Dynamic model of anisotropic x-ray refraction

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General mechanisms of anisotropic x-ray refraction at the resonance energy are investigated on the basis of dynamic-scattering theory. The deductions show that x rays within the crystals that have anisotropic susceptibility are completely polarized and have two elliptical polarization states. Analytical expressions of the elliptical axes, refractive indices, and absorption coefficients for these two types of polarized waves are obtained in terms of the anisotropic components of the susceptibility tensor. Anisotropic birefringence and dichroism effects associated with the polarization properties of the x-ray waves are also illustrated theoretically. [S0163-1829(97)06034-7]

I. INTRODUCTION

The conventional theories of x-ray refraction and diffraction have been well elaborated on the general assumption that the electric susceptibility of crystals is isotropic in the x-ray frequency range.[1,2] While this assumption is usually justified, it may be invalid when the x-ray energy is close to an atomic absorption edge. It has been shown both experimentally and theoretically that the x-ray susceptibility near an absorption edge is slightly anisotropic, mainly as a consequence of the anomalous resonance effect related to the chemical bonds and local environment of the atom in crystals. It is this small anisotropy of susceptibility that gives rise to a series of anisotropic anomalous-scattering phenomena, such as the energy-dependent dichroism, birefringence, and forbidden-reflection diffraction.[3–7]

As anisotropic scattering occurs, the x-ray susceptibility can be represented by a second-rank tensor $\hat{\boldsymbol{\chi}}(\mathbf{r})$ (within the dipole approximation) which gives at every point $\mathbf{r}$ the local relationship between the electric field $\mathbf{E}(\mathbf{r})$ and the induced polarization $\mathbf{P}(\mathbf{r})$: $\mathbf{P}(\mathbf{r})\propto\hat{\boldsymbol{\chi}}(\mathbf{r})\mathbf{E}(\mathbf{r})$.[5] Generally, $\mathbf{P}(\mathbf{r})$ is no longer parallel to $\mathbf{E}(\mathbf{r})$, and similar to the optics of visible light, only x-ray waves with specific polarization may propagate through the crystal. Since the Fourier components $\hat{\boldsymbol{\chi}}_g$ of the periodic $\hat{\boldsymbol{\chi}}(\mathbf{r})$ also have tensorial forms, the polarization phenomena may be explained by the anisotropic structure amplitudes $\hat{\mathbf{F}}_g$ (proportional to $\hat{\boldsymbol{\chi}}_g$) in the framework of kinematic-scattering theories, as demonstrated in some previous investigations.[5,8,9] Nevertheless, the kinematic model mainly deals with global refracted and diffracted intensities; it cannot provide information concerning the wave phase, propagation, and interaction, though it is such information that indicates the intrinsic mechanism of x-ray polarization.[10,11] To fully understand the wave behaviors in anisotropic x-ray refraction and diffraction, therefore, one has to resort to dynamic-scattering theories.

It is known that the dynamic theory can give explicitly the properties of propagating x-ray waves by taking into account all the interactions between the refracted and diffracted waves and the crystals.[2] For anisotropic scattering, the interaction between the waves and the crystals [represented by the susceptibility tensor $\hat{\boldsymbol{\chi}}(\mathbf{r})$] is essential: It determines the polarization and anisotropic absorption of the x rays propagating along any directions. In fact, all the elements of $\hat{\boldsymbol{\chi}}(\mathbf{r})$ are energy-dependent complex quantities near the absorption edge due to the complex anomalous-dispersion corrections. This makes the x-ray waves within the crystal elliptically (or at least linearly) polarized. Meanwhile, the anisotropic absorption phenomena are directly related to the imaginary part of the susceptibility.[5,12,13] In the viewpoint of dynamic scattering, therefore, the only task is to find the actual forms of the x-ray waves in crystals.

It is our purpose to show in the present work how the general principles governing the anisotropic x-ray refraction and absorption can be deduced from dynamic wave theories. In fact, we give an exact description of the elliptical polarization properties of anisotropically refracted x rays propagating in crystals. Analytical expressions for the refractive indices and absorption coefficients of x-ray waves are obtained in terms of the anisotropic parts of the susceptibility, from which the intrinsic mechanisms of x-ray birefringence and dichroism are revealed explicitly.

