

phys. stat. sol. (a) **66**, 219 (1981)

Subject classification: 10.2; 22.5.1; 22.5.2

*Institut für Metallphysik der Universität Göttingen<sup>1)</sup>*

## Plasticity of Polycrystalline Ionic Solids

By

W. SKROTZKI, G. FROMMEYER, and P. HAASEN

Alkali and silver halide polycrystals prepared by extrusion are deformed in compression and in tension at different temperatures. The results show that the critical flow stress of these materials is determined by the activation of secondary  $\{100\} \langle 110 \rangle$  slip as long as there is no strong  $[100]$  fibre texture. This is necessary to fulfill the von Mises criterion. On the other hand, the ductility is correlated with the change of the slip mode on  $\{110\}$  planes from planar to wavy. Waviness is generated by the cross slip of dissociated screw dislocations and the motion of edge dislocation jogs produced on the cross slip plane. Of these processes the one dominates which requires the strongest activation. The experimental results show contrary to previous statements that the change in slip mode does not necessarily occur at the temperature at which secondary slip becomes easy.

Durch Extrusion hergestellte Alkali- und Silberhalogenidpolykristalle werden durch Druck- und Zugbeanspruchung bei verschiedenen Temperaturen deformiert. Die Ergebnisse zeigen, daß die kritische Fließspannung dieser Materialien solange durch die Aktivierung sekundärer  $\{110\} \langle 110 \rangle$ -Gleitung bestimmt wird, wie keine starke  $[100]$ -Fasertextur vorhanden ist. Dies ist für die Erfüllung des von Mises-Kriterium notwendig. Andererseits ist die Duktilität mit der Änderung der Gleitmode auf  $\{110\}$ -Ebenen von planar zu wellig verknüpft. Welligkeit wird durch Quergleitung aufgespaltener Schraubenversetzungen und der Bewegung von Sprüngen von Stufenversetzungen auf der Quergleitebene erzeugt. Von diesen Prozessen dominiert derjenige, der die stärkste Aktivierung erfordert. Die experimentellen Ergebnisse zeigen im Gegensatz zu früheren Feststellungen, daß die Änderung der Gleitmode nicht notwendigerweise bei Temperaturen auftritt, bei denen sekundäre Gleitung einfach wird.

### 1. Introduction

A homogeneous plastic deformation of a polycrystal with a random distribution of grain orientations is only possible if each grain is capable of a perfectly general strain. This means that each grain has to realize five components of the strain tensor by shear in different slip systems. In 1928 von Mises [1] recognized that five independent slip systems are necessary for this. In ionic crystals with the NaCl-structure slip is restricted to the primary  $\{110\} \langle 110 \rangle$  and the secondary  $\{100\} \langle 110 \rangle$  systems. Six physically distinct slip systems of each exist, but only two and three, respectively, are independent of each other [2]. Therefore, the activation of both types of slip systems is necessary to deform a polycrystalline ionic solid without texture.

Recently Skrotzki and Haasen [3] have shown by a comprehensive investigation of the plastic anisotropy that below a critical temperature the  $\{110\} \langle 110 \rangle$  and  $\{100\} \langle 110 \rangle$  slip systems differ strongly in their critical resolved shear stresses. This characteristic feature of ionic crystals is demonstrated in Fig. 1. An analysis of the low-temperature results showed that the steep increase of the plastic anisotropy below  $T_0^{(100)}$  is due to a high Peierls potential for cubic slip [4].

It is the aim of the present work to reveal the role of this plastic anisotropy in the deformation of ionic polycrystals. In particular it should be checked whether the plastic anisotropy determines the ductility of these materials.

<sup>1)</sup> Hospitalstr. 12, D-3400 Göttingen, BRD.

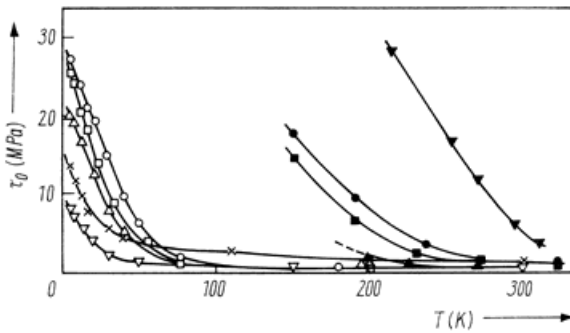


Fig. 1. Temperature dependence of the critical resolved shear stresses for slip on  $\{110\}$  and  $\{100\}$  planes. LiF:  $\times$   $\{110\}$ ; NaCl:  $\nabla$   $\{110\}$ ,  $\blacktriangledown$   $\{100\}$ ; KCl:  $\circ$   $\{110\}$ ,  $\bullet$   $\{100\}$ ; KBr:  $\square$   $\{110\}$ ,  $\blacksquare$   $\{100\}$ ; KI:  $\triangle$   $\{110\}$ ,  $\blacktriangle$   $\{100\}$

