

Laue diagrams: investigating the lattice structure of monocrystals

Objects of the experiment

- Evaluating the *Laue* diagrams of an NaCl and an LiF monocrystal.
- Investigating the symmetry and lattice structure of both crystals.

Principles

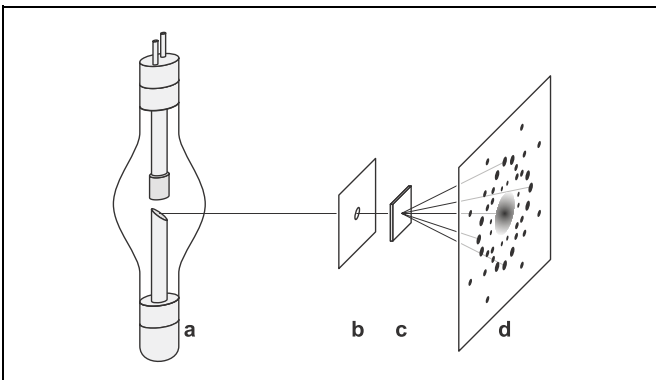


Fig. 1 Scheme of the setup for taking a *Laue* diagram of a monocrystal
a X-ray tube
b Collimator
c Crystal
d X-ray film

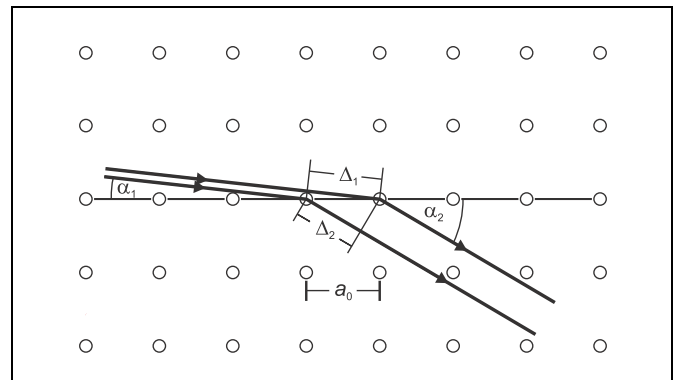


Fig. 2 Two-dimensional representation for calculating the difference of path $\Delta = \Delta_1 - \Delta_2$ between two neighbouring X-rays which are diffracted at a row of points in a cubic crystal.

In 1912, *Max von Laue* proposed to provide evidence for the supposed wave character of X-rays by diffraction at crystals. *Friedrich* and *Knipping* took up his proposal and exposed a crystal to a collimated ray from an X-ray tube. On a photographic plate behind the crystal they observed – as expected – discrete reflections. For the first time they also confirmed the spatial lattice structure of crystalline substances with this experiment.

Laue condition:

In his interpretation of these findings, *von Laue* considered the crystal to be a lattice built up of three groups of one-dimensional equidistant rows of points. When an X-ray is diffracted at a row of points,

$$\Delta = \Delta_1 - \Delta_2 = a_0 \cdot \cos \alpha_1 - a_0 \cdot \cos \alpha_2 \quad (I)$$

a_0 : distance between the points

α_1 : angle between the incoming X-ray and the row of points

α_2 : angle between the diffracted X-ray and the row of points

is the difference of path of partial rays scattered at two neighbouring points (lattice elements, see Fig. 2). There is construc-

tive interference between the partial rays if Δ is an integer multiple of the wavelength λ . This condition has to be fulfilled in all three spatial directions.

In a cubic crystal, the rows of points associated with the three spatial directions are all perpendicular to each other the distance a_0 between the points always being the same. The *Laue* condition for constructive interference therefore reads:

$$a_0 \cdot \cos \alpha_1 - a_0 \cdot \cos \alpha_2 = h \cdot \lambda$$

$$a_0 \cdot \cos \beta_1 - a_0 \cdot \cos \beta_2 = k \cdot \lambda$$

$$a_0 \cdot \cos \gamma_1 - a_0 \cdot \cos \gamma_2 = l \cdot \lambda \quad (II)$$

with integer values of h, k, l

Here α_1, β_1 and γ_1 are the angles between the incoming ray and the rows of points whereas α_2, β_2 and γ_2 are the angles between the diffracted ray and the rows of points. The quantities h, k, l are called *Laue* or extended *Miller* indices. As they are small integer numbers, the *Laue* condition cannot be fulfilled for arbitrary wavelengths λ , but only for particular ("appropriate") ones.