II. WAVE EQUATION OF ANISOTROPIC REFRACTION

For anisotropic x-ray optics in single crystals, the electric susceptibility is described by a second-rank tensor $\hat{\boldsymbol{\chi}}$ which is invariant against the symmetry transformations of the space group and has the symmetry $\chi^{ij}=\chi^{ji}$.[5] Because all the susceptibility elements χ^{ij} are small (typically less than 10^{-4}) in the x-ray frequency range, the electrically induced polarization can be written as

$$\mathbf{P}=\mathbf{D}-\mathbf{E}=[\hat{\mathbf{I}}-(\hat{\mathbf{I}}+\hat{\boldsymbol{\chi}})^{-1}]\mathbf{D}\simeq\hat{\boldsymbol{\chi}}\mathbf{D}, \tag{1}$$

where $\hat{\mathbf{I}}$ is the unit matrix and $\mathbf{D}$ is the electric displacement. Based on this relation, Maxwell's equations have the general solutions

$$\mathbf{D}_j=\frac{1}{K^2-\mathbf{K}_j\cdot\mathbf{K}_j}\sum_n \mathbf{K}_j\times[\mathbf{K}_j\times(\hat{\boldsymbol{\chi}}_{j-n}\mathbf{D}_n)], \tag{2}$$

where K is the vacuum wave vector, the $\hat{\chi}_n$ are the Fourier components of the susceptibility which have the same tensorial form as $\hat{\chi}$, and $\mathbf{D}_j$ and $\mathbf{K}_j$ are the amplitude and wave vector of the jth Bloch wave, respectively. Equation (2) represents the basic wave equations for anisotropic x-ray scattering,[10] from which a series of polarization phenomena can be deduced.

In the case that there are only refracted waves in the crystal, Eq. (2) reduces to the ''one-beam'' equation

$$\mathbf{D}_0 = \frac{1}{2\eta_0}(\hat{\chi}_0 \mathbf{D}_0)_{[K_0]}, \tag{3}$$

where

$$\eta_0 = \frac{K_0^2 - K^2}{2K_0^2} \tag{4}$$

is a small wave vector correction associated with x-ray refraction and $(\hat{\chi}_0 \mathbf{D}_0)_{[K_0]}$ is the perpendicular component of $\hat{\chi}_0 \mathbf{D}_0$ with respect to the wave-propagating direction $\mathbf{K}_0$. As mentioned in Sec. I, anisotropic x-ray refraction and diffraction usually occur near the atomic absorption edges. In such frequency ranges, all the elements of the susceptibility are complex and the susceptibility tensor may be simply written as $\hat{\chi}_0 + i\hat{\chi}_0'$. As a consequence, the wave vector $\mathbf{K}_0$ is complex and has the form

$$\mathbf{K}_0 = \mathbf{K}_0^r + i\mathbf{K}_0^i, \tag{5}$$

where the small imaginary part $\mathbf{K}_0^i$ is perpendicular to the entrance plane according to the boundary condition.[2] Therefore, Eqs. (3) and (4) are rigorous only when $\mathbf{K}_0^i$ is parallel to $\mathbf{K}_0^r$. This actually corresponds to the case of perpendicular incidence in which $K_0 = K_0^r + iK_0^i$. For the general cases of inclined incidence, one can prove that, to a good approximation, $\mathbf{K}_0$ is nearly parallel to $\mathbf{K}_0^r$ with $K_0 \simeq K_0^r + iK_0^i \cos(\mathbf{K}_0^r \cdot \mathbf{K}_0^i)$.

As η_0 and the susceptibility tensor are complex, the wave obtained from Eq. (3) is, in general, *elliptically* polarized. Then the electric displacement of the elliptical wave has the form[11]

$$\mathbf{D}_0 = D_0(\mathbf{d}_1 + ip\mathbf{d}_2), \tag{6}$$

where the unit vectors $\mathbf{d}_1$ and $\mathbf{d}_2$ are parallel to the major and minor axes of the ellipse, respectively ($\mathbf{d}_1 \cdot \mathbf{d}_2 = 0$), and the ellipticity is described by $p(|p| \leqslant 1)$ which also determines the rotation direction of the electric displacement. Making the substitution $2\eta_0 = a + ib$ (a and b are real numbers) and inserting Eq. (6) into Eq. (3), one can obtain the scalar forms of Eq. (3) as

$$a = (\hat{\chi}_0 \mathbf{d}_1)_{[K_0]} \cdot \mathbf{d}_1 - p(\hat{\chi}_0' \mathbf{d}_2)_{[K_0]} \cdot \mathbf{d}_1, \tag{7a}$$