## 2. Experimental

Alkali and silver halide polycrystals have been prepared from reagent-grade powders (Merck, pro analysi) in two steps. First the powders (grain size in the order of  $100\ \mu\text{m}$  for the AH and  $10\ \mu\text{m}$  for AgCl) have been compressed into cylindrical blocks of a density  $\rho \approx 0.98\rho_{\text{theor}}$ . Secondly these blocks have been extruded at different temperatures  $T_{\text{ex}}$  to rods of 2 cm diameter and about 30 cm length with an extrusion ratio of 14/1 leading to a density  $\rho \approx 0.99\rho_{\text{theor}}$ . The microstructures of these rods, illustrated in Fig. 2, reveal that the grains are deformed plastically and elongated in the extrusion direction. A preferred orientation in the extruded materials was found by neutron diffraction in the form of a  $[100][111]$  double fibre texture characteristic of extruded f.c.c. metals (Table 1). Details about the texture investigations will be published

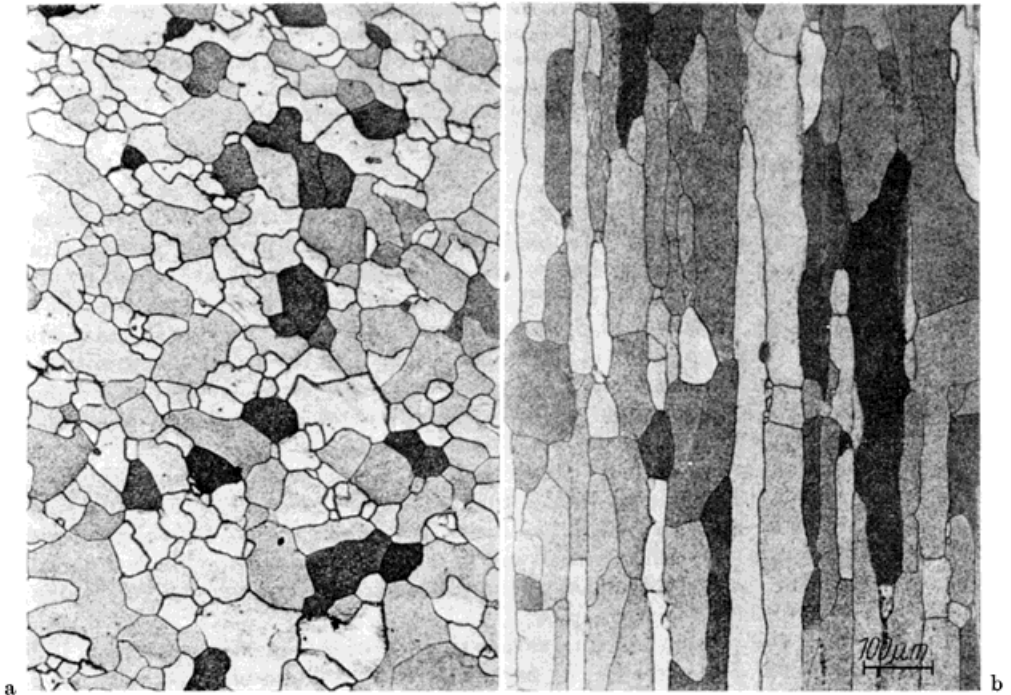


Fig. 2. Effect of plastic flow on the microstructure of KCl extruded at 570 K. a) Perpendicular to and b) parallel to the direction of extrusion

Table 1  
Results of texture determination

		NaCl	KCl	KBr	AgCl
$T_{\text{ex}}/T_{\text{m}}$		0.53	0.55	0.56	0.41
intensity in units	[100]	16.9	9.4	7.0	5.5
of a random orientation distribution	[111]	1.9	1.3	1.7	8.9

Extrusion temperature reduced to the melting temperature ( $T_{\text{ex}}/T_{\text{m}}$ ) and intensity of the [100] and [111] orientation in units of a random orientation distribution.

separately [5]. Cylindrical specimens for compression tests (diameter  $d$ : AH 4 mm AgCl 3 mm; length  $l$ : AH 16 mm, AgCl 12 mm) and standard specimens for tension tests ( $d$ : AH 5 mm, AgCl 3 mm and  $l$ : AH 16 mm, AgCl 12 mm for the reduced section) have been machined from the extruded rods by a lathe. To get preferred deformation in the middle of the samples the tension specimens had an additional reduction 0.2 mm in diameter for a length of 5 mm. Before testing all specimens have been chemically polished [6]. The deformation experiments have been performed in an Instron machine at an initial strain rate of  $(5 \text{ to } 7) \times 10^{-5} \text{ s}^{-1}$ . The test temperatures have been achieved by cooling and heating in different inert liquids [6].

