Resonant scattering and suppression of inelastic channels in a two-dimensional crystal

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An inelastic-channel suppression mechanism not connected with Bragg diffraction in a three-dimensional lattice is investigated using a two-dimensional model. To this end, coherent scattering by a monatomic crystalline film is considered, with account taken of the mutual irradiation of its sites by scattered radiation. The same problem is considered simultaneously on the basis of a modification, formulated in the paper, of the optical theorem for a planar lattice. The result is a general formula for the amplitude q_0 of planar scattering in terms of the individual amplitude f_0 of the free site, as well as an expression for the change $f_0 \to \tilde{f}_0$ of the free amplitude when the sites combine to form a lattice. The relations derived predict an abrupt growth of the coefficient of reflection from one atomic plane at incident-radiation frequencies close to the resonant atomic ones. If inelastic channels are present, they may be suppressed, so that the same radiation can be almost totally reflected backward from a two-dimensional crystal, even though it is almost completely absorbed when scattered by an isolated site. A connection is obtained between the resonant-radiation linewidth and the band structure of a two-dimensional crystal. The possibility of observing the effect is discussed.

1. INTRODUCTION

When radiation interacts with a solid, an important role can be played by effects connected with coherent scattering by an individual atomic plane of the crystal. Besides Bragg diffraction,[1] this scattering can cause, for example, channeling between the planes.[2,3] It is also of interest for investigation of crystalline planes of atomic thickness, which can be produced on the surface[4–6] or in the interior[7,8] of material transparent to the radiation in question.

The known description of planar scattering[1] is restricted to an approximation in which the scattering is regarded as weak. This approximation leads to the formula

$$q_0 = i\frac{\lambda N_\sigma}{\sin\theta} f_0, \tag{1.1}$$

that express the amplitude q_0 of elastic planar forward scattering in terms of the wavelength λ, the site surface density N_σ, the glancing angle θ, and the amplitude f_0 of the elastic forward scattering by an individual site. The dimensionless amplitude q_0 is obtained by summing the scattered waves from all the sites of the planar lattice under the condition that the sites themselves are excited only by the incident wave. This condition means that $|q_0| \ll 1$, i.e.,

$$|f_0| \ll \sin\theta/\lambda N_\sigma \approx (a^2/\lambda)\sin\theta, \tag{1.2}$$

where a is a quantity of the order of the planar-lattice constant. The approximation indicated is regarded as sufficient, for in contrast to a three-dimensional lattice, where a small scattering effect can build up over a stack of planes, there is here, so to speak, nothing to build up on (if the case $\theta \to 0$ is disregarded). The problem of taking exact account of multiple scattering by sites of an isolated atomic plane has, to our knowledge, not even been raised.

It is just this problem which is the subject of the present paper. It is shown that in a number of important cases multiple scattering by a planar lattice can play a decisive role. A new formula is then obtained for q_0. In the weak-scattering limit it includes Eq. (1.1), but in the general case it describes qualitatively new phenomena. At resonance, in particular, an atomic plane becomes optically rigid, and the coefficient of reflection from it can reach almost unity even for certain x-ray frequencies at arbitrary θ. If inelastic channels are present, they may become suppressed, so that a system of atoms that absorb the particular radiation differently even in a three-dimensional lattice, can reflect this radiation elastically without absorption if the atoms are ordered in a single plane. A simple relation is found between the linewidth of such resonant reflection and the lattice characteristics, so that information can be obtained on the energy band structure of a two-dimensional crystal.

2. STATEMENT OF PROBLEM

Consider an isolated atomic plane xy constituting an ideal two-dimensional crystal with constant lattice vectors $\mathbf{a}_1$ and $\mathbf{a}_2$. The properties of a single lattice site are characterized by the atomic elastic-scattering amplitude $f_0(\omega)$, which, in contrast to (1.2), is not assumed to be small. The lattice is irradiated by a plane wave $\mathbf{E}_0 \exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$ of frequency ω that can coincide with an arbitrary resonant atomic frequency ω_0, i.e., be located inside the absorption band for the corresponding bulk crystal. It is required to determine the field coherently scattered by the atomic plane, and find the general form of the function $q_0(f_0)$ that describes both weak and strong (including resonant) scattering.

To solve this problem, exact account must be taken of the effect of mutual irradiation of the lattice sites by the scattered radiation. This is done for the case when the atomic dimension a_0 and the atom-oscillation frequency amplitude are small compared with a_1, a_2, and the wavelength λ. The

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scattering site can then be regarded as pointlike, and the scattering itself as dipolar.

We derive an expression for the total field $E^{\alpha}(\mathbf{r}, t)$ at an arbitrary point of space $\mathbf{r} = (x, y, z) = (\boldsymbol{\rho}, z)$

$$E^{\alpha}(r, t) = E_0^{\alpha} e^{i(\mathbf{kr}-\omega t)} + \sum_m \sum_n Q_{\beta}^{\alpha}(\boldsymbol{\rho}_{mn}-\mathbf{r}) E^{\beta}\left(\boldsymbol{\rho}_{mn}, t - \frac{|\boldsymbol{\rho}_{mn}-\mathbf{r}|}{c}\right), \quad (2.1)$$

