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Many-wave optics of blue phases

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A theory of many-wave optics of cholesteric blue phases is developed. The solution of the phase problem (i.e. the determination of the relative phases of the Fourier harmonics of the blue-phase order parameter) by means of many-wave diffraction is discussed. In the framework of many-wave blue-phase optics, the experimentally observed intensities of the Kossel lines are described. It is shown that the (111) and (200) Bragg reflections, observed in BPII, may be caused by coherent multiple diffraction.

1. Introduction

Optical methods are widely used to study liquid-crystalline blue phases (BPs) with their three-dimensional periodicity of the order parameter [1–3]. A detailed theory of BP optics is now available [2, 4, 5], but some observed features remain unexplained. The most challenging problem is the existence of Bragg reflections with unusual polarization properties, which contradict the well established BP structure [6–12].

Here it is shown that the properties of the unusual reflections find their natural explanation in the framework of the many-wave theory of BP optics. Moreover, many-wave optics gives a practical method for a solution of the phase problem [13]: the relative phases of the Fourier harmonics of the BP order parameter can be obtained by means of optical measurements under the conditions of many-wave diffraction. This method is similar to the method of the solution of the phase problem in the case of multiple X-ray diffraction [14]. Note that consideration of many-wave effects may be also essential for the description of diffraction-induced birefringence and optical rotation in BP [15].

2. Basic equations

To begin, we discuss the general description of many-wave diffraction. It is well known that in BPs (as in any periodic medium) a plane wave is not a solution of the Maxwell equations. The proper solution is given by the Bloch wave, i.e. by the superposition of the plane waves:

$$\mathbf{E}(\mathbf{r}, t) = \exp\left[i(\mathbf{k}_0 \cdot \mathbf{r} - \omega t)\right] \sum_{\tau} \mathbf{E}_\tau \exp\left(i\tau \cdot \mathbf{r}\right), \tag{1}$$

where τ are the reciprocal lattice vectors, $\mathbf{k}_0$ is the wavevector, ω is the frequency, and $\mathbf{E}_\tau$ are the amplitudes of the partial waves. The Bloch waves (see equation (1)) are excited inside the BP crystal by an incident wave with wavevector $\boldsymbol{\kappa}_0$, which becomes $\mathbf{k}_0$ after refraction at the crystal surface. We can consider equation (1) as a result of diffraction of the incident wave of amplitude $\mathbf{E}_0$ into an infinite number of diffracted waves of amplitudes $\mathbf{E}_\tau$ and with wavevectors $\mathbf{k}_\tau = \mathbf{k}_0 + \tau$. Inserting equation (1) into the Maxwell equations, we obtain an infinite set of equations for the partial

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amplitudes $\mathbf{E}_\tau$:

$$\left(\varepsilon_0 - \frac{k_\tau^2}{\kappa_0^2}\right)\mathbf{E}_\tau + \frac{\mathbf{k}_\tau(\mathbf{k}_\tau \cdot \mathbf{E}_\tau)}{\kappa_0^2} + \sum_{\tau'} \hat{\boldsymbol{\varepsilon}}_{\tau-\tau'} \cdot \mathbf{E}_{\tau'} = 0, \tag{2}$$

where $\tau = 0, \tau_1, \tau_2, \tau_3, \ldots, \mathbf{k}_\tau = \mathbf{k}_0 + \tau$, $\kappa_0^2 = (\omega/c)^2$, ε_0 is the average dielectric constant of the BP, and $\hat{\boldsymbol{\varepsilon}}_\tau$ are the (tensorial) Fourier harmonics of the BP dielectric constant ($\hat{\boldsymbol{\varepsilon}}_\tau$ are also the Fourier harmonics of the BP order parameter [2, 4]).

