2a). By using a tint plate, one can work near the sensitive colour; then blue tints appear in one band and red tints in the neighbouring one showing that stresses are respectively compressive and extensive. When the crossed polarizers are parallel (or perpendicular) to the $\langle 100 \rangle$ direction the birefringent bands are no longer seen. Similar observations have previously been made on alkali halides and interpreted as slip bands, with a dislocation structure [22, 23, 24].

The stress field produced by a planar distribution of periodic edge dislocations is the sum of two contributions [25, 26] : a uniform term σ' changing its sign

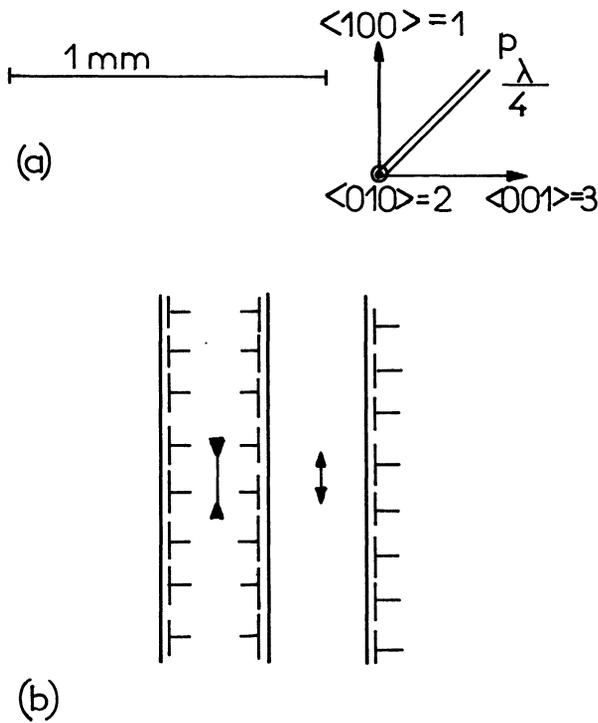
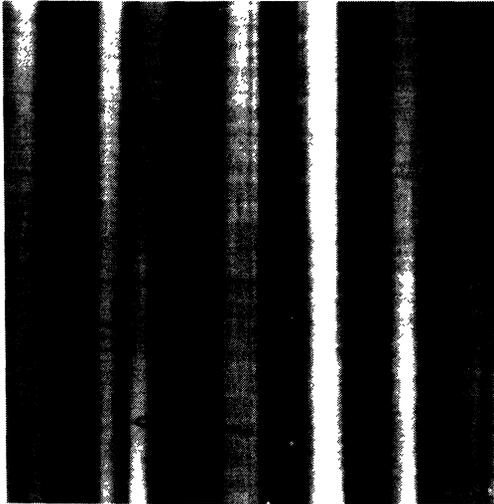


FIG. 2. — *a*) Stress birefringent bands in a thin (0.5 mm) plate of NH₄Cl single crystal observed by de Senarmont's method. The retardation angle is constant in a band and changes sign in successive bands indicating alternate compressive and extensive uniform stresses. *b*) Model of edge dislocation distribution explaining internal stresses observed in figure 2*a*.

when crossing the glide plane and a modulated term σ'' decreasing exponentially with the distance from the glide plane but diverging when approaching the dislocation core.

Taking axis 1 parallel to the Burger's vector, axis 2 parallel to the dislocation line and axis 3 perpendicular to the glide plane (Fig. 2*b*), the uniform stress σ' has only two components σ'_1 and σ'_2 . Choosing the origin on a dislocation line one has :

$$\sigma_1(x = 0, y, z) = \frac{\mu}{1 - \nu} \cdot \frac{b}{h} \times \left[1 + \frac{2}{e^{2\pi z/h} - 1} - \frac{2 \pi z/h \cdot e^{2\pi z/h}}{(e^{2\pi z/h} - 1)^2} \right] \quad (1)$$

$$\sigma_2(x = 0, y, z) = \frac{\nu \cdot \mu}{1 - \nu} \cdot \frac{b}{h} \left[1 + \frac{2}{e^{2\pi z/h} - 1} \right] \quad (2)$$

where μ is the shear modulus, ν the Poisson modulus, b the Burger's vector modulus and h the distance between two dislocations. Neglecting σ'' , the stress discontinuity across the glide plane is $2 \sigma'$. Edge dislocations of opposite signs must exist in successive glide planes in order to generate the alternate uniform compressive and extensive stresses.