where $E^{\alpha}(\mathbf{r}, t)$ is the sum of the incident and all scattered waves. Each of the latter depends in turn on the total field $E(\boldsymbol{\rho}_{mn}, t')$ at the corresponding site (the scatterer). The coordinates $\boldsymbol{\rho}_{mn} = m\mathbf{a}_1 + n\mathbf{a}_2$ with integer m and n number the sites, and the origin is at the site with $(m, n) = (0, 0)$. The indices α and β (for which the Einstein summation rule is assumed) number the wave-function components of the scattering particles (in our case—the components of the electric vector $\mathbf{E}$). The quantities $Q_{\beta}^{\alpha}(\boldsymbol{\rho}_{mn} - \mathbf{r})$ describe the contribution of the β component of the total field at the (m, n) site to the α component of the scattered radiation at the point $\mathbf{r}$. Given the frequency ω, this contribution is proportional to the amplitude $\tilde{f}_0(\omega)$ of the elastic scattering of this frequency by the bound site. In fact, the field $\mathbf{E}_{\omega}(\boldsymbol{\rho}_{mn}) \times e^{-i\omega t}$ induces at the site (m, n) a dipole moment

$$\mathbf{d}_{mn} = \frac{e^2 \mathbf{E}_{\omega}(\boldsymbol{\rho}_{mn})}{M(\omega_0^2 - \omega^2 - i\omega\omega_\Gamma)} e^{-i\omega t}, \quad (2.2)$$

where M is the mass, ω_0 is the natural frequency, and ω_Γ is the width of the resonant line of the corresponding bound oscillator. It is assumed for simplicity that the atomic-polarizability tensor of the lattice material reduces to a scalar, and that the elementary scatterer is two-level, i.e., equivalent to one oscillator. Using the expression for the amplitude of forward scattering by an isotropic oscillator

$$\tilde{f}_0 = \frac{\omega^2}{\omega_0^2 - \omega^2 - i\omega\omega_\Gamma} r_e, \qquad r_e \equiv \frac{e^2}{Mc^2}, \quad (2.3)$$

we rewrite (2.2) in the form

$$\mathbf{d}_{mn} = \frac{\tilde{f}_0}{k^2} \mathbf{E}_{\omega}(\boldsymbol{\rho}_{mn}) e^{-i\omega t}. \quad (2.4)$$

Substituting this in the known equation[9] for the field produced at the point $\mathbf{r}$ by the oscillating dipole moment $\mathbf{d}_{mn}(\rho_{mn})$, we obtain

$$Q_{\beta}^{\alpha}(\boldsymbol{\rho}_{mn}-\mathbf{r}) = \xi_{\beta}^{\alpha}(\boldsymbol{\rho}_{mn}-\mathbf{r}) \tilde{f}_0, \quad (2.5)$$

where

$$\xi_{\beta}^{\alpha}(\boldsymbol{\rho}_{mn}-\mathbf{r}) \equiv \frac{\delta_{\beta}^{\alpha} - n_{mn}^{\alpha} n_{\beta mn}}{|\boldsymbol{\rho}_{mn}-\mathbf{r}|} + \frac{i}{k} \frac{\delta_{\beta}^{\alpha} - 3n_{mn}^{\alpha} n_{\beta mn}}{|\boldsymbol{\rho}_{mn}-\mathbf{r}|^2} - \frac{1}{k^2} \frac{\delta_{\beta}^{\alpha} - 3n_{mn}^{\alpha} n_{\beta mn}}{|\boldsymbol{\rho}_{mn}-\mathbf{r}|^3}, \quad (2.6)$$

$\mathbf{n}_{mn}$ is the unit vector in the direction from the site (m, n) to the point $\mathbf{r}$, and $k = \omega/c$. At $|\boldsymbol{\rho}_{mn} - \mathbf{r}| \lesssim a_0$, allowance for the finite dimensions of the site leads to the substitution $|\boldsymbol{\rho}_{mn} - \mathbf{r}| \to a_0$ in the denominators of (2.6).

The elastic scattering is described by a Fourier amplitude $\mathbf{E}_{\omega}(\mathbf{r})\exp(-i\omega t)$, for which (2.1) with allowance for (2.5) takes the form (we omit the index ω)

$$E^{\alpha}(\mathbf{r}) = E_0^{\alpha} e^{i\mathbf{kr}} + \tilde{f}_0 \sum_m \sum_n \xi_{\beta}^{\alpha}(\boldsymbol{\rho}_{mn}-\mathbf{r}) \times \exp\{ik|\boldsymbol{\rho}_{mn}-\mathbf{r}|\} E^{\beta}(\boldsymbol{\rho}_{mn}). \quad (2.7)$$

We use (2.7) to calculate the fields at the lattice sites. Since each site (m', n') is acted upon by an external field produced by all sites except by itself, the sums of the right-hand side of (2.7) are taken at $\boldsymbol{\rho} = \boldsymbol{\rho}_{m'n'}$ without the term with $(m, n) = (m', n')$. In the upshot we obtain for the values of $\mathbf{E}(\boldsymbol{\rho}_{mn})$ a system of inhomogeneous equations with a specified external "source"

$$E^{\alpha}(\boldsymbol{\rho}_{m'n'}) - \tilde{f}_0 \sum_{(m,n)\neq(m',n')} \sum \xi_{\beta}^{\alpha}(\boldsymbol{\rho}_{mn,m'n'}) \exp(ik\rho_{mn,m'n'}) E^{\beta}(\boldsymbol{\rho}_{mn}) = E_0^{\alpha} \exp(ik\rho_{mn}), \quad (2.8)$$

where

$$\boldsymbol{\rho}_{mn,\,m'n'} \equiv \boldsymbol{\rho}_{mn} - \boldsymbol{\rho}_{m'n'}, \quad \rho_{mn,\,m'n'} \equiv |\boldsymbol{\rho}_{mn,\,m'n'}|, \quad m', n' = 0, \pm 1, \pm 2, \ldots.$$

The scattering effect of interest to us is described by a particular solution of the system (2.8); this solution enables us to express the values of $E(\boldsymbol{\rho}_{mn})$, and by the same token also the amplitudes $q_0 \sim |\mathbf{E} - \mathbf{E}_0|/|E_0|$ of the waves scattered by the lattice, in terms of the atomic amplitude $\tilde{f}_0$.