An approximate solution of equation (2) can be found from a truncated set of N equations for those N amplitudes $\mathbf{E}_\tau$ for which the wavevectors $\mathbf{k}_\tau$ satisfy or approximately satisfy the Bragg condition (see equation (3)). In general, the solutions of the N equations can be found only with the aid of a computer. However, some qualitative features of many-wave diffraction are evident without any calculations. Because of the amplitudes in equation (2) are mutually connected, the diffracted wave (say, $\mathbf{E}_\tau$) can be excited both directly and indirectly via many-wave diffraction (the excitation via many-wave diffraction is called *umweganregung* [14]). Thus the amplitude $\mathbf{E}_\tau$ has the qualitative form

$$\mathbf{E}_\tau = \left(A\,\hat{\boldsymbol{\varepsilon}}_\tau + B\sum_{\tau'} \hat{\boldsymbol{\varepsilon}}_{\tau-\tau'} \cdot \hat{\boldsymbol{\varepsilon}}_{\tau'} + \ldots\right) \cdot \mathbf{E}_0,$$

where A and B are some coefficients. Two effects of many-wave diffraction are evident from this equation. First, the intensities and polarization properties of the diffracted waves should depend on the phases of the triple products $\hat{\boldsymbol{\varepsilon}}_\tau^* \cdot \hat{\boldsymbol{\varepsilon}}_{\tau-\tau'} \cdot \hat{\boldsymbol{\varepsilon}}_{\tau'}$ (i.e. on the relative phases of the Fourier harmonics); as a result, the polarization selection rules [2, 4] may be violated. Secondly, the diffracted wave can be excited, even if direct diffraction is absent ($\hat{\boldsymbol{\varepsilon}}_\tau = 0$ or very small).

3. Many-wave diffraction conditions

To decide how many partial waves are important in equations (1) and (2), we should find those τ that satisfy (at least approximately) the Bragg condition

$$\tau^2 + 2\mathbf{k}_0 \cdot \tau = 0. \tag{3}$$

N-wave diffraction takes place if $N - 1$ different reciprocal lattice vectors ($\tau_1, \tau_2, \ldots, \tau_{N-1}$) satisfy equation (3) simultaneously. Because of the high symmetry of the BP cubic lattice, the conditions for many-wave diffraction are satisfied rather regularly. Note that there is a threshold (maximum wavelength) for many-wave diffractions; this may be used to discriminate between many-wave effects and two-wave effects. We shall present some examples of many-wave diffraction in BP without going into calculational details.

For BPII (O^2 space group) many-wave diffraction is possible if the light wavelength (*inside* the BP crystal) satisfies the inequality $\lambda \leqslant \lambda_{100}/\sqrt{2} = a\sqrt{2}$, where a is the period of the BPII lattice and λ_{100} is the maximum wavelength for two-wave diffraction. If $\mathbf{k}_0$ is parallel to the [110] direction and $\lambda = a\sqrt{2}$ then four-wave diffraction occurs: $\tau_1 = (110), \tau_2 = (100), \tau_3 = (010)$. If $\lambda = 2a/\sqrt{3}$ and $\mathbf{k}_0$ is parallel to the cubic space diagonal ([111] direction) then eight-wave diffraction occurs: $\tau_i = (111), (100), (010), (001), (110), (011), (101)$.

For BPI (O^8 space group) the many-wave conditions can be satisfied if $\lambda \leqslant \frac{1}{2}\lambda_{110}\sqrt{3}$, where $\lambda_{110} = a\sqrt{2}$ (λ_{110} is the maximum wavelength for two-wave diffraction in BPI). Note only two cases: $\lambda = a$ and $\mathbf{k}_0 \parallel [100]$ (six-wave diffraction); $\lambda = a\sqrt{\frac{3}{2}}$ and $\mathbf{k}_0 \parallel [211]$ (three-wave diffraction).

As the analysis of equation (2) shows, the partial amplitudes $\mathbf{E}_\tau$ are comparable to the amplitude $\mathbf{E}_0$ of the incident wave even if the Bragg condition is satisfied only approximately. Namely, $|\mathbf{E}_\tau| \approx |\mathbf{E}_0|$ if the parameter $\alpha_\tau = (\tau^2 + 2\mathbf{k}_0 \cdot \boldsymbol{\tau})/k_0^2$ is sufficiently small: $|\alpha_\tau| \lesssim \varepsilon_a$, where ε_a is a typical value of the local dielectric anisotropy in the BP (usually, $|\varepsilon_a| \approx 0{\cdot}1$–$0{\cdot}01$). If $|\alpha_\tau| \gg |\varepsilon_a|$ then the amplitude $\mathbf{E}_\tau$ is small in comparison with $\mathbf{E}_0$: $|\mathbf{E}_\tau|/|\mathbf{E}_0| \approx |\varepsilon_a|/|\alpha_\tau|$.