$$b = p(\hat{\chi}_0 \mathbf{d}_2)_{[K_0]} \cdot \mathbf{d}_1 + (\hat{\chi}_0' \mathbf{d}_1)_{[K_0]} \cdot \mathbf{d}_1, \tag{7b}$$

$$bp = p(\hat{\chi}_0' \mathbf{d}_2)_{[K_0]} \cdot \mathbf{d}_2 - (\hat{\chi}_0 \mathbf{d}_1)_{[K_0]} \cdot \mathbf{d}_2, \tag{7c}$$

$$ap = p(\hat{\chi}_0 \mathbf{d}_2)_{[K_0]} \cdot \mathbf{d}_2 + (\hat{\chi}_0' \mathbf{d}_1)_{[K_0]} \cdot \mathbf{d}_2. \tag{7d}$$

Equations (7a), (7b), (7c), and (7d) are the wave equations of anisotropic x-ray refraction, from which the wave parameters a, b, p, and $\mathbf{d}_1$ (or $\mathbf{d}_2$) can be obtained.

III. TWO POLARIZATION STATES OF REFRACTED WAVES

The susceptibility tensor $\hat{\chi}_0 + i\hat{\chi}_0'$ of a crystal is usually expressed with respect to the conventional Cartesian coordinate system (here called the ''$x_0y_0z_0$ system'') of the related crystallographic system. In order to deduce the actual form of the refracted wave from Eq. (7), it is convenient to build up a new Cartesian coordinate system xyz in the following two steps. First, the z_0 axis is rotated to be parallel to the wave-propagating direction $\mathbf{K}_0$ (or $\mathbf{K}_0^r$ for inclined incidence), so that an intermediate system $x'y'z'$ is established. If the corresponding rotation matrix is $\mathbf{R}_1$, the susceptibility tensor in the $x'y'z'$ system becomes $\hat{\chi}_p + i\hat{\chi}_q = \mathbf{R}_1 \hat{\chi}_0 \mathbf{R}_1^{-1} + i\mathbf{R}_1 \hat{\chi}_0' \mathbf{R}_1^{-1}$. It is evident that the transformed susceptibility is still symmetric (i.e., $\chi_p^{ij} + i\chi_q^{ij} = \chi_p^{ji} + i\chi_q^{ji}$). Then one can further rotate the z' axis by an angle

$$\theta_z = \frac{1}{2}\tan^{-1}\frac{2\chi_p^{12}}{\chi_p^{22} - \chi_p^{11}} \tag{8}$$

to generate the xyz system in which the susceptibility tensor is transformed to have the form

$$\begin{pmatrix} \chi_r^{11} & 0 & \chi_r^{13} \\ 0 & \chi_r^{22} & \chi_r^{23} \\ \chi_r^{13} & \chi_r^{23} & \chi_r^{33} \end{pmatrix} + i \begin{pmatrix} \chi_s^{11} & \chi_s^{12} & \chi_s^{13} \\ \chi_s^{12} & \chi_s^{22} & \chi_s^{23} \\ \chi_s^{13} & \chi_s^{23} & \chi_s^{33} \end{pmatrix}, \tag{9}$$

with $\chi_r^{12} = 0$. If the transformation matrix corresponding to the second rotation is written as $\mathbf{R}_2$, $\hat{\chi}_r + i\hat{\chi}_s$ is related to the original susceptibility by

$$\hat{\chi}_r + i\hat{\chi}_s = \mathbf{R}_2 \mathbf{R}_1 \hat{\chi}_0 \mathbf{R}_1^{-1} \mathbf{R}_2^{-1} + \mathbf{R}_2 \mathbf{R}_1 \hat{\chi}_0' \mathbf{R}_1^{-1} \mathbf{R}_2^{-1}. \tag{10}$$

It is noticeable that for the transformation associated with an arbitrary propagating direction $\mathbf{K}_0$, the imaginary off-diagonal element χ_s^{12} does not necessarily vanish. In the following, we generally assume that $\chi_s^{12} \neq 0$.

