DEPENDENCE OF THE ABSORPTION COEFFICIENT OF THE $1s$ BAND OF SINGLE CRYSTALS ON THE POLARIZATION AND ORIENTATION OF THE RADIATION

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L. K. IZRAILEVA

DEPENDENCE OF THE ABSORPTION COEFFICIENT OF THE $1s$ BAND OF SINGLE CRYSTALS ON THE POLARIZATION AND ORIENTATION OF THE RADIATION

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Taking into account the quasi-atomic character of the $1s$ states, their localization near the nucleus ($\chi r_{1s} \ll 1$, where χ is the wave vector of the x-ray photon, r_{1s} is the radius of the $1s$ state), and the translational symmetry of the crystal, the general form for the absorption coefficient τ_k of the $1s$ band of a simple lattice can be written in the form ([1])

$$\tau_k(\omega) \sim \sum_{i=1}^{l} \left| \int \psi_i(x, y, z) \mathbf{E}^0 \nabla \psi_{1s}(r)\, dr \right|^2 =$$

$$= \left| \int \psi_i(\mathbf{r}) \nabla \psi_{1s}(r)\, dr \right|^2 \sum_{t=1}^{l} (\mathbf{E}^0 \mathbf{n}_t)^2, \qquad (1)$$

where $\psi_i(x, y, z)$ is one of the l states with energy $\mathcal{E}_i = \mathcal{E}_{1s} + \hbar\omega$, and summation over t denotes summation over all directions of the vectors $|\int \psi_t \nabla \psi_{1s} dr|\, \mathbf{n}_t$, which transform into one another under transformations of the crystal symmetry (the length of the vector $|\int \psi_t \nabla \psi_{1s} dr|$ does not depend on t). The first factor in formula (1) is proportional to the corresponding absorption coefficient for a polycrystalline specimen

$$\vec{\tau}_k \sim \frac{l}{3} \left| \int \psi_i(\mathbf{r}) \nabla \psi_{1s}(r)\, dr \right|^2, \qquad (2)$$

while the second, depending on the polarization $\mathbf{E}^0$,

$$\sum_{t=1}^{l} (\mathbf{E}^0 \mathbf{n}_t)^2 = \sum_{i,j=1}^{3} E_i^0 E_j^0 \sum_{t=1}^{l} n_{ti} n_{tj}, \qquad (3)$$

is characteristic of the crystal; it is this factor that we shall study (generally speaking, (3) must also depend on the energy $\mathcal{E}_i$). For a crystal with several atoms in the unit cell, the expression for $\tau_k(\omega)$ will contain a sum of terms of type (1). In the short-wavelength region of the K absorption spectrum of a single crystal ($\hbar\omega > |\mathcal{E}_{1s}|$), in first-order perturbation theory, the influence of polarization, according to (2), is described by an additional term, relative to τ_k of a polycrystalline specimen, $\Delta\tau_k^{E^0}$:

$$\Delta\tau_k^{E^0} = \tau_k^0 \frac{2m}{\hbar^2 p} \int_{r_0}^{\infty} dr\, \sin(2pr + 2\eta_1) \left[-\frac{1}{\sqrt{5\pi}} V_{2,0}^{E^0}(r) \right], \tag{4}$$

where τ_k^0 and η_1 are, respectively, the absorption coefficient and the phase of the electron wave in the field of the "quasi-atom" for $l = 1$; r_0 is the radius of the "quasi-atom" ;

$$\hbar^2 p^2/2m = \mathcal{E}_i = \mathcal{E}_{1s} + \hbar\omega; \quad V_{2,0}^{E^0}(\mathbf{r})$$

is a component of the expansion of the crystal potential $V(\mathbf{r})$ in a series of spherical functions ($\mathbf{E}^0$ is the axis of rotation):

$$\begin{aligned} V_{2,0}^{E^0}(r) &= \iint V(r) Y_{20}^*(\theta,\varphi)\, d\Omega = \sqrt{\frac{5}{16\pi}} \iint V(r)(1 - 3\cos^2 \mathbf{E}^0\hat{\mathbf{r}})\, d\Omega \\ &= \sqrt{\frac{5}{16\pi}} \iint_S \frac{V(x,y,z)}{x^2+y^2+z^2} \left(1 - 3 \sum_{i,j=1}^{3} E_i^0 E_j^0 \frac{x_i x_j}{x^2+y^2+z^2} \right) dS. \end{aligned} \tag{5}$$

(In the last expression (5) we have changed to integration over the surface of a sphere of radius $|\mathbf{r}|$: $dS = r^2 d\Omega$.)