4. Many-wave diffraction for two strong waves

Many-wave effects can be described analytically if we assume that there are only two strong waves—the incident one $\mathbf{E}_0$ and the diffracted one $\mathbf{E}_\tau$—and that the others (the intermediate waves) are weak, but not negligible (the conditions limiting this assumption are given later). With this assumption, an approximate solution of the many-wave set of equations (2) may be found from effective two-wave equations for $\mathbf{E}_0$ and $\mathbf{E}_\tau$. To obtain the two-wave equations, the amplitudes of the weak waves, $\mathbf{E}_\mathbf{w}$, should be determined approximately from equation (2) as functions of the strong amplitudes $\mathbf{E}_0$ and $\mathbf{E}_\tau$:

$$\mathbf{E}_\mathbf{w} = \frac{\hat{\boldsymbol{\varepsilon}}_\mathbf{w} \cdot \mathbf{E}_0 - \kappa^{-2}\mathbf{k}_\mathbf{w}(\mathbf{k}_\mathbf{w} \cdot \hat{\boldsymbol{\varepsilon}}_\mathbf{w} \cdot \mathbf{E}_0) + \hat{\boldsymbol{\varepsilon}}_{\mathbf{w}-\tau} \cdot \mathbf{E}_\tau - \kappa^{-2}\mathbf{k}_\mathbf{w}(\mathbf{k}_\mathbf{w} \cdot \hat{\boldsymbol{\varepsilon}}_{\mathbf{w}-\tau} \cdot \mathbf{E}_\tau)}{\varepsilon_0 \alpha_\mathbf{w}}, \quad (4)$$

where $\kappa^2 = \kappa_0^2 \varepsilon_0$, $\mathbf{k}_\mathbf{w} = \mathbf{k}_0 + \mathbf{w}$, $\mathbf{w}$ is the reciprocal lattice vector of the intermediate (weak) wave (the analogous approximation has been used earlier [15, 16]). The approximation in equation (4) is valid if $\hat{\boldsymbol{\varepsilon}}_\mathbf{w}$ and $\hat{\boldsymbol{\varepsilon}}_{\mathbf{w}-\tau}$ are small in comparison with $\varepsilon_0 \alpha_\mathbf{w}$, i.e. if the intermediate waves do not satisfy the Bragg conditions. Inserting equation (4) into equation (2) for $\mathbf{E}_0$ and $\mathbf{E}_\tau$, we can find the effective two-wave equations

$$\left.\begin{aligned} \left[\left(\varepsilon_0 - \frac{k_0^2}{\kappa_0^2}\right)\mathbf{I} + \Delta\hat{\boldsymbol{\varepsilon}}_{00}\right] \cdot \mathbf{E}_0 + (\hat{\boldsymbol{\varepsilon}}_{-\tau} + \Delta\hat{\boldsymbol{\varepsilon}}_{0\tau}) \cdot \mathbf{E}_\tau &= 0, \\ (\hat{\boldsymbol{\varepsilon}}_\tau + \Delta\hat{\boldsymbol{\varepsilon}}_{\tau 0}) \cdot \mathbf{E}_0 + \left[\left(\varepsilon_0 - \frac{k_\tau^2}{\kappa_0^2}\right)\mathbf{I} + \Delta\hat{\boldsymbol{\varepsilon}}_{\tau\tau}\right] \cdot \mathbf{E}_\tau &= 0 \end{aligned}\right\} \quad (5)$$

(here $\mathbf{I}$ is the unit tensor), where the many-wave corrections $\Delta\hat{\boldsymbol{\varepsilon}}_{\mathbf{pn}}$ to the Fourier harmonics are given by

$$\Delta\hat{\boldsymbol{\varepsilon}}_{\mathbf{pn}} = \sum_\mathbf{w} \frac{\hat{\boldsymbol{\varepsilon}}_{\mathbf{p}-\mathbf{w}} \cdot \hat{\boldsymbol{\varepsilon}}_{\mathbf{w}-\mathbf{n}} - \kappa^{-2}(\hat{\boldsymbol{\varepsilon}}_{\mathbf{p}-\mathbf{w}} \cdot \mathbf{k}_\mathbf{w}) \otimes (\mathbf{k}_\mathbf{w} \cdot \hat{\boldsymbol{\varepsilon}}_{\mathbf{w}-\mathbf{n}})}{\varepsilon_0 \alpha_\mathbf{w}}; \quad (6)$$

