Multiwave resonance in fast-electron diffraction in thin crystals

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The problems of the diffraction and inelastic scattering of nonrelativistic fast electrons in crystals are solved for the case in which the angle (ϑ) between the momentum vector of the particle and a crystallographic axis is many times the Lindhard angle. The solutions are analyzed. At certain values of ϑ, a resonant coupling may arise between states of quasifree motion of the particle and Bloch states from the low-lying narrow band in the energy spectrum of an electron in the $2D$ transverse periodic potential of the crystal. The onset of a resonance of this type in the final state of the inelastic-scattering problem may be responsible for the anomalies which have recently been seen in experiments on the transmission and reflection of electrons from crystalline targets.

INTRODUCTION

In research on the motion of fast electrons in crystals it is generally assumed that the periodic potential has a strong effect on the shape of the trajectory only if the angle between the particle momentum **p** and the atomic plane or axis is less than or equal to the Lindhard angle:

$$\vartheta \lesssim \vartheta_L = (\overline{U}/\varepsilon_p)^{1/2}, \qquad (1)$$

where $\overline{U}$ is on the order of the internal crystal potential, and ε_p is the energy of the particle.[1] In a quantum-mechanical formulation of the scattering problem, inequality (1) is the condition of a pronounced restructuring of a plane wave incident on a crystal, accompanied by simultaneous excitation of a large number of Bragg diffraction reflections.[2-6] In particular, when condition (1) holds, the whole set of the coefficients in the expansion of the electron wave function in a perturbation-theory series in Fourier components of the periodic crystal potential, which correspond to a certain plane of the reciprocal-lattice vectors orthogonal to the particle momentum **p** become anomalously large (Ref. 2). From the geometric standpoint, condition (1) is equivalent to the configuration in Fig. 1a, in which the Ewald sphere[7] $\mathbf{p}^2 = 2m\varepsilon_p$ is tangent to this plane of reciprocal-lattice vectors.

However, it is not difficult to show that the relative positions of the Ewald sphere and the three-dimensional system of reciprocal-lattice sites in Fig. 1a do not account for all the cases which might correspond to a pronounced dynamic restructuring of the wave function of the particles incident on the crystal. In particular, the configuration in Fig. 1b also corresponds to the case of the simultaneous excitation of many diffraction reflections—a number of reflections comparable in order of magnitude to the number of features in case 1a. We denote by g the distance between successive planes of reciprocal-lattice vectors. It is a straightforward matter to derive the angle ϑ_r, between the momentum vector of the electron and the crystal axis, for the case corresponding to Fig. 1b:

$$\vartheta_r = 2 \arcsin\,[(g/2p)^{1/2}]. \qquad (2)$$

The probability for the excitation of Bragg reflections in case 1b can be estimated in the following way. The angle (2) through which the electron is deflected in the course of the scattering corresponds to a momentum transfer on the order of $\Delta p \sim (2pg)^{1/2}$. Since the matrix element of the periodic crystal potential corresponding to a diffractive reflection by a certain reciprocal-lattice vector **G** contains a Debye-Waller factor[2] $\exp(-1/2\langle(\mathbf{Gu})^2\rangle)$, all processes with a momentum transfer $\Delta p \gtrsim (\langle u^2\rangle)^{-1/2}$ are suppressed (here and below, we are using a system of units with $\hbar = 1$). Comparison of the latter inequality with (2) leads to an upper limit on the energy of the electrons:

$$\varepsilon_p \lesssim 1/mg^2(\langle u^2\rangle)^2. \qquad (3)$$

Under ordinary conditions the right side of (3) would be some tens of kiloelectron volts, suggesting that a special motion of the fast electrons in the crystal might arise at nonrelativistic energies in the scattering geometry in Fig. 1b.

As we will see below, for a motion of this type a multiwave resonant coupling typically arises between (on the one hand) free semiclassical states of the particle which correspond to relatively large angles between **p** and the lat-

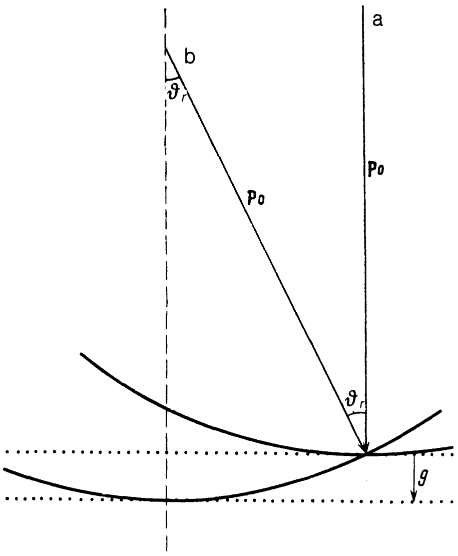

FIG. 1. Relative positions of the Ewald sphere and the system of reciprocal-lattice sites of the crystal. a—The electrons are incident approximately normally on the crystal surface, and inequality (1) holds; b—a multiwave resonance arises in the solution of the diffraction problem.

