Shock Wave Compression of NaCl Single Crystals observed by Flash-X-Ray Diffraction

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Flash-x-ray-diffraction patterns (FXD) with an exposure time of 4 ns of NaCl single crystals compressed by plane shock waves are obtained at pressures of about 30 kbar. From the diffraction patterns the compression is determined and compared with Hugoniot data. During shock load the lattice shows an uniaxial compression. While in case of measurements at the free surface an observation time of only a few nanoseconds is available, this experimental set-up allows an observation time of two microseconds.

I. Introduction

The problem of getting flash-x-ray-diffraction patterns (FXD) from shock loaded crystals has already been discussed many years ago [1, 2, 3]. The development of high performance flash-x-ray tubes in recent years, however, has allowed to take up the problem once more.

Johnson and coworkers [4] found from FXD, that for a shock loaded single crystal of LiF the crystal is compressed isotropically, and that the crystal orientation is preserved. The investigation was made at pressures of about 300 kbar. At about 240 kbar deviations from the isotropic compression have been found by Kondo, Sawaoka, and Saito [5]. Discussing experiments at about 10 kbar Jamet and Thomer [6] pointed out the possibility of non-isotropic compression. The non-isotropic compression may be expected with regard to the material strength in this low pressure range.

In the experiments the following considerable difficulties arise:

1. An x-ray source of high intensity with a pulse duration of only a few nanoseconds must be used.
2. The arrival of the shock wave at the crystal surface under investigation and the time of the x-ray pulse must be synchronized with an accuracy of 10 ns.
3. The incident shock wave is disturbed by rarefaction waves from the surface of the crystal.

Egorov and coworkers [7] partially avoid these difficulties by using a plane plate which covers the surface under investigation and is transparent to x-rays. By this, rarefaction waves from the surface

are avoided, and the time of undisturbed observation is extended. Till now flash-x-ray-diffraction patterns obtained by such an arrangement have been evaluated only qualitatively.

Following Egorov and coworkers, in this paper we discuss an experiment in which the deformation of a single crystal of NaCl is determined quantatively by FXD and compared to Hugoniot data obtained from the same experiment.

II. Experimental Conditions

a) A General Plan of the Experiments

The experimental set-up is shown schematically in Figure 1. In a thin plate of a NaCl single crystal (dimensions $(20 \times 10 \times 1)$ mm^3) a plane shock wave is generated by impact of a plane plate at the surface I. At the surface B of the crystal the distance of the atomic planes under shock load is measured by FXD. To prevent disturbances by rarefaction waves this surface is covered by a plate with a high transmission factor for x-rays.

By means of K-x-ray pulses time resolved Bragg-reflections from the crystal are recorded on film. In each experiment a diffraction pattern is taken after a certain delay time measured from the moment of impact of the flyer-plate on the surface of the crystal. In a series of experiments the time dependence of interplanar spacing under shock loading is determined.

b) Generation of Plane Shock Waves

A plane plate of PMMA (dimensions $(100 \times 20 \times 2)$ mm^3) is accelerated by an exploding copper foil to a velocity of 1 mm/µs [8, 9]. The flyer-plate is adjusted parallel to the surface of the crystal by spacing pieces of 4.5 ± 0.005 mm. Thus during impact the plate is sufficiently parallel to the
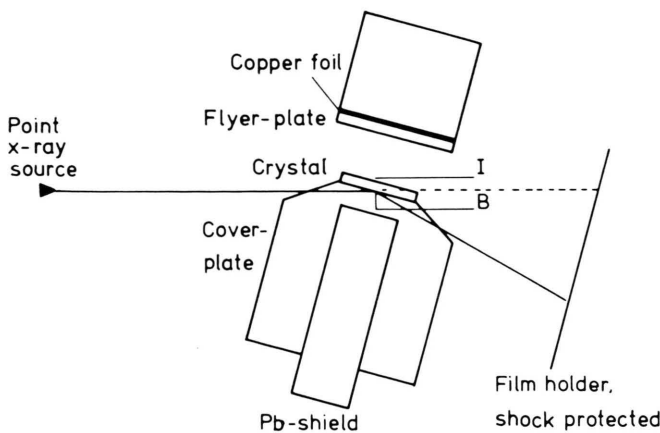
Fig. 1. Experimental set-up.