Dislocations emerging at the crystal surface were indeed observed by etching (Fig. 3). The crystal was placed in a solution of 50% ethanol in water for a few seconds and then in silicon oil.

The etch pits are aligned periodically along a $\langle 100 \rangle$ direction and must correspond to dislocation

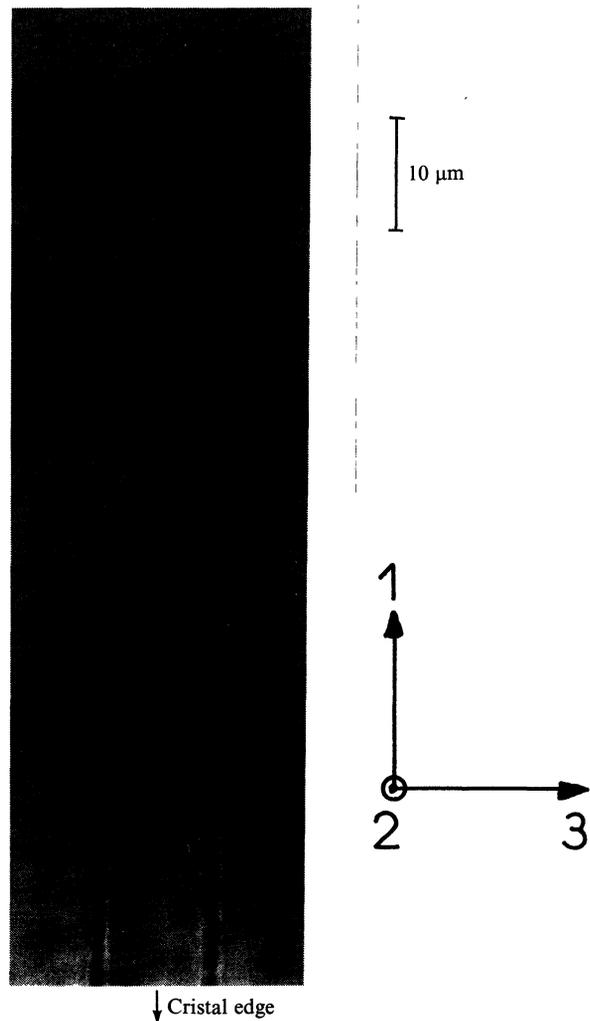


FIG. 3. — Etching of the surface of a NH₄Cl single crystal revealing the emergence of dislocations periodically distributed in the glide plane. Near a crystal edge there is a pile up phenomenon. These dislocation lines coincide with the separation between birefringent bands.

lines parallel to $\langle 010 \rangle$ with a $\{001\}$ glide plane. They are separated by a distance $h \sim 1.5 \mu\text{m}$ which is typical of the crystals investigated. Using this value of h , and knowing the elastic constants [27] we can calculate $(\sigma'_1)_{\text{th}} \sim 30 \times 10^6 \text{ dynes/cm}^2$ (with

$$\mu = c_{44} = 10^{11} \text{ dynes/cm}^2, \\ \nu = c_{12}/(c_{11} + c_{12}) = 0.13, \quad b = a = 3.86 \text{ \AA}.$$

This value of σ'_1 can be compared to that deduced from a measurement of the birefringence discontinuity

$$\Delta n = \frac{n^3}{2} (q_{12} - q_{11}) \cdot 2 \sigma'_1$$

where n is the refractive index and q_{ij} photo-elastic constants. From the value $\Delta n = 5.2 \times 10^{-5}$ measured by de Senarmont's method and the reported values $n = 1.666$ and $q_{12} - q_{11} = 3.06 \times 10^{-13} \text{ dynes/cm}^2$ [11] we find :

$$(\sigma'_1)_{\text{exp}} = 37 \times 10^6 \text{ dynes/cm}^2$$

which is in good agreement with the above value.

