J. Phys. A: Math. Gen. **31** (1998) 3493–3502. Printed in the UK PII: S0305-4470(98)89941-1

Lop-sided diffraction by absorbing crystals

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Received 11 December 1997

Abstract. Diffraction of atoms by a particular absorbing 'crystal of light', that has been studied experimentally, is described by the complex potential $\mathrm{i}V_0(\exp(\mathrm{i}Kx)-1)$. The diffracted beam intensities I_n can be calculated exactly, as a function of (dimensionless) potential strength σ, angle of incidence α, and crystal thickness ζ. Only the beams $n \geqslant 0$ have nonzero intensity; this 'lop-sidedness' is a dramatic violation of Friedel's law. The nth beam is strong at Bragg angles $\alpha = -\frac{1}{2}, -1, \ldots, -(n-\frac{1}{2})$, and the peaks get sharper with increasing ζ; they correspond to degeneracies of the (non-Hermitian) governing matrix. For normal incidence ($\alpha = 0$), the I_n are periodic in ζ, and as n increases they approach a self-similar function of ζ.

1. Introduction and basic equation

Recent discoveries in the optics of atoms diffracted by standing waves of light ('light crystals') (Oberthaler *et al* 1996) have made it possible to create complex potentials V with a variety of forms, and study in detail how they affect the behaviour governed by the Schrödinger equation whose solution describes the position coordinates of the atoms. The (negative) imaginary part of V describes absorption: loss of atoms by decay into a state different from the two 'working levels' responsible for the coherent refraction and diffraction by the light described by the real part of V. An extreme case is

$$V(x) = \mathrm{i}V_0(\exp\{\mathrm{i}Kx\} - 1) \tag{1}$$

(the -1 ensures $\mathrm{Im}\, V \leqslant 0$). Theory and experiment (Keller *et al* 1997) reveal dramatic violations of Friedel's law (valid in the kinematic and two-beam approximations for transparent crystals) that the diffracted beam intensities should be invariant to reversal of the crystal, and thus extend the small violations observed for slightly absorbing crystals in the diffraction of x-rays and electrons.

My aim here is to point out that (1) is one of the rare potentials for which the strengths of the diffracted beams can be calculated exactly (that is, in the full dynamical diffraction theory, rather than perturbatively) and to explore the solution. This extends recent work on the peculiarities of purely imaginary potentials (Berry and O'Dell 1998).

For atoms with mass m and energy E, travelling in the x, z plane, the Schrödinger equation for the wavefunction $\Psi(x, z)$ is

$$-(\partial_x^2 + \partial_z^2)\Psi + \mathrm{i}U_0(\exp\{\mathrm{i}Kx\} - 1)\Psi = k^2\Psi \tag{2}$$

where

$$k^2 = \frac{2mE}{\hbar^2} \qquad U_0 = \frac{2mV_0}{\hbar^2}. \tag{3}$$

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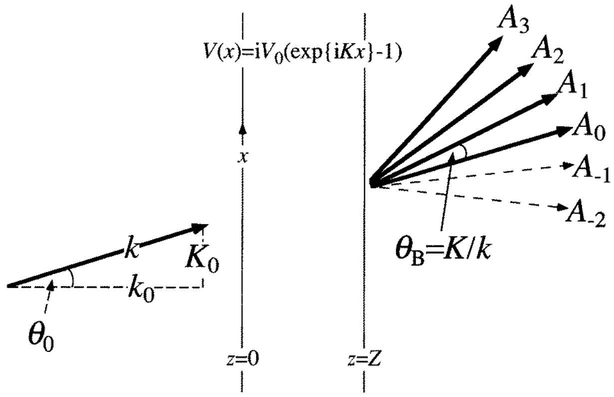

Figure 1. Geometry and notation for lop-sided diffraction of an atomic beam from a crystal of light with the complex potential indicated. Only diffracted beams with non-negative orders (amplitudes A_n with $n \geqslant 0$—full lines) exist.

