Absorption of X-rays by Coplanar Four-beam Reflections

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Z. Naturforsch. **37 a**, 617—625 (1982); received January 18, 1982

Dedicated to Prof. G. Hildebrandt on the occasion of his 60th birthday

The absorption effect in dispersion and reflection curves is shown for the σ and π polarizations and for some wavelengths in the small wavelength interval in which the coplanar four-beam diffraction of x-rays exists. The investigated coplanar Ge[000, 440, 260, $\bar{2}$20] reflections appear in a Ge[440, $\bar{2}$20] monolithic monochromator working with CoKα_1 radiation. It is shown that the absorption not only reduces the intensity in a complicated way but also changes the energy distribution between the participating reflections.

Introduction

In this paper we deal with the influence of absorption on the reflection curves for the coplanar four-beam x-ray diffraction Ge/000, 440, 260, $\bar{2}$20/.

Our results are part of a preliminary treatment of the monolithic monochromator Ge[440, $\bar{2}$20] proposed by Deslattes [1]. In this monochromator the reflections 440, $\bar{2}$20 appear successively on the walls of the single perfect monocrystal Ge. If the Bragg condition is fulfilled for the successive reflections 440, $\bar{2}$20 on the single monocrystal, then it must also be fulfilled for both reflections simultaneously. Hence on the walls of the dispersive monolithic monochromator many-beam reflection occurs instead of the conventional two-beam reflection. Beaumont and Hart [2] were the first to mention this fact. In the case of the monolithic monochromator Ge[440,, $\bar{2}$20] the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}$20] takes place. To get some idea about the properties of the Ge[440, $\bar{2}$20] monochromator we had to study first the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}$20].

Ewald's geometrical construction for the Ge[000, 440, 260, $\bar{2}$20] diffraction is shown in Figure 1. Depending on the orientation of the crystal surface the reflection pairs 440, 260 or $\bar{2}$20, 260 are available, which we have investigated previously. We have solved the problem both experimentally and theoretically. To compare the computed results with experiment, we established a simplified model of the experiment. The calculated and experimental dependences turned out to have the same shape. The influence of absorption may not be confirmed directly by experiment because there is no Ge crystal without absorption. Nevertheless the experiment confirms the main procedure of calculation.

To our knowledge, the only contributions to the theoretical investigation of a many-beam diffraction in the Bragg case was published by Mayer [3] and later by Graeff and Bonse [4]. Both papers treated the three-beam diffraction without taking absorption into account. The first part of the experimental and theoretical treatment of the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}$20] was shown on the

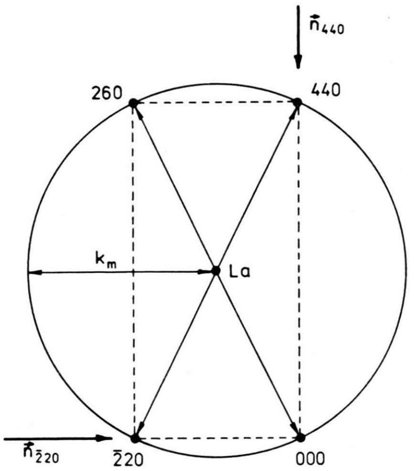

Fig. 1. Ewald's geometrical construction for the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}$20]. The drawn normal vectors $\boldsymbol{n}_{440}$, $\boldsymbol{n}_{\bar{2}20}$ are identical with the inward facing surface normals of the plane crystal boundary (surface (110), surface ($\bar{1}$10)) at the studied reflections.

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XI. International Congress of Crystallography in Warszaw in 1978 [5]. The details of our experiment and calculation will be published elsewhere.

Principles of Calculation

We take the fundamental equations of the dynamical solution of a many-beam diffraction from the paper of Penning and Polder [6]. For our purpose we give only their formulas for coplanar diffractions.

The two mutually perpendicular components $\boldsymbol{X}$, $\boldsymbol{Y}$ of the dielectric displacement $\boldsymbol{D}$ inside the diffracting crystal ($\boldsymbol{X}$ lies in the plane of incidence and thus creates the polarization component π, $\boldsymbol{Y}$ then coincides with the component σ) are independent for any coplanar diffraction and the amplitudes X_i, Y_i ($i = 1, 2, \ldots, n$ in an n-beam diffraction) satisfy the equations

$$\sum_{j=1}^{n} A_{ij} Y_j = 0; \quad \sum_{j=1}^{n} A_{ij} (\boldsymbol{u}_i \cdot \boldsymbol{u}_j) X_j = 0,$$
$$i = 1, 2, \ldots, n.$$