Putting this value in the exponential term σ'' one finds that the stress at 250 \AA from a dislocation core is about $500 \times 10^6 \text{ dynes/cm}^2$. As a hydrostatic pressure of 10^9 dynes/cm^2 increases the transition temperature by $10 \text{ }^\circ\text{C}$ [1], one may expect some influence of the internal stresses on transition phenomena, which are indeed described in part 2.

2. Heterophase structure. — Optical observations of the NH_4Cl transition were made in a specially constructed cryostat : the sample is placed inside a glass cell filled with silicon oil to reduce surface scattering and improve thermal contact. This cell is enclosed in a large copper block so as to maintain good temperature stability.

Type A crystals were slowly cut with a diamond saw and polished on a silk cloth wetted with ethanol, to have $\{110\}$ faces. A He-Ne or a white light beam was incident in the $\langle 110 \rangle$ direction. Visual observations were made with a polarizing microscope with a long focal objective. At room temperature one sees only the $\{001\}$ slip bands with the crossed polarizers at 45° from $\langle 001 \rangle$. On withdrawing the polarizers, these bands vanish. When cooling, new features appear near $-28 \text{ }^\circ\text{C}$, that is, about $2 \text{ }^\circ\text{C}$ above the transition temperature.

First small needles, perpendicular to $\langle 111 \rangle$ and $\langle 1\bar{1}\bar{1} \rangle$, appear in the more compressed regions of the slip bands (Fig. 4a). As the temperature is lowered new nucleations appear and the needles grow throughout the crystal volume (Fig. 4b, c). The measured angle between the needles is $109^\circ \pm 2^\circ$, which is the angle between $\langle 111 \rangle$ and $\langle 1\bar{1}\bar{1} \rangle$. With a tint plate and polarizers, broad differently coloured regions appear during the coexistence state. At the same time

there is such an intense light scattering that virtually no light remains in the beam of the He-Ne laser in the $\langle 110 \rangle$ direction. Near the forward direction the scattered light forms a cross with arms perpendicular to the needle directions, on a screen perpendicular to the beam. With further cooling this heterophase structure disappears near $-31 \text{ }^\circ\text{C}$ but some traces visible in polarized light remain, showing some plastic deformations (Fig. 4d). On heating, the same phenomena are observed between $-30 \text{ }^\circ\text{C}$ and $-27 \text{ }^\circ\text{C}$, the temperature being displaced by about $1 \text{ }^\circ\text{C}$ due to hysteresis. If the crystal is kept for several days in the low or high temperature phase, without further transition, the contrast of these traces decreases slowly. The observed needles are probably the section of $\{111\}$ plates seen in projection along $\langle 110 \rangle$. As suggested by Bruins *et al.* [3] $\{111\}$ orientation comes from elastic energy considerations. Following Khachaturyan [28] one may consider that the local free energy $f(\mathbf{r})$ of a crystal with a single inclusion is given by :

$$f(\mathbf{r}) = - \sum_i \sigma_i^0 \cdot \varepsilon_i \theta(\mathbf{r}) + \frac{1}{2} \sum_{i,j} c_{ij} \cdot \varepsilon_i \cdot \varepsilon_j \quad (3)$$

where $\theta(\mathbf{r}) = 1$ inside the inclusion and 0 outside,

$$\sigma_i^0 = \sum_j (c_{ij} \varepsilon_j^0)$$

with ε_j^0 the transformation strain tensor. In our case $\varepsilon_1^0 = \varepsilon_2^0 = \varepsilon_3^0 = \Delta a/a$ is the relative variation of the lattice parameter and $\varepsilon_4^0 = \varepsilon_5^0 = \varepsilon_6^0 = 0$.

By writing the equilibrium condition one can calculate the total elastic energy due to the presence of an inclusion of a given volume as a function of its shape $\theta(r)$.

In the cubic symmetry the minimum energy shape is a $\{100\}$ plane if $\Delta = C_{11} - C_{12} - 2C_{44}$ is negative and a $\{111\}$ plane if Δ is positive. In NH_4Cl the latter case prevails [27]. As explained by Katchaturyan the preceding calculation does not take into account the surface energy of the inclusion, which, if taken alone, would favour a spherical shape. This may explain why the nuclei first appear with an elliptical section and become more and more elongated when growing, since the surface effect becomes less important as the volume increases.

