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THE RELATIONSHIP BETWEEN THE ANOMALOUS TRANSMISSION AND THE NEGATIVE EXTINCTION IN THE BRAGG CASE

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The relationship between the anomalous transmission and the negative extinction in the symmetrical Bragg case near the absorption edge is studied in detail. The negative extinction results from the anomalous transmission and takes the largest value when the diffraction is induced only by the imaginary part of the atomic scattering factor near the absorption edge. The appearance of the negative extinction also depends on temperature, which can be used for the identification of the theoretical model for calculating the Debye-Waller factor.

1. Introduction

X-ray diffraction theories are called dynamical or kinetic, according to whether the multi-scattering effect is taken into account or not [1-5]. The observation of Borrmann effect and *Pendellösung* fringes proves the validity of the dynamical theory.

With X-rays from a synchrotron radiation source, it is possible to make $|F_{hr}|$ smaller than $|F_{hi}|$, even $F_{hr} = 0$, near an absorption edge of a constitute atom of a perfect crystal

[6-7]. Here, F_{hr} and F_{hi} are the contributions of the real and imaginary parts, respectively, of the atomic scattering factor to the crystal structure factor. In 1992, Kato presented the general formulae for calculating the reflectivity for a semi-infinite crystal, which is valid even when $F_{hr} = 0$ [8]. The corresponding formulae for a finite parallel-plane crystal were derived by Xu Zhangcheng et al. in 1995 [9]. More recently, the present authors found that the famous Kato-Ewald theorem is not valid when $|F_{hi}|$ is comparative to $|F_{hr}|$ near the absorption edge [10,11].

It is well known, the extinction effect in the Bragg case under normal experimental conditions is one of the most important dynamical effects. This effect manifests itself not only in the integrated intensity, but also in the secondary fluorescence emission [3,12]. However, the anomalous transmission can take place at the exact Bragg angle in the Bragg case when $|F_{hr}|$ is smaller than $|F_{hi}|$ near the absorption edge, as predicted in Ref. 9. The purpose of the present work is to show how such an effect affects the reflectivity, especially the integrated intensity for the perfect crystal.

2. Theoretical basis

The parameters used in the present work are physically the same with those in Ref. 1, avoiding the problem of infinity when $F_{hr} = 0$ [8,9].

2.1. The reflection and transmission coefficients for a finite perfect crystal [9]

The reflectivity in the Bragg case for a finite perfect crystal is given by

$$\frac{P_h}{P_0} = (1 - 2p\sin\delta)\frac{\cosh(2AI_1) - \cos(2AR_1)}{P_1}, \tag{1}$$

where

$$P_1 = (y^2 + g^2 + R_1^2 + I_1^2)\cosh(2AI_1) - 2(yR_1 + gI_1)\sinh(2AI_1) - \tag{2}$$

$$(y^2 + g^2 - R_1^2 - I_1^2)\cos(2AR_1) + 2(yI_1 - gR_1)\sin(2AR_1),$$

$$R_1 = \pm\sqrt{(L_1 + B)/2}, \tag{3}$$

$$I_1 = \pm\sqrt{(L_1 - B)/2}, \tag{4}$$

$$L_1 = \sqrt{B^2 + C^2}, \tag{5}$$

$$B = y^2 - g^2 - 1 + a^2, \tag{6}$$

$$C = 2(gy - p\cos\delta), \tag{7}$$

$$g = \frac{(1+|b|)\,\chi_{0i}}{2\sqrt{b}\,K\,|\overline{\chi_h}|}, \tag{8}$$

$$y = \frac{|b|\,(\theta-\theta_B)\sin 2\theta_B + \frac{1}{2}(1+|b|)\,\chi_{0r}}{\sqrt{b}\,K\,|\overline{\chi_h}|}, \tag{9}$$

$$b = -\sin(\Theta_B+\alpha)/\sin(\Theta-\alpha), \tag{10}$$

$$A = \frac{\pi K|\chi_h|}{\sqrt{\sin(\Theta_b+\alpha)\sin(\Theta_B-\alpha)}}, \tag{11}$$

$$p = \frac{|\chi_{hr}\,\chi_{hi}|}{|\overline{\chi_h}|^2}, \tag{12}$$

$$a = \frac{\sqrt{2}\,\chi_{hi}}{|\overline{\chi_h}|^2}, \tag{13}$$

$$|\overline{\chi_h}|^2 = \sqrt{|\chi_{hr}|^2+|\chi_{hi}|^2}, \tag{14}$$

$$\chi_{hr} = |\chi_{hr}|\,\mathrm{e}^{\mathrm{i}\alpha_{hr}} = -\frac{e^2}{V\pi m\nu^2}F_{hr}, \tag{15}$$