The notation is as follows:

$A_{ij} (i \neq j)$ $A_{ij} = \psi_{ji}$,

A_{ii} $A_{ii} = \psi_0 - 2 (\boldsymbol{u}_i \cdot \boldsymbol{\Delta}) / k_{\mathrm{m}}$,

ψ_{ji} the term in the Fourier sum of the susceptibility ψ corresponding to the reciprocal lattice vector $\boldsymbol{b}_{ji} = \boldsymbol{k}_i - \boldsymbol{k}_j$,

$\boldsymbol{u}_i$ the unit vector parallel to the vacuum wave vector $\boldsymbol{k}_i$, $\boldsymbol{k}_i = k_{\mathrm{m}} \boldsymbol{u}_i$,

$\boldsymbol{\Delta}$ the deviation of the centre of the circle of reflection from the Laue point La.

The vector $\boldsymbol{\Delta}$ is the only unknown quantity which appears in the coefficients of the fundamental equations. It defines the so called dispersion surface in the reciprocal space. Since our problem is coplanar, we use the concept of the plane dispersion curve.

Our calculation of the Ge[000, 440, 260, $\bar{2}$20] diffraction is done under the following assumptions:

1. The incident x-ray wave is monochromatic and plane-parallel.
2. The diffracting crystal is assumed to be perfect and bounded by the only plane boundary. The boundary is oriented in two ways: parallel to the (440) or ($\bar{2}$20) diffracting planes, respectively. Both these orientations are indicated in Fig. 1 by their normal vectors $\boldsymbol{n}_{440}$, $\boldsymbol{n}_{\bar{2}20}$.
3. The coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}$20] is supposed to take place even if the Bragg condition is not exactly satisfied for all the participating diffractions 440, 260, $\bar{2}$20 at the same time. Then the coplanar Ge[000, 440, 260, $\bar{2}$20] diffraction holds not only for $\lambda_{\mathrm{m}} = 2\pi / k_{\mathrm{m}}$, but for some interval of λ including the value λ_{m}.
4. a) The diffracting monocrystal does not absorb the radiation.
   b) The diffracting monocrystal absorbs the radiation.

In our calculation we employed some common practice in solving of a general problem of diffraction. When absorption is of primary importance in our case we shall pay attention to its mathematical description.

Introduction of Absorption

In a non-absorbing crystal the susceptibility ψ is real and its Fourier coefficients ψ_{ij} satisfy the condition $\psi_{ij} = \psi_{ji}^{*}$. For an absorbing crystal the susceptibility is no longer real and may be written $\psi^{(\mathrm{c})} = \psi' + i \psi''$. Both real ψ' and ψ'' can be expressed as Fourier sum. We have

$$\psi_{ij}^{(\mathrm{c})} = \psi_{ij}' + i \psi_{ij}'',$$
$$\psi_{ji}^{(\mathrm{c})} = \psi_{ji}' + i \psi_{ji}'', \quad \text{where } \psi_{ij}', \psi_{ij}'', \psi_{ji}', \psi_{ji}''$$

are complex values. In this case $\psi_{ij}^{(\mathrm{c})} \neq \psi_{ji}^{(\mathrm{c})*}$.

The argument of ψ_{ij} depends on the choice of the origin of the position vector $\boldsymbol{r}$. If the crystal structure has inversion symmetry and the origin is placed into the inversion centre, then ψ_{ij}, ψ_{ij}' and ψ_{ij}'' are real and $\psi_{ij} = \psi_{ji}$ not only for a non-absorbing crystal but also $\psi_{ij}^{(\mathrm{c})} = \psi_{ji}^{(\mathrm{c})}$ for an absorbing crystal. $\psi_{ij}^{(\mathrm{c})}$, however, is complex.

The values of $\psi_{ij}^{(\mathrm{c})}$ are proportional to the structure factor according to known relations (James [7], Hildebrandt, Stephenson, and Wagenfeld [8]).

The introduction of a complex $\psi_{ij}^{(\mathrm{c})}$ leads to a complex resulting amplitude D_i^{r} of the diffracted waves in vacuum. From the point of view of experimental results we are interested in the real intensity

$$I_i^{\mathrm{r}} = |\boldsymbol{D}_i^{\mathrm{r}}|^2 = D_i^{\mathrm{r}} \cdot D_i^{\mathrm{r}*}.$$

The values of the necessary physical constants have been taken from [9], and the absorption correction of the structure factors from the paper of Cromer [10].

Results of Calculation

The reflection curves in this paper present the intensities I_i^{r}, expressed as fractions of the incident intensity, plotted as functions of the incidence angle θ.