We consider the light crystal as a volume grating, extending from $z = 0$ to $z = Z$, and infinite in the x direction (figure 1). Atoms are incident from $z < 0$, in a direction $\sin\theta_0 = K_0/k$, where

$$k_0^2 + K_0^2 = k^2. \tag{4}$$

We assume $\theta_0 \ll 1$, that is, paraxial propagation. Writing

$$\Psi(x, z) \equiv \exp\{\mathrm{i}k_0 z\}\psi(x, z) \tag{5}$$

we can regard ψ as varying slowly with z, so that $\partial_z^2\psi$ can be neglected. In the periodic crystal potential, and incorporating the initial condition, we can write

$$\psi(x, z) = \sum_{n=-\infty}^{\infty} A_n(z)\exp\{\mathrm{i}(nK + K_0)x\} \qquad A_n(0) = \delta_{n,0} \tag{6}$$

where n labels the diffracted beams, spaced in direction by the Bragg angle $\theta_\mathrm{B} = K/k$, whose intensities

$$I_n(Z) = |A_n(Z)|^2 \tag{7}$$

we will determine.

Natural dimensionless variables describing the strength of the potential, propagation distance and angle of incidence are

$$\sigma \equiv \frac{U_0}{K^2} = \frac{2mV_0}{\hbar^2 K^2} \qquad \zeta \equiv \frac{K^2 z}{2k_0} \qquad \alpha \equiv \frac{K_0}{K} = \frac{\theta_0}{\theta_\mathrm{B}}. \tag{8}$$

In terms of these, the amplitudes $A_n(\zeta, \alpha)$ satisfy

$$\mathrm{i}\partial_\zeta A_n = (n^2 + 2\alpha n - \mathrm{i}\sigma)A_n + \mathrm{i}\sigma A_{n-1}. \tag{9}$$

This resembles the Raman–Nath equation (Raman and Nath 1936, Berry 1966), first written for the diffraction of light by ultrasound. However, there is the crucial difference that in (9) each amplitude is coupled to one, rather than both, of its neighbours, because the potential (1) has only one Fourier component; this is what makes the equations exactly solvable.

2. Exact solution

An immediate consequence of (9), with the initial condition in (6), is that $A_n = 0$ for all $n < 0$, for all incidence angles α. This 'lop-sidedness' of the diffraction pattern (figure 1) embodies the violation of Friedel's law, and is a consequence of the non-Hermitian nature of the operator in (2). For convenience, the form of Friedel's law appropriate for transparent light crystals is derived in the appendix. It also follows from (9) that the dependence on the potential strength σ can be extracted explicitly:

$$A_n(\zeta, \alpha) = \sigma^n \exp(-\sigma\zeta) B_n(\zeta, \alpha) \tag{10}$$

where $B_n(\zeta, \alpha)$ satisfies

$$\mathrm{i}\partial_\zeta B_n = (n^2 + 2\alpha n) B_n + \mathrm{i} B_{n-1} \qquad B_n(0) = \delta_{n,0}. \tag{11}$$

The exponential simply reflects the -1 in the potential (1), and guarantees absorption.

One of several ways to solve this equation is by taking its Laplace transform. This leads to a first-order difference equation, and thence to

$$B_n(\zeta, \alpha) = \frac{\mathrm{i}}{2\pi} \int_{c-\mathrm{i}\infty}^{c+\mathrm{i}\infty} \mathrm{d}s \, \frac{\exp(s\zeta)}{\prod_{j=0}^{n} (s + \mathrm{i} j(j + 2\alpha))} \qquad (\mathrm{Re}\, c > 0) \tag{12a}$$

$$= (-\mathrm{i})^n \sum_{j=0}^{n} \frac{\exp\{-\mathrm{i}\zeta j(j + 2\alpha)\}}{\prod_{m \neq j = 0}^{n} (m - j)(m + j + 2\alpha)} \tag{12b}$$

$$= 2(-\mathrm{i})^n \sum_{j=0}^{n} \frac{\exp\{-\mathrm{i}\zeta j(j + 2\alpha)\}(-1)^j (j + \alpha)(j + 2\alpha - 1)!}{j!(n - j)!(n + j + 2\alpha)!}. \tag{12c}$$

Another form of solution starts by defining

$$B_n(\zeta, \alpha) \equiv \exp\{-\mathrm{i}\zeta(n^2 + 2\alpha n)\} C_n(\zeta, \alpha) \tag{13}$$

whence

$$C_n(\zeta, \alpha) = \int_0^\zeta \mathrm{d}\zeta' \, C_{n-1}(\zeta', \alpha) \exp\{\mathrm{i}\zeta'(2n - 2\alpha - 1)\}. \tag{14}$$

Yet another way of seeing that (9) or (11) can be solved exactly is to write the solution as a superposition of 'Bloch waves' that oscillate with ζ with 'frequencies' given by the eigenvalues of the matrices in the equations. Since the elements of the matrix in (11) are nonzero only on the principal and one next-to-principal diagonal, it is triangular; therefore the eigenvalues are simply the diagonal elements.