in this equation $\mathbf{p}, \mathbf{n} = 0, \tau$ and $\mathbf{w} \neq 0, \tau$, the symbol $\otimes$ denotes the outer (tensor) product of two vectors). Note that in the general case, the many-wave corrections $\Delta\hat{\boldsymbol{\varepsilon}}_{\mathbf{pn}}$ are asymmetric complex tensors with non-zero traces. The correction $\Delta\hat{\boldsymbol{\varepsilon}}_{00}$ is responsible for the many-wave birefringence, which has been discussed for two-wave diffraction [2, 5, 16] in connection with optical rotation in BPs.

It is possible to find the specific expression for $\Delta\hat{\boldsymbol{\varepsilon}}_{\mathbf{pn}}$, if the restrictions on $\hat{\boldsymbol{\varepsilon}}_\mathbf{w}$ and $\hat{\boldsymbol{\varepsilon}}_{\tau-\mathbf{w}}$ known from theory [2, 4, 17] and experiment [1, 9, 16] are taken into account. Namely, just one tensor mode (with $m = 2$ or $m = -2$) gives the main contribution to the largest Fourier harmonics (see [2, 4, 9]). If we take into account only this mode (e.g. with $m = 2$) then the Fourier harmonic $\hat{\boldsymbol{\varepsilon}}_\mathbf{w}$ can be written as

$$\hat{\boldsymbol{\varepsilon}}_\mathbf{w} = \varepsilon(\mathbf{w}, 2)\mathbf{m}_\mathbf{w} \otimes \mathbf{m}_\mathbf{w}, \quad (7)$$

where $\varepsilon(\mathbf{w}, 2)$ is the mode amplitude, $\mathbf{m}_w = (\mathbf{m}_{1w} - i\mathbf{m}_{2w})/\sqrt{2}$, and the unit vectors $\mathbf{m}_{1w}$, $\mathbf{m}_{2w}$ and $\mathbf{w}/|\mathbf{w}|$ form a right-handed triad. Inserting equation (7) into equation (6), we obtain

$$\Delta\hat{\varepsilon}_{pn} = \sum_{w} \frac{(\mathbf{m}_{p-w} \otimes \mathbf{m}_{w-n})\varepsilon(\mathbf{p} - \mathbf{w}, 2)\varepsilon(\mathbf{w} - \mathbf{n}, 2)\,[\mathbf{m}_{p-w} \cdot \mathbf{m}_{w-n} - \kappa^{-2}(\mathbf{k}_w \cdot \mathbf{m}_{p-w})(\mathbf{k}_w \cdot \mathbf{m}_{w-n})]}{\varepsilon_0 \alpha_w} \tag{8}$$

This expression shows that usually the mode with $m = 2$ is present in the $\Delta\hat{\varepsilon}_{pn}$ (with the exception of those cases where this mode is forbidden by symmetry; for cubic structures this mode is absent only for exact backward diffraction along the three-fold or four-fold rotation axes).

5. Forbidden reflections

It is very important for BP structural investigations that there are the many-wave corrections to those coefficients in equations (5) that connect $\mathbf{E}_0$ and $\mathbf{E}_\tau$. This is the reason for the excitation of forbidden reflections for which $\hat{\varepsilon}_\tau$ is zero or very small. Therefore observation of the forbidden reflection is not a decisive argument in the determination of the BP space group. To clarify this question, we should determine whether the reflection is excited far from the many-wave diffraction conditions. As an example, we consider the (111) reflection in BPII. For symmetry reasons the Fourier harmonic $\hat{\varepsilon}_{111}$ contains only one tensor mode with $m = 0$ [4], and for two-wave diffraction this reflection should be weak and achiral [2]. However, for nearly backward diffraction eight-wave diffraction occurs and the polarization properties of the (111) reflection become rather unusual.