The computed reflection curves 440, 260 resulting from the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}20$] with the crystal boundary parallel to the (110) lattice planes are for some values of $\Delta k = k - k_{\mathrm{m}}$ shown in Fig. 2 (a: reflection 440, b: reflection 260). The drawn extent of the reflection curves corresponds to a difference of the angle of incidence of 1 minute of arc, the marked point being the position of the Bragg angle for the diffraction Ge[000, 440]. The distance of the horizontal axes for the particular values of Δk is unity, the value of the total reflection.

The reflection curves $\bar{2}20$, 260 corresponding to the surface ($\bar{1}10$) are shown in Fig. 3 (a: reflection $\bar{2}20$, b: reflection 260). All what was said about Fig. 2 can be repeated for Figure 3.

The reflection curves were computed for a high number of Δk in the interval $(-10 \times 10^6\ \mathrm{m}^{-1}, 10 \times 10^6\ \mathrm{m}^{-1})$. In the ranges $\Delta k < -0.5 \times 10^6\ \mathrm{m}^{-1}$ and $\Delta k > 3 \times 10^6\ \mathrm{m}^{-1}$ the reflections 440, $\bar{2}20$ are similar to those for the two-beam reflection Ge[000, 440], Ge[000, $\bar{2}20$]. Both these ranges have their representative in Fig. 2 ($\Delta k = 10 \times 10^6\ \mathrm{m}^{-1}$) and Fig. 3 ($\Delta k = -10 \times 10^6\ \mathrm{m}^{-1}$). In the range $0 < \Delta k < 2 \times 10^6\ \mathrm{m}^{-1}$ the four-beam reflections 440, $\bar{2}20$ are notably distinct from the corresponding two-beam reflections. Figures 2 and 3 show those reflection curves from this range of Δk which demonstrate most expressively the influence of absorption. The wavelength belonging to the wave vector $k_{\mathrm{m}} = 3.51204 \times 10^{10}\ \mathrm{m}^{-1}$ is $\lambda_{\mathrm{m}} = 0.178904\ \mathrm{nm} \sim \mathrm{CoK}\alpha_1$. The difference $\Delta\lambda = \lambda - \lambda_{\mathrm{m}}$ is expressed by means of Δk as $\Delta\lambda = -(2\pi/k_{\mathrm{m}}^2) \cdot \Delta k$. The value of k_{m} leads to the following numerical relation:

$$\Delta\lambda = -5.1 \times 10^{-6}\ \mathrm{nm} \quad \text{for}$$
$$\Delta k = 1 \times 10^6\ \mathrm{m}^{-1}.$$