The intensities corresponding to these solutions do not satisfy any sum rule that would be analogous to $\sum |A_n|^2 = 1$ for transparent gratings, or to the alternating-sign sum rule (Berry and O'Dell 1998) for imaginary gratings.

3. Bright Bragg beams

For certain angles of incidence α, some of the diffracted beams $n \geqslant 0$ are much brighter than others, because of zeros in the denominator of (12*b*). For the nth beam, there are poles for

$$\alpha = -\tfrac{1}{2}, -1, \ldots - (n - \tfrac{1}{2}). \tag{15}$$

Alternatively stated, when α is one of the Bragg angles, that is $\alpha = -l/2$, the strong beams are those (figure 2) with

$$n \geqslant \mathrm{int}(\tfrac{1}{2}l + 1) \qquad (l \geqslant 1). \tag{16}$$

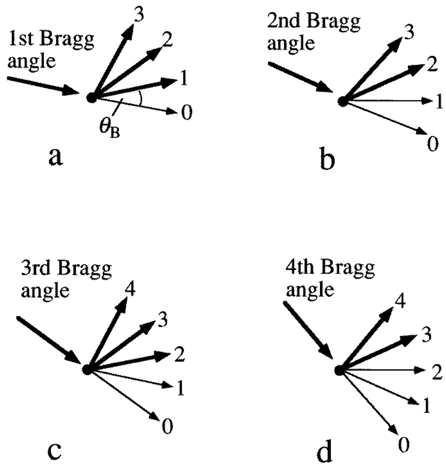

Figure 2. Strong beams (bold arrows) for incidence at different Bragg angles with negative θ_0: (*a*) $\alpha = -\frac{1}{2}$, (*b*) $\alpha = -1$, (*c*) $\alpha = -\frac{3}{2}$, (*d*) $\alpha = -2$.

Three points should be noted. First, for incidence at each Bragg angle $\alpha = -l/2$, the strong beams are all those emerging at positive angles, that is on the same side of the z-axis as the incident beam, and not just the single beam $n = l$ corresponding to Bragg reflection.

Second, phase coherence between the terms in (12*b*) cause cancellations, so that the strengths of the strong beams are all finite; this can be seen explicitly in the first few amplitudes:

$$\begin{aligned} B_1(\zeta,\alpha) &= -\mathrm{i}\left(\frac{1-\exp\{-\mathrm{i}\zeta(1+2\alpha)\}}{1+2\alpha}\right) \\ B_2(\zeta,\alpha) &= -\left(\frac{1}{4(1+2\alpha)(1+\alpha)} - \frac{\exp\{-\mathrm{i}\zeta(1+2\alpha)\}}{(1+2\alpha)(3+2\alpha)} + \frac{\exp\{-4\mathrm{i}\zeta(1+\alpha)\}}{4(3+2\alpha)(1+\alpha)}\right) \\ B_3(\zeta,\alpha) &= \mathrm{i}\bigg(\frac{1}{12(1+2\alpha)(1+\alpha)(3+2\alpha)} - \frac{\exp\{-\mathrm{i}\zeta(1+2\alpha)\}}{4(1+2\alpha)(3+2\alpha)(2+\alpha)} \\ &\qquad + \frac{\exp\{-4\mathrm{i}\zeta(1+\alpha)\}}{4(3+2\alpha)(1+\alpha)(5+2\alpha)} - \frac{\exp\{-3\mathrm{i}\zeta(3+2\alpha)\}}{12(3+2\alpha)(2+\alpha)(5+2\alpha)}\bigg). \end{aligned} \tag{17}$$

Figure 3 shows the intensities of these beams, and their increasing strength (as a function of thickness) at the Bragg directions. Figure 4 shows sample rocking curves, that is intensities as a function of incidence α (or, equivalently, crystal orientation).