3. SOLUTION

When a transition is made from a three-dimensional to a planar crystal, the translational symmetry is preserved in the planes $z = \text{const}$. The field of a two-dimensional crystal is such a plane should therefore have the form of a Bloch wave

$$\mathbf{E}(\mathbf{r}) = e^{i\mathbf{k}_\sigma \boldsymbol{\rho}} \mathbf{E}_f(\boldsymbol{\rho}, z) \quad (3.1)$$

with a doubly periodic function

$$\mathbf{E}_f(\boldsymbol{\rho}, z) = \mathbf{E}_f(\boldsymbol{\rho} + m\mathbf{a}_1 + n\mathbf{a}_2, z) = \mathbf{E}_0 e^{ik_z z} + \sum_j \sum_l \mathbf{E}_{jl}(z) \exp\{2i(j\mathbf{k}_1 + l\mathbf{k}_2)\boldsymbol{\rho}\}. \quad (3.2)$$

Here $\mathbf{k}_\sigma$ and k_z are the $\mathbf{k}$ components parallel and perpendicular to the lattice, respectively, while $\mathbf{k}_1$ and $\mathbf{k}_2$ are the reciprocal-lattice vectors of the two-dimensional crystal. The amplitudes $\mathbf{E}_{jl}(z)$ are determined by the inverse Fourier transform of the scattered field $\mathbf{E}_s(r) = \mathbf{E}(r) - \mathbf{E}_0 \times \exp(i\mathbf{kr})$ in the plane $z = \text{const}$:

$$\mathbf{E}_{jl}(z) = \frac{1}{\sigma_A} \iint [\mathbf{E}(\mathbf{r}) - \mathbf{E}_0 e^{i\mathbf{kr}}] \exp\{-i[\mathbf{k}_\sigma + 2(j\mathbf{k}_1 + l\mathbf{k}_2)]\boldsymbol{\rho}\} d\sigma, \quad (3.3)$$

where the integral is taken over the area σ_A of the two-dimensional unit cell.

Since the periodic function $\mathbf{E}_f(\rho, 0)$ assumes the same value

$$\mathbf{E}_f(\boldsymbol{\rho}_{mn}, 0) = \mathbf{E}_f(0, 0, 0) = \mathbf{E}_0 + \sum_j \sum_l \mathbf{E}_{jl}(0) \equiv \mathbf{E}_f, \quad (3.4)$$

at the sites $\boldsymbol{\rho} = \boldsymbol{\rho}_{mn}$, we can seek the particular solution of (2.8) in the form

$$\mathbf{E}(\rho_{mn})=\mathbf{E}_f e^{i\mathbf{k}\rho_{mn}} \tag{3.5}$$

with a specified $\mathbf{k}$ and an unknown "amplitude" $\mathbf{E}_f$. Substituting this in (2.8) we get

$$E_f^\alpha-\mathscr{f}_0 \sum\sum_{(m,n)\neq(m'n')} \xi_\beta{}^\alpha(\rho_{mn,m'n'}) \exp\{i(\mathbf{k}\rho_{mn,m'n'}+k\rho_{mn,m'n'})\}E_f^\beta=E_0{}^\alpha. \tag{3.6}$$

By virtue of the same lattice periodicity that leads to the Bloch function, the sum

$$-i\xi_\beta{}^\alpha\equiv \sum\sum_{(m,n)\neq(m',n')} \xi_\beta{}^\alpha(\rho_{mn,m'n'})\exp\{i(\mathbf{k}\rho_{mn,m'n'}+k\rho_{mn,m'n'})\} \tag{3.7}$$

is independent of the number (m', n') of the site for which it is calculated. The infinite system (3.6) degenerates then into the finite (three-dimensional for photons)

$$\mathbf{E}_f{}^\alpha+i\mathscr{f}_0\xi_\beta{}^\alpha E_f{}^\beta=E_0{}^\alpha. \tag{3.8}$$

Hence

$$\mathbf{E}_f{}^\alpha=\frac{\Delta_\beta{}^\alpha}{\Delta}E_0{}^\beta,\quad E^\alpha(\rho_{mn})=\frac{\Delta_\beta{}^\alpha}{\Delta}E_0{}^\beta\exp(i\mathbf{k}\rho_{mn}), \tag{3.9}$$

where Δ is the determinant of the system (3.8)

$$\Delta\equiv|\delta_\beta{}^\alpha+i\mathscr{f}_0\xi_\beta{}^\alpha|, \tag{3.10}$$

and Δ_β^α are the corresponding algebraic components. The tensor factors $\Delta_\beta^\alpha/\Delta$ in (3.9), because of which the amplitude $\mathbf{E}_f$ of the field acting on the sites can differ greatly from $\mathbf{E}_0$, describe the result of the multiple scattering.