While under the exact eight-wave Bragg conditions the equations (5) are not valid, there are nearby angle regions where only the $\mathbf{E}_0$ and $\mathbf{E}_{111}$ amplitudes are strong, and equations (5) may be applied. Using equations (8) and (5), we can calculate the ratio of the scattering amplitude for right-hand polarized light into right-hand polarized light to the scattering amplitude for left-hand polarized light into left-hand polarized light. For nearly backward diffraction this ratio coincides with $\Delta\varepsilon(111, 2)/\Delta\varepsilon(111, -2)$. For every individual many-wave channel (say, 001/110), this ratio may be estimated from

$$\frac{\Delta\varepsilon(111, 2)}{\Delta\varepsilon(111, -2)} = \frac{(\mathbf{m}^*_{111} \cdot \mathbf{m}_{110})(\mathbf{m}^*_{111} \cdot \mathbf{m}_{001})}{(\mathbf{m}_{111} \cdot \mathbf{m}_{110})(\mathbf{m}_{111} \cdot \mathbf{m}_{001})} = \frac{(\sqrt{3} + \sqrt{2})(\sqrt{3} + 1)}{(\sqrt{3} - \sqrt{2})(\sqrt{3} - 1)} \approx 37, \tag{9}$$

where * denotes the complex conjugate. It is clear from this result that the many-wave contribution to (111) *nearly* backward diffraction is strongly chiral. However, *exact* backward diffraction is an exception. In this case the right-hand polarized wave diffracts only into a left-hand polarized wave and vice versa. Such polarization properties of *exact* backward diffraction can be obtained both from symmetry considerations (for n-fold rotation axes with $n \geqslant 3$) and from specific calculations (see [8], where the cancellation of the individual chiral terms was demonstrated for the 111 exact backward diffraction). Nevertheless, for real experimental conditions, even a small deviation of the beam from the [111] direction destroys the cancellation of individual chiral terms. Thus the observed predominant [9] scattering of beams with circular polarization (for (111) backward reflection) can be explained by the small deviations from the [111] direction (as small as the angular widths of the reflection bands of intermediate waves).

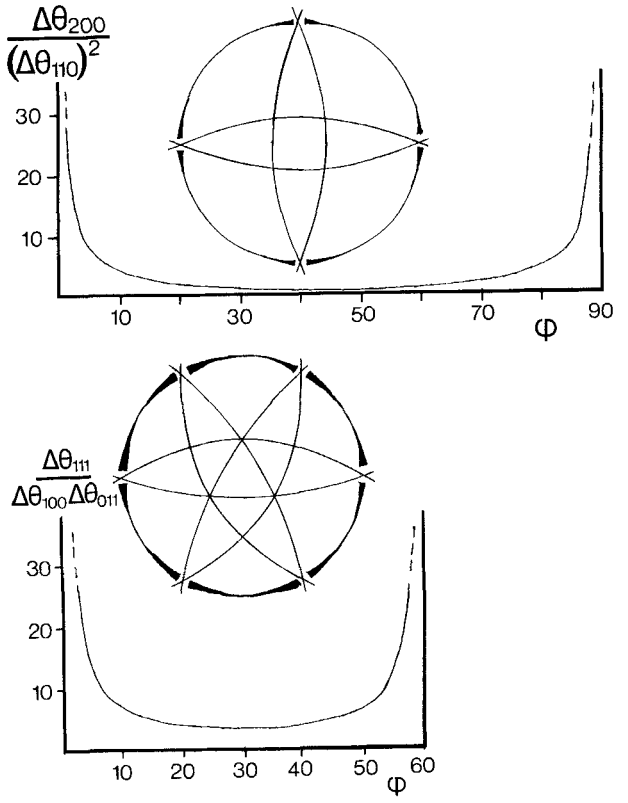

Figure 1. Calculated azimuthal dependences of the angular widths of (*a*) (200) and (*b*) (111) reflections in BPII. The corresponding Kossel diagrams are also shown (the angular widths are shown for (200) and (111) reflections only).

The simplest way to discriminate between many-wave and two-wave reflections consists in the examination of the azimuthal dependence of the intensity and polarization properties of the reflections during crystal rotation around τ. The many-wave diffraction depends strongly on the azimuthal angle φ (see figure 1) because the many-wave Bragg conditions (see equation (3)) are met at definite values of φ, while the two-wave diffraction depends very smoothly on φ (if it depends on φ at all). For perfect BP crystals the azimuthal dependences of the angular widths of (200) and (111) reflections are given approximately by

$$\Delta\theta_{200} \approx \frac{(\Delta\theta_{110})^2}{|\sin 2\varphi|},$$

$$\Delta\theta_{111} \approx \frac{\Delta\theta_{100}\Delta\theta_{011}}{|\sin 3\varphi|}.$$

The experimentally observed Kossel lines for the (200) and (111) reflections have azimuthal dependences (similar to those in figure 1) with a steep growth of linewidths at the points of many-wave diffraction [18]. These azimuthal dependences confirm that the many-wave mechanism is predominant for both reflections.