Third, these strong beams can be understood as degeneracies of the matrix in (11). This is because eigenvalues labelled m and n coincide when $m^2 + 2m\alpha = n^2 + 2n\alpha$, that is $\alpha = -(m+n)/2$. Because these matrices are non-Hermitian, their degeneracies are different from the more familiar ones for Hermitian matrices, where the two degenerating eigenvectors can be chosen as any orthogonal pair. Here, in contrast, the two eigenvectors become parallel, and at the degeneracy there is only a single eigenvector. A consequence of this is that the amplitudes of the strong beams grow linearly in the light crystal (roughly, the $\exp\{\mathrm{i}\lambda\zeta\}$ of the single eigenvector, with eigenvalue λ, is accompanied by $\zeta\exp\{\mathrm{i}\lambda\zeta\}$). The quadratic increase of the intensities is accompanied by oscillations (figure 5). These non-Hermitian degeneracies have implications elsewhere in physics (Berry and O'Dell 1998, Berry and Klein 1997, Berry 1994).

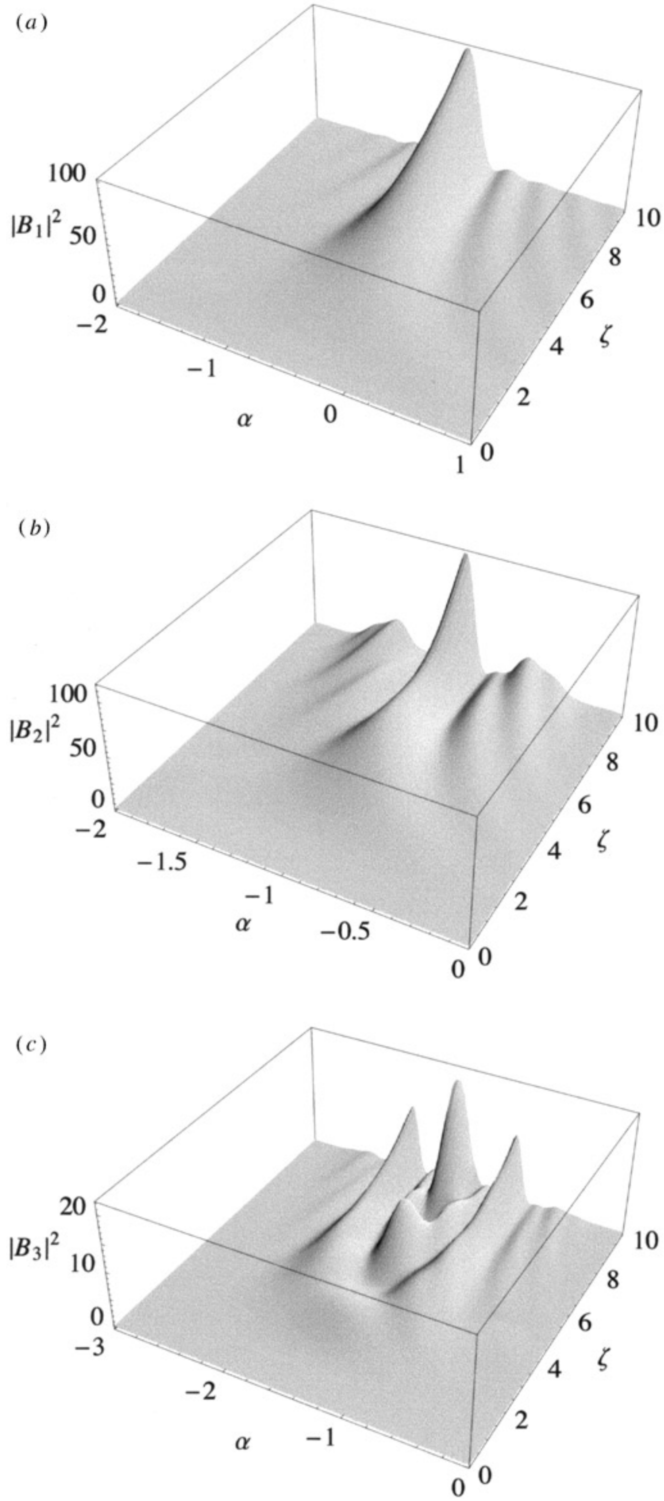

Figure 3. Intensity $|B_n|^2$ of diffracted beams, as a function of angle of incidence α (orientation) and crystal thickness ζ. (*a*) $n = 1$; (*b*) $n = 2$; (*c*) $n = 3$.