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tice axes and (on the other) "bound" Bloch states corresponding to narrow bands of transverse motion in the $2D$ periodic potential of the crystal. In particular, under the conditions of this multiwave resonance there may be a complete "capture" of a nonrelativistic electron from a plane wave into a bound state of transverse motion, localized near atomic axes of the crystal (by way of comparison, the probability for the filling of bound states in the case in which electrons are incident normally on a target is no greater than $\sim 50\%$; Ref. 8).

Special motivation for analyzing multiwave resonant scattering comes from some anomalies recently seen experimentally in the angular distributions of electrons scattered inelastically through relatively large angles with respect to the lattice axes of a crystal.[9-13] Some nearly regular intensity rings were observed in Refs. 9–13 in addition to the customary angular distribution of the scattered particles (a pattern of intersecting lines and bands[3,4]). These rings appeared both in experiments on the passage of fast electrons through thin crystals[11] and in experiments on reflection[9,12] and backscattering[13] from bulk samples. The expression derived below for the differential cross section for inelastic collisions of fast electrons in a thin crystal makes it possible to work from ideas concerning a multiwave resonance to explain the experimental results of Refs. 9–13. In several cases, it becomes possible to find a good quantitative agreement between the theoretical intensity profiles and the observed angular distributions of inelastically scattered particles.

1. WAVE FUNCTION IN THE ELASTIC-SCATTERING PROBLEM

The motion of a fast electron in matter can be represented as a diagram sequence of events of elastic scattering by a periodic crystal potential averaged over thermodynamic equilibrium,

$$U_0(\mathbf{r})=\frac{1}{\mathscr{Z}}\sum_n e^{-E_n/T}\langle n|U(\mathbf{r})|n\rangle, \tag{4}$$

and of inelastic collisions in exciting internal degrees of freedom of the target.[2] The summation in (4) is over the eigenstates of the electron and phonon subsystems of the crystal, $|n\rangle$, with energies E_n; T is the absolute temperature; and $\mathscr{Z}=\Sigma_n\exp(-E_n/T)$. In accordance with the usual experimental situation,[10,11] we restrict the analysis below to the case in which electrons pass through a relatively thin crystal, whose dimension (L) along the direction of motion of the particles does not exceed the mean free path (l) of the electrons with respect to inelastic interactions (at $\varepsilon_p\sim 10^5$ eV, for example, the mean free path of the electrons with respect to the excitation of the phonon subsystem of the crystal is[14] 10^3 Å).

The wave function of the problem of elastic scattering in a thin crystal is the solution of the equation

$$-\frac{1}{2m}\frac{\partial^2}{\partial\mathbf{r}^2}\Psi+U_0(\mathbf{r})\Psi=\varepsilon_p\Psi \tag{5}$$

with a boundary condition for the incident wave,

$$\psi_{inc}(\mathbf{r})=e^{i\mathbf{pr}}. \tag{5a}$$

We assume that the crystal occupies the spatial region $0<z<L$ and that the vector $\mathbf{p}$ makes a small angle ϑ with the z axis. Under condition (1) for fast particles, with

$$pg/m\gg|\bar{U}|, \tag{6}$$

in solving Eq. (5) we can restrict the Fourier expansion of potential (4) to reciprocal-lattice vectors $\{\mathbf{G}_h\}$ which lie in the xy plane.[2] We seek the wave function of the fast electron in (5), (5a) as a superposition of transmitted and diffracted waves with slowly varying amplitudes:

$$\Psi(\mathbf{r})=\sum_h\varphi_h(z)\exp[i(\mathbf{p}+\mathbf{G}_h)\mathbf{r}]. \tag{7}$$

Substituting (7) into (5), we find a system of coupled differential equations for the amplitudes $\varphi_h(z)$:

$$v\cos\vartheta\frac{\partial}{\partial z}\varphi_h+i(\varepsilon_h-\varepsilon_0)\varphi_h+i\sum_s\Lambda_{hs}\varphi_s=0, \tag{8}$$

where v is the velocity of the electron, $\varepsilon_h=(\mathbf{p}+\mathbf{G}_h)^2/2m$, and the quantities Λ_{hs} are the Fourier components of the periodic crystal potential,

$$\Lambda_{hs}=\Lambda(\mathbf{G}_h-\mathbf{G}_s)=\frac{1}{\Omega_0}\int_{\Omega_0}d^3r U_0(\mathbf{r})\exp[-i(\mathbf{G}_h-\mathbf{G}_s)\mathbf{r}], \tag{9}$$

where Ω_0 is the volume of the unit cell.