Using the reported values

$$\frac{\Delta a}{a} \approx 2 \times 10^{-3} [29],$$

$$C_{11} = 3.5 \times 10^{11} \text{ dynes/cm}^2$$

and

$$C_{12} = 0.5 \times 10^{11} \text{ dynes/cm}^2 [27]$$

one calculates the value of the transformation stress :

$$\sigma^0 = (C_{11} + 2C_{12}) \varepsilon^0 = 8.8 \times 10^8 \text{ dynes/cm}^2 \quad (4)$$

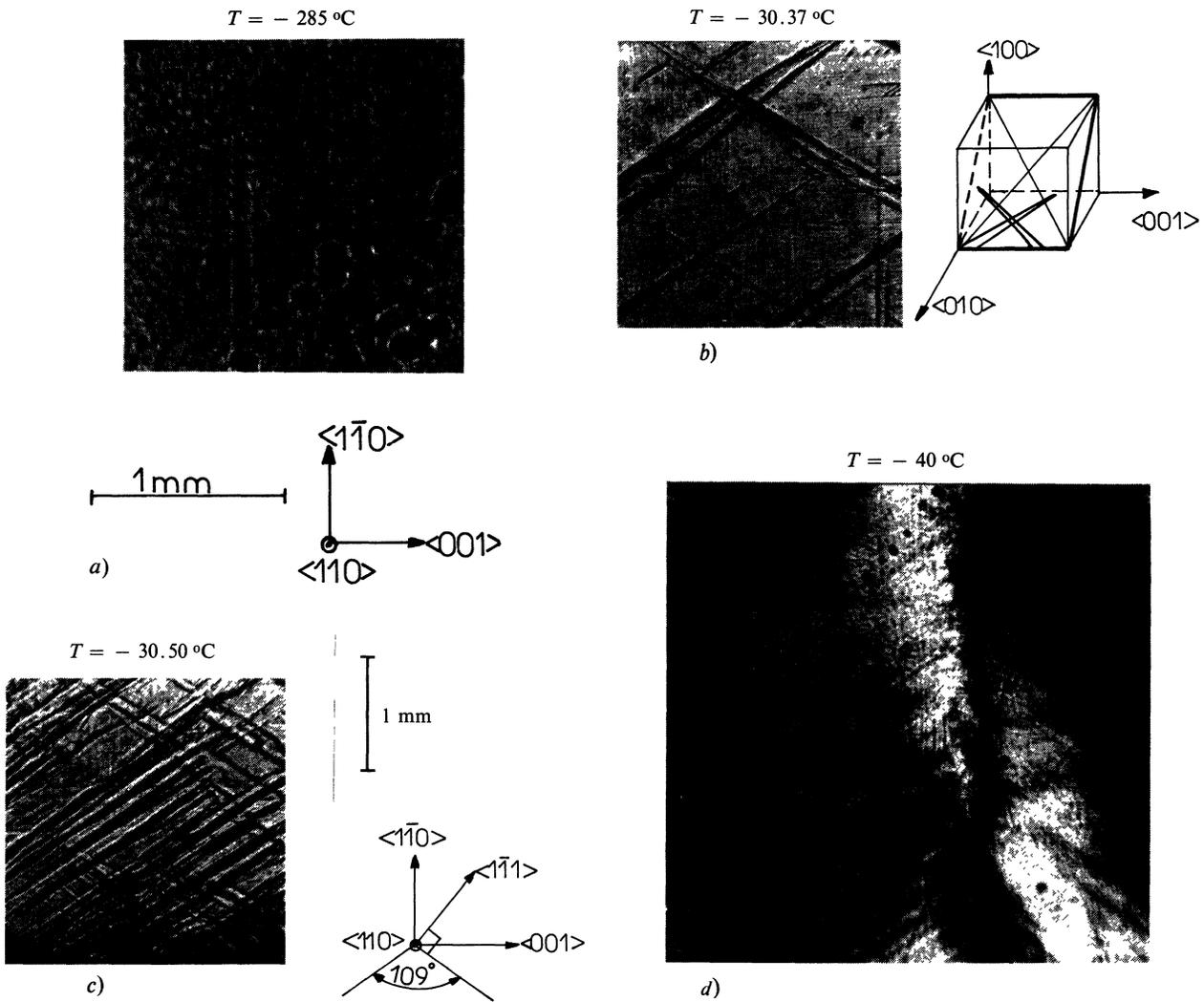


FIG. 4. — *a*) Nucleation of the low temperature phase in compressive bands about two degrees above the transition temperature. *b*) Heterophase structure at the beginning of the phase coexistence showing the growth of the nuclei as plates parallel

to {111} planes. *c*) Heterophase structure extending over the whole crystal. *d*) Internal stresses produced by the heterophase structure, observed between crossed polarizers several degrees below the transition.