$$\chi_{hi} = |\chi_{hi}|\,\mathrm{e}^{\mathrm{i}\alpha_{hi}} = -\frac{e^2}{V\pi m\nu^2}F_{hi}, \tag{16}$$

$$\delta = \alpha_{hi}-\alpha_{hr}, \tag{17}$$

$$\chi_h = \chi_{hr}+\mathrm{i}\,\chi_{hi}. \tag{18}$$

In (15) and (16), V is the volume of a crystal cell, ν the X-ray frequency, and m and e the mass and the charge of an electron. In (11), Θ_B is the Bragg angle, α the angle between the diffraction plane and the crystal surface, and K the wavenumber in vacuum. In (3) the negative sign is taken only when $B<0$ and $C<0$, but in (4) only when $B>0$ and $C<0$. The transmission coefficient is

$$\frac{P_d}{P_0} = \frac{L_1\exp\{2Ag(1+b)/(1-b)\}}{P_1}. \tag{19}$$

2.2. Integrated intensity

The integrated intensity for a finite perfect crystal is given by

$$R_{dyn} = \frac{\sqrt{|b|}\ |\overline{\chi_h}|}{\sin 2\theta} R^y_{dyn}(A_0, g_0, k', b) = \frac{\sqrt{|b|}\ |\overline{\chi_h}|}{\sin 2\theta} \int_{-\infty}^{\infty} \frac{P_h}{P_0} \mathrm{d}y. \tag{20}$$

The integrated intensity for an ideally imperfect crystal is given by

$$R_{kin} = \frac{\sqrt{|b|}\ |\overline{\chi_h}|}{\sin 2\theta} R^y_{kin}(A_0, g_0, k', b), \tag{21}$$

where

$$R^y_{kin}(A_0, g_0, k', b) = (1 - \mathrm{e}^{4Ag}) \frac{\pi}{4} \frac{(1 - 2p \sin\delta)}{|g|}. \tag{22}$$

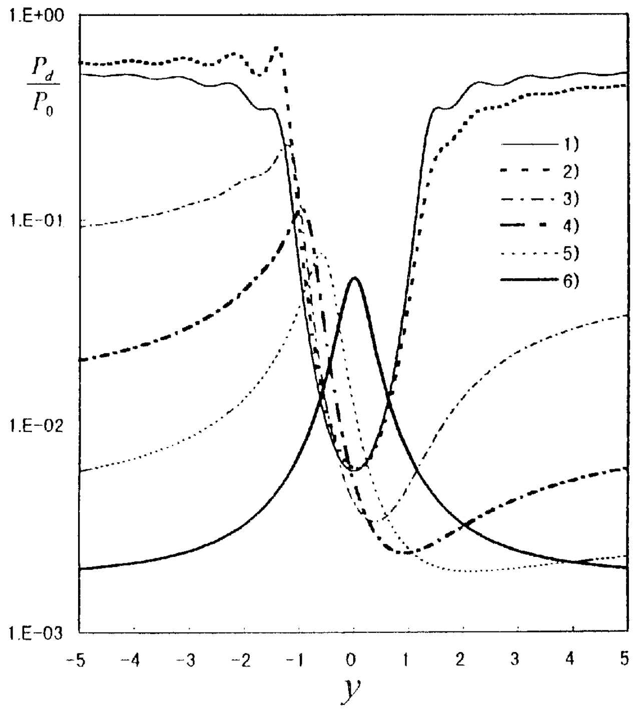

Fig. 1. The rocking curves of the transmitted beam of a finite parallel-plane crystal. The parameters are: $A = \pi, \delta = 0$, (1) $\chi_{hi} = 0, g = -0.1$, (2) $|\chi_{hr}| = 10\ |\chi_{hi}|, g = -1.0$, (3) $|\chi_{hr}| = 6\ |\chi_{hi}|, g = -1.0$, (4) $|\chi_{hr}| = 2\ |\chi_{hi}|, g = -1.0$, (5) $|\chi_{hr}| = \ |\chi_{hi}|, g = -1.0$ and (6) $\chi_{hr} = 0, g = -1.0$.