### 3. Results

The onset of plastic deformation of the polycrystalline samples has been determined from parabolic stress-strain curves by taking the flow stress at 0.1% strain, termed  $\sigma_{0.1}$ . In Fig. 3a to d the temperature dependence of  $\sigma_{0.1}$  is shown in comparison with the temperature dependence of the critical resolved shear stresses for slip on {110} and {100} planes. It is evident that at the temperature at which cubic slip becomes more difficult there is an increase of  $\sigma_{0.1}$ , too. The fact that  $\sigma_{0.1}(T)$  does not increase as strongly as  $\tau_0^{(100)}(T)$  presumably means that the deformation is mainly carried by slip on {110} planes especially at low temperatures. It should be pointed out that our polycrystals have a strong texture which depends on the ionicity of the material (see Table 1). For NaCl the fibre axis of the texture is mainly [100], so the predominance of {110} slip is to be expected. For AgCl, on the other hand, we found a [100] [111] double fibre texture. In agreement with this the critical flow stress rises steeply at  $T_0^{(100)}$ . The critical flow stresses measured in compression and tension agree quite well with each other as is demonstrated for AgCl in Fig. 3d. For the alkali halides the critical flow stresses could not be determined in tension at low temperatures because these materials are considerably less plastic in tension than in compression ( $\epsilon_{\text{tension}} < 0.1\%$ ,  $\epsilon_{\text{compression}} > 3\%$  at room temperature). At high temperatures there is a strong increase of the plastic strain to fracture of about three orders in magnitude depending on the material (Fig. 4). The transition to ductile behaviour occurs in a relatively narrow temperature range. The temperatures of the brittle-ductile transition  $T_{\text{bd}}$  are listed in Table 2. Comparable results have been obtained by Stoloff et al. [10] and Stokes [11] using polycrystals prepared by recrystallization of extruded single crystals.

A comparison with results from single crystals [6] leads to the following conclusions:

1.  $T_{\text{bd}}$  becomes smaller with decreasing ionicity of the material, i.e. in the given series from NaCl to AgCl.
2.  $T_{\text{bd}}$  does not correlate with the temperature  $T_0^{(100)}$  at which cubic slip becomes more difficult.

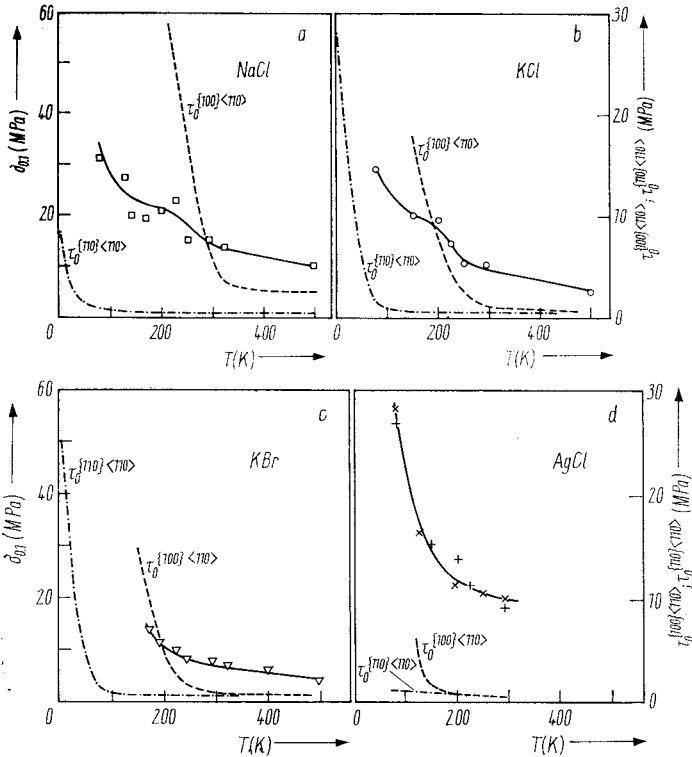


Fig. 3. a) to d) Temperature dependence of the flow stress of polycrystalline ionic solids at 0.1% strain; + and x symbols refer to compression and tension, respectively. The dashed lines represent the temperature dependence of the critical resolved shear stresses for slip on {110} and {100} planes

Table 2

Comparison of the brittle-ductile transition temperature ( $T_{bd}$ ), the temperature at which slip of screw dislocations on {110} becomes wavy at the beginning of deformation ( $T_w$ ), and the temperature below which slip on {100} becomes difficult ( $T_0^{(100)}$ )