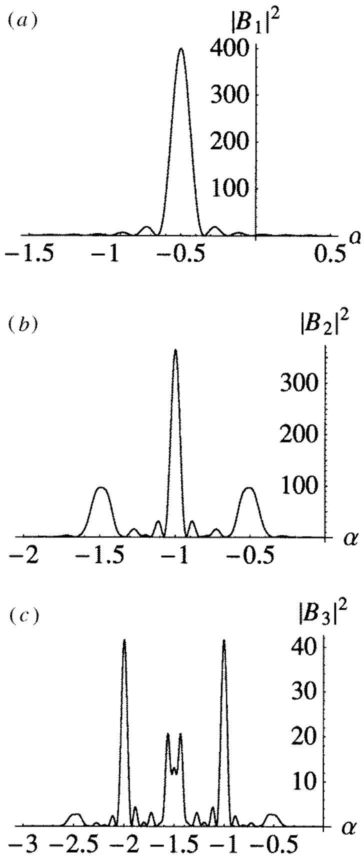

Figure 4. Rocking curves (diffracted intensities as a function of orientation α), calculated for thickness $\zeta = 20$ for (*a*) $n = 1$; (*b*) $n = 2$; (*c*) $n = 3$. For the nth beam, there are Bragg peaks near $\alpha = -\frac{1}{2}, -1, \ldots (n - \frac{1}{2})$.

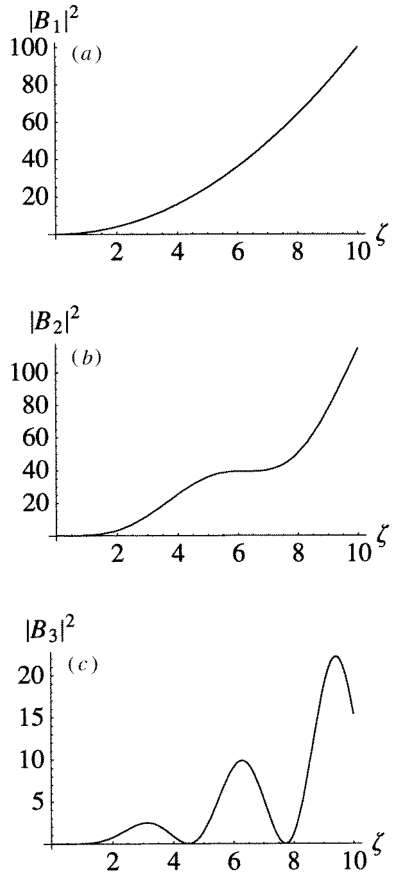

Figure 5. Diffracted intensities $|B_n|^2$ at Bragg angles $\alpha = -n/2$, as a function of thickness ζ. Because the governing matrix is degenerate and non-Hermitian, the intensities increase quadratically on the average, for (*a*) $n = 1$; (*b*) $n = 2$; (*c*) $n = 3$.

As a function of α, the intensity of the nth beam is symmetric about its own Bragg angle. This symmetry of the rocking curves can be made obvious by writing the amplitudes in terms of

$$2\alpha \equiv -n + \beta. \tag{18}$$

Then, from equation (12*c*) and after some algebra,

$$\begin{aligned} B_n(\zeta, \tfrac{1}{2}(-n+\beta)) &= \mathrm{i}^n \exp(-\tfrac{1}{2}\mathrm{i}n\beta\zeta)\beta!(-\beta)! \\ &\quad\times\left\{ \sum_{j=0}^{\mathrm{int}((n-1)/2)} \frac{\exp\{\mathrm{i}j(n-j)\zeta\}}{j!(n-j)!} \frac{[g_{n,j}(\beta,\zeta) - g_{n,j}(-\beta,\zeta)]}{\beta} \right. \\ &\quad\left. + \frac{\exp\{\frac{1}{4}\mathrm{i}n^2\zeta\}\delta_{n,2\,\mathrm{int}(n/2)}}{n!^2(\frac{1}{2}n+\beta)!(\frac{1}{2}n-\beta)!} \right\} \end{aligned} \tag{19}$$