(100)-surface. Deviations from the exact parallelism may arise from non uniform explosion of the foil. Experiments with such deviations can be detected clearly and eliminated [10]. Details of the impact arrangement and the crystal mounting are shown in Figure 2. Two pin contacts mounted closely to the crystal monitor the velocity of the flyer-plate and its arrival on the crystal surface.
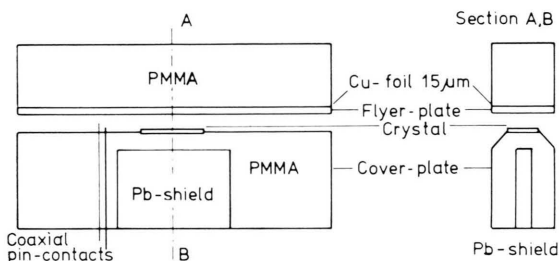

Fig. 2. Mounting of the Crystal.

c) Recording X-Ray-Diffraction Patterns

Unfiltered K_α-radiation of Molybdenium is used to determine the distance of the atomic planes by Bragg-reflections at the surface B of the crystal. The experimental set-up (Fig. 1) is a modification of Seeman's edge-spectrograph. In our case, however, the edge disturbs the reflection of the shock wave at the surface under investigation. Therefore the Pb-shielding is separated by about 5 mm from the surface B of the crystal. Moreover the possibility is given to observe the shock wave, transmitted in the cover plate, by optical methods.

The flash-x-ray equipment encompasses an x-ray tube constructed by the authors; the details of the device are described elsewhere [11]. It produces an x-ray pulse with a half amplitude pulse duration of 4 ns. Figure 3 shows diffraction patterns obtained by an edge spectrograph (a) and by the modified spectrograph described above (b). The patterns are recorded on film, sticking in a shock-proof film holder and covered by intensifying screens on both sides. In each experiment one half of the film is exposed by a diffraction pattern with the crystal at rest, the other half by a pattern with the crystal under shock load.

d) Streak Photography of the Shock Wave Propagation

The shock wave transmitted from the crystal into the PMMA cover plate is observed by high
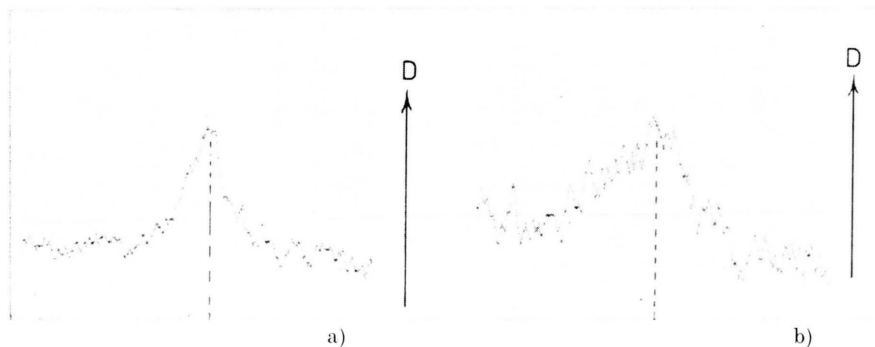

Fig. 3. Diffraction patterns with an exposure time of 4 ns obtained by a) edge spectrograph, b) modification of the edge spectrograph. (Enlargement factor of the photometric density measurement: 20).
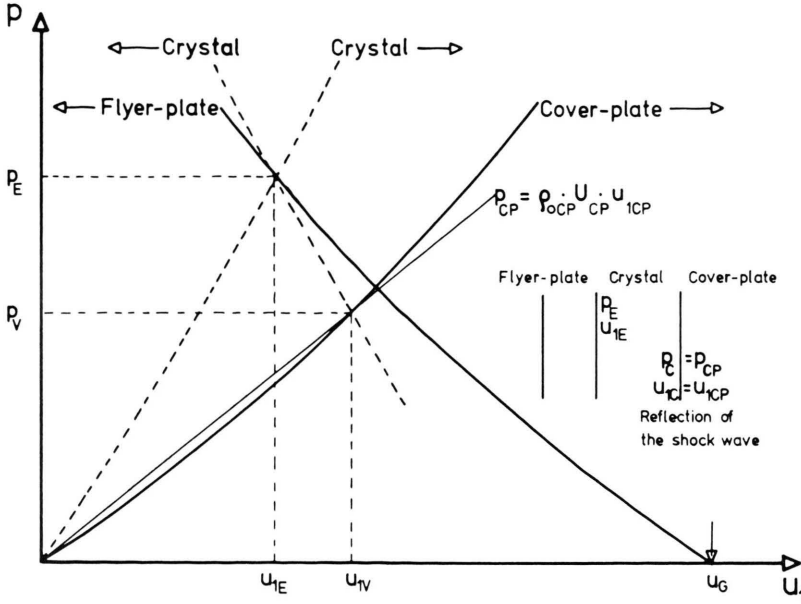
Fig. 4. Pressure and particle velocity behind the incident and the reflected shock wave.