When

$$A \to \infty \qquad (\text{if } g \neq 0), \qquad e^{4Ag} \to 0 \tag{23}$$

$$k' = \frac{|\chi_{hr}|}{|\chi_{hi}|}, \tag{24}$$

$$p = k'/(1+k'^2), \tag{25}$$

$$a = \sqrt{2/(1+k'^2)}. \tag{26}$$

3. The rocking curves of the transmitted beams

According to Eq. (19), the rocking curves of the transmitted beam of a finite parallel-plane crystal in the symmetrical Bragg case are calculated for all kinds of absorption cases. When $\chi_{hi} = 0$ (Fig. 1, curve 1), the curve shows a symmetrical dip with respect to $y = 0$; when $\chi_{hr}\chi_{hi} \neq 0$ (curves 2 to 5), the curve is not symmetrical to $y = 0$, which is due to the asymmetry of the absorption coefficient. The anomalous transmission in the region $y < 0$ and the anomalous absorption in the region $y > 0$ are clearly seen. When $|\chi_{hr}| >> |\chi_{hi}|$, the anomalous transmission is

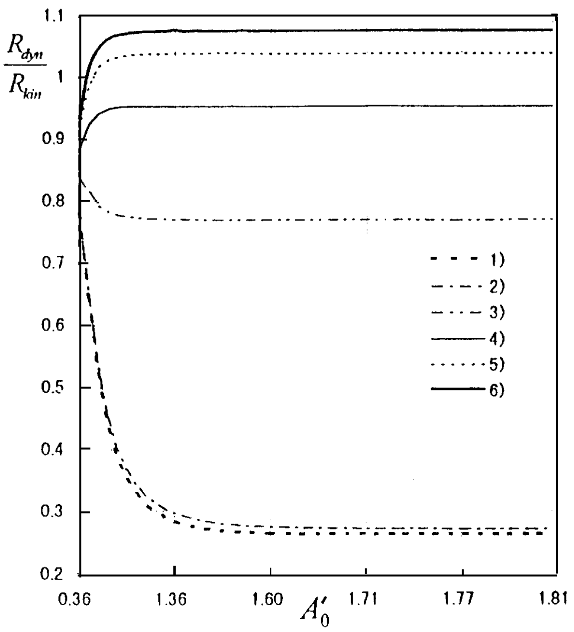

Fig. 2. The dependence of R_{dyn}/R_{kin} on the ratio of $|\chi_{hr}|$ and $|\chi_{hi}|$. The parameters are: $g_0 = -1, A'_0 = 2A/(A+\pi)$, $\delta = 0$, (1) $|\chi_{hr}| = 10\,|\chi_{hi}|$, (2) $|\chi_{hr}| = 8\,|\chi_{hi}|$, (3) $|\chi_{hr}| = 6\,|\chi_{hi}|$, (4) $|\chi_{hr}| = 4\,|\chi_{hi}|$, (5) $|\chi_{hr}| = |\chi_{hi}|$, (6) $|\chi_{hr}| = 0$.

very small. Note that the anomalous transmission occurs in the so-called "total reflection" region, when $|\chi_{hr}| < |\chi_{hi}|$. When $\chi_{hr} = 0$ (curve 6), the rocking curve shows a symmetrical peak with respect to $y = 0$, which means the anomalous transmission is the largest at exact Bragg angle.

4. Primary extinction and negative primary extinction

Darwin first predicted the primary extinction phenomena, when he constructed his dynamical diffraction theory. That is to say, when reflection occurs, the transmission depth of X-rays is much shallower than with normal absorption. Only several thousands of atomic layers near the crystal surface contribute to the diffraction and the contribution of the lower part is nearly zero. However, the anomalous transmission occurs in the so-called "total reflection" region, which means more atomic layers contribute to the diffraction in the perfect crystal than in the corresponding ideally imperfect crystal. This will result in the appearance of negative primary extinction - the integrated intensity for a perfect crystal being larger than that for a corresponding ideally imperfect crystal.