Apparatus

1 X-ray apparatus	554 811
or	
1 X-ray apparatus	554 812
1 X-ray film holder	554 838
1 filmpack 2 (X-ray film)	554 892
1 LiF crystal for Laue diagrams	554 87
1 NaCl crystal for Laue diagrams	554 88

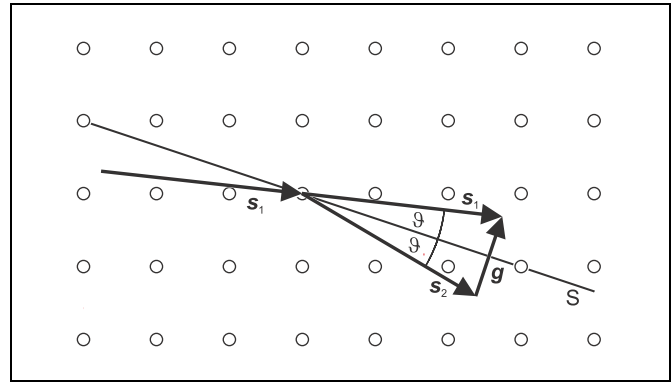


Fig. 3 Geometric connection between the unit vectors s_1 and s_2 and the vector $g = \lambda G$.

The unit vectors

$$s_1 = (\cos \alpha_1, \cos \beta_1, \cos \gamma_1)$$

and

$$s_2 = (\cos \alpha_2, \cos \beta_2, \cos \gamma_2) \quad (III)$$

are frequently introduced. They point in the direction of the incoming X-ray and the outgoing X-ray, respectively. The *Laue* condition (II) then takes the form

$$s_1 - s_2 = \lambda \cdot G \text{ with } G = (h, k, l) \cdot \frac{1}{a_0} \quad (IV)$$

G is called the vector of the reciprocal lattice. The *Laue* condition is fulfilled if h , k and l are integers.

Bragg condition:

As s_1 and s_2 are unit vectors, they have equal magnitudes. Therefore the vector G is perpendicular to the bisectrix S

between the incoming and the diffracted X-ray (see Fig. 3). From this

$$\lambda \cdot |G| = |s_1 - s_2| = 2 \cdot \sin \vartheta$$

follows and after inserting the magnitude of G

$$\lambda = 2 \cdot \sin \vartheta \cdot \frac{a_0}{\sqrt{h^2 + k^2 + l^2}} \quad (V)$$

Eq. (V) is identical with *Bragg's* law if the spacing of the lattice planes is

$$d = \frac{a_0}{\sqrt{h^2 + k^2 + l^2}} \quad (VI)$$

From *Bragg's* point of view the crystal lattice thus consists of a set of lattice planes that have the spacing d (see Fig. 4 and experiment P6.3.3.1). These lattice planes are parallel to the bisectrix S and perpendicular to the vector G of the reciprocal lattice.

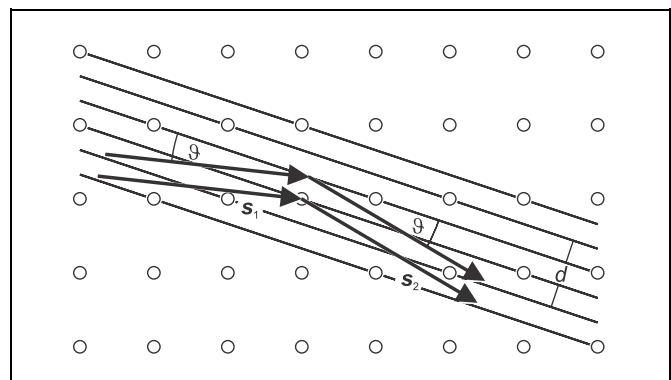
The conditional equation of the set of lattice planes in the co-ordinate system spanned by the crystal axes reads

$$x \cdot h + y \cdot k + z \cdot l = m \cdot a_0 \quad (VII)$$

m : running number in a set of lattice planes

h^{-1} , k^{-1} and l^{-1} are the axis sections of the first lattice plane ($m = 1$) measured in units of the lattice constant (see Fig. 5). As the indices h, k, l represent the set of lattice planes uniquely, the set is assigned the symbol $(h \ k \ l)$.