	NaCl	KCl	KBr	AgCl		
$T_{bd}$ (K)	550	450	450	80		
$T_w$ (K)	630 [7]	570 [8]	470	420	370	100
$T_0^{(100)}$ (K)	530 [9]	320	250	230	215	165

3. There is a correlation between  $T_{bd}$  and  $T_w$ , the temperature at which the {110} slip mode changes from planar to wavy. This effect is demonstrated in Fig. 5 and 6. At low temperatures the slip bands of screw dislocations on {110} planes are straight, while above a certain temperature slip becomes wavy. Cubic slip bands by screw dislocations are wavy at all investigated temperatures (Fig. 7 and 8). The preferred cross slip plane for screw dislocations on {100} is the {110} plane. Just the opposite is generally observed for {110} slip.

4. The change of the slip mode does not generally occur at the same temperature at which secondary slip becomes easy ( $T_0^{(100)} \approx T_w$ ) contrary to what has been stated so far [2, 11].

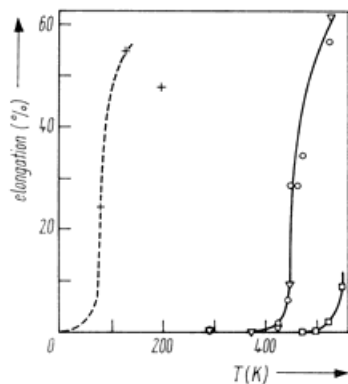


Fig. 4. Influence of temperature on the ductility of polycrystalline ionic solids deformed in tension.  $\square$  NaCl,  $\circ$  KCl,  $\nabla$  KBr,  $+$  AgCl

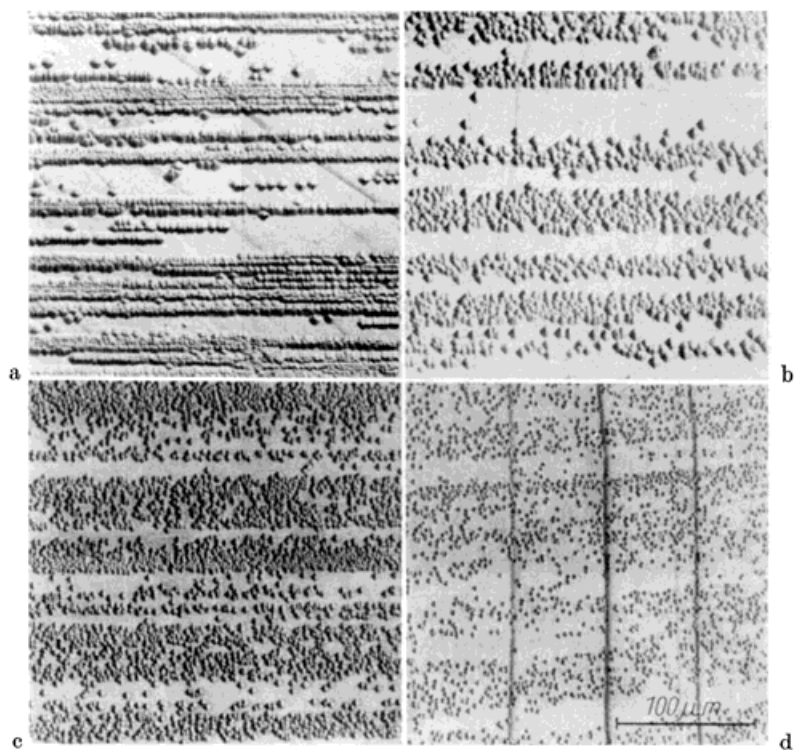


Fig. 5. Influence of temperature on the slip band structure of screw dislocations on  $\{110\}$ . The KCl crystals ( $\approx 56$  ppm  $\text{Sr}^{2+}$ ) have been deformed in  $\langle 100 \rangle$  orientation. a)  $T = 77$ , b) 293, c) 423, d) 473 K

## 4. Discussion

### 4.1 Role of the plastic anisotropy

The increase of the critical flow stress of polycrystalline ionic solids below  $T_0^{\{100\}}$  shows clearly that the plasticity of these materials is determined by the activation of cubic slip as required by the von Mises criterion. It is not so clear to which extent the  $\{100\} \langle 110 \rangle$  systems contribute to the total plastic strain. The step-like increase of