Substituting the obtained values $\mathbf{E}^\alpha(\rho_{mn})$ in the right-hand side we obtain an explicit expression for the field at any point $\mathbf{r}$:

$$E^\alpha(\mathbf{r})=E_0{}^\alpha e^{i\mathbf{r}\mathbf{k}}+\mathscr{f}_0\sum_m\sum_n\xi_\beta{}^\alpha(\rho_{mn}-\mathbf{r}) \times\exp\{i(k|\rho_{mn}-\mathbf{r}|+\mathbf{k}\rho_{mn})\}\frac{\Delta_\delta{}^\beta}{\Delta}E_0{}^\delta. \tag{3.11}$$

It is easy to verify that the right-hand side of (3.11), multiplied by $\exp(-i\mathbf{k}_\sigma\rho)$, satisfies the condition (3.2). We obtain then $\mathbf{E}_{jl}^\alpha(z)$ from (3.3). Inasmuch as the field scattered by the lattice is described at $|z|\to\infty$ by a discrete set of plane waves, the coefficients $\mathbf{E}_{jl}(z)$ take the asymptotic form

$$\mathbf{E}_{jl}(z\to\pm\infty)\approx\mathbf{E}_{jl}{}^\pm\exp\{\pm ik_{z,jl}z\}, \tag{3.12}$$

where

$$k_{z,jl}=[k^2-|k_\sigma+2(jk_1+lk_2)|^2]^{1/2}. \tag{3.13}$$

At sufficiently large j and l the harmonics (3.12) describe surface waves, with imaginary $k_{z,jl}$, which travel over the lattice.

Taking (3.3), (3.11), and (3.12) into account we obtain general equations for the amplitudes of any order of diffraction

$$E_{jl}{}^{\pm\alpha}=i\eta_{\beta,jl}^{\pm\alpha}\mathscr{f}_0\frac{\Delta_\delta{}^\beta}{\Delta}E_0{}^\delta, \tag{3.14}$$

where as $|z|\to\infty$ we have

$$i\eta_{\beta,jl}^{\pm\alpha}=\frac{1}{\sigma_A}\iint\sum_m\sum_n\xi_\beta{}^\alpha(\rho_{mn}-\mathbf{r}) \times\exp i\{k|\rho_{mn}-\mathbf{r}|+\mathbf{k}\rho_{mn}-[\mathbf{k}_\sigma+2(j\mathbf{k}_1+l\mathbf{k}_2)]\rho\pm k_{z,jl}z\}d\sigma \tag{3.15}$$

and the signs + and − correspond to scattering into the half-spaces $z>0$ and $z<0$. These relations can be simplified, since the tensor ξ_β^α has a symmetry that permits it to be diagonalized by an appropriate choice of the axes $(x, y)\leftrightarrow(1, 2)$ in the lattice plane. The tensor $\Delta_\beta^\alpha/\Delta$ is diagonalized simultaneously with ξ_β^α. As a result, the system (3.8) breaks up into three independent equations:

$$E_f^\alpha+i\mathscr{f}_0\xi_\alpha{}^\alpha E_f{}^\alpha=E_0{}^\alpha,\quad \alpha=1, 2, 3,$$

so that

$$E_f^\alpha=\frac{E_0{}^\alpha}{1+i\xi_\alpha{}^\alpha\mathscr{f}_0}. \tag{3.8a}$$

It follows in this case from (3.14) that

$$E_{jl}{}^{\pm\alpha}=i\eta_{\beta,jl}^{\pm\alpha}\mathscr{f}_0E_0{}^\beta/(1+i\xi_\beta{}^\beta\mathscr{f}_0). \tag{3.14a}$$

Let the incidence plane pass through one of the principal axes (the 2 axis) of the lattice, and let the incident photon be polarized in the direction of the 1 axis, so that $E_0^\beta=E_0\delta_1^\beta$ and

$$E_{jl}{}^{\pm\alpha}=i\eta_{1,jl}^{\pm\alpha}\mathscr{f}_0E_0/(1+i\xi_1{}^1\mathscr{f}_0). \tag{3.16}$$

For photons that are scattered forward or are scattered in accordance with the usual law [$j=l=0$ in (3.12)] we have $\alpha=1$ and it follows from (3.16) that

$$q_0\equiv q_{00}{}^+=q_{00}{}^-=i\eta\mathscr{f}_0/(1+i\xi\mathscr{f}_0), \tag{3.17}$$

where $\xi\equiv\xi_1^1$ is the corresponding principal value of the tensor ξ_β^α, while $\eta\equiv\eta_{1,00}^{+1}=\eta_{1,00}^{-1}$.

Let us calculate the parameter η for the case $\lambda\gg a$. As $|z|\to\infty$ and at $j=l=0$ the value of the double sum in (3.15) averaged over the area σ_A can be replaced by its value on the site (0, 0) i.e., at $\rho=0$ and as $|z|\to\infty$ we have

$$i\eta=\sum_m\sum_n\xi_1{}^1(\rho_{mn}-z)\exp i\{k(\rho_{mn}{}^2+z^2)^{1/2}+k_\sigma\rho_{mn}-k_z|z|\}. \tag{3.18}$$

We replace the remaining summation over the sites by integration over the lattice plane, letting $\rho_{mn}\to\rho$ and introducing the integration element

$$dN_\sigma=N_\sigma\rho d\rho d\varphi=N_\sigma\tilde{r}d\tilde{r}d\varphi,$$

where ρ and φ are polar coordinates in the xy plane with center opposite the observation point P, and $\tilde{r}=(\rho^2+z^2)^{1/2}$. Then

$$i\eta=N_\sigma\int_{|z|}^{\infty}\xi_1{}^1(\tilde{r})e^{ik\tilde{r}}\int_0^{2\pi}\exp\{ik(\tilde{r}^2-z^2)^{1/2}\cos\theta\cos\varphi -|z|\sin\theta\}\tilde{r}d\tilde{r}d\varphi. \tag{3.19}$$