In the xyz coordinate system, both $\mathbf{d}_1$ and $\mathbf{d}_2$ are perpendicular to $\mathbf{K}_0$ (z axis), and they can be represented as

$$\mathbf{d}_1 = \cos\theta \hat{\mathbf{i}} + \sin\theta \hat{\mathbf{j}},$$

$$\mathbf{d}_2 = -\sin\theta \hat{\mathbf{i}} + \cos\theta \hat{\mathbf{j}}, \tag{11}$$

respectively, where $\hat{\mathbf{i}}$, $\hat{\mathbf{j}}$, and $\hat{\mathbf{k}}$ are the unit vectors along the x, y, and z axes, respectively. Introducing the present $\mathbf{d}_1, \mathbf{d}_2$, and susceptibility tensor into Eq. (7) enables one to rewrite the equations as

$$a = \chi_r^{11}\cos^2\theta + \chi_r^{22}\sin^2\theta + \frac{p}{2}v\sin 2\theta - pw\cos 2\theta, \tag{12a}$$

$$b=-\frac{p}{2}u\sin 2\theta+\chi_s^{11}\cos^2\theta+\chi_s^{22}\sin^2\theta+w\sin 2\theta, \tag{12b}$$

$$bp=p(\chi_s^{11}\sin^2\theta+\chi_s^{22}\cos^2\theta-w\sin 2\theta)+\frac{1}{2}u\sin 2\theta, \tag{12c}$$

$$ap=p(\chi_r^{11}\sin^2\theta+\chi_r^{22}\cos^2\theta)-\frac{1}{2}v\sin 2\theta+w\cos 2\theta, \tag{12d}$$

where

$$u=\chi_r^{11}-\chi_r^{22},$$

$$v=\chi_s^{11}-\chi_s^{22},$$

$$w=\chi_s^{12}. \tag{13}$$

After a and b are eliminated, Eqs. (12a), (12b), (12c), and (12d) reduce to

$$(u-wP)\cos 2\theta+\tfrac{1}{2}vP\sin 2\theta=0,$$

$$v\cos 2\theta+\tfrac{1}{2}(4w-uP)\sin 2\theta=0, \tag{14}$$

where P is defined as

$$P=p+\frac{1}{p}. \tag{15}$$

It follows from Eq. (14) that

$$P^2-\frac{u^2+w^2+4w^2}{uw}P+4=0. \tag{16}$$

For $uw\neq 0$, this equation has two real roots P_1 and P_2 with $|P_1|\geqslant 2$ and $|P_2|\leqslant 2$. In view of Eq. (15), $|P_2|<2$ has no physical meaning. Therefore, the only solution is expressed by

$$P=\frac{u^2+v^2+4w^2\pm\sqrt{(u^2+v^2+4w^2)-16u^2w^2}}{2uw}, \tag{17}$$

where the plus and minus signs before the radical correspond to the two cases $uw>0$ and $uw<0$, respectively. Taking into account that $|p|<1$, one obtains from Eq. (15) the corresponding values of p for the two cases:

$$p=\frac{P\mp\sqrt{P^2-4}}{2}. \tag{18}$$

As P is known, one can derive from Eq. (14) two different values of θ:

$$\theta^o=\frac{1}{2}\tan^{-1}\frac{2wP-2u}{vP},$$

$$\theta^e=\theta^o+\pi/2. \tag{19}$$

These two values correspond to two elliptical polarization states of the waves propagating along the wave vector $\mathbf{K}_0$ within the crystal. Similar to visible-light optics, the two waves here are called ''ordinary'' (o) and ''extraordinary'' (e) waves, respectively, with their major (minor) axes perpendicular to each other:

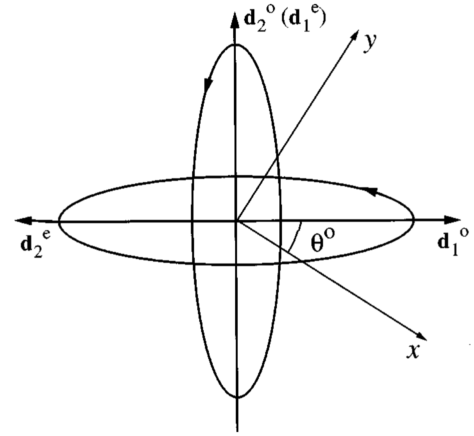

FIG. 1. Two elliptical polarization states (o and e waves) of anisotropically refracted x rays in crystals. $\mathbf{d}_1^i$ and $\mathbf{d}_2^i$ represent the directions of the major and minor axes of the i wave, respectively ($i=o,e$). The two ellipses have the same ellipticity and the arrows indicate the rotating directions of the electric displacements.

$$\mathbf{d}_1^o=\cos\theta^o\hat{\mathbf{i}}+\sin\theta^o\hat{\mathbf{j}},$$

$$\mathbf{d}_2^o=\mathbf{d}_1^e=-\sin\theta^o\hat{\mathbf{i}}+\cos\theta^o\hat{\mathbf{j}},$$

$$\mathbf{d}_2^e=-\mathbf{d}_1^o. \tag{20}$$

However, the two polarized waves have the same ellipticity p expressed in Eq. (18), indicating the similarity of the two ellipses in shape. Moreover, the electric displacements associated with the two waves are rotated in the same direction during propagation, as represented schematically in Fig. 1.

