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Imaginary Potential for Electrons of CaF_2 from a Bloch-Wave Analysis

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Abstract

In a previous analysis of Bloch waves in electron diffraction experiments with a CaF_2 single-crystal wedge [Ishida, Johnson & Lehmpfuhl (1975). *Z. Naturforsch. Teil A*, **30**, 1715-1729] absorption coefficients of the strong Bloch waves were determined. The experiments were performed with two types of crystal wedges, produced by (111) cleavage faces. One type had a wedge angle of 109° and the other type an angle of 71°. In the experiment with the blunt wedge the direction of incidence was close to [100] and in the sharp-wedge experiment close to [110]. The two sets of absorption coefficients were not consistent with an imaginary potential describing the absorption. From reexamination of the data a consistent absorption potential, expressed by an analytical formula, could be determined, which confirmed the absorption coefficients for the blunt wedge and modified the former values for the sharp wedge. Agreement between calculation and experiment could be achieved by convoluting the diffracted beams with the profile of the incident beam. This effect is important for the sharp wedge and negligible for the blunt wedge.

The current density of electrons travelling through a crystal can be described by Bloch waves. The various Bloch waves show characteristic density distributions over the crystal unit cell, depending on the direction of the incident beam (Lehmpfuhl, 1973). Each of the Bloch waves is absorbed with its own characteristic absorption coefficient as predicted by von Laue (1953). For localized inelastic scattering processes this results in an orientation dependence of absorption as shown experimentally (Taftø & Lehmpfuhl, 1982). The orientation-dependent absorption of the electrons can be described by introducing an imaginary part into the scattering potential, according to Molière (1939), in the form

$$V(r) = V(r)^{\text{real}} + iV(r)^{\text{im}}. \quad (1)$$

For this complex crystal potential, which is no longer Hermitian, the eigenvalues τ_j become complex:

$$\tau_j = \tau_j' + i\tau_j''. \quad (2)$$

The imaginary part represents the absorption coefficient μ_j of the corresponding Bloch wave,

$$\mu_j = 4\pi\tau_j''/\lambda. \quad (3)$$

A Bloch-wave analysis can be performed for electron diffraction from a single-crystal wedge. In such an experiment the Bloch waves are dispersed into their partial waves, and the strongest ones can be identified in the diffraction spots as a fine structure. Owing to absorption the fine-structure spots are broadened and overlap coherently. The resulting intensity profile $I_g(\mathbf{S})$ can be analysed, giving information on eigenvalues and absorption coefficients (Molière & Lehmpfuhl, 1962). $\mathbf{S}$ is a unit vector in the direction of observation. This intensity distribution,

$$I_g(\mathbf{S}) = U_g(\mathbf{S})\,U_g^*(\mathbf{S}), \quad (4)$$

results from the Fourier transform of the amplitude distribution of the electron waves $\psi_g^{(j)}$ in the exit surface of the crystal wedge given by equation (5) of Ishida, Johnson & Lehmpfuhl (1975),

$$U_g(\mathbf{S}) = (\lambda/2\pi)\sum_j \psi_g^{(j)}[(\mathbf{S}_e + \mathbf{B}_g - \mathbf{S})\mathbf{N} + \tau_j'\mathbf{N}\mathbf{N}_e + i\mu_j(\lambda/4\pi)\mathbf{N}\mathbf{N}_e]^{-1}. \quad (5)$$

$\mathbf{S}_e$ is a unit vector in the direction of the incident electron beam, $\mathbf{N}_e$ a unit vector perpendicular to the entrance surface of the crystal, $\mathbf{N}$ a unit vector in the exit surface and perpendicular to the edge, and $\mathbf{B}_g$ is the reciprocal-lattice vector $\mathbf{g}$ multiplied by the wavelength λ.

In the previous analysis of Bloch waves in electron diffraction experiments with a CaF_2 single-crystal wedge (Ishida, Johnson & Lehmpfuhl, 1975), absorption coefficients were determined by fitting the profile of the diffraction spots with calculated intensity profiles according to (4) with the absorption coefficients as adjustable parameters. The eigenvectors $\psi_g^{(j)}$ and the real eigenvalues τ_j' were obtained from a 42-beam calculation with a structure potential after Doyle & Turner (1968). The experiments were

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Table 1. *Parameters of equation* (6) *for* CaF_2