In what follows we shall consider (3) and (5) in parallel. First of all, let us note that (3), as well as (5), leads to the same result for all $\mathbf{E}^0$ related to one another by symmetry transformations of the crystal point group. Let us consider the dependences of (3) and (5), and consequently of the absorption coefficient τ_k (1), (4), on the direction of $\mathbf{E}^0$ for various crystal systems. For the triclinic system, symmetry gives no simplifications of (3) and (5)[3].

1. **Monoclinic system.** We place the axes x, y in the plane of the base of the parallelepiped, and z is the monoclinic axis perpendicular to the base. Since in this coordinate system $V(x,y,z) = V(x,y,-z)$, the terms in (3) and (5) with $E_i^0 E_z^0$ $(i = x, y)$ vanish, and we obtain:

$$\sum_{t=1}^{l} (\mathbf{E}^0 \mathbf{n}_t)^2 = \sum_{i,j=1}^{2} E_i^0 E_j^0 \sum_{t=0}^{l} n_{ti} n_{tj} + (E_z^0)^2 \sum_{t=1}^{l} n_{tz}^2, \tag{6}$$

$$V_{2,0}^{E^0}(r) = \sqrt{\frac{5}{16\pi}} \iint_S \frac{V(x,y,z)}{x^2+y^2+z^2} \left[1 - 3\sum_{i,j=1}^{2} E_i^0 E_j^0 \frac{x_i x_j}{r^2} - 3(E_z^0)^2 \frac{z^2}{r^2}\right] dS. \quad (7)$$

2. **Orthorhombic system.** The axes x, y, z are parallel to the edges of the base parallelepiped. Proceeding now from formulas (6) and (7) and additionally taking into account $V(x,y,z) = V(-x,y,z)$, $V(x,y,z) = V(x,-y,z)$, we arrive at the following expressions:

$$\sum_{t=1}^{l} (\mathbf{E}^0 \mathbf{n}_t)^2 = \sum_{i=1}^{3} (E_i^0)^2 \sum_{t=1}^{l} n_{ti}^2, \quad (8)$$

$$V_{2,0}^{E^0}(r) = \sqrt{\frac{5}{16\pi}} \iint_S \frac{V(r)}{r^2} \left(1 - 3\sum_{i=1}^{3} (E_i^0)^2 \frac{x_i^2}{r^2}\right) dS. \quad (9)$$

Let us note that for $|E_i^0|^2 = 1/3$, $V_{2,0}^{E^0} \equiv 0$, i.e. $\Delta\tau_k^{E^0} = 0$, while the sum (8), in view of

$$\sum_{i=1}^{3} n_{ti}^2 = 1,$$

is simply equal to $l/3$, and, consequently, τ_k in the case of such $\mathbf{E}^0$ is equal to τ_k (2) for a polycrystalline specimen.

3. For the rhombohedral, tetragonal, and hexagonal systems (z is a symmetry axis of order 3, 4, or 6; the axes x, y are arbitrary in the plane perpendicular to the z-axis), owing to the presence of the symmetry axis, the relations

$$\sum_{t=1}^{l} n_{tx}^2 = \sum_{t=1}^{l} n_{ty}^2, \qquad \iint_S V(r) \frac{x^2}{r^4}\, dS = \iint_S V(r) \frac{y^2}{r^4}\, dS;$$

will be valid, as is not difficult to verify, and therefore (8) and (9) take the following form:

$$\sum_{t=1}^{l} (\mathbf{E}^0 \mathbf{n}_t)^2 = (E_\parallel^0)^2 \sum_{t=1}^{l} n_{tz}^2 + (E_\perp^0)^2 \frac{1}{2} \sum_{t=1}^{l} (n_{tx}^2 + n_{ty}^2), \quad (10)$$

$$V_{2,0}^{E^0}(r) = \sqrt{\frac{5}{16\pi}} \iint_S \frac{V(r)}{r^2} \left[1 - 3(E_\parallel^0)^2 \frac{z^2}{r^2} - 3(E_\perp^0)^2 \frac{1}{2} \frac{x^2+y^2}{r^2}\right] dS. \quad (11)$$