As $|z|\to\infty$ we can confine ourselves to the first term of ξ_1^1 in (2.6)

$$\xi_1{}^1(\tilde{r})\approx\tilde{r}^{-1}(1-n_1{}^2)\approx\tilde{r}^{-1}.$$

Substituting this in (3.19) and recognizing that integration with respect to φ yields the zeroth-order Bessel function $I_0(k(\tilde r^2 - z^2)^{1/2}\cos\theta)$, we obtain[10]

$$i\eta = 2\pi N_\sigma \exp(ik|z|\sin\theta)\int_{|z|}^{\infty} e^{ik\tilde r} J_0\left(k(\tilde r^2 - z^2)^{1/2}\cos\theta\right) d\tilde r \xrightarrow[|z|\to\infty]{} i\,\frac{\lambda N_\sigma}{\sin\theta}. \tag{3.20}$$

The result (3.20) coincides with the coefficient in (1.1). Therefore in the weak-scattering limit ($f_0 \to 0$) Eq. (3.17) goes over into (1.1), as it should.

The calculation of the parameter ξ is much more complicated, inasmuch as an essential role is played in (3.7) at $z=0$ by the interaction described by the second and third terms of (2.6), and the dependence on φ is significant in each term. Therefore depends strongly on the actual geometry of the lattice.

The obtained relations (3.14)–(3.17) connect the picture of the diffraction by a two-dimensional crystal with the geometric (vectors $\mathbf{a}_1$ and $\mathbf{a}_2$) and physical (amplitude $\tilde f_0$) properties of the crystal. They are valid also if the atomic scattering includes inelastic processes with change of frequency or with photon absorption, since the summary contribution made to (2.8) by the corresponding inelastic amplitudes $E_{\omega'\neq\omega}$ is zero. Substitution of these amplitudes in (2.1) yields an independent system of homogeneous equations, that leads to a dispersion relation between the natural frequency ω' and the quasimomentum $\hbar k'_\sigma$ of the "photons" in the plane lattice. Therefore the form, specified by the system (2.8), of the connection between q_0 and $\tilde f_0$ is insensitive to the presence of inelastic channels in the atomic scattering: information on these channels is contained in the amplitude $\tilde f_0$ itself, but not in the expression that connects q_0 with $\tilde f_0$.

The method developed here determines q_0 via the amplitude $\tilde f_0$ of the bound site. This amplitude, generally speaking, differs from the amplitude f_0 of the scattering by an isolated site. To determine $\tilde f_0$ we must take into account the influence of the lattice on the individual properties of each site taken separately.

4. OPTICAL THEOREM FOR TWO-DIMENSIONAL CRYSTAL

We solve this problem by a new method that gets around the computational difficulties in a number of problems, since it takes into account the interactions between sites automatically. The gist of the new approach is that the entire lattice is regarded as one compound scatterer, for which is formulated an analog of the optical theorem known for quasipointlike scatterer.

For the amplitude f_0 of scattering by an isolated site, the optical theorem can be written in the form

$$\mathrm{Im}\, f_0 = k\Phi|f_0|^2, \tag{4.1}$$

with $\Phi = \mathrm{const} = 2/3$ in the case of pure elastic dipole scattering. Relation (4.1) yields for each ω a connection between $\mathrm{Im}\, f_0$ and $\mathrm{Re}\, f_0$.

Let us formulate the analog of the optical theorem for a two-dimensional crystal. In accordance with the initial assumption that the lattice thermal vibrations are small, we neglect the possible dissipation of the incident energy into the additional inelastic channels that are produced when individual sites in the lattice are combined to form a lattice, and are connected, for example, with photon production.

Scattering by such a two-dimensional crystal results in a finite number of plane waves departing from the crystal and having amplitudes $q_{jl}^\pm = E_{jl}^\pm/E_0$. The amplitude of the transmitted (incident + forward-scattered) wave is equal to $1+q_0$. In the case of pure elastic scattering we have

$$|1+q_0|^2+|\bar q_{00}|^2+\sum_{(j,l)\neq(0,0)}\sum |q_{jl}^\pm|^2=1. \tag{4.2}$$

The summation in (4.2) is bounded by the values j and l for which the component $k_{z,jl}$ in (3.13) in real, inasmuch as in the steady state the surface waves add nothing to the energy balance.

Introducing the relative intensities

$$S_{jl}^{\pm 2}\equiv|q_{jl}^\pm|^2/|q_0|^2,$$

we write for the left-hand side of (4.2)

$$1+2\,\mathrm{Re}\, q_0+|q_0|^2\sum_j\sum_l S_{jl}^{\pm 2},$$

after which, assuming

$$\sum_j\sum_l S_{jl}^{\pm 2}\equiv 2\tilde\Phi, \tag{4.3}$$

we rewrite (4.2) in the form

$$\mathrm{Re}\, q_0=-\tilde\Phi|q_0|^2, \tag{4.4a}$$

or

$$(\mathrm{Re}\, q_0)^2+\tilde\Phi^{-1}\,\mathrm{Re}\, q_0+(\mathrm{Im}\, q_0)^2=0. \tag{4.4b}$$

This is lattice analog of the optical theorem: the scattered energy fraction $Q\equiv 2\tilde\Phi|q_0|^2$ plays the role of the (dimensionless) total cross section and is expressed in terms of the real part of the lattice amplitude q_0.