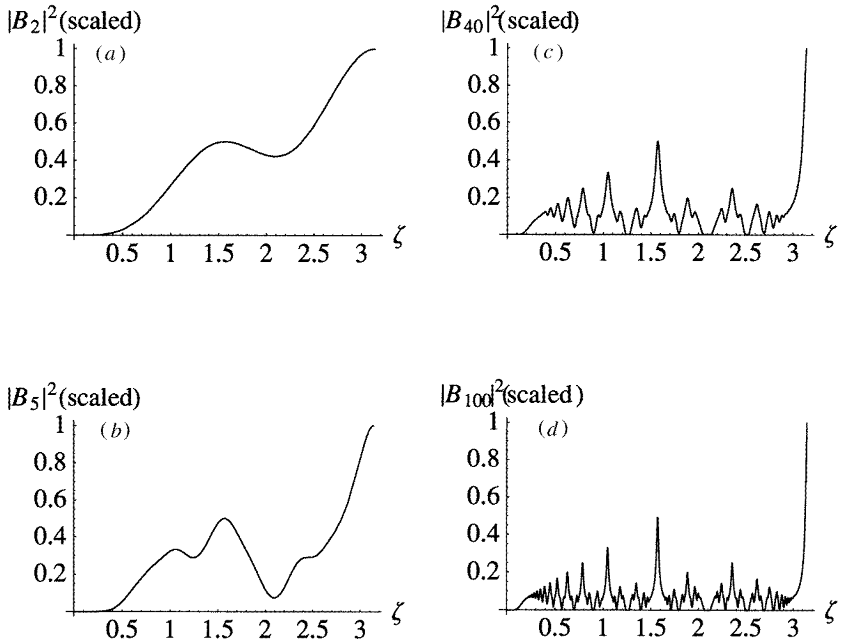

Figure 6. Diffracted intensities $|B_n|^2$ at normal incidence $\alpha = 0$, over a half-period $(0 \leqslant \zeta \leqslant \pi)$, scaled by division by its maximum value $2^{4n}/(2n)!^2$ (equation (23)), for (*a*) $n = 2$; (*b*) $n = 5$; (*c*) $n = 40$; (*d*) $n = 100$.

where

$$g_{n,j}(\beta, \zeta) = \exp\{\mathrm{i}\beta(\tfrac{1}{2}n - j)\zeta\}\frac{(2n - j + \beta)}{(j + \beta)!(n - j - \beta)!}. \tag{20}$$

4. Normal incidence

From (12*c*), and being careful with the $\alpha \to 0$ limit of the term $j = 0$, we find, for normal incidence,

$$B_n(\zeta, 0) = (-\mathrm{i})^n \left\{ \frac{1}{n!^2} + 2\sum_{j=1}^{n} \frac{\exp\{-\mathrm{i}\zeta j^2\}(-1)^j}{(n-j)!(n+j)!} \right\}. \tag{21}$$

By changing the summation variable to $n - j$, this can be written in the more symmetrical form

$$B_n(\zeta, 0) = \mathrm{i}^n \sum_{j=0}^{2n} \frac{\exp\{-\mathrm{i}\zeta(n-j)^2\}(-1)^j}{(j)!(2n-j)!}. \tag{22}$$

It is clear that these amplitudes are periodic in ζ, in contrast to what happens for real periodic potentials, where the beating of Bloch waves with incommensurate eigenvalues (of the Mathieu equation) (Berry 1966) makes the amplitudes nonrepeating. The period is $\Delta\zeta = 2\pi$ and it is easy to see that the extremes of destructive and constructive interference in the sum (22) occur at $\zeta = 2l\pi$ and $\zeta = (2l + 1)\pi$, when

$$B_n(2l\pi, 0) = \delta_{n,0} \qquad B_n((2l+1)\pi, 0) = (-\mathrm{i})^n \frac{2^{2n}}{(2n)!}. \tag{23}$$