As is seen from (10) and (11), the absorption coefficient τ_k of single crystals of the indicated three systems is completely independent of the direction $E_\perp^0(E_x^0, E_y^0)$—the projection of $\mathbf{E}^0$ onto the plane perpendicular to the symmetry axis. At the same time, for all polarization directions of $\mathbf{E}^0$ ($E_\perp^{02}=2/3, E_\parallel^{02}=1/3$), forming a cone with angle χ ($|\cos\chi|=1/\sqrt{3}$) about the symmetry axis, the absorption coefficient of the single crystal τ_k coincides with τ_k for a polycrystalline specimen, since

$$\sum_{t=1}^{l}(\mathbf{E}^0\mathbf{n}_t)^2=\frac{1}{3}\sum_{t=1}^{l}\sum_{i=1}^{3}n_{ti}^2=\frac{l}{3},\qquad V_{2,0}^{E^0}(r)=0\quad\text{for }\mathbf{E}^0(E_\perp^{02}=2/3,\ E_\parallel^{02}=1/3).$$

Moreover, it follows from (10) and (11) that, when the radiation is directed parallel to the symmetry axis ($E_\parallel^0=0$, $(E_\perp^0)^2=1$), τ_k of the single crystal depends neither on the direction of polarization nor on whether the incident radiation is polarized. Indeed, (10) and (11) in this case are respectively equal to

$$E_\parallel^0=0,\qquad E_\perp^{0\,2}=1,$$

$$\sum_{t=1}^{l}(\mathbf{E}^0\mathbf{n}_t)^2=\frac{1}{2}\sum_{t=1}^{l}\left(n_{tx}^2+n_{ty}^2\right)=\frac{1}{2}\left(l-\sum_{t=0}^{l}n_{tz}^2\right), \qquad (10')$$

$$V_{2,0}^{E^0}(r)=-\frac{1}{2}\sqrt{\frac{5}{16\pi}}\iint\limits_{S}\frac{V(r)}{r^2}\left(1-3\frac{z^2}{r^2}\right)dS. \qquad (11')$$

In the case of unpolarized radiation, τ_k of the single crystal depends on the direction of the wave vector $\vec{\chi}$; the form of this dependence is obtained by averaging (3) in the plane perpendicular to $\vec{\chi}$:

$$\overline{\sum_{t=1}^{l}(\mathbf{E}^0\mathbf{n}_t)^2}=\sum_{t=1}^{l}\sin^2\vec{\chi}\,\hat{\mathbf{n}}_t\,\overline{\cos^2\varphi_{E^0}}=\frac{1}{2}\left[l-\sum_{t=1}^{l}(\vec{\chi}\mathbf{n}_t)^2\right] \qquad (12)$$

(In the crystal systems considered by us, the sum in (12) has the form, respectively, of (6), (8), and (10), if E_i^0 is replaced in them by χ_i.) For $\vec{\chi}$ ($\chi_x=\chi_y=0$, $\chi_z^2=1$) (radiation parallel to the symmetry axis), formula (12) is identical to (10′).

In the short-wavelength region of the K-absorption spectrum of a single crystal, in the case of unpolarized radiation, the additional term analogous to (4) has the form [2]

$$\Delta\tau_k^{\vec{\chi}} = \tau_k^0 \frac{2m}{\hbar^2\rho} \int_{r_0}^{\infty} dr\, \sin(2\rho r + 2\eta_1) \left[\frac{1}{2\sqrt{5\pi}} V_{2,0}^{\vec{\chi}}(r) \right], \tag{13}$$

where $V_{2,0}^{\vec{\chi}}(r)$ is the term of the series for $V(r)$ analogous to (5) ($\vec{\chi}$ is the axis of rotation).

In particular, from (11), for $\vec{\chi}$ ($\chi_x = \chi_y = 0,\ \chi_z^2 = 1$), it follows that

$$V_{2,0}^{\vec{\chi}}(r) = \sqrt{\frac{5}{16\pi}} \iint\limits_S \frac{V(r)}{r^2} \left(1 - 3\frac{z^2}{r^2}\right) dS. \tag{14}$$