Fig. 4 Two-dimensional representation of *Bragg* reflection of X-rays at a set of lattice planes in a cubic crystal. The lattice planes are parallel to the bisectrix S between the incoming and the diffracted X-ray.



Safety notes

The X-ray apparatus fulfils all regulations governing an X-ray apparatus and fully protected device for instructional use and is type approved for school use in Germany (NW 807 / 97 Rö).

The built-in protection and screening measures reduce the local dose rate outside of the X-ray apparatus to less than $1 \mu\text{Sv/h}$, a value which is on the order of magnitude of the natural background radiation.

- Before putting the X-ray apparatus into operation, inspect it for damage and make sure that the high voltage is shut off when the sliding doors are opened (see instruction sheet for X-ray apparatus).
- Keep the X-ray apparatus secure from access by unauthorized persons.

Do not allow the anode of the X-ray tube Mo to overheat.

- When switching on the X-ray apparatus, check to make sure that the ventilator in the tube chamber is turning.

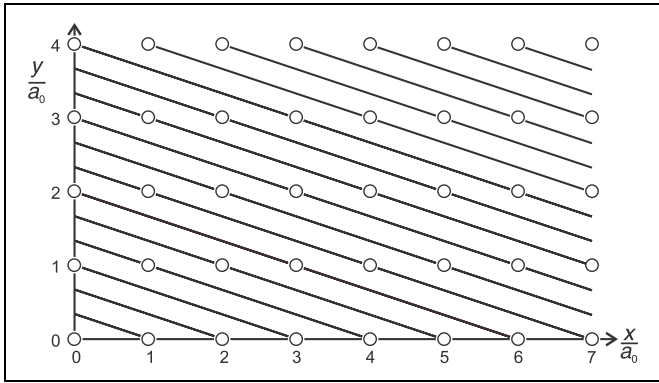


Fig. 5 Two-dimensional representation of the set of lattice planes (1 3 0) in a cubic crystal.

Crystals with NaCl structure:

In the case of crystals with NaCl structure, the condition for constructive interference turns out to be more complicated than in simple cubic crystals because alkali atoms (e.g. Na) and halogenide atoms (e.g. Cl) take turns in a cubic lattice. The spatial lattice is no longer built up of rows of points with the distance a_0 , but it is a series of cubic unit cells with an edge length a_0 (see Fig. 6). Every unit cell contains four alkali atoms with the co-ordinates

$$r_1 = (0,0,0), r_2 = \left(\frac{a_0}{2}, \frac{a_0}{2}, 0\right), r_3 = \left(\frac{a_0}{2}, 0, \frac{a_0}{2}\right), r_4 = \left(0, \frac{a_0}{2}, \frac{a_0}{2}\right)$$

and four halogenide atoms with the co-ordinates

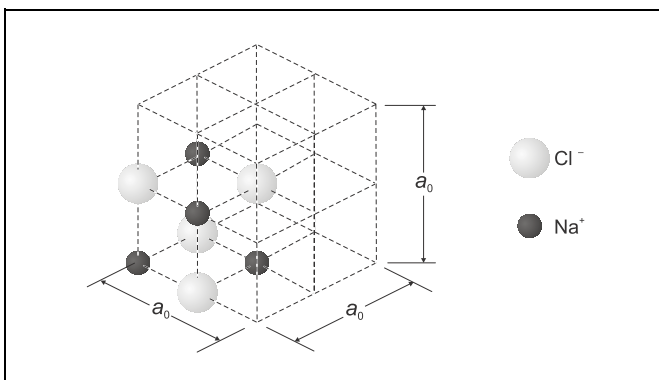
$$r_5 = \left(\frac{a_0}{2}, 0, 0\right), r_6 = \left(0, \frac{a_0}{2}, 0\right), r_7 = \left(0, 0, \frac{a_0}{2}\right), r_8 = \left(\frac{a_0}{2}, \frac{a_0}{2}, \frac{a_0}{2}\right)$$

At each atom of the unit cell the incoming X-ray is scattered, whereby the amplitudes of the scattered partial waves depend on the atomic number of the atom. The differences of path Δ_i of the partial waves can be calculated from the co-ordinates r_i of the atoms:

$$\Delta_i = (\mathbf{s}_1 - \mathbf{s}_2) \cdot \mathbf{r}_i \tag{VIII}$$

The partial waves scattered at the alkali atoms A and the halogenide atoms H interfere to form a common wave that is