Within the hypothesis of an elastic isotropic medium, Eshelby [30] has calculated the stress inside an ellipsoidal inclusion. In our case one obtains :

$$\sigma_1 = \sigma_2 = \sigma_3 = (C_{11} + 2 C_{12}) \times \left(\frac{1}{3} \times \frac{1 + \nu}{1 - \nu} - 1 \right) \varepsilon^0 \sim 0.57 \sigma^0 . \quad (5)$$

This value is surely far beyond the elastic limit of the material so that a correct estimate must include the plastic behaviour of the crystal.

This remark explains our observations of traces of the heterophase structure well outside the coexistence region and the *memory effects* observed by several authors [3, 16] and the particular behaviour of *virgin* samples which have never passed through the transition point.

3. Domain structure. — In many solid state phase-transitions, several equivalent states can be found in the low temperature phase, producing a domain structure. In NH₄Cl the order parameter η can take two opposite values $\pm \eta_0$ corresponding to the two orientations of the NH₄ tetrahedra. The symmetry operation which transforms one state into the other is an inversion around the cube centre, and in NH₄Cl this inversion can affect only physical properties described by an odd rank tensor [31, 32]. As NH₄Cl does not have spontaneous polarization (first rank tensor) the smallest rank tensor which can exist is third rank and related to such effects as the piezoelectricity, electro-optic effect or second harmonic generation, properties which indeed appear in the low temperature phase of NH₄Cl. The effect of the domain structure has been observed in piezoelectric measurements [3, 33], and in second harmonic generation

where it produces the non-linear diffraction reported by Freund [20].

These domains can be moved by simultaneous application of stress and electric field, and one can even obtain a single domain crystal [33].

We have been able to make, for the first time, a direct observation of these domains by using the electro-optic effect. Gold electrodes were evaporated on {001} faces for electric field application and an He-Ne laser beam was passed in the $\langle 110 \rangle$ direction. As the electro-optic coefficient r_{41} changes its sign from one domain to the other, the electric field produces opposite optical retardation on the polarized light beam in two different domains. These phase changes can be transformed into intensity variations by introducing a birefringent element into the light beam. The picture of a NH_4Cl obtained by this method is shown in figure 5. The domains were also found to be parallel to {111} planes, so that the light beam propagating along $\langle 110 \rangle$ can remain in the same domain throughout the sample thickness for domain families {111} and $\{\bar{1}\bar{1}\bar{1}\}$. For domains parallel to $\{\bar{1}\bar{1}\bar{1}\}$ and $\{\bar{1}\bar{1}\bar{1}\}$ a light beam will encounter positive and negative domains, and they can not be observed. One expects that in general the four {111} domain families will be present in a given sample. But by chance sometimes only one family exists and gives a good contrast picture with a d.c. field of 20 kV/cm (Fig. 5). Electro-optic measurements with an a.c. field (3 kV/cm peak to peak) actually show a change of sign of the effect when the laser beam is scanned across a domain wall, while the absolute value of the effect is uniform inside one domain and nearly the same in opposite domains. The measured value of the electro-optic coefficient r_{41} at -44°C is about 1.46×10^{-10} cm/V.

This result is in good agreement with the only reported value $r_{41} = 1.4 \times 10^{-10}$ cm/V [34].