speed streak photography. The streak photographs show the impact of the flyer plate on the crystal, the propagation of the shock wave in the crystal and in the cover plate. Details of the arrangement are given by Schulte [10].

III. Theoretical Description of the Experiments

a) General Considerations

The Hugoniot relations can be applied to determine the state of the crystal behind the shock front. For a plane wave moving with the velocity U into the undisturbed medium these relations are

$$\varrho_0 U = \varrho_1 (U - u_1),\tag{1}$$

$$p_1 - p_0 = \varrho_0 U u_1,\tag{2}$$

$$E_1 - E_0 = (1/2)(V_0 - V_1)(p_1 + p_0),\tag{3}$$

p being the pressure, ϱ the density, u_1 the particle velocity, $V = 1/\varrho$ the specific volume and E the specific energy; the subscripts 0 and 1 stand for the regions in front of and behind the shock front, respectively [12]. A combination of Eqs. (1) and (2) gives:

$$p_1 - p_0 = \varrho_0 U^2 (1 - V_1/V_0).\tag{4}$$

If the conditions in front of the shock wave are known, measurements of U and V_1 are necessary for the determination of the pressure. In the case of a crystal the ratio V_1/V_0 is obtained by x-ray diffraction patterns. Since in our experiment the

interplanar spacing is measured only in the direction of the shock propagation, the ratio of V_1/V_0 is given by

a) $V_1/V_0 = d_1/d_0$ or   b) $V_1/V_0 = (d_1/d_0)^3$. (5)

d = spacing of the atomic planes. Equations (5a) and (5b) are valid for a uniaxial or isotropic compression of the lattice, respectively. The type of compression has to be determined by further measurements.

b) Shock Compression of the Crystal

The pressure p_E and the particle velocity u_{1E} of the shock wave propagating in the crystal are given in the $p - u_1$ plane by the point of intersection of the Hugoniot curve of the flyer plate and the Hugoniot curve of the crystal [12] (Figure 4). After reflection of the shock wave at the surface B of the crystal the pressure p_{CP} and velocity u_{1CP} in the cover plate are given by the point of intersection of the reflected Hugoniot curve of the crystal and the Hugoniot curve of the cover plate. With the well known Hugoniot curve of PMMA [8, 13, 14, 15], p_{CP} is determined by a measurement of U_{CP}:

$$p_{CP} = \varrho_{0CP} U_{CP} u_{1CP}.\tag{6}$$

Because the pressures p_C and p_{CP} as well as the velocities u_{1C} and u_{1CP} after reflection are equal, the pressure in the crystal may be written

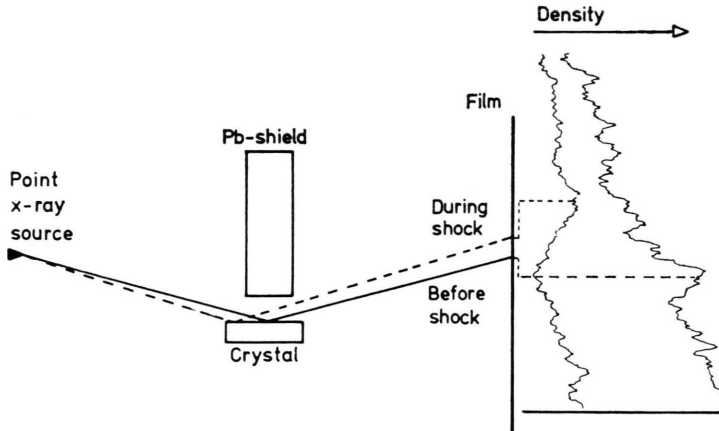$$p_C = \varrho_{0C} u_{1CP}^2 / (1 - \varrho_{0C}/\varrho_{1C}).\tag{7}$$

Fig. 5. Reflex-shift measurement by photometry.