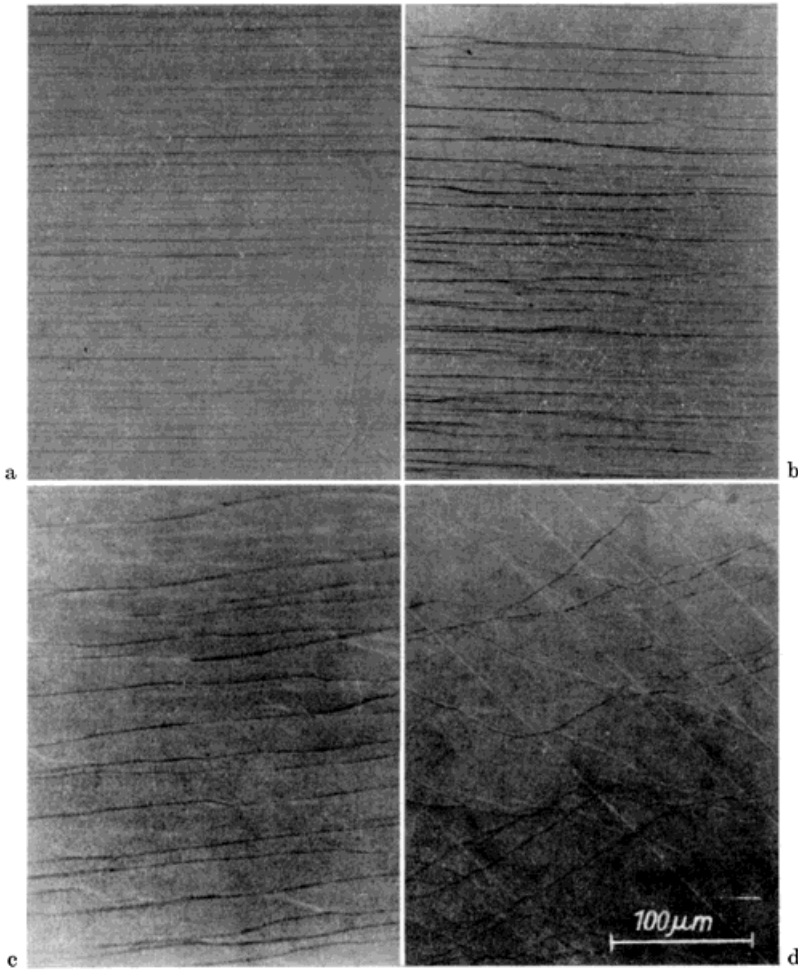


Fig. 6. Change of slip lines of screw dislocations on  $\{110\}$  with temperature. The  $\langle 100 \rangle$  oriented AgCl crystals ( $\approx 100$  ppm  $\text{Ca}^{2+}$ ) have been deformed to 3% strain. The straight slip lines in d) have been produced by edge dislocations on an oblique  $\{110\} \langle 110 \rangle$  system (observation plane nearly  $\{100\}$ ). a)  $T = 77$ , b) 140, c) 200, d) 293 K

$\sigma_{0.1}(T)$  below  $T_0^{\{100\}}$  also observed for LiF [12] and MgO [13], indicates that at low temperatures the plasticity is mainly due to the activation of  $\{110\} \langle 110 \rangle$  slip. This may be caused by the strong  $[100]$  fibre texture of the polycrystalline alkali halides used. For AgCl, on the other hand, which shows the  $[100] [111]$  double fibre texture  $\{100\}$  slip is necessary in polycrystalline deformation and this is evident in the strong rise of  $\sigma_{0.1}(T)$  below  $T_0^{\{100\}}$ . Brittle behaviour is to be expected, when the stress to activate the required independent slip systems is higher than the cleavage stress. This consideration assumes, however, homogeneity of slip and neglects the banded nature of the slip process. The effect of the slip mode on the plasticity of polycrystalline ionic solids will be discussed next.