Fig. 6 Unit cell of an NaCl crystal



“scattered at the unit cell”. The amplitude of this wave has the form

$$A = A_A + A_H \tag{IX}$$

with

$$A_A = f_A \left(\cos\left(\frac{2\pi}{\lambda}\Delta_1\right) + \cos\left(\frac{2\pi}{\lambda}\Delta_2\right) + \cos\left(\frac{2\pi}{\lambda}\Delta_3\right) + \cos\left(\frac{2\pi}{\lambda}\Delta_4\right) \right)$$

and

$$A_H = f_H \cdot \left(\cos\left(\frac{2\pi}{\lambda}\Delta_5\right) + \cos\left(\frac{2\pi}{\lambda}\Delta_6\right) + \cos\left(\frac{2\pi}{\lambda}\Delta_7\right) + \cos\left(\frac{2\pi}{\lambda}\Delta_8\right) \right)$$

All waves that start from the unit cells interfere constructively if the *Laue* condition (IV) is fulfilled. By inserting (IV) and (VIII) in (IX) one obtains

$$A_A = f_A \cdot (1 + \cos((h+k) \cdot \pi) + \cos((h+l) \cdot \pi) + \cos((k+l) \cdot \pi))$$

and

$$A_H = f_H \cdot (\cos(h \cdot \pi) + \cos(k \cdot \pi) + \cos(l \cdot \pi) + \cos((h+k+l) \cdot \pi)).$$

A short calculation shows that

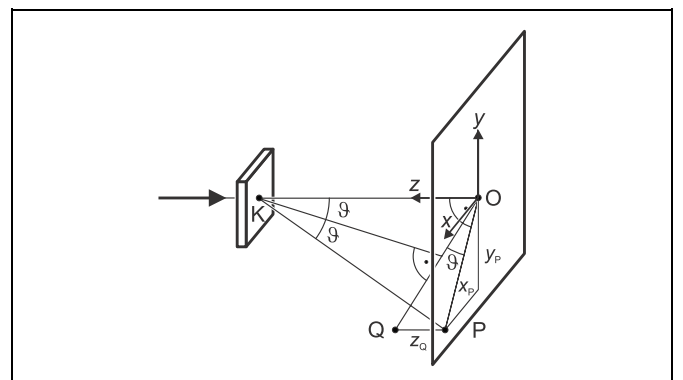
$$A = \begin{cases} 4 \cdot f_A + 4 \cdot f_H, & \text{if } h, k \text{ and } l \text{ even} \\ 4 \cdot f_A - 4 \cdot f_H, & \text{if } h, k \text{ and } l \text{ odd} \\ 0, & \text{if } h, k \text{ and } l \text{ mixed} \end{cases} \tag{X}$$

The amplitudes A of the waves starting from the unit cells thus only are different from zero if all indices h, k, l are even or if they are all odd.

Evaluating a *Laue* diagram:

The object of the evaluation of a *Laue* diagram is to assign the set of lattice planes that causes the scattering to one of the reflections observed on the X-ray film. For this the co-ordinate system is chosen so that its origin O corresponds to the space point of the incoming X-ray on the X-ray film. The X-ray film is perpendicular to the ray, i.e., it lies in the x-y-plane (see Fig. 7). The orientation of the z-axis is opposite to the propagation direction of the X-ray. The X-ray penetrates the flat crystal at the point K; its undiffracted part impinges on the X-ray film at the point O. The part of the X-ray which is scattered at K and which fulfils the *Laue* condition (II) leaves the crystal with an