The accurate temperature dependence of this coefficient has been studied and will be published elsewhere. We only mention here that the electro-optic effect is still found above the transition temperature but that

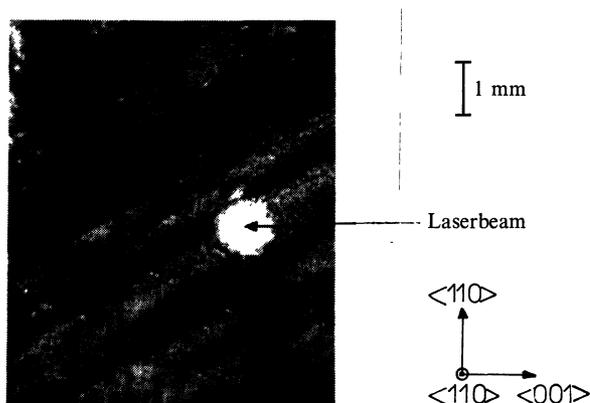


FIG. 5. — Domain pattern in the ordered phase of a NH_4Cl crystal revealed by electro-optic effect with a d.c. field of 20 kV/cm applied along the $\langle 001 \rangle$ direction.

it is spatially very inhomogeneous and stronger in the more highly stressed slip bands. It disappears gradually several degrees above the transition point. This indicates clearly the importance of the defects in studying the transition in this crystal.

Concerning the presence of {111} plane domain boundaries, two explanations may be given :

a) These boundaries are influenced by the shape of the heterophase structure and in particular by the plastic deformations that it leaves in the lower temperature phase.

b) The inhomogeneous striction [35] also favours {111} type domain boundaries. This effect may easily be derived with the formalism previously used to describe the heterophase structure. The local free energy is written :

$$f(\mathbf{r}) = \frac{a}{2} \eta^2(\mathbf{r}) + \frac{b}{4} \eta^4(\mathbf{r}) + \frac{c}{2} (\nabla \eta)^2 - q \cdot \eta^2(\mathbf{r}) \cdot \sum_{i=1}^3 \varepsilon_i(\mathbf{r}) + \frac{1}{2} \sum_{i,j} c_{ij} \cdot \varepsilon_i(\mathbf{r}) \cdot \varepsilon_j(\mathbf{r}) \quad (6)$$

where $\eta(\mathbf{r})$ is the order parameter, a , b and c the usual coefficients of the Landau expansion of the free energy and q the striction coefficient.

For $a < 0$, and discarding the weak striction coupling, the equilibrium state is

$$\eta(\mathbf{r}) = \pm \left(\frac{a}{b} \right)^{1/2} = \pm \eta_0$$

except in the domain boundary where it varies proportionally to $\tanh |x \sqrt{a/c}|$ (where the x axis is normal to the boundary). In the cubic symmetry the gradient term cannot introduce any anisotropy of the boundary. The striction term in the free energy may be decomposed into a homogeneous part

$$- q \eta_0^2 \sum_{i=1}^3 \varepsilon_i(\mathbf{r})$$

and an inhomogeneous part

$$- q(\eta^2(\mathbf{r}) - \eta_0^2) \sum_{i=1}^3 \varepsilon_i(\mathbf{r}) .$$

This last term is non-vanishing only inside the domain boundary and plays exactly the same role as

$$- \sigma^0 \sum_{i=1}^3 \varepsilon_i(\mathbf{r}) \theta(\mathbf{r})$$

in the inclusion problem (in particular its tensorial form is the same). Therefore it is easily demonstrated that for a given thickness (determined by $(c/a)^{1/2}$) the optimal shape of the domain boundary is a {111} plane.

A possible test of this last hypothesis will be provided by the observation of the domain pattern under

high pressure : the transition is then 2nd order and phase coexistence no longer exists so that the first mechanism *a.* cannot be invoked.

4. **Conclusion.** — In this paper we have described our observations of very easily created crystallographic defects in NH₄Cl : slip bands parallel to { 100 } planes. We have shown their importance in the nucleation process during the order-disorder phase transformation. The nucleation on this type of defect provides an attractive explanation of the *long-range order* observed in the high temperature phase in various experiments and in particular in the non-linear diffraction reported by Freund [16]. We have described the heterophase structure which is present during the

coexistence at the transition and we have shown that it imposes plastic deformations which are responsible for some *memory effects* observed after the crystal has passed through the transition. High pressure decreases the jump in the lattice parameter and probably increases the elastic limit of the material : these effects must influence the nucleation process. Experiments are currently under way to investigate this point and in particular how the phase coexistence disappears when approaching the tricritical point. Finally we have reported the first pictures of the domain structure in the ordered phase as observed by the electro-optical effect. Extensive electro-optical and non-linear optical measurements under high pressure are also in progress in our laboratory.

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