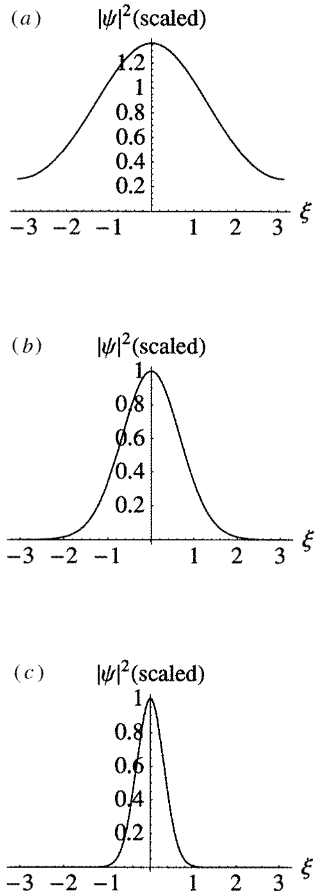

Figure 7. Wave intensity $|\psi(\xi+\pi/2)|^2$ inside the crystal (equation (25)) for normal incidence, at a depth $\zeta=(2l+1)\pi$ of maximum contrast, scaled by the asymptotic maximum in (26), for potential strength (*a*) $\sigma=0.2$; (*b*) $\sigma=5$; (*c*) $\sigma=100$.

Figure 6 shows the intensities over a half-period, for several values of n. A self-similar structure appears to emerge as $n\to\infty$, as might be expected from the 'almost theta function' (22); this could repay further study.

For these special values of ζ, it is easy to calculate the form of the wave ψ inside the light crystal. From (6) and (10), and defining $\xi\equiv Kx$, we have, for any ζ,

$$\psi(\xi,\zeta)=\exp(-\sigma\zeta)\sum_{n=0}^{\infty}\sigma^n B_n(\zeta,0)\exp(\mathrm{i}n\xi). \tag{24}$$

Thus

$$\begin{aligned}\psi(\xi,2l\pi)&=\exp\{-2l\pi\sigma\}\\ \psi(\xi,(2l+1)\pi)&=\exp\{-(2l+1)\pi\sigma\}\cosh\{2\sqrt{\sigma}\exp(\tfrac{1}{2}\mathrm{i}(\xi-\tfrac{1}{2}\pi))\}.\end{aligned} \tag{25}$$

Thus, the form of the wave in the crystal alternates: between the initial constant, and peaks concentrated on the focusing minima $\xi=(2n+\frac{1}{2})\pi$ of the real part of the potential. The concentration is strongest in the 'semiclassical' limit of large σ, where

$$|\psi(\xi,(2l+1)\pi)|^2\approx\tfrac{1}{4}\exp\{-(2l+1)2\pi\sigma+4\sqrt{\sigma}\}\exp\{-\tfrac{1}{2}\sqrt{\sigma}(\xi-\tfrac{1}{2}\pi)^2\}\qquad(\sigma\gg1). \tag{26}$$

Figure 7 shows how the peaks get sharper as σ increases.

Acknowledgments

I would like to thank C Keller, S Bernet, J Scmiedmayer and A Zeilinger for introducing me to this problem, and the British Council in Austria for financial support.

Appendix. Friedel's law

For a one-dimensional light-crystal potential $V(x)$, Friedel's law is the assertion that the diffracted beam intensities I_n are invariant under spatial inversion of the crystal, that is $x \to -x$ (this is equivalent to, but easier to think about than, reversing the angle of incidence, that is $\alpha \to -\alpha$, and at the same time considering I_{-n} instead of I_n). Here I show that the law holds for crystals with a centre of symmetry, and for noncentrosymmetric transparent crystals only in the kinematic (weak diffraction) and two-beam approximations; it does not hold, in general, for absorbing crystals.

If the potential has dimensionless Fourier coefficients v_n, so that

$$V(x) = \frac{\hbar^2 K^2}{2m} \sum_{-\infty}^{\infty} v_n \exp\{inKx\} \tag{A1}$$

the intensities $I_n \equiv |A_n|^2$ are determined by the analogue of the Raman–Nath (dynamical diffraction) equation (9), namely

$$i\partial_\zeta A_n = (n^2 + 2\alpha n)A_n + \sum_{m=-\infty}^{\infty} v_{n-m} A_m. \tag{A2}$$

Reversal of the crystal corresponds to replacing v_n by v_{-n} in (A1). If the crystal has a centre of symmetry, and this is chosen as the origin of $V(x)$, then $v_n = v_{-n}$ and Friedel's law is true both for transparent and absorbing crystals; this trivial case will not be discussed further.