According to (4.4), all the nonzero values of $q_0(\omega)$ lie in the left complex half-plane of $q_0 = q_{0r} + iq_{0i}$. The maximum values of $|q_0|$ are bounded by the condition $|1+q_0|^2\leqslant 1$ that follows from (4.2), so that

$$|q_0|\leqslant 2. \tag{4.5}$$

Figure 1 shows plots of $f_{0i}(f_{0r})$ and $q_{0i}(q_{0r})$ (at

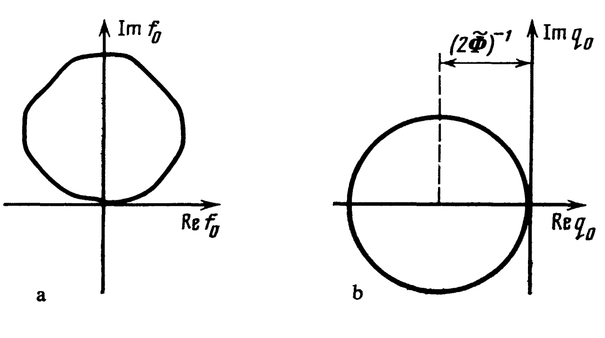

FIG. 1.

$\tilde{\Phi}=1$) corresponding to pure elastic scattering, with the values of f_0 determined from an expression of the type (2.3) with $\omega_\Gamma=\omega_\gamma=\frac{2}{3}r_e\omega^2/c$ (Ref. 9), where ω_γ is the elastic-scattering width.

The simplest mapping $q_0\leftrightarrow f_0$ relating the points of the planes f_0 and q_0, which would reduce to (1.1) as $f_0\to 0$ and would amount to $|q_0|\leqslant 2$ at $\eta|f_0|\gg 1$, is of the form

$$q_0=i\eta f_0/(1+i\tilde{\xi}f_0), \quad (4.6)$$

which coincides with the independently obtained relation (3.17). This equation, however, contains the amplitude for scattering not by a bound but by a free site, so that the value of $\tilde{\xi}$ can differ from the corresponding ξ. Optical theorems for f_0 and q_0 allow us to express the real part of $\tilde{\xi}$ in terms of η, k, Φ, and $\tilde{\Phi}$. Substituting (4.6) and (4.4) and stipulating that the resultant relation for f_0 coincide with (4.1) [i.e., that the mapping (4.6) transform the curve (4.1) into (4.4)], we get

$$\operatorname{Re}\tilde{\xi}=\Phi k-\tilde{\Phi}\eta. \quad (4.7)$$

The imaginary part $\operatorname{Im}\tilde{\xi}\equiv\tilde{\xi}_i$ remains in this case arbitrary in accord with the fact that the optical theorem determines $q_0(\omega)$ only accurate to an arbitrary mapping $q_0(\omega)\to q_0'(\omega)$ that transforms the curve (4.4) into itself. The uncertainty is eliminated by determining $\tilde{\xi}_i(k)$ from the dispersion relations between the real and imaginary parts of the amplitudes f_0 and q_0. In the present paper the exact form of $\tilde{\xi}_i(k)$ is immaterial and the expression sought for $q_0(f_0)$ can be written as

$$q_0=i\eta f_0/\{1-\tilde{\xi}_i f_0+i(\Phi k-\tilde{\Phi}\eta)f_0\}. \quad (4.8)$$

Equation (4.8), just as (3.17), remains valid in the presence of inelastic processes in the atomic scattering.

Comparison of (3.17) and (4.8) yields a relation between the free and bound amplitudes

$$\tilde{f}_0=f_0/\{1+i(\Phi k-\tilde{\Phi}\eta+i\tilde{\xi}_i-\xi)f_0\}. \quad (4.9)$$

Thus, the lattice optical theorem, first, allows us to express q_0 and $\tilde{f}_0$ in terms of f_0 and, second, leaves [recalling the remark concerning $\tilde{\xi}_i(k)$] only one unknown parameter $\tilde{\Phi}(k)$ that can be frequently determined from symmetry considerations (see below).

5. RESONANT SCATTERING

We consider now the following situation: 1) the entire scattering is pure elastic, i.e., the amplitudes $f_0(\omega)$ and $q_0(\omega)$ are mapped by points lying on the curves (4.1) and (4.4). 2) $\lambda>2a$, so that (4.2) and (4.3) contain only the amplitudes q_0 and $\bar{q}_{00}$. 3) the direction of the induced dipole moments $\mathbf{d}_{mn}$ is either perpendicular to the incidence plane, or is the bisector of the angle between the scattering directions $\mathbf{n}_{00}^+$ and $\mathbf{n}_{00}^-$ (the latter is possible at high polarizability for the given frequency[11,12]). In this case $\tilde{\Phi}$ can be easily determined from the symmetry of the problem. Since the amplitudes of scattering in directions that are symmetric about $\mathbf{d}$ are equal, it follows that $q_0=\bar{q}_{00}$, $S_{00}{}^{+2}=S_{00}{}^{-2}=1$ and $\tilde{\Phi}=\frac{1}{2}(1+S_{00}{}^{-2})=1$. All the values of $q_0(\omega)$ that are possible in (4.4) lie an the unity-diameter circle (Fig. 1b). The point of intersection of this circle with the real axis corresponds to the value $q_0=-1$ at which the transmitted wave $q_{tr}=1+q_0$ is completely damped and the entire incident energy goes over into the reflected wave ($\bar{q}_{00}=q_0=-1$). In other words, at a certain frequency one should observe total reflection of the radiation from one atomic plane at arbitrary θ. This effect follows directly from the optical theorem (4.4), although it says nothing about the corresponding frequency. It is physically clear, however, that this frequency should correspond to the resonance $\omega=\omega_0$ in scattering by a single site. In fact, an isolated dipole moment oscillates at resonance with a maximum amplitude $\mathbf{d}_{mn}(\omega_0)$. In pure elastic scattering, the resonant amplitude $f_0(\omega_0)=k_0^2 d_{mn}(\omega_0)/E_0$ reaches its unitary limit $i/\Phi k_0$, while the corresponding cross section is $\sigma=(\pi\Phi)^{-1}\lambda_0^2$. Under the condition $\lambda_0>2a$ this cross section is larger than σ_A, and it is this which leads to total reflection.