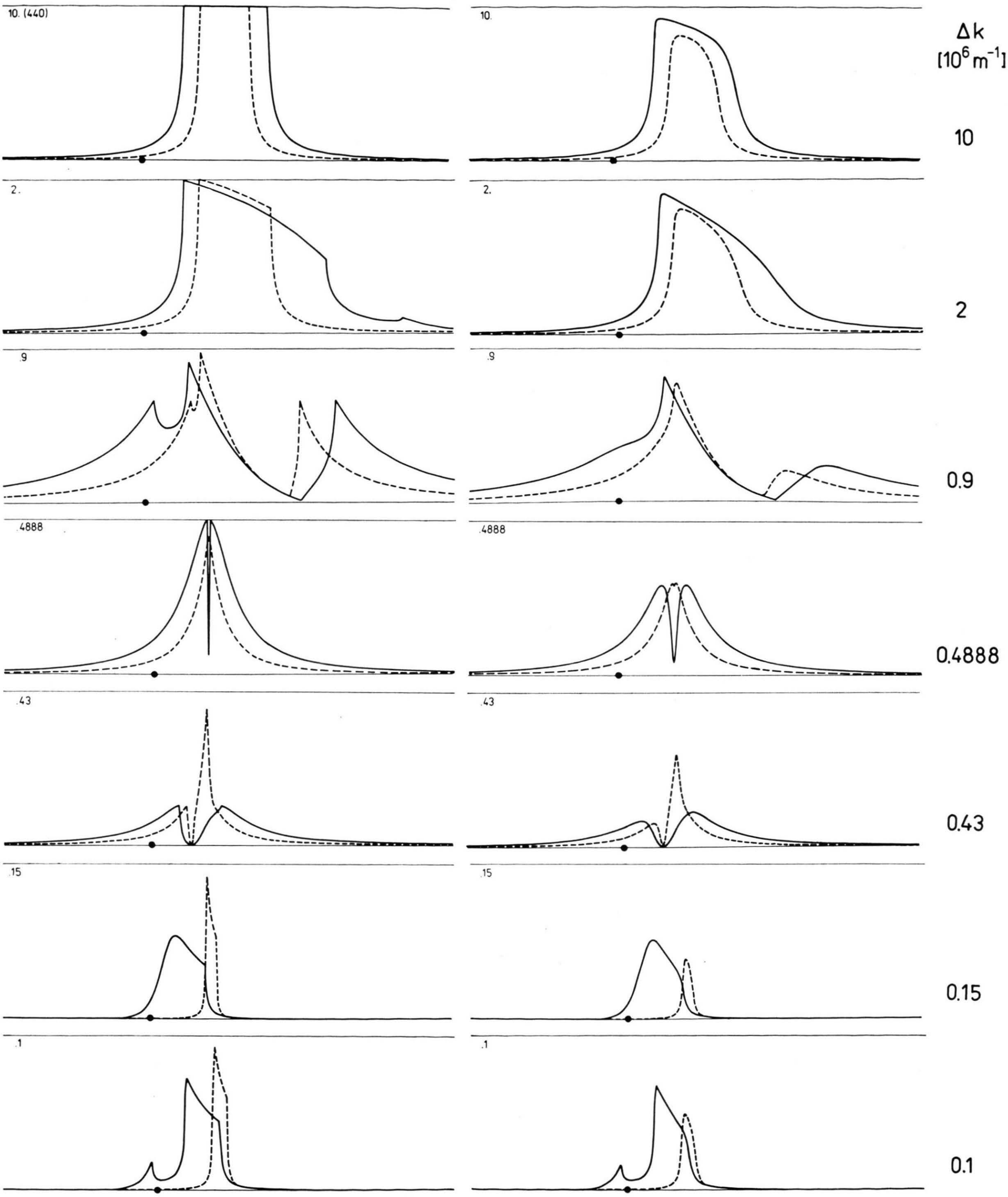
From Figs. 2 and 3 it can be seen that the influence of absorption is very complicated. The peaks of a given reflection curve are influenced differently. We may also see the increase of some tails or even the appearance of a new peak due to the absorption. This effect is noticeable especially in the case of the ($\bar{1}10$) surface (see Fig. 3a, $\Delta k \sim 0.5 \times 10^6\ \mathrm{m}^{-1}$, π polarization). The detailed discussion of the whole reflected energy divided into two reflected beams suggests a redistribution of energy between particular reflections in the Ge[000, 440, 260, $\bar{2}20$] diffraction due to absorption.

In Fig. 4 we present the dispersion curves drawn in the plane of incidence for five values of Δk. The region near the value of $\Delta k \sim 0.5 \times 10^6\ \mathrm{m}^{-1}$ is particularly interesting because of the form of the dispersion curves computed for the whole considered interval of $\Delta k \sim (-10 \times 10^6\ \mathrm{m}^{-1},\ 10 \times 10^6\ \mathrm{m}^{-1})$. The dispersion curve for the polarization σ is created by two points only, and for the polarization π no dispersion curve exists in the plane of incidence. Further, the redistribution of energy accompanying the absorption is most apparent for the region shown in Figure 4.

We chose the orthogonal coordinate system with respect to the orientation of the crystal boundary. The origin is placed to the Laue point La (see Figure 1). The axis for the independent variable, which expresses the angle of incidence θ of the incident wave, is parallel to the boundary. In Fig. 4 the perpendicular axis is marked by 0. The role of the axes θ, 0 is changed for both considered boundaries (110) and ($\bar{1}10$).

The equation for possible values of $\mathbf{\Delta}$ is of the fourth order in $|\mathbf{\Delta}|$ for both polarizations σ and π. Ignoring absorption, the four solutions are of the type: $A, B, -A, -B$; $A, ia, -A, -ia$; $ia, ib, -ia, -ib$; $A+ia, A-ia, -A+ia, -A-ia$, where A, B, a, b are real values. When absorption is taken into account, the above solutions have the form $A+ia, B+ib, -A-ia, -B-ib$.