Fig. 7 Geometrical description of an X-ray that is diffracted at a point K of the crystal and that penetrates the film plane in the point P



angle 2ϑ relative to the direction of the primary ray and impinges on the X-ray film at the point $P = (x_P, y_P, 0)$. Therefore:

$$\tan 2\vartheta = \frac{\sqrt{x_P^2 + y_P^2}}{L} \quad (\text{XI})$$

L : distance between crystal and X-ray film

The direction of the set of lattice planes ($h k l$), which leads to the reflection, is given by the bisectrix of the angle 2ϑ (see Fig. 3). The angle between the perpendicular to the bisectrix which passes O and the straight line OP is ϑ . This perpendicular intersects a parallel to the z -axis through P in the point Q . The vector OQ has the co-ordinates (x_Q, y_Q, z_Q) with

$$\tan \vartheta = \frac{z_Q}{\sqrt{x_Q^2 + y_Q^2}} \quad (\text{XII})$$

and is parallel to the vector G of the reciprocal lattice. Because of $x_P = x_Q, y_P = y_Q$ and Eq. (XI),

$$z_Q = \sqrt{x_Q^2 + y_Q^2 + L^2} - L \quad (\text{XIII})$$

The crystals used in this experiment are cut parallel to the (100) plane. Their crystal axes thus coincide with the laboratory co-ordinate system. From the fact that the vectors G and OQ are parallel, it therefore follows that

$$h : k : l = x_Q : y_Q : z_Q \quad (\text{XIV})$$

The indices h, k, l which are looked for therefore are the smallest unmixed triple of integers which fulfil Eq. (XIV). They allow all parameters of the diffraction that leads to the reflection to be calculated: the spacing of lattice planes d is obtained from Eq. (VI), the wavelength λ from Eq. (V) and the Bragg angle ϑ is, according to Eqs. (XII) and (XIV),

$$\vartheta = \arctan \left(\frac{l}{\sqrt{h^2 + k^2}} \right) \quad (\text{XV})$$

Setup and carrying out the experiment

The experimental setup is illustrated in Fig. 8.

- If necessary, remove the goniometer or the plate capacitor X-ray.

Remark:

NaCl and LiF crystals are hygroscopic and brittle: keep the crystals in a place as dry as possible, avoid mechanical stress to the crystals, and only touch the front side of a crystal.

a) Laue diagram at NaCl:

- Carefully attach the NaCl crystal for Laue diagrams **(b)** to the pinhole diaphragm **(a)** (from the scope of supply of the X-ray film holder) with transparent adhesive tape.
- Attach the collimator, and cautiously turn it so that the outside edges of the crystal are aligned as horizontally (or vertically) as possible.
- Clamp the X-ray film **(c)** at the film holder so that it is centred, and see to it that the entire surface of the film is planar.
- Clamp the film holder onto the experiment rail, and mount the experiment rail in the experiment chamber of the X-ray apparatus.
- Make a 15 mm long spacer from paper board and shift the film holder so that the distance between the monocrystal and the film is 15 mm (by varying the distance between the crystal and the film the area covered in the diagram is changed).
- Set the tube high voltage $U = 35$ kV, the emission current $I = 1.0$ mA and $\Delta\beta = 0.0^\circ$.
- Select the measuring time $\Delta t = 1800$ s, and start the exposure timer with the key SCAN.

If the exposure time is longer, the reflections near the centre are blurred by the unscattered X-rays; however structures which are farer away from the centre become discernable.

- When the exposure time is over, take the film holder with the experiment rail out of the experiment chamber.
- Remove the X-ray film from the holder, and develop it according to the instruction sheet for the X-ray film.

b) Laue diagram at LiF:

- Exchange the NaCl crystal with the LiF crystal, and align the LiF crystal.
- Clamp a new X-ray film in the film holder, and mount the experiment rail with the film holder once more.
- Shift the film holder so that the distance L between the monocrystal and the film is 11 mm.
- Select the measuring time $\Delta t = 1200$ s, and start the exposure timer with the key SCAN.
- When the exposure time is over, take the X-ray film from the film holder and develop it.