$V_1 = 4{\cdot}0$ V	$T_1 = 1{\cdot}0$ V	$B_1 = 0{\cdot}6$ Å^2	$A = 2{\cdot}0$	1:Ca
$V_2 = 1{\cdot}8$ V	$T_2 = 0{\cdot}3$ V	$B_2 = 0{\cdot}8$ Å^2		2:F

$$G^{(i)}_{hkl} = \tfrac{1}{24} \sum_j \exp\left(2\pi i g_{hkl} r_j^{(i)}\right)$$

where $r_j^{(i)}$ is the position of an atom of type (i).

performed with two types of crystal wedges, produced by (111) cleavage faces. One type had a wedge angle of ~109° and the other type an angle of ~71°. In the experiment with the blunt wedge the direction of the incident beam was close to [100], and in the sharp-wedge experiment close to [110]. In these two directions the atom strings consist of only one kind of atom. However, the density of atoms along the strings is different for the two cases, which is of great interest with respect to elastic and inelastic scattering. For the two different wedges two sets of absorption coefficients were obtained. In a later test (not published) it was found that they were not consistent with an imaginary potential describing the absorption corresponding to (1) and (6). Only separate absorption potentials could be determined for each wedge type.

Table 2. *Fourier coefficients of the imaginary potential* (V) *for* CaF_2

$$V^{\mathrm{im}} = \frac{1}{6}\left[\frac{3}{1+2x} + \exp(-0{\cdot}01x)\right] + \frac{\gamma}{3}\left[\frac{1{\cdot}5}{1+2x} + 0{\cdot}3\exp(-0{\cdot}013x)\right]$$

$$x = h^2 + k^2 + l^2.$$

$\gamma = 1$ for *hkl* even, multiple of 4

$\gamma = -1$ for *hkl* even, multiple of 4+2

$\gamma = 0$ for *hkl* odd.

hkl	x	V^{im}(V)
000	0	1·27
111	3	0·23
200	4	0·065
220	8	0·30
311	11	0·17
222	12	0·06
400	16	0·25
331	19	0·15
420	20	0·06
422	24	0·22
333	27	0·13
440	32	0·20
442	36	0·05
444	48	0·17