The dispersion curve is defined as the curve which gives real values of $\mathbf{\Delta}$. In Fig. 4 we show not only the dispersion curves, but also the real parts of the complex values of $\mathbf{\Delta}$. Because of the symmetry of all dependences around the axis 0, it is sufficient to illustrate in Fig. 4 always only one half of the whole angular region. Thus both cases with or without absorption can be drawn in one picture. In those parts of Fig. 4 concerning the case without absorption the dispersion curves for the polarization σ are drawn by solid lines, for the polarization π, as far as it exists, we use dashed lines and for the real parts of complex values of $\mathbf{\Delta}$ we use dotted lines. For the case with absorption in Fig. 4 the solid or dashed lines determine the real parts of $\mathbf{\Delta}$ for the polarizations σ and π, respectively. From the four



a)

Fig. 2. The reflection curves 440 (Fig. 2a) and 260 (Fig. 2b) of the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}20$] for a non-absorbing crystal (left) and for an absorbing crystal (right). Surface (110). Polarization σ: solid lines, polarization π: dashed lines. The curves for $\Delta k = 10 \times 10^6$ m^{-1} are similar to those of the two-beam diffraction Ge[000, 440].

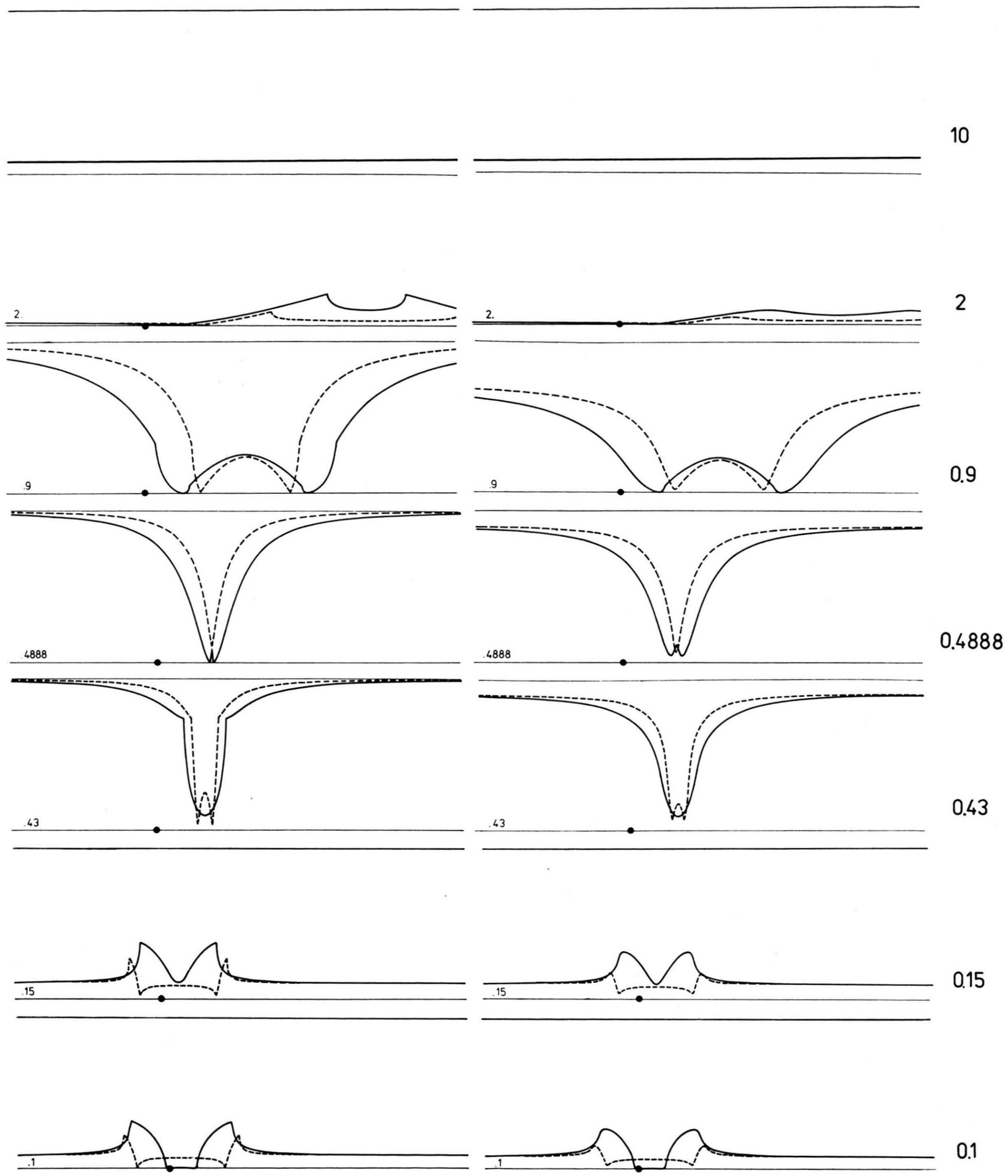


Fig. 2b.