Transparent crystals are those with $V(x)$ real, so that

$$v_{-n} = v_n^*. \tag{A3}$$

The amplitudes $A_{n,\mathrm{rev}}$ in the reversed crystal are therefore governed by

$$i\partial_\zeta A_{n,\mathrm{rev}} = (n^2 + 2\alpha n)A_{n,\mathrm{rev}} + \sum_{m=-\infty}^{\infty} v_{n-m}^* A_{m,\mathrm{rev}}. \tag{A4}$$

Thus

$$A_{n,\mathrm{rev}}(\zeta) = A_n^*(-\zeta) \qquad \text{i.e. } I_{n,\mathrm{rev}}(\zeta) = I_n(-\zeta) \tag{A5}$$

so Friedel's law in a transparent crystal is equivalent to the assertion that the intensities are even functions of ζ.

The solution of (A2) is

$$A_n(\zeta) = [\exp\{-i\zeta \mathbf{M}\}]_{n0} \tag{A6}$$

where $\mathbf{M}$ is the matrix

$$\mathbf{M} = \{M_{nm}\} \qquad M_{nm} = (n^2 + 2\alpha n)\delta_{nm} + v_{n-m}. \tag{A7}$$

For transparent crystals, $\mathbf{M}$ is Hermitian, because of (A3). Expanding (A6) to order ζ^3, the intensity of the nonforward beams is, to the same order,

$$I_{n\neq 0} = \zeta^2 |v_n|^2 + \zeta^3 \,\mathrm{Im}\left\{ v_n^* \sum_{m=-\infty}^{\infty} v_{n-m} v_m \right\} + O(\zeta^4). \tag{A8}$$

The kinematic (weak-diffraction) approximation consists of retaining just the first term, in ζ^2. Friedel's law holds because this is an even function of ζ (the law can also be seen as a direct consequence of (A3)). However, the first term not even in ζ, namely the term in ζ^3, is not zero in general, even for transparent crystals, so Friedel's law is false. However, in the important special case of two-beam diffraction from transparent crystals, Friedel's law does hold. Let the nonzero beams be $n = 0$ and $n = 1$, and write $2\alpha = -1 + \beta$ (cf (18)). Then the solution (A6) is, using (A3),

$$\begin{pmatrix} A_0 \\ A_1 \end{pmatrix} = \exp\left\{-\mathrm{i}\zeta \begin{pmatrix} v_0 & v_1^* \\ v_1 & v_0+\beta \end{pmatrix}\right\} \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

$$= \exp\{-\mathrm{i}\zeta(v_0 + \tfrac{1}{2}\beta)\}$$

$$\times \left[\cos\left\{\zeta\sqrt{|v_1|^2 + \tfrac{1}{4}\beta^2}\right\}\begin{pmatrix} 1 \\ 0 \end{pmatrix} - \mathrm{i}\frac{\sin\left\{\zeta\sqrt{|v_1|^2 + \frac{1}{4}\beta^2}\right\}}{\sqrt{|v_1|^2 + \frac{1}{4}\beta^2}}\begin{pmatrix} -\frac{1}{2}\beta \\ v_1 \end{pmatrix}\right]. \quad \text{(A9)}$$

From this, the two intensities are

$$I_1 = 1 - I_0 = \frac{|v_1|^2 \sin^2\left\{\zeta\sqrt{|v_1|^2 + \frac{1}{4}\beta^2}\right\}}{|v_1|^2 + \frac{1}{4}\beta^2}. \quad \text{(A10)}$$

These are even in ζ; hence Friedel's law.

For absorbing noncentrosymmetric crystals, (A3) does not hold and Friedel's law fails even in the kinematic approximation (first term of (A8)), because this term need not be invariant under $v_n \to v_{-n}$. A simple example is $V(x) = 2(a\cos Kx + \mathrm{i}b\sin Kx)$ (a and b real), where $v_1 = a + b$ and $v_{-1} = a - b$, so that in (A8) $|v_1|^2 \neq |v_{-1}|^2$. Of course another example is the potential (1) studied here, for which the only nonzero coefficients are $v_0 = -\mathrm{i}\sigma$ and $v_1 = \mathrm{i}\sigma$.

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