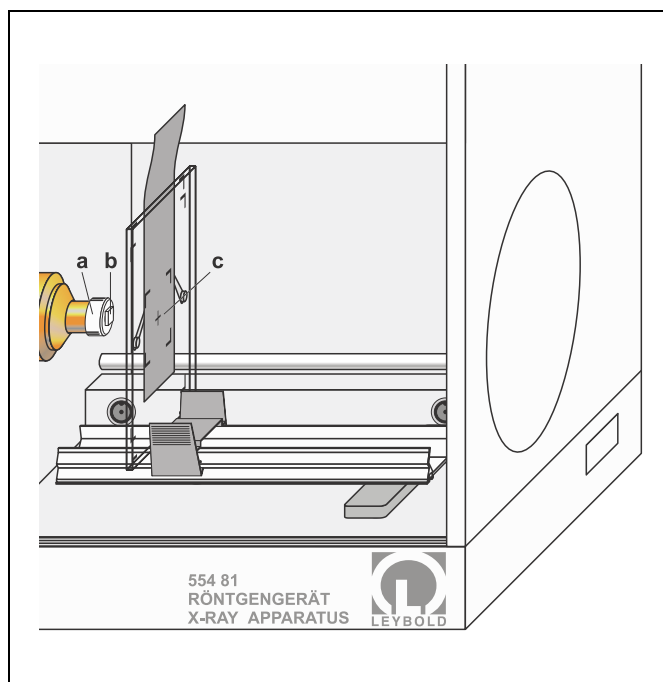


Fig. 8 Experimental setup for taking a Laue diagram at crystals

Measuring example

a) *Laue* diagram at NaCl:

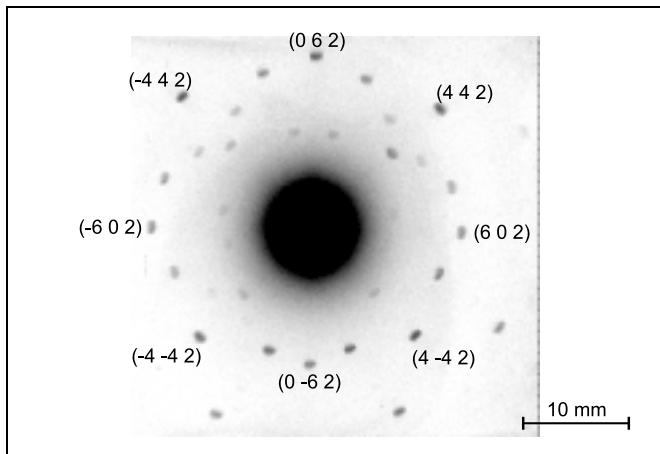


Fig. 9 *Laue* diagram at NaCl,
 $U = 35 \text{ kV}$, $I = 1 \text{ mA}$, $L = 15 \text{ mm}$, $\Delta t = 1800 \text{ s}$
 (for identifying the reflections refer to Table 1)

b) *Laue* diagram at LiF:

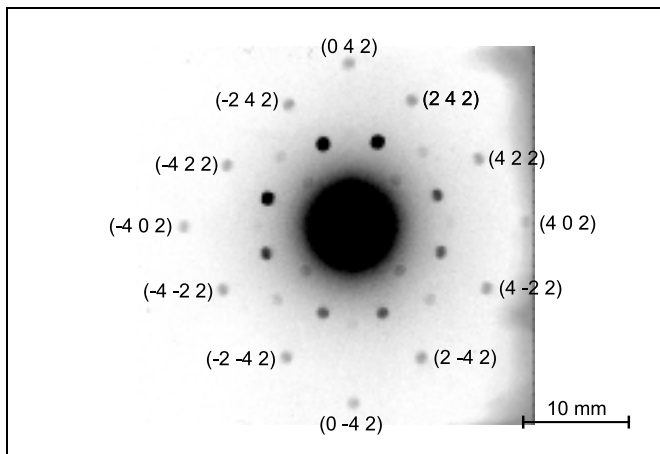


Fig. 10 *Laue* diagram at LiF,
 $U = 35 \text{ kV}$, $I = 1 \text{ mA}$, $L = 11 \text{ mm}$, $\Delta t = 1200 \text{ s}$
 (for identifying the reflections refer to Table 3)

a) *Laue* diagram at NaCl:

Tab. 1: Coordinates and extended *Miller* indices of the reflections in the *Laue* diagram of NaCl (*: calculated)