Under the condition that u or w approach zero, one can obtain from Eqs. (17), (18), and (19) the following results:

$$\lim_{u\to 0}p=0,\qquad \lim_{u\to 0}\theta^o=\frac{1}{2}\tan^{-1}\frac{2w}{v},$$

$$\lim_{w\to 0}p=0,\qquad \lim_{w\to 0}\theta^o=0. \tag{21}$$

In these two extreme cases, the refracted waves are linearly polarized along $\mathbf{d}_1^o$ or $\mathbf{d}_1^e$.

Using the present θ^i and p, one may obtain the other two parameters a^i and b^i ($i=o$ or e) from Eqs. (12a) and (12b). Especially, it can be found that a^o is different from a^e by

$$\Delta a=(u-2pw)\cos 2\theta^o+pv\sin 2\theta^o, \tag{22}$$

while the difference between b^o and b^e is

$$\Delta b=(2w-pv)\sin 2\theta^o+u\cos 2\theta^o. \tag{23}$$

The two wave vectors expressed in terms of the derived a^i and b^i are

$$K_0^o = K\left[1 + \frac{1}{2}(a^o + ib^o)\right],$$

$$K_0^e = K\left[1 + \frac{1}{2}(a^e + ib^o)\right]. \tag{24}$$

It follows immediately that the refractive indices of the two waves are $n^o = 1 + a^o/2$ and $n^e = 1 + a^e/2$, respectively. Owing to the anisotropy of the susceptibility, n^o and n^e are generally not equal, which gives rise to the birefringence of the crystal along $\mathbf{K}_0$:

$$\Delta n = \frac{\Delta a}{2}. \tag{25}$$

Meanwhile, the linear absorption coefficients of the o and e waves are represented by $\mu^o = -2\pi K|b^o|$ and $\mu^e = -2\pi K|b^e|$, respectively, along $\mathbf{K}_0$, and the associated dichroism is

$$\Delta\mu = 2\pi K|\Delta b|. \tag{26}$$

In an actual anisotropic refraction case, an incident wave generally splits into two elliptical waves with the $\mathbf{K}_0$-related o and e polarization states, respectively, with the initial amplitudes and relative phases of the two waves determined by the boundary condition. For example, when a linearly polarized plane wave with amplitude $\mathbf{D} = D_1\mathbf{d}_1^o + D_2\mathbf{d}_1^e$ is incident on the crystal (note that an x-ray beam in vacuum can always be expanded into a set of such waves), the boundary condition is

$$D_o e^{i\alpha} - ipD_e^{i\beta} = D_1,$$

$$ipD_o e^{i\alpha} + D_e e^{i\beta} = D_2, \tag{27}$$

where D_o and D_e are the amplitudes of the o and e waves along their major axes, respectively [see Eq. (6)], and α and β are their relative phases. Solving Eq. (27) yields

$$\alpha = \tan^{-1}\frac{pD_2}{D_1}, \quad \beta = -\tan^{-1}\frac{pD_1}{D_2},$$

$$D_o = \frac{\sqrt{D_1^2 + p^2 D_2^2}}{1 - p^2}, \quad D_e = \frac{D_2}{|D_2|}\frac{\sqrt{D_2^2 + p^2 D_1^2}}{1 - p^2}. \tag{28}$$

It is interesting that for $p \neq 0$, the o and e waves are always excited simultaneously by the linearly polarized incident wave. Inside the crystal, the two waves propagate independently. But as the waves leave the crystal, their amplitudes and relative phases are slightly changed due to the small birefringence and dichroism. Consequently, the two waves couple into an elliptical wave behind the exit surface.