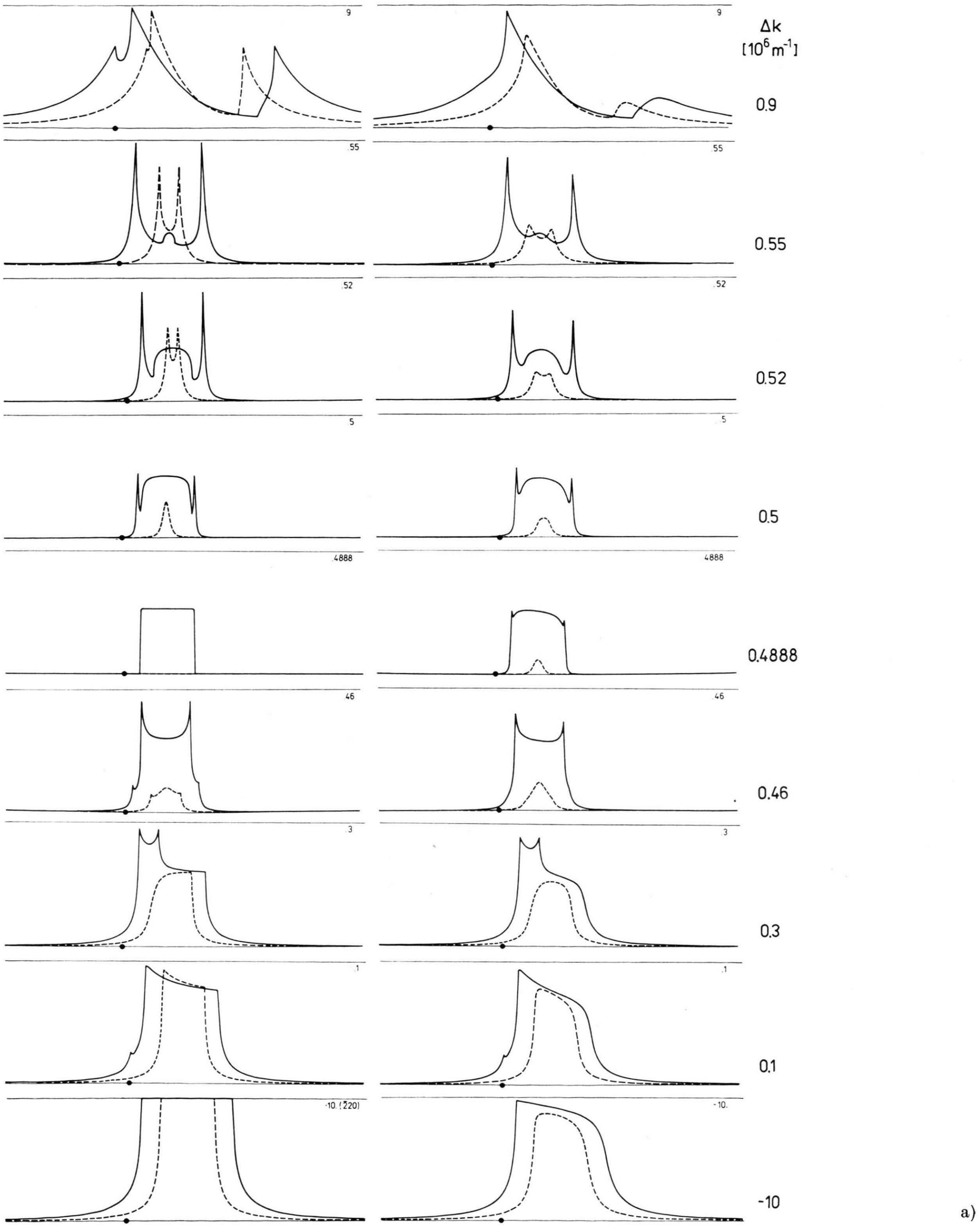


Fig. 3. The reflection curves $\bar{2}20$ Fig. 3a) and 260 (Fig. 3b) of the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}20$] for a non-absorbing crystal (left) and for an absorbing crystal (right). Surface ($\bar{1}10$). See text to Figure 2.