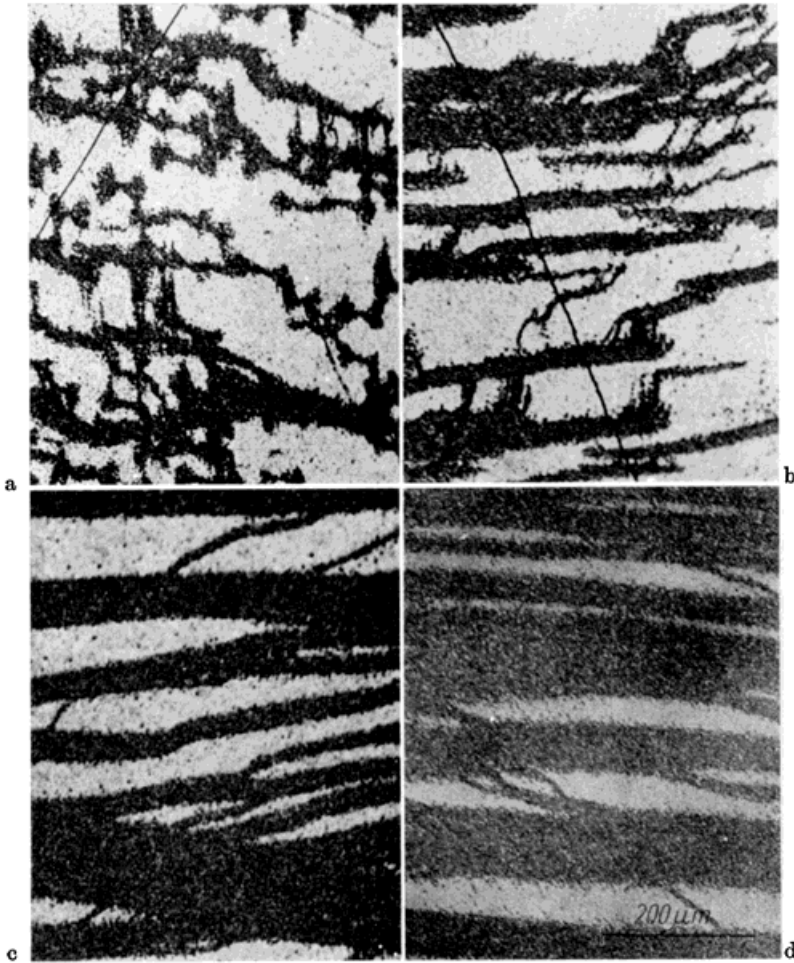


Fig. 7. Influence of temperature on the slip band structure of screw dislocations on  $\{100\}$ . The NaCl crystals ( $\approx 110$  ppm  $\text{Sr}^{2+}$ ) have been deformed in  $\langle 557 \rangle$  orientation. a)  $T = 293$ , b) 323, c) 373, d) 473 K

#### 4.2 Slip mode and fracture

The investigation of slip bands of single crystals deformed on  $\{110\}$  and  $\{100\}$  planes, respectively, showed that the slip mode changes with temperature and the Burgers vector of the dislocations relative to the surface. While slip bands of screw dislocations gliding on  $\{100\}$  are always wavy, those of screws on  $\{110\}$  are straight below a temperature  $T_w$ . The disappearance of planar slip on  $\{110\}$  is correlated with the brittle-ductile transition. This observation can be qualitatively explained by crack nucleation theories.

According to Stroh [14] crack nuclei can be formed by dislocation pile-ups at barriers like grain boundaries. This prediction has been confirmed on MgO [15] and NaCl [11]. Cottrell [16] considers as crack nuclei the coalescence of dislocations on intersecting slip planes to sessile superdislocations forming a large dilated volume below the slip plane. Possible dislocation reactions in ionic crystals result from the inter-