6. The phase problem

Many-wave effects in the azimuthal dependences of the intensities and polarization properties of reflections can also be used to determine the relative phases of different Fourier harmonics $\hat{\mathbf{e}}_\tau$. For simplicity, we examine the situation where only one many-wave channel is excited, i.e. the case of three-wave diffraction. In this case the coefficients in equation (5) that describes the diffraction from the wave $\mathbf{E}_0$ into the wave $\mathbf{E}_\tau$ has the form

$$\hat{\mathbf{e}}_\tau + \frac{\hat{\mathbf{e}}_{\tau-\mathbf{w}} \cdot \hat{\mathbf{e}}_\mathbf{w} - \kappa^{-2}(\hat{\mathbf{e}}_{\tau-\mathbf{w}} \cdot \mathbf{k}_\mathbf{w}) \otimes (\mathbf{k}_\mathbf{w} \cdot \hat{\mathbf{e}}_\mathbf{w})}{\varepsilon_0 \alpha_\mathbf{w}}. \tag{10}$$

Note that this coefficient depends both on the phase of the triple product $\mathbf{e}_\tau^* \cdot \hat{\mathbf{e}}_{\tau-\mathbf{w}} \cdot \hat{\mathbf{e}}_\mathbf{w}$ and on the parameter $\alpha_\mathbf{w}$ (in particular, on its sign). Let us suppose that the two-wave conditions for τ reflection is valid:

$$\tau^2 + 2\mathbf{k}_0 \cdot \tau = 0.$$

Then the form of the dependence of $\alpha_\mathbf{w}$ on φ and the corresponding dependences of the intensity and polarization parameters are determined by the geometry of the exact three-wave Bragg conditions. If the exact three-wave condition corresponds to the vector $\mathbf{k}_0$ being non-coplanar with the plane formed by τ and $\mathbf{w}$ then the denominator in equation (10) depends linearly on φ:

$$\alpha_\mathbf{w} = 2(\varphi - \varphi_0) \frac{\mathbf{w} \cdot (\tau \times \mathbf{k}_0)}{k_0^2}, \tag{11}$$

where φ_0 is the azimuthal angle of $\mathbf{k}_0$ for the exact three-wave Bragg condition, and $|\varphi - \varphi_0| \ll 1$.

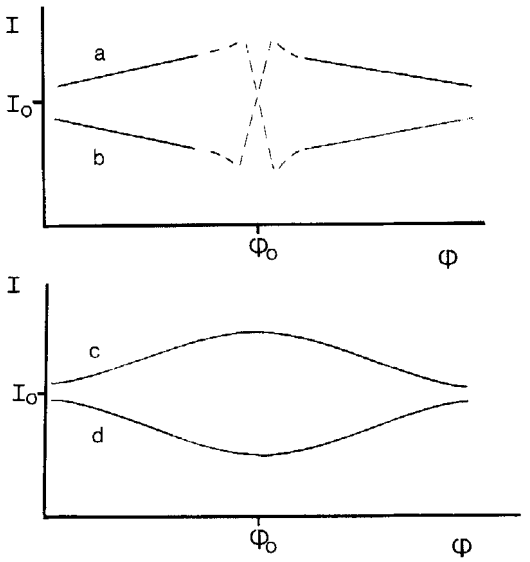

Figure 2. Qualitative form of the possible azimuthal dependence of reflection intensity for the different phases of the triple products $\hat{\mathbf{e}}_{\tau_1}^* \cdot \hat{\mathbf{e}}_{\tau_2} \hat{\mathbf{e}}_{\tau_3}$; I_0 is the reflection intensity far from the many-wave point. Curves (*a*) and (*b*) correspond to the non-coplanar three-wave Bragg condition; curves (*c*) and (*d*) correspond to the coplanar condition (with the vectors $\mathbf{k}_0$, τ and $\mathbf{w}$ lying in the same plane). The dashed lines shown those regions where equations (5) are inapplicable.