The values in the crystal and in the cover plate are marked by the subscripts C and CP, respectively. The model of compression (uniaxial or isotropic) is checked by Equation (7).

IV. Evaluation and Interpretation of Experimental Results

a) The Shift of the Reflex

The positions of the Bragg-reflexes for the shock loaded crystal and the crystal at rest are determined photometrically. The shift of the Bragg-reflex is given by the difference between the two values. A typical example of an evaluation is given in Figure 5. The reflex under shock loading is broadened in comparison to the reflex with the crystal at rest. The shift of the reflex as a function of time is given in Figure 6. For a time of 500 ns the shift is very low. It then increases to a nearly constant value. This shift is the sum of two components: 1) a shift caused by the change of inter-

planar spacing in the crystal under shock load; 2) a shift caused by the motion of the interface between crystal and cover plate with the velocity u_{1CP}. The first component can be determined quantitatively (Figure 7). A point x-ray source R is positioned at a distance z from the point of reflection A. Θ_0 is the angle of reflection for the atomic planes parallel to the surface of the crystal. The film is positioned at a distance a from the point A. Under shock loading the distance of the atomic planes and thereby the angle of reflection changes to Θ. The reflex being due to the compressed state is to be found on the film in a distance Δr_1 from the normal reflex. From Fig. 7 can be seen

$$\Delta r_1 = r_2 - r_1 = a(\tan \Theta - \tan \Theta_0). \qquad (8)$$

The point of reflection A on the crystal surface is shifted by a distance of x_1 and thereby the reflex on the film by

$$\Delta r_w = z(\cos \Theta_0 \tan \Theta - \sin \Theta_0). \qquad (9)$$
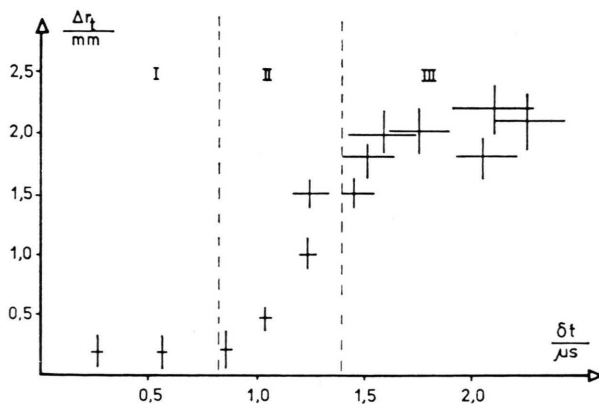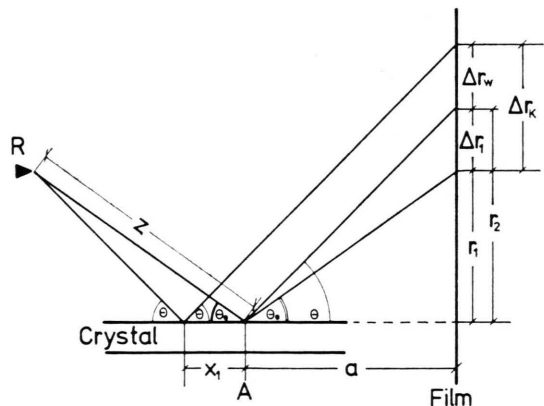

Fig. 6. Shift of the reflex vs. time.

Fig. 7. Shift of the reflex by change of the interplanar spacing and the motion of the reflecting surface.

The shift of the reflex being due to the shock compressed state is

$$\Delta r_k = \Delta r_1 + \Delta r_w \,. \tag{10}$$

During transit of the shock wave through the interface between crystal and cover plate this plane is moving with the velocity u_{1CP}. An additional shift Δr_u of the reflex on the film is the result. As the velocity u_{1CP} is not constant, Δr_u is given by

$$\Delta r_u = \int_0^{\delta t} 2\, u_{1CP}\, \mathrm{d}t \,. \tag{11}$$

The total shift of the reflex due to the crystal under shock load as compared to the reflex due to the crystal at rest is

$$\Delta r_t = \Delta r_k + \Delta r_u \,. \tag{12}$$

The angle of reflection Θ is given by

$$\Theta = \arctan\left[(\Delta r_t + a \tan \Theta_0 + z \sin \Theta_0 - 2 \int_0^{\delta t} u_{1CP}\, \mathrm{d}t) / (a + z \cos \Theta_0)\right]. \tag{13}$$