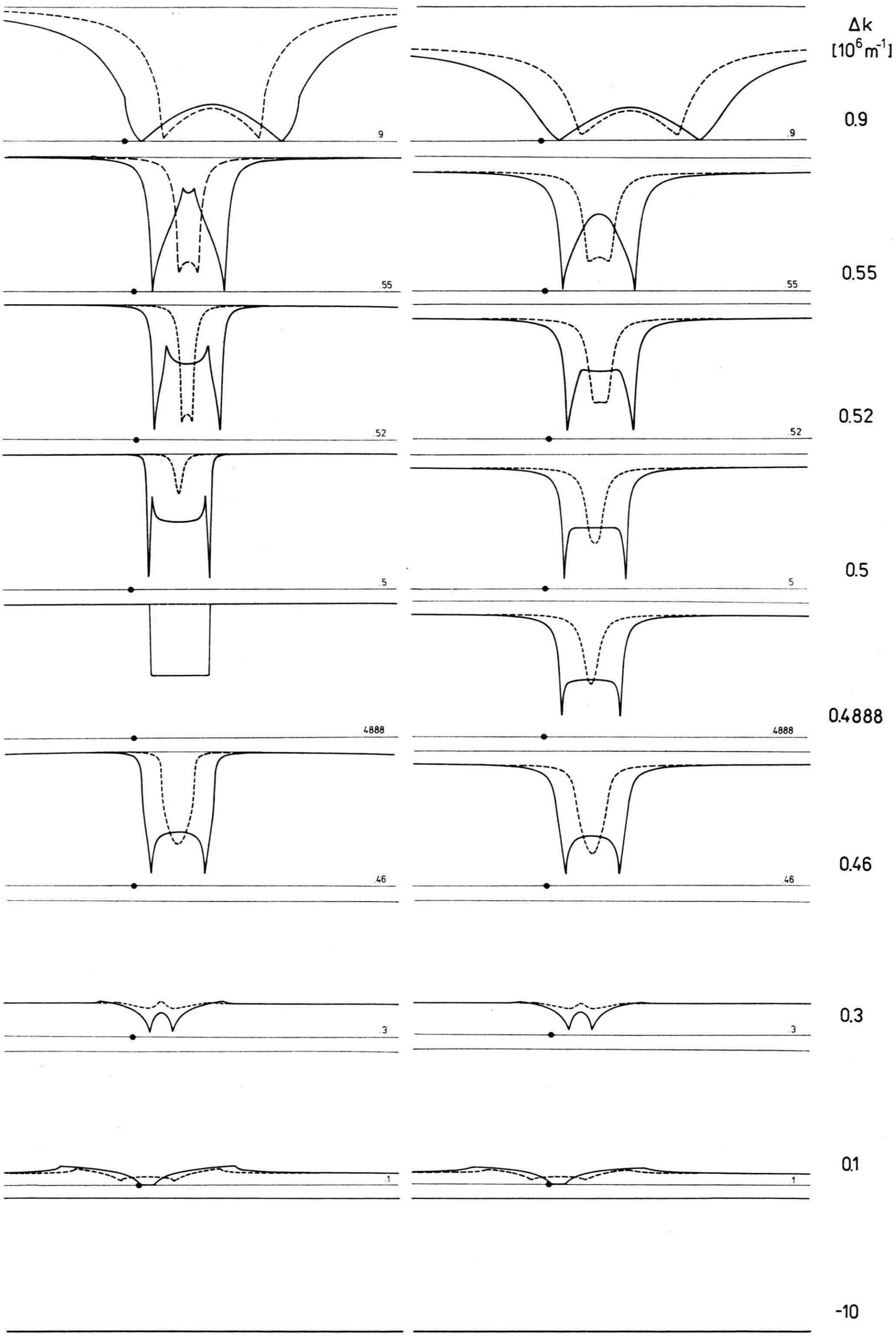
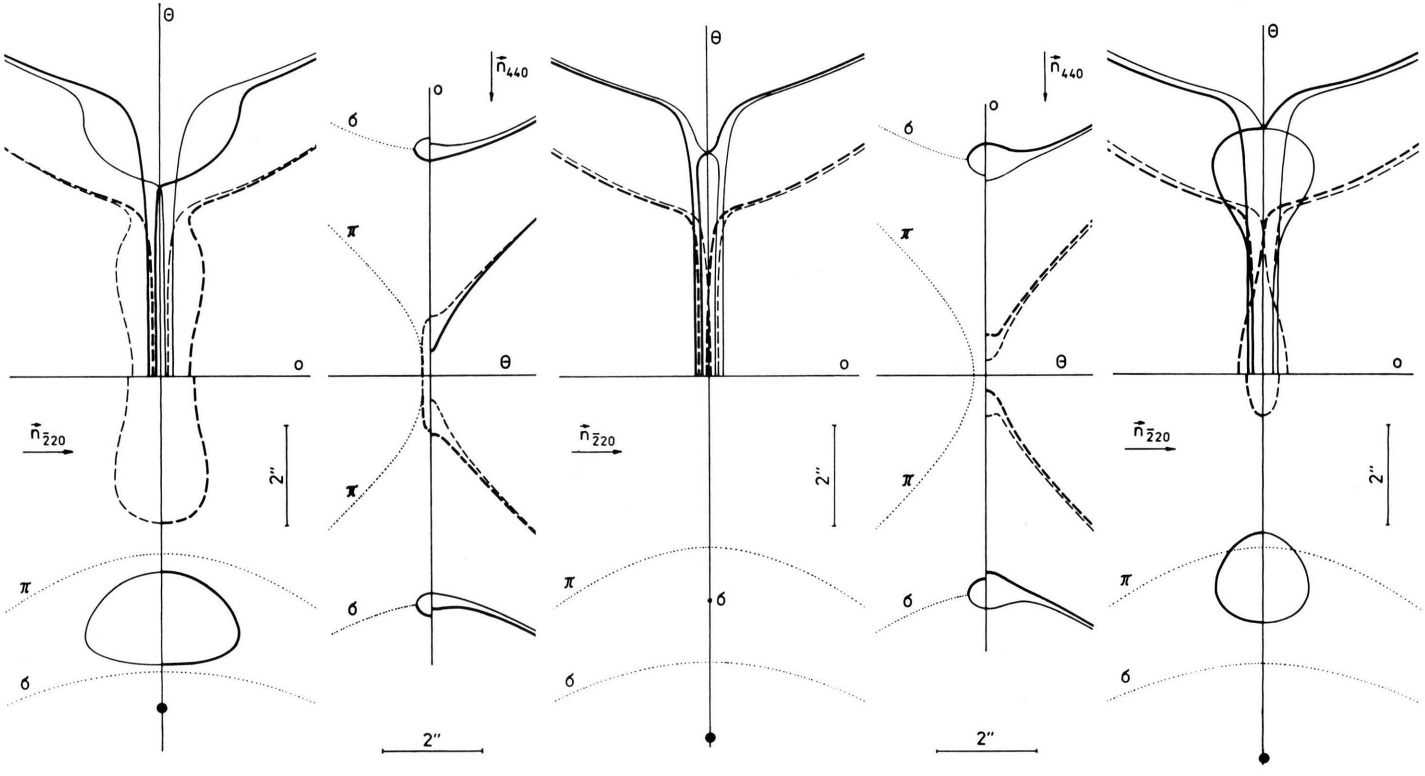