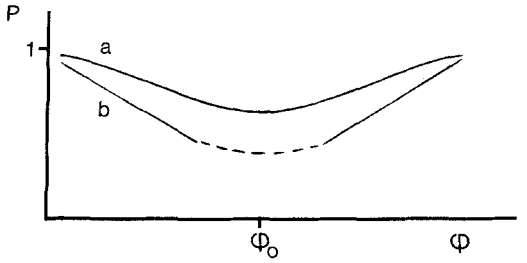

Figure 3. Possible azimuthal dependence of the degree of polarization: (*a*) for coplanar and (*b*) for non-coplanar three-wave diffraction. The dashed line shows the region where equations (5) are inapplicable.

If the exact three-wave Bragg condition (or the closest to it) corresponds to the vector $\mathbf{k}_0$ being coplanar with the plane formed by τ and $\mathbf{w}$ then the denominator in equation (10) is an even function of φ:

$$\alpha_{\mathbf{w}} = \alpha_{\mathbf{w}}^{\min} + (\varphi - \varphi_0)^2 \frac{|\mathbf{w} \times \tau| (4k_0^2 - \tau^2)^{1/2}}{2k_0^2}, \tag{12}$$

where $\alpha_{\mathbf{w}}^{\min}$ is the minimum possible deviation from the three-wave point:

$$\alpha_{\mathbf{w}}^{\min} = \left[w^2 - \boldsymbol{\tau} \cdot \mathbf{w} - |\tau \times \mathbf{w}| \left(\frac{4k_0^2}{\tau^2} - 1 \right)^{1/2} \right] \Big/ k_0^2, \quad \alpha_{\mathbf{w}}^{\min} \geqslant 0.$$

The intensity of the τ reflection is obtained from equations (10) and (5) (for example, in the framework of dynamical diffraction theory [2]). Typical forms of the azimuthal dependence of the reflection intensity are shown in figure 2. Since the azimuthal dependence of $\alpha_{\mathbf{w}}$ is known, we can obtain the phases for all the triple products $\hat{\boldsymbol{\varepsilon}}_{\tau_1}^* \cdot \hat{\boldsymbol{\varepsilon}}_{\tau_2} \cdot \hat{\boldsymbol{\varepsilon}}_{\tau_3}$ with $\boldsymbol{\tau}_1 = \boldsymbol{\tau}_2 + \boldsymbol{\tau}_3$ (these phases are called the structure invariants). The solution of this phase problem is of great importance for the complete determination of BP structure [2].

The azimuthal dependence of the polarization properties of reflections near the many-wave diffraction condition can be found in a similar way. One quantitative effect should be noted: for the reflected beam averaged over the polar angle the degree of polarization is less than unity and depends on the azimuthal angle φ (see figure 3).

7. Conclusion

We have shown that interesting phenomena in BP optics can be observed in the case of many-wave diffraction and, moreover, can be used for the delicate structural investigations of BPs. Some additional effects should also be noted. In particular, for the solution of the phase problem, the experimentally observed many-wave points on the Kossel diagrams [18, 19] may be used. It is also worth noting some peculiarities in the transmission spectra [20] (kinks and dips at definite wavelengths), which can be explained as manifestations of the wavelength thresholds for many-wave diffraction in BP polycrystals. The form of the transmission spectra at the thresholds (dip or kink) may be connected with the relative phases of the corresponding Fourier harmonics of the dielectric tensor. In the light of this discussion, it seems quite reasonable that the kink observed at 535 nm in the BPI transmission spectra was attributed [20] to the threshold for the three-wave diffraction ($\lambda = a\sqrt{\frac{3}{2}}$, $\tau_1 = (110)$, $\tau_2 = (0\bar{1}1)$). Moreover, the small dip observed at 306 nm in the BPII spectra [20] may also be

attributed to the same threshold, but in BPII. In the general case, the three-wave diffraction threshold is determined by the expression

$$\lambda = \frac{4\pi|\tau_1 \times \tau_2|}{|\tau_1||\tau_2||\tau_1 + \tau_2|},$$

where λ is the wavelength inside the polycrystal sample.

Note added during revision.—After submission of this paper, we received the work [21], where another mechanism (*incoherent* multiple scattering) had been used for the description of (111) reflection. We are grateful to R. M. Hornreich for the preprints.

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