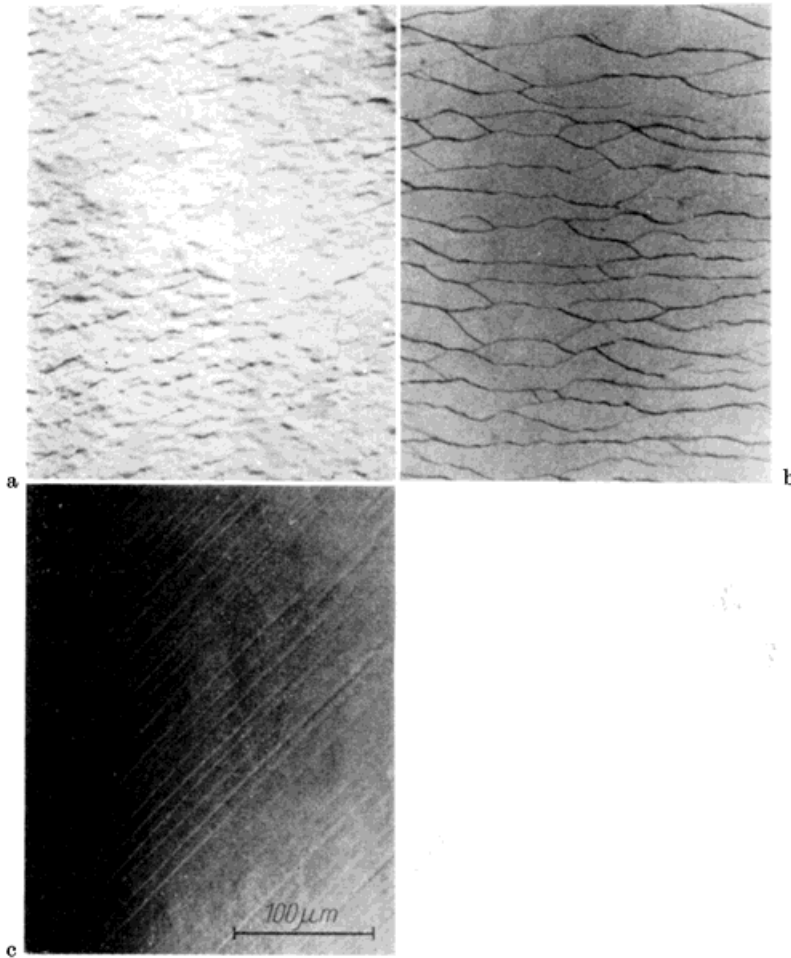


Fig. 8. Change of the slip lines of screw dislocations on  $\{100\}$  with temperature. The  $\langle 557 \rangle$  oriented AgCl crystals ( $\approx 10$  ppm  $\text{Ca}^{2+}$ ) have been deformed to 3% strain. c) shows the slip lines of the edge dislocations (observation plane nearly  $\{110\}$  in c)). a)  $T = 150$ , b) 293, c) 293 K

section of orthogonal and oblique  $\{110\}$  slip planes [17]. Cottrell cracks have been observed in MgO [17]. These pile-up induced crack nucleation mechanisms become more important at low temperatures and in doped crystals because of strongly concentrated slip. Thus it is clear why ionic crystals behave semi-brittle below  $T_w$ . The plastic strains to fracture are much larger in compression because there no tensile stresses exist to open microcracks. Above  $T_w$  pile-ups are dispersed by cross slip and the crystals behave more ductile. We assume here that  $T_w$  lies below the temperatures where grain boundary sliding and recrystallization contribute to polycrystalline plasticity [11].

In the following the mechanism of the formation of wavy slip bands will be discussed in detail. It is assumed that wavy slip of screw dislocations results in two steps:

1. Cross slip of dissociated dislocation segments,
2. motion of edge dislocation jogs produced on the cross slip plane (QSP).



For  $\{100\} \langle 110 \rangle$  slip the preferred QSP is  $\{110\}$ . Slip on this plane is always easier than on the cubic plane. Furthermore recent calculations of stacking fault energies [18] indicate that screw dislocations on  $\{100\}$  are even less dissociated than on  $\{110\}$ . Therefore, it seems clear why slip lines of screw dislocations gliding on  $\{100\}$  are always wavy in the investigated temperature range.

For  $\{110\} \langle 110 \rangle$  slip at the beginning of stage III of the stress-strain curve Matucha [19] has observed cross slip steps from  $\{110\}$  to  $\{111\}$  and further to  $\{100\}$ . Slip of edge dislocations on cubic planes is more difficult than slip on  $\{110\}$  due to the higher Peierls barrier as well as due to the electrostatic interaction with aliovalent impurities [3]. In addition screw dislocations gliding on  $\{110\}$  are hindered from cross slipping by their expected dissociation on this plane. This explains, why slip bands of screw dislocations on  $\{110\}$  at the beginning of deformation are wavy only above a temperature  $T_w$ . In the following it will be checked which of the two above-mentioned processes determines  $T_w$ .

Assuming cubic slip to be stopping just below  $T_0^{(100)}$  then wavy slip should be observed around  $T_0^{(100)}$ , if the motion of edge dislocations is the controlling process. It is evident from Table 2 that  $T_0^{(100)}$  and  $T_w$  agree only approximately for LiF and AgCl whereas for the other materials  $T_w$  lies significantly higher. In these cases cubic slip obviously cannot control the waviness. For  $\{110\} \langle 110 \rangle$  slip it is well known that cross slip determines the onset of stage III of the stress-strain curve [19]. This is supported by the dependence of  $\tau_{III}$  on temperature and hydrostatic pressure as well as by the cross slip sequence  $\{110\} \rightarrow \{111\} \rightarrow \{100\}$  [20]. If  $T_w$  is determined by cross slip then this temperature must agree with that at which  $\tau_{III}$  decreases to the CRSS  $\tau_0$ . According to Hesse [21]  $\ln \tau_{III}$  of NaCl decreases linearly with increasing temperature as follows from the cross slip theory of Seeger et al. [22]. Extrapolation of  $\tau_{III}(T)$  to high temperatures yields  $\tau_{III} = \tau_0$  at  $T = 620$  K. This value agrees within 10% with  $T_w$  of NaCl (Table 2). For the other materials  $\tau_{III}(T)$  is not known.