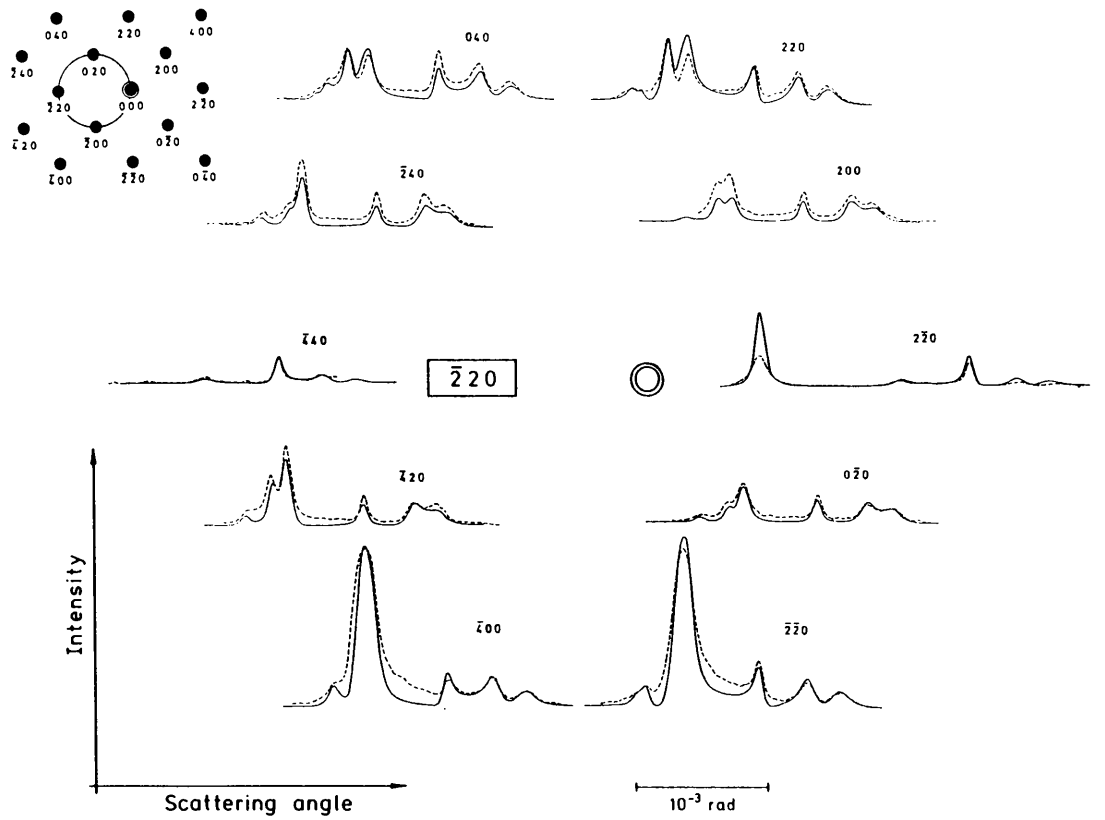

Fig. 1. Intensity distributions of different reflections from the blunt wedge of CaF_2 with 220, 200 and 020 simultaneously excited. Comparison of experimental curves (dashed) with calculations (solid) using the imaginary potential (6). Incident beam close to the [001] zone axis. Intensity in arbitrary units. (The strongly excited beams are not shown because they are heavily overexposed.) The experimental curves correspond to the curves in Fig. 11 of Ishida, Johnson & Lehmpfuhl (1975). Energy of the electrons 60·85 keV.

From a reexamination of the data in a new analysis we found that it was possible to describe the absorption in both cases by one imaginary potential. However, the profile of each partial wave of the reflected beam had to be convoluted by the profile of the incident beam. This convolution had a very small effect on the absorption coefficients for the blunt wedge. Owing to the strong absorption, the profile of the partial waves is already broadened, so that the profile of the primary beam has only little influence. But the partial waves from the sharp wedge showed only a small broadening due to the weaker absorption effect, so that the primary-beam profile has a great influence on the resulting diffraction spot profile.

The new analysis was carried out by comparing the experimental intensity distribution with calculated intensity distributions as described in previous papers (Lehmpfuhl & Reissland, 1968; Ichimiya & Lehmpfuhl, 1978) but with a non-Hermitian complex crystal potential (1) and by convolution with the Gaussian profile of the incident beam with an angular half-width of 2×10^{-5} rad. The complex crystal potential was used in a 50-beam calculation leading to eigenvalues, eigenvectors and absorption coefficients for the different Bloch waves. As imaginary part for the non-Hermitian potential a parametric expression was used which includes in its general form the absorption model due to Humphreys & Hirsch (1968) and Ichimiya (1985):

$$V_{hkl}^{\mathrm{im}}=\sum_{i=1}^{2}\left\{\frac{(V_i-T_i)}{1+A(h^2+k^2+l^2)}+T_i\exp\left(-0{\cdot}5B_i\frac{h^2+k^2+l^2}{a^2}\right)\right\}G_{hkl}^{(i)}. \quad (6)$$

In an earlier paper (Ichimiya & Lehmpfuhl, 1978) a similar model was found to fit the experimental