	$\frac{x_Q}{\text{mm}}$	$\frac{y_Q}{\text{mm}}$	$\frac{z_Q^*}{\text{mm}}$	h	k	l	$\frac{x^*}{\text{mm}}$	$\frac{y^*}{\text{mm}}$
1	17.2	8.4	9,3	4	2	2	15.0	7.5
	-7.8	-15.2	7,7	-2	-4	2	-7.5	-15.0
	7.8	-15.2	7,7	2	-4	2	7.5	-15.0
	15.8	-8.0	8,2	4	-2	2	15.0	-7.5
2	12.6	-0.4	4,6	6	0	2	11.2	0.0
	0.2	14.5	5,9	0	6	2	0.0	11.2
	-13.2	-0.1	5,0	-6	0	2	-11.2	0.0
	0.0	-11.2	3,7	0	-6	2	0.0	-11.2
3	11.5	3.6	4,2	6	2	2	10.0	3.3
	4.2	12.4	4,9	2	6	2	3.3	10.0
	-4.3	13.0	5,3	-2	6	2	-3.3	10.0
	-12.2	4.0	4,7	-6	2	2	-10.0	3.3
	-11.2	-3.9	4,1	-6	-2	2	-10.0	-3.3
	-3.3	-10.2	3,4	-2	-6	2	-3.3	-10.0
	3.2	-10.0	3,3	2	-6	2	3.3	-10.0
	10.5	-3.9	3,7	6	-2	2	10.0	-3.3
4	9.0	9.2	4,8	4	4	2	8.6	8.6
	-11.0	11.0	6,6	-4	4	2	-8.6	8.6
	-9.0	-9.2	4,8	-4	-4	2	-8.6	-8.6
	9.0	-9.0	4,7	4	-4	2	8.6	-8.6
5	9.0	5.8	3,4	6	4	2	7.5	5.0
	6.3	9.2	3,7	4	6	2	5.0	7.5
	-6.5	9.8	4,1	-4	6	2	-5.0	7.5
	-9.5	6.2	3,8	-6	4	2	-7.5	5.0
	-8.2	-5.5	3,0	-6	-4	2	-7.5	-5.0
	-5.2	-8.0	2,8	-4	-6	2	-5.0	-7.5
6	6.4	6.2	2,4	3	3	1	5.3	5.3
	-6.8	6.8	2,8	-3	3	1	-5.3	5.3
	-5.8	-5.8	2,1	-3	-3	1	-5.3	-5.3
	5.3	-5.3	1,8	3	-3	1	5.3	-5.3
7	6.8	1.3	1,5	5	1	1	6.0	1.2
	1.8	7.8	2,0	1	5	1	1.2	6.0
	-1.2	8.0	2,0	-1	5	1	-1.2	6.0
	-7.2	1.2	1,7	-5	1	1	-6.0	1.2
	-7.0	-1.5	1,6	-5	-1	1	-6.0	-1.2

Tab. 2: Spacing of lattice planes d , Bragg angle ϑ and wavelength λ associated with the sets of lattice planes of NaCl, $a_0 = 564.02 \text{ pm}$ [1]

h	k	l	$\frac{d}{\text{pm}}$	ϑ	$\frac{\lambda}{\text{pm}}$
4	2	2	115.1	24.1°	94.0
6	0	2	89.2	18.4°	56.3
6	2	2	85.0	17.5°	51.1
4	4	2	94.0	19.5°	62.8
6	4	2	75.4	15.5°	40.3
3	3	1	129.4	13.3°	59.5
5	1	1	108.5	11.1°	41.8

Evaluation

- Lay a piece of millimetre-square graph paper on the X-ray film, read the x_Q - and y_Q -co-ordinates of the reflections, and calculate the z_Q -co-ordinate according to Eq. (XIII).
- Identify the indices h , k and l according to Eq. (XIV).
- Calculate the spacing of the lattice planes d according to Eq. (VI), the Bragg angle ϑ according to Eq. (XV) and the wavelength λ according to Eq. (V).
- Now apply Eqs. (XI) and (XIV) to calculate the co-ordinates x_P and y_P or x_Q and y_Q , respectively, and compare them with the measured values.

b) *Laue* diagram at LiF:Tab. 3: Co-ordinates and extended *Miller* indices of the reflections in the *Laue* diagram of LiF (*: calculated)