Extrapolation of Hesse's  $\tau_{III}(T)$  data to  $T = 0$  K yields  $\tau_{III_0}/\mu \approx 3 \times 10^{-3}$  ( $\mu$  shear modulus). This value is smaller than the reduced Peierls stress  $\tau_p^{(100)}/\mu \approx 10^{-2}$  for NaCl [4]. Thus there is the possibility that below a certain temperature  $\tau_{III}$  may be determined by the conservative motion of edge dislocation jogs on  $\{100\}$ . The expected course of  $\ln(\tau_{III}(T)/\mu)$  is presented schematically in Fig. 9. However, a prediction of the transition temperature is not possible because  $\tau_{III}$  is connected with  $\tau_0^{(100)}$  by an unknown effective Schmid factor with respect to internal stress (the Schmid factor of the applied stress for  $\{100\}$  slip is zero for  $\langle 100 \rangle$  oriented crystals). As  $\tau_0^{(100)}$  should not depend on hydrostatic pressure  $p$  while cross slip becomes easier,  $\tau_{III}$  decreases with increasing  $p$ , it can be qualitatively understood why  $\tau_{III}$  may remain constant above a certain temperature [23].

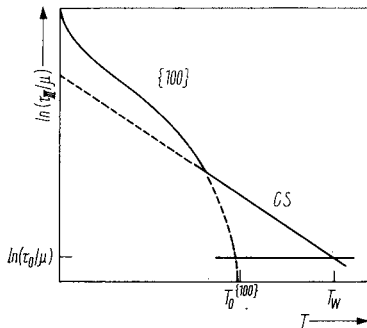


Fig. 9. Schematic representation of the expected temperature dependence of  $\tau_{III}/\mu$  when cross slip of screw dislocations (CS) or the motion of edge dislocation jogs on  $\{100\}$  dominate

Finally it should be mentioned that our analysis of the strength and ductility of pure polycrystalline ionic solids may be relevant for deformation problems of rock salt domes considered for the storage of nuclear waste in geological formations.

### *Acknowledgements*

Thanks are due to Dr. P. Welch (TU Clausthal) for the texture analysis and due to the DFG for support.

### **References**

- [1] R. VON MISES, *Z. angew. Mech.* **8**, 161 (1928).
- [2] G. W. GROVES and A. KELLY, *Phil. Mag.* **8**, 877 (1963).
- [3] W. SKROTZKI and P. HAASEN, *J. Physique*, in the press.
- [4] T. SUZUKI, W. SKROTZKI, and P. HAASEN, *phys. stat. sol. (b)* **103**, 763 (1981).
- [5] G. FROMMEYER, W. SKROTZKI, and P. WELCH, in preparation.
- [6] W. SKROTZKI, Ph. D. Thesis, Göttingen 1980.
- [7] H. L. FOTEDAR and T. G. STOEBE, *Phil. Mag.* **23**, 859 (1971).
- [8] B. H. KEAR, C. E. SILVERSTONE, and P. L. PRATT, *Proc. Brit. Ceram. Soc.* **6**, 269 (1966).
- [9] Z. LIU, Diploma Work, Göttingen, in preparation.
- [10] N. S. STOLOFF, D. K. LEZIUS, and T. L. JOHNSTON, *J. appl. Phys.* **34**, 3315 (1963).
- [11] R. J. STOKES, *Proc. Brit. Ceram. Soc.* **6**, 189 (1966).
- [12] D. W. BUDWORTH and J. A. PASK, *J. Amer. Ceram. Soc.* **46**, 560 (1963).
- [13] C. O. HULSE, S. M. COPLEY, and J. A. PASK, *J. Amer. Ceram. Soc.* **46**, 317 (1963).
- [14] A. N. STROH, *Proc. Roy. Soc. A* **223**, 404 (1954); **A 232**, 548 (1955); *Adv. Phys.* **6**, 418 (1957).
- [15] R. J. STOKES, T. L. JOHNSTON, and C. H. LI, *Phil. Mag.* **3**, 718 (1958).
- [16] A. H. COTTRELL, *Trans. MS AIME* **212**, 192 (1958).
- [17] A. S. KEH, J. C. M. LI, and Y. T. CHOU, *Acta metall.* **7**, 694 (1959).
- [18] P. W. TASKER and T. J. BULLOUGH, *Phil. Mag. A* **43**, 313 (1981).
- [19] K.-H. MATUCHA, *phys. stat. sol.* **9**, 209 (1965).
- [20] P. HAASEN, *J. Physique* **35**, C7-167 (1974).
- [21] J. HESSE, *phys. stat. sol.* **9**, 209 (1965).
- [22] A. SEEGER, R. BERNER, and H. WOLF, *Z. Phys.* **155**, 247 (1959).
- [23] E. ALADAG, L. A. DAVIS, and R. B. GORDON, *Phil. Mag.* **21**, 469 (1970).

*(Received May 4, 1981)*