Substituting (11′) into (4), and (14) into (13), we arrive at the conclusion that the formulas for the short-wavelength region, (4) and (13), as well as the general expressions (10′) and (12), are identical in the cases of radiation polarized or unpolarized parallel to the symmetry axis. Moreover, for unpolarized radiation, from (12) and (13), taking (10) and (11) into account, it follows that τ_k does not depend on the direction $\chi_\perp(\chi_x, \chi_y)$—the projection of $\vec{\chi}$ onto the plane perpendicular to the symmetry axis. Therefore, when the radiation is directed

perpendicular to the axis of symmetry ($\chi_\parallel = 0,\ \chi_\perp^2 = 1$), τ_k does not depend on the orientation of the beam

$$\chi_\parallel = 0, \quad \chi_\perp^2 = 1,$$

$$\overline{\sum_{t=1}^{l} (\mathbf{E}^0 \mathbf{n}_t)^2} = \frac{1}{2}\left[l - \frac{1}{2}\sum_{t=1}^{l}(n_{tx}^2 + n_{ty}^2) \right] = \frac{1}{4}\left(l + \sum_{t=1}^{l} n_{tz}^2 \right), \tag{15}$$

$$V_{2,0}^{\vec{\chi}}(r) = -\frac{1}{2}\sqrt{\frac{5}{16\pi}} \iint_S \frac{V(r)}{r^2}\left(1 - 3\frac{z^2}{r^2}\right) dS. \tag{16}$$

If the radiation is polarized parallel to the axis of symmetry ($E_\perp^0 = 0$), (10) and (11) take the form

$$E_\perp^0 = 0, \quad (E_\parallel^0)^2 = 1, \quad \sum_{t=1}^{l} (\mathbf{E}^0 \mathbf{n}_t)^2 = \sum_{t=1}^{l} n_{tz}^2, \tag{10”}$$

$$V_{2,0}^{\mathbf{E}^0}(r) = \sqrt{\frac{5}{16\pi}} \iint_S \frac{V(r)}{r^2}\left(1 - 3\frac{z^2}{r^2}\right) dS. \tag{11”}$$

Separating in formulas (10′), (10″), and (15) the constant $l/3$, characteristic of a polycrystalline sample, we find that between the additional terms $\Delta\tau_k = \tau_k - \overline{\tau_k}$

in the three cases considered by us there hold the relations (17), independently of the photon energy $\hbar\omega$:

$$\Delta\tau_k(E^0_\parallel = 0,\ E^{02}_\perp = 1) = -\frac{1}{2}\Delta\tau_k(E^{02}_\parallel = 1,\ E^0_\perp = 0) =$$

$$= \Delta\tau_k(\chi^2_\parallel = 1,\ \chi_\perp = 0) = -2\Delta\tau_k(\chi_\parallel = 0,\ \chi^2_\perp = 1). \tag{17}$$

Formulas (4) and (13) for the short-wavelength region lead to the same relations if one substitutes (11′) and (11″) into (14), and (16) into (13).

4. For crystals of the cubic system, from (11) it is easy to obtain that $V^{\mathbf{E}^0}_{2,0}(r) = 0$ for any $\mathbf{E}^0$; at the same time in (4) $\Delta\tau^E_k = 0$, and, consequently, our formulas for the short-wavelength region in this case are identical for single crystals and polycrystalline samples, in full agreement with assertion (1).

We note that all the results obtained for crystals with an axis of symmetry retain their meaning also in the case when the axis is a screw axis; only instead of (1) one must consider the sum over atoms of one kind that are related to one another by transformations about the axis of symmetry.

The principal results of the latest experiments (4) on the study of the K-absorption spectrum of a single crystal of gallium (rhombic system) consist in the fact that, as the angle between the polarization direction $\mathbf{E}^0$ and the base plane of the parallelepiped—the plane (a, b) (or between the plane perpendicular to $\vec{\chi}$ and the plane (a, b) in the case of unpolarized radiation)—increases, there occurs a noticeable decrease in the contrast of the fine structure of the K-spectrum. This is apparently a consequence of the change in the components $V^{\mathbf{E}^0}_{2,0}(r)$ or $V^{\vec{\chi}}_{2,0}(r)$ of the gallium potential according to formulas (4) and (13). We note that in the case of complete linear polarization one can change the direction of the beam in the crystal without changing the direction of $\mathbf{E}^0$; in this case, as is seen from (1), $\tau_k(\omega)$ does not change. The weak change, observed in (4), of the contrast of the fine structure in going from $\mathbf{E}^0 \parallel a,\ \vec{\chi} \parallel c$ to $\mathbf{E}^0 \parallel a,\ \widehat{\chi c} = 52^\circ$ may be connected with quadrupole transitions.

Institute of Geology of Ore Deposits,
Petrography, Mineralogy, and Geochemistry

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