Δk
[10⁶m⁻¹]
0.9
.9
.9
0.55
.55
.55
0.52
.52
.52
0.5
.5
.5
0.4888
.4888
.4888
0.46
.46
.46
0.3
.3
.3
0.1
.1
.1
-10

Fig. 3b.



(a) $\Delta k = 0.45 \times 10^6\ \mathrm{m}^{-1}$ (b) $0.48 \times 10^6\ \mathrm{m}^{-1}$ (c) $0.4888 \times 10^6\ \mathrm{m}^{-1}$ (d) $0.5 \times 10^6\ \mathrm{m}^{-1}$ (e) $0.52 \times 10^6\ \mathrm{m}^{-1}$

Fig. 4. The dispersion curves of the coplanar four-beam diffraction Ge[000, 440, 260, $\bar{2}20$]. — Parts ((a), (c) and (e): surface ($\bar{1}10$), parts (b) and (d): surface (110). — Non-absorbing crystal: (a), (c) and (e) below, (b) and (d) left .See text.

possible tie points only two are excited at an arbitrary angle of incidence of the incident wave. In Fig. 4 the excited branches are shown by thick lines, the unexcited by thin lines. The angular measure drawn corresponds, in reciprocal space units, to $2'' \Leftrightarrow 0.304 \times 10^6\ \mathrm{m}^{-1}$ in case of the (110) crystal surface and to $2'' \Leftrightarrow 0.152 \times 10^6\ \mathrm{m}^{-1}$ in case of the $(\bar{1}10)$ surface.

Conclusions

The influence of absorption for the coplanar four-beam diffraction $[000, 440, 260, \bar{2}20]$ in a perfect halfinfinite germanium monocrystal was studied.

It was found that the absorption causes not only a complicated reduction of intensity but also a redistribution of energy between the participating reflections. As a result of this redistribution of energy caused by the absorption some parts of a particular reflection curve may increase and also some new peaks may appear.

Acknowledgements

The authors would like to thank Prof. G. Hildebrandt and Dr. A. Fingerland for helpful and stimulating discussions.

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