As shown in Fig. 2, R_{dyn}/R_{kin} is calculated for typical absorption cases by direct integration of the reflected rocking curves. The parameter $g_0 = \chi_{0i}/|\chi_{hi}|$ is set to – 1, which means the effect of temperature is not taken into account. It can

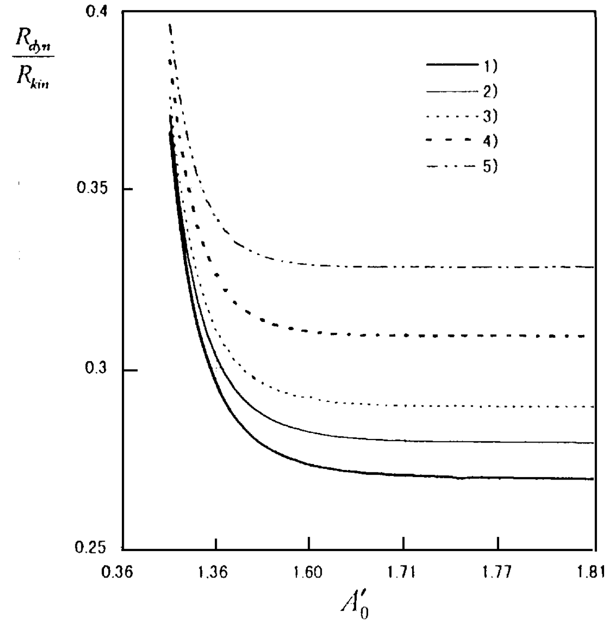

Fig. 3a. The dependence of R_{dyn}/R_{kin} on temperature. The parameters are: $A'_0 = 2A/(A+\pi)$, $\delta = 0$, $|\chi_{hr}| = 10\ |\chi_{hi}|$, (1) $g_0 = -1.0$, (2) $g_0 = -1.05$, (3) $g_0 = -1.1$, (4) $g_0 = -1.2$, and (5) $g_0 = -1.5$.

be seen that R_{dyn}/R_{kin} increases with the decreasing of k'. When $|\chi_{hr}| \geq |\chi_{hi}|$ (curves 1 to 4), $R_{dyn}/R_{kin} < 1$, which is called the primary extinction. When $|\chi_{hr}| < |\chi_{hi}|$, (curves 5,6), $R_{dyn}/R_{kin} > 1$, which is called the negative primary extinction. When diffraction is induced only by the imaginary part the atomic scattering factor ($\chi_{hr} = 0$), the negative primary extinction takes the largest value. As shown in Fig. 3, the effect of temperature on R_{dyn}/R_{kin} is studied. When $|\chi_{hr}| >> |\chi_{hi}|$, (Fig. 3a), R_{dyn}/R_{kin} increases and tends to 1 when the temperature increases (g_0 increasing). However, when $\chi_{hr} = 0$ (Fig. 3b), R_{dyn}/R_{kin} decreases and tends to 1 when the temperature increases. If the temperature is high enough, the negative primary extinction will disappear, which may be used for the identification of the theoretical model for calculating the Debye-Waller factor.

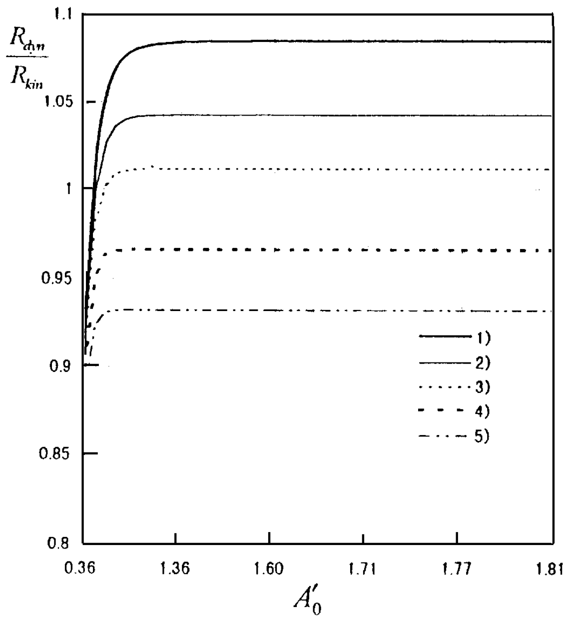

Fig. 3b. The dependence of R_{dyn}/R_{kin} on temperature. The parameters are the same as of Fig. 3a, except for $\chi_{hr} = 0$.

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ODNOS ANOMALNE TRANSMISIJE I NEGATIVNOG GAŠENJA U BRAGGOVOM SLUČAJU

Podrobno se razmatra odnos anomalne transmisije i negativnog gašenja za simetričan Braggov slučaj. Negativno gašenje je posljedica anomalne transmisije i najveće je kada je difrakcija uzrokovana samim imaginarnim dijelom atomskog faktora raspršenja u blizini apsorpcijskog ruba. Pojava negativnog gašenja ovisi takoder o temperaturi, što se može primijeniti za prepoznavanje teorijskog modela pri izračunavanju Debye-Wallerovog faktora.