	$\frac{x_0}{\text{mm}}$	$\frac{y_0}{\text{mm}}$	$\frac{z_0}{\text{mm}}$ *	<i>h</i>	<i>k</i>	<i>l</i>	$\frac{x}{\text{mm}}$ *	$\frac{y}{\text{mm}}$ *
1	14.2	-0.1	7.0	4	0	2	14.7	0.0
	0.2	13.3	6.3	0	4	2	00	14.7
	-14.2	0.0	7.0	-4	0	2	-14.7	0.0
	0.1	-15.2	7.8	0	-4	2	0.0	-14.7
2	10.5	5.0	5.0	4	2	2	11.0	5.5
	5.5	10.0	4.9	2	4	2	5.5	11.0
	-5.8	10.3	5.2	-2	4	2	-5.5	11.0
	-10.5	5.2	5.1	-4	2	2	-11.0	5.5
	-11.0	-5.8	5.6	-4	-2	2	-11.0	-5.5
	-5.6	-11.5	5.9	-2	-4	2	-5.5	-11.0
	5.8	-11.2	5.7	2	-4	2	5.5	-11.0
	11.0	-5.5	5.5	4	-2	2	11.0	-5.5
3	8.0	0.0	2.6	6	0	2	8.3	0.0
	-0.5	7.2	2.2	0	6	2	0.0	8.3
	-9.1	-0.3	3.3	-6	0	2	-8.3	0.0
	0.5	-8.0	2.6	0	-6	2	0.0	-8.3
4	7.2	2.0	2.3	6	2	2	7.3	2.4
	2.2	6.5	2.0	2	6	2	2.4	7.3
	-2.0	7.4	2.4	-2	6	2	-2.4	7.3
	-7.0	2.2	2.2	-6	2	2	-7.3	2.4
	-7.2	-2.3	2.4	6	-2	2	-7.3	-2.4
	-2.2	-7.8	2.7	2	-6	2	-2.4	-7.3
	2.7	-7.6	2.6	2	-6	2	2.4	-7.3
	7.2	-2.8	2.4	6	-2	2	7.3	-2.4
5	6.0	5.8	2.8	4	4	2	6.3	6.3
	-6.0	6.0	2.9	-4	4	2	-6.3	6.3
	-6.2	-6.3	3.1	-4	-4	2	-6.3	-6.3
	6.5	-6.5	3.3	4	-4	2	6.3	-6.3
6	4.0	3.5	1.2	3	3	1	3.9	3.9
	-3.5	3.6	1.1	-3	3	1	-3.9	3.9
	-3.8	-4.0	1.3	-3	-3	1	-3.9	-3.9
	4.2	-4.0	1.4	3	-3	1	3.9	-3.9

Tab. 4: Spacing of lattice planes *d*, *Bragg* angle ϑ and wavelength λ associated with the sets of lattice planes of LiF, $a_0 = 402.80 \text{ pm}$ [1]

<i>h</i>	<i>k</i>	<i>l</i>	$\frac{d}{\text{pm}}$	ϑ	$\frac{\lambda}{\text{pm}}$
4	0	2	90.1	26.6°	80.6
4	2	2	82.2	24.1°	67.1
6	0	2	63.7	18.4°	40.3
6	2	2	60.7	17.5°	36.5
4	4	2	67.1	19.5°	44.8
3	3	1	92.4	13.3°	42.5

Results

A *Laue* diagram is a diffraction photograph of a monocrystal taken with a continuous ("white") spectrum of X-rays. From the continuum of the X-rays only those wavelengths (cf. Tables 2 and 4) contribute to the diffraction pattern taken on a plane film which fulfil the *Bragg* condition for a particular set of lattice planes.

The symmetry of the *Laue* diagrams is in accordance with the cubic structure of the NaCl and the LiF crystal.

Literature

[1] Handbook of Chemistry and Physics, 52nd Edition (1971–72), The Chemical Rubber Company, Cleveland, Ohio, USA.

Supplementary Remarks for Analyzing the Laue Patterns

1. From the size of the film, calculate the maximum Bragg angle θ that can be observed.
2. Instead of trying to determine xyz coordinates of the spots, it is simpler to just measure radii (R) or diameters. The Bragg angle θ can then be determined from $\tan 2\theta = R/L$ where L is the crystal film distance.
3. For Laue patterns the allowed Bragg angles will only depend on the Miller indices as $\theta = \tan^{-1}\left(\frac{l}{\sqrt{h^2+k^2}}\right)$. This enables you to match up Bragg angle predictions with the measured θ of the spots.
4. Once you match up some Miller indices with Bragg angles you should be able to determine the absolute xy orientation of the pattern with respect to the crystal orientation.
5. Determine the Miller indices of as many spots as you can.
6. Check your results with the Laue Applet. The Laue Applet may also give you some guidance for steps 2-4.
7. Calculate the spacing of the lattice planes from the crystal lattice constant and Miller indices. Then use the Bragg formula to calculate the wavelength λ for each of your spots.