The foregoing is exactly described by Eq. (4.8), but according to this equation the resonant value $q_0=-1$ is attained by the lattice amplitude at a frequency $\tilde{\omega}_0$ somewhat different from the resonant atomic frequency ω_0. Substituting in the equation $q_0(f_0)=-1$ with $\tilde{\Phi}=1$ the expression for $f_0(\omega)$ near resonance

$$f_0(\omega)=(\Phi k)^{-1}\frac{\omega_\gamma/2}{\omega_0-\omega-i\omega_\gamma/2}, \quad (5.1)$$

we get

$$\tilde{\omega}_0=\omega_0+\delta\omega_0=\omega_0-\frac{\tilde{\xi}_i(\tilde{k}_0)}{2\Phi\tilde{k}_0}\omega_\gamma,\quad \tilde{k}_0\equiv\tilde{\omega}_0/c\approx\omega_0/c. \quad (5.2)$$

The shift of the resonant frequency is due to the shift of the atomic levels of the isolated sites when the latter are combined into a lattice, and is automatically taken into account by the term with $\tilde{\xi}_i(k)$ in (4.8).

It is possible, without calculating $\tilde{\xi}_i(k)$, to estimate the order of magnitude of $\delta\omega$ by using for $\xi(k)$ Eqs. (2.6) and (3.7), in which the main contribution to $\operatorname{Im}\xi$ is made at $\lambda>2a$ by the terms λ^2/a^3 from the nearest sites. Therefore

$$|\delta\omega|\approx{}^1/_2\frac{\tilde{\xi}_i(k_0)}{\Phi k_0}\omega_\gamma\sim\frac{\xi_i(k_0)}{k_0}\omega_\gamma\sim\left(\frac{\lambda_0}{a}\right)^3\frac{r_e}{c}\omega_0{}^2\sim\left(\frac{r_e c}{a^3}\right)\lambda_0. \quad (5.3)$$

Equation (4.8) contains as a particular case the known effect of total reflection of light and radiowaves form an ideally conducting surface. In (4.8) this effect corresponds to scattering near the "resonance" $\omega_0=0$ (scattering by the free electrons of the metal). In the general case, however, (4.8) describes the possibility of total reflection from a system of bound electrons. This reflection can be observed all the way to the x-ray band. It is effected not by the interface between two media and not by a stack of planes, as in the Bragg mechanism, but by a single atomic plane. Thus, we are dealing in fact with a new modification of total reflection, connected with purely elastic atomic resonance with sites of a two-dimensional crystal. This effect can be called total resonant reflection.

6. SUPPRESSION OF INELASTIC CHANNELS

We proceed now to the general case, when inelastic channels exist in atomic scattering. The corresponding expression for $f_0(\omega)$ near an isolated resonance is

$$f_0(\omega)=(\Phi k)^{-1}\frac{\omega_\gamma/2}{\omega_0-\omega-i\omega_\Gamma/2}, \tag{6.1}$$

where $\omega_\Gamma=\omega_\gamma+\omega_r$ is the total width, and ω_γ and ω_r are the width of the elastic and inelastic scattering. Substituting this in (4.8) at $\tilde{\Phi}=1$, we get

$$q_0=\bar{q}_{00}=\frac{i\tilde{\omega}_\gamma/2}{\tilde{\omega}_0-\omega-i\tilde{\omega}_\Gamma/2}, \tag{6.2}$$

where

$$\tilde{\omega}_\gamma\equiv\frac{\eta}{\Phi k}\omega_\gamma,\quad \tilde{\omega}_\Gamma\equiv\omega_\Gamma-\left(1-\frac{\eta}{\Phi k}\right)\omega_\gamma=\omega_r+\frac{\eta}{\Phi k}\omega_\gamma. \tag{6.3}$$

and $\tilde{\omega}_0$ is determined by (5.2).

Expression (6.2) has the same form as (6.1) and describes resonance with a scatterer having a natural frequency $\tilde{\omega}_0$ and effective widths $\tilde{\omega}_\gamma$ and $\tilde{\omega}_\Gamma$; these widths play the role of the elastic and total width of the planar-scattering resonance line. Inasmuch as at $\lambda_0>2a$ we have

$$\tilde{\omega}_\gamma/\omega_\gamma=\eta/\Phi k\approx(2\pi\Phi\sin\theta)^{-1}(\lambda_0/a)^2>1. \tag{6.4}$$

Equation (6.3) describes the broadening of the scattering line. This broadening corresponds to merging of the resonant levels of individual sites into a band when the two-dimensional crystal is formed. The width of the produced band is proportional, in accord with the band theory, to N_σ and increases together with λ_0. The last circumstance is due to the fact that the low transition frequency ω_0 couples close atomic levels, meaning high-lying atomic levels that are subject to large broadening.

Since lattic formation results not only in broadening but also in a shift of the atomic levels, the center of the resultant band is shifted relative to the initial level E_n. This leads to a resonant-frequency change $\omega_0\to\tilde{\omega}_0$, the value of which is the same for pure elastic as well as for inelastic scattering, and the relative shift $\delta\omega/\omega_0$ depends on λ_0 in the same manner as the broadening $\tilde{\omega}_\gamma/\omega_\gamma$.