From the Bragg's law (first order) d_0 and d_1 are given by

$$d_0 = \lambda/(2 \sin \Theta_0); \quad d_1 = \lambda/(2 \sin \Theta) \,. \tag{14}$$

Thereby the interplanar spacing under shock load perpendicular to the shock front is determined.

b) The Compression of the Crystal

Under shock load in an NaCl single crystal a two-wave structure develops; the crystal is compressed by an elastic wave up to the yield point and by a following plastic wave to the peak pressure. The region I in Fig. 6 corresponds to the compression in the elastic wave, the region II to the rise of compression, and the region III to the constant compression by the elastic-plastic wave. About 800 ns after reaching the peak pressure the x-ray reflex disappears.

After the reflection of the wave at the surface B the pressure in the crystal is equal to the pressure in the cover plate. Therefore the pressure in the crystal may be determined in two different ways: on the one hand from the velocity of the transmitted shock wave according to Equation (6); the pressure range is marked in Fig. 8a in the Hugoniot curve of NaCl; on the other hand from the compression of the crystal following Eqs. (5) and (7) and measured values of region III in Figure 6. Only on the assumption of uniaxial compression of
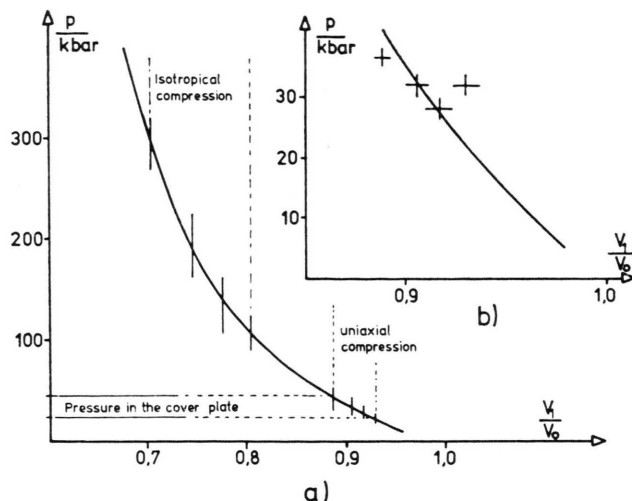

Fig. 8. Compression of the crystal in comparison to Hugoniot data. (Hugoniot data from L. V. Al'shuler et al. [16] and E. Schulte [10]).

the lattice according to Eq. (5) and (7) the pressure values obtained agree with those obtained from (6). These pressure values for an uniaxial compression of the lattice are plotted once more in an experimentally determined Hugoniot curve of NaCl in Figure 8 b [10, 16].

c) Discussion of the Results

The profile of the Bragg-reflex under shock load is broadened compared to the normal profile, and the intensity of the reflex is smaller (Figure 6). The intensity however is clearly higher than in the case of powder patterns. An explanation for the broadening may be the great increase in dislocation density in the elastic wave [18] and the appearance of mosaic structure in the crystal [5]. The increase in temperature during shock and the corresponding broadening of the reflex can be neglected in the pressure range considered.

A phase transition in the NaCl crystal under shock load and thereby a shift of the x-ray reflex may be excluded, because phase transitions are expected to take place only at about 300 kbar [17].

A comparison of x-ray measurements to Hugoniot data shows that a uniaxial compression of the lattice perpendicular to the shock front takes place. The disappearance of the x-ray reflex at about 800 ns after the peak pressure is reached indicates additional strong disturbances of the lattice, probably caused by plastic deformation. The influence of

rarefaction waves on the disappearance of the x-ray reflex may be excluded. The further transient development of the lattice cannot be observed, because the experiment allows only an observation time of two microseconds.

V. Conclusions

The existence of crystalline order behind the shock front, sufficient for the registration of x-ray-diffraction patterns, is confirmed. This order is maintained for a time of about 1 µs.

Since the distance of the atomic planes is measured in one direction only, it is impossible to describe the transient phenomena in the unit cell completely. Therefore an explanation cannot yet be given for the following problems: Isotropic compression in case of high pressure [4], deviations from the isotropic compression [5], and uniaxial compression in the low pressure range [6] as observed in this work. Final clearness will be gained, if the distance of different atomic planes of a crystal under shock load could be determined by FXD simultaneously.

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