IV. DISCUSSION AND CONCLUSION

Based on the dynamic model of x-ray refraction, we have demonstrated that elliptical polarized x rays are the fundamental wave forms in anisotropic x-ray scattering. It should be noted that the present theory is valid for all the crystallographic systems with a symmetric susceptibility tensor. For the least symmetric crystal, the energy-dependent susceptibility $\hat{\boldsymbol{\chi}}_0 + i\hat{\boldsymbol{\chi}}_0'$ contains at most six different real elements (χ_0^{ij}) and six imaginary ones ($\chi_0'^{ij}$) in the conventional coordinate system $x_0y_0z_0$. According to Eq. (10), the transformed elements χ_r^{ij} in the special system xyz are linear combinations of the elements χ_0^{ij} with the transformation coefficients determined by the direction of $\mathbf{K}_0$, and similar relations exist between χ_s^{ij} and $\chi_0'^{ij}$. If the refraction is viewed in the system $x_0y_0z_0$, the anisotropic parameters u, v, and w which determine the wave characteristics are generally related to all the susceptibility elements χ_0^{ij} and $\chi_0'^{ij}$. Meanwhile, the dependence of the three parameters on the direction of $\mathbf{K}_0$ implies that the birefringence and dichroism are also anisotropic, changing with the variation in wave-propagating directions. Therefore, the actual forms of the refracted waves inside the crystal are tunable in experiments.

The other feature of anisotropic refraction is that the parameters u, v, and w are only related to the energy-dependent anomalous-dispersion corrections and independent of the large isotropic part of the susceptibility which is conventionally written as[2]

$$\chi_0^{\mathrm{isotropic}} = -\frac{\lambda^2 e^2}{\pi m c^2}N \tag{29}$$

(N the number of electrons in a unit cell). As the magnitude of the real anomalous-dispersion correction is, in most cases, comparable to that of the imaginary part,[12,13] we believe that the three parameters can be of the same order near the atomic absorption edges. Under this condition, Eqs. (17) and (18) yield definite values for P and p. Only in the special case that u or v is extremely small does the ellipticity tend to zero, but the crystal is still birefringent or dichroic [see Eq. (21)].

Experimental data for anisotropic x-ray susceptibility are sparse at present, but the phenomena of x-ray birefringence and dichroism have been observed in many crystals. For instance, Petcov, Kirfel, and Fischer[7] have measured directly the birefringence and dichroism of lithium niobate with monochromatized synchrotron beam in the transmission case. Making the incident beam along the [10.0] direction, they found that the maximum values of the dichroism and birefringence are 53.6 cm^{-1} and -0.11×10^{-7}, respectively, in the vicinity of the Nb K-absorption edge. [Note that the birefringence and dichroism termed in Ref. 7 may be slightly different from that defined in Eqs. (25) and (26) of the present paper.] Compared with the isotropic x-ray refractive index (typically several percent) and absorption coefficient (a few hundreds or greater in cm^{-1}), the birefringence and dichroism are very small, but the results indeed gave direct proof of the occurrence of anisotropic refraction as well as the anisotropic interaction between x-ray waves and crystals. It was also indicated that, for the [10.0] wave-propagating direction, the maximum values of the anisotropic parameters u and v are $-0.30(8)$ and $0.70(1)$ (in electron), respectively. Unfortunately, the absolute value of w was not provided in Ref. 7, but the results are consistent with our assumption that u, v, and w are generally of the same order.

Although the detectable birefringence and dichroism of the transmitted beam are very weak in the transmission geometry, x rays within the crystal are, based on our deductions, completely polarized even if the anisotropic elements of the susceptibility are very small. The problem is that one can hardly separate the two kinds of elliptically (or at least linearly) polarized waves in experiments. In the diffraction case, however, the polarization state of the reflected wave may be totally different from that of the incident beam. Especially in ‘‘forbidden’’ or weak reflections where the isotropic susceptibility associated with the diffracted waves becomes insignificant and the resonant scattering is mainly related to the anisotropic interaction, crystals can act as efficient x-ray polarizers to change linearly polarized radiation into an elliptically polarized one or *vice versa*. In fact, highly polarized x rays have many applications in materials science, crystallography, chemistry, biology, etc.[11,14–16] The availability of highly polarized and tunable synchrotron radiation has made anisotropic refraction and diffraction an attractive tool for a broad range of studies of these fields in both transmission and diffraction cases. In a later work, we will illustrate explicitly the principles of anisotropic x-ray diffraction on the basis of the ‘‘two-beam’’ dynamic scattering model.

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