Thus, the relations obtained connect the scattering picture with the energy band structure of a monatomic layer.

According to (6.3), the elastic and inelastic elementary widths enter in $\tilde{\omega}_\Gamma$ with different weight factors. This can lead to a unique effect of suppression of the inelastic channels in planar scattering. In fact, assume that for an isolated site we have

$$\beta\equiv\omega_\gamma/\omega_\Gamma\ll1, \tag{6.5}$$

i.e., only one particle out of $N=\beta^{-1}$ is elastically scattered. Nonetheless, under the condition

$$\tilde{\omega}_\gamma\equiv(\eta/\Phi k)\,\omega_\gamma\gg\omega_r,\ \text{i.e.}\ (\lambda_0/a)^2\gg2\pi\Phi\beta^{-1}\sin\theta \tag{6.6}$$

we obtain $\tilde{\omega}_\Gamma\approx\tilde{\omega}_\gamma$, and for a two-dimensional crystal almost the entire resonant scattering turns out to be elastic with amplitude $q_0=\bar{q}_{00}=-1$. If (6.6) is satisfied for $\theta=\pi/2$, almost total reflection from the atomic plane will be observed at resonance for arbitrary θ, although the same radiation is almost totally absorbed in the case of scattering by free sites. The physical reason is that in strong scattering and when condition (6.6) is satisfied the resultant field at the lattice sites becomes weak. Indeed, at resonance expression (3.8) for the field E_f takes, if (2.6), (3.7), and (6.6) are taken into account, the form

$$|E_f|\approx\left|\frac{E_0}{1-i\xi_i(k)(\beta/\Phi k)}\right|\approx\frac{E_0}{(\lambda_0/a)^3\beta}\ll E_0. \tag{6.7}$$

This restructuring of the field prevents the appearance of inelastic channels.

The optical theorem provides a geometrically lucid interpretation of this effect. According to (4.1) and (6.1), the total cross section in the presence of inelastic channels is a fraction β of the unitary limit $\lambda_0^2/\Phi\pi$. The elastic cross section, on the other hand, is a fraction β of the total cross section, i.e., it is equal to $\beta^2(\lambda_0^2/\Phi\pi)$ (Ref. 13). Under condition (6.6) even these small fractions become comparable with the area σ_A of the unit cell and "overlap" the lattice.

We note that suppression of inelastic channels is a known effect in diffraction by a three-dimensional lattice.[14,17] The process considered here pertains to one atomic plane and is brought about by an entirely different mechanism. The wave-field redistribution, which causes the field at the sites to be weak, is due here to the resonant growth of the elastic cross section at the individual site. Therefore the entire effect is connected with the discrete atomic frequencies and can be observed at any value of θ.

To observe the effect in experiment it suffices to have a perfect lattice of monatomic thickness on a substrate transparent to the radiation in the given band. We consider by way of example a monatomic Li film on the surface of a suitable sample. Let the incident particles be photons that are resonably scattered by the $1s\leftrightarrow2p$ transition of the Li atom, and resonant wavelengths $\lambda_0=220$ Å for the unexcited atom and 200 Å for an atom with an excited $2s$ electron.[18] The probability of the radiative $2p\leftrightarrow1s$ transition in Li with elastic scattering of the photon is $\omega_\gamma\approx(e^2/\hbar c^3)a_0^2\,\omega_0^3\approx2.5\cdot10^{11}\,s^{-1}$. The probability of the main inelastic process, viz., emission of an Auger electron from the L shell, is $\omega_r\approx1.86\times10^{14}\,s^{-1}$ (Ref. 19). Allowance for the fact that the Li outer electron is strongly shifted to the substrate yields $\omega_r\approx10^{13}\,s^{-1}$. Using by way of estimate the intermediate value $\omega_r\approx2.5\cdot10^{13}\,s^{-1}$, we obtain $\beta=10^{-2}$ (one photon out of 100 is scattered elastically). In scattering by a two-dimensional crystal with $a=3$ Å we have $(\lambda_0/a)^2=5\cdot10^3$, and the condition (6.6) is satisfied for all θ. Equation (6.2) yields for the parameters considered $q_0=\bar{q}_{00}\approx-0.9$, so that the reflection coefficient at normal incidence is ≈0.8, and the transmission coefficient is $(1+q_0)^2\approx10^{-2}$. Approximately 20% of the total radiation is dissipated at the lattice sites.

7. CONCLUSION

The method used in the paper can be generalized to take into account the possible transfer of energy to collective excitations. For the case of phonon excitations this allowance should lead to the appearance, in (3.14), of factors of the type $\exp(-\overline{u^2}/2\lambda^2)$, where $\overline{u^2}$ is the mean squared site displacement from the equilibrium position.

The problem considered and the main results pertain in principle to scattering of particles of all kinds. The main equation (4.6) can be used not only for an individual plane, but also to a layer of a substance. In this case, however, account must be taken of the effects of damping and phase shift between the oscillations of the induced dipoles at different depths of the layer. Owing to these effects, elementary radiators on opposite faces of the layer cannot be regarded on a par, and the symmetry considerations used in Sec. 5, which lead to the equalities $q_{00}{}^{+} = \bar{q}_{00}$, $\widetilde{\Phi} = 1$ and by the same token to the possibility of total reflection, are no longer valid. Therefore an individual atomic plane can, under certain conditions, reflect radiation better than a whole crystal. This can be used, for example, to develop frequency-selective radiation reflectors, and to investigate a number of surface effects.

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