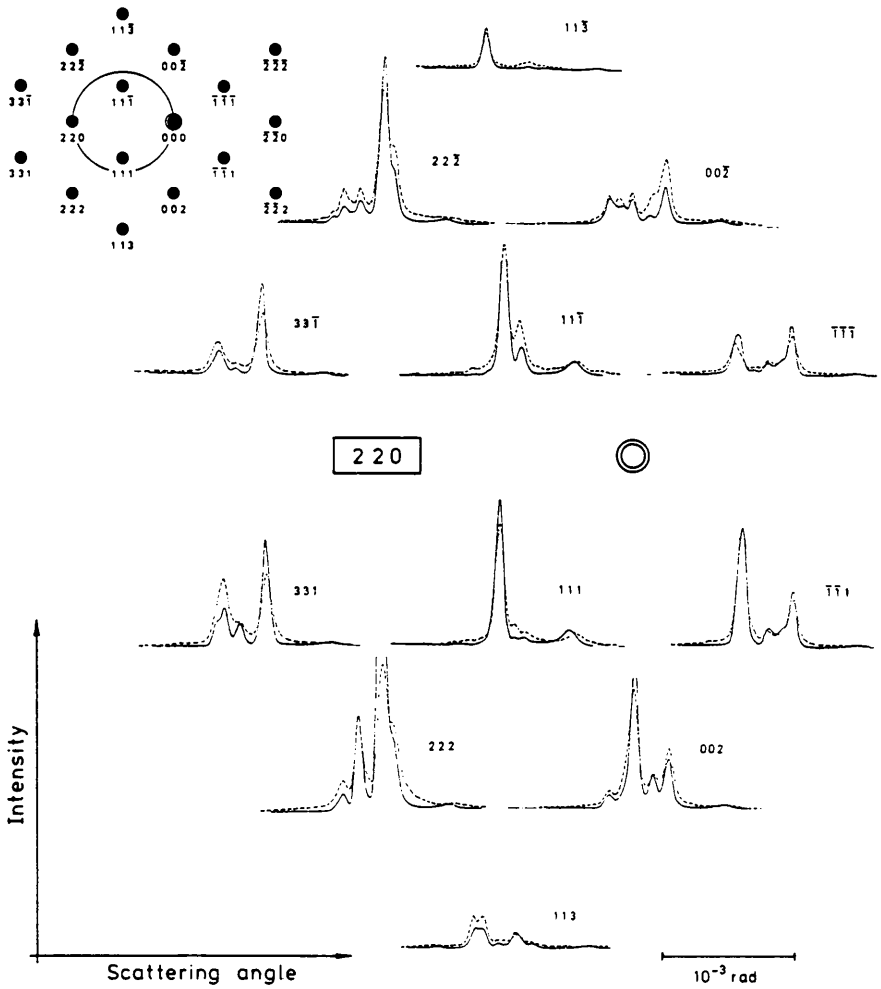

Fig. 2. Intensity distributions of different reflections from the sharp wedge of CaF_2 with 220 excited near the [$\bar{1}$10] zone axis. Comparison of experimental curves (dashed) with calculations (solid) using the same imaginary potential as for the blunt wedge in Fig. 1. The experimental curves correspond to the curves in Fig. 17 of Ishida, Johnson & Lehmpfuhl (1975).

Table 3. *Absorption coefficients* ($\times 10^3$ Å^{-1})

Bloch wave	109° wedge		71° wedge	
	Ishida *et al.* (1975)	Present results	Ishida *et al.* (1975)	Present results
1	11·0	11·9	18·0	12·1
2	8·0	8·1	7·0	3·8
3	4·0	3·5	7·0	4·3
4	(4·0)*	3·5	5·5	3·9
5	6·0	6·4	5·5	3·1
6	5·5	5·1	5·5	3·8
7	4·5	4·5	4·5	2·8
8	4·5	4·6	5·0	2·7
9	4·5	3·3	5·5	3·1
10	4·5	3·5	5·0	3·0
11	-	3·5	6·0	2·7
12	-	3·4	4·5	3·0
13	-	4·3	5·0	2·7
14	-	4·7	5·5	2·7
15	-	4·5	—	3·8

* Bloch waves 3 and 4 are nearly identical.

observations. The exponential term represents the square of the Debye-Waller factor. Table 1 shows the parameters for the equation giving the best fit with the experiment. $i = 1, 2$ stands for the two kinds of atoms in the unit cell. $G_{hkl}^{(i)}$ is a part of the structure factor for the type (i) of atoms in the unit cell. With these parameters some Fourier coefficients of the imaginary potential are calculated and shown in Table 2. As shown in Table 1, the Debye parameters B_i for Ca and F were 0·6 and 0·8 Å^2 respectively. These values are in good agreement with the values obtained by X-ray diffraction (*International Tables for X-ray Crystallography*, 1968). Therefore we conclude that the analytical formula (6) for the imaginary potential can be valuable for dynamical calculations of electron diffraction.

The degree of agreement between calculated diffraction spot profiles and experimental densitometer records is similar to that in the previous work (Ishida, Johnson & Lehmpfuhl, 1975). Two examples are shown in Figs. 1 and 2 for the two types of wedges. They correspond to Figs. 11 and 17 of Ishida, Johnson & Lehmpfuhl (1975). The agreement of the profiles for other experimental conditions is of the same order when using the absorption potential (6) with the data of Table 1.

The resulting absorption coefficients for the first strong Bloch waves are shown in Table 3, together with the previously determined absorption coefficients. For the blunt wedge the new data show only a little change, while for the sharp wedge the data show a remarkable change owing to the effect of convolution with the profile of the primary beam.

This investigation has shown the possibility of describing the absorption of electrons in a crystal by an imaginary part of the crystal potential, which is in agreement with the absorption model of Humphreys & Hirsch (1968). Consistent results were obtained for two different directions of projection of the atoms. In the two projections atoms of only one kind are lying on top of each other but with different densities along the atomic strings.

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