The condition $\varepsilon_p\gg|\bar{U}|$ allows us to ignore the reflected waves in formulating the boundary conditions on (8), so we can write[15]

$$\varphi_h(0)=\delta_{h0}. \tag{10}$$

We can solve Eqs. (9) by transforming the set of functions $\{\varphi_h(z)\}$ to the new representation

$$\varphi_j(z)=\sum_h(C^+(\mathbf{q}))_{jh}\varphi_h(z), \tag{11}$$

where the unitary matrix $C_{hj}(\mathbf{q})$ is fixed by the condition

$$\sum_{s,h}(C^+(\mathbf{p}))_{js}\left\{\frac{(\mathbf{q}+\mathbf{G}_h)^2}{2m}\delta_{sh}+\Lambda_{sh}\right\}C_{hj'}(\mathbf{p})=E_j\delta_{jj'}=E_j(\mathbf{q})\delta_{jj'}. \tag{12}$$

A direct comparison of (11) and (12) with Eqs. (3.8) and (3.9) of Ref. 2 easily reveals that the matrix $C_{hj}(\mathbf{q})$, which depends on only the projection $\mathbf{q}$ of the momentum $\mathbf{p}$ onto the xy plane, determines a basis of Bloch functions of the transverse motion in the $2D$ potential $U_0(x,y)$:

$$b_j(\mathbf{q},\boldsymbol{\rho})=\sum_h C_{hj}(\mathbf{q})\exp[i(\mathbf{q}+\mathbf{G}_h)\boldsymbol{\rho}], \tag{13}$$

where $\boldsymbol{\rho}$ is a $2D$ vector with the components x, y. In representation (11) and (12), the solution (10) becomes

$$\varphi_j(z)=C_{0j}^*(\mathbf{q})\exp\left[\frac{iz}{v\cos\vartheta}\left(\frac{\mathbf{q}^2}{2m}-E_j\right)\right]. \tag{14}$$

The value of $\varphi_j(z)$ at $z=0$ is the same as the known result from the sudden-perturbation theory:[1,2]

$$\varphi_j(0)=C_{0j}^*(\mathbf{q})=\frac{1}{S_0}\int_{S_0}d^2\rho\, b_j^*(\mathbf{q},\boldsymbol{\rho})e^{i\mathbf{q}\boldsymbol{\rho}}, \tag{15}$$

where S_0 is the area of the $2D$ unit cell of the crystal in the xy plane.

Using (13) and (14), we can write the wave function of scattering problem (5) as

$$\Psi(\mathbf{r})=e^{ikz}\sum_j b_j(\mathbf{q},\boldsymbol{\rho})C_{0j}^*(\mathbf{q})\exp\left[\frac{iz}{v\cos\vartheta}\left(\frac{\mathbf{q}^2}{2m}-E_j\right)\right], \tag{16}$$

where $k=p\cos\vartheta$. In particular, for the density of particles in the crystal we find [see Eq. (4.5) of Ref. 2]

$$|\Psi(\mathbf{r})|^2=\sum_{j,j'}C_{0j}^*(\mathbf{q})C_{0j'}(\mathbf{q})b_j(\mathbf{q},\boldsymbol{\rho})b_{j'}^*(\mathbf{q},\boldsymbol{\rho})$$
$$\times\exp\left[-\frac{iz}{v\cos\vartheta}(E_j-E_{j'})\right]. \tag{17}$$

Equations (7)–(16) completely solve the problem of the scattering of a fast electron in a crystal under the condition that the angle made by the momentum of the particle with the crystal axis is no greater in order of magnitude than ϑ_L in (1) (Ref. 5).

We turn now to an analysis of the scattering geometry in Fig. 1b. At angles in the intermediate region,

$$\vartheta_L\ll\vartheta\ll\vartheta_r,$$

the electron wave function is a plane wave perturbed slightly by Bragg diffraction reflections of higher orders. A new and radical change occurs in the nature of the electron motion when the angle ϑ reaches the value ϑ_r. At $\vartheta\approx\vartheta_r$, reflections corresponding to reciprocal-lattice vectors $\{\mathbf{G}_h\}$ with a nonzero z projection, for which we have

$$\mathbf{G}_h\mathbf{e}_z=g, \tag{18}$$

play an important role in shaping the wave function.

In describing the electron motion in this case, it is not possible to distinguish between independent "longitudinal" and "transverse" degrees of freedom of the particle. We now have to take account of the z dependence of potential (4) in Eq. (5). It might appear at first glance that at $\vartheta\sim\vartheta_r$ the diffraction problem would become substantially three-dimensional and that the solution of the problem, like that given in Ref. 16, should contain a set of arbitrarily oriented reciprocal-lattice vectors $\{\mathbf{G}_h\}$. As it turns out, however, at $\vartheta\approx\vartheta_r$ inequality (6) allows us to restrict the analysis to three terms in the expansion of the potential,

$$U_0(\mathbf{r})=U_{-1}(\boldsymbol{\rho})e^{-igz}+U_0(\boldsymbol{\rho})+U_1(\boldsymbol{\rho})e^{igz}, \tag{19}$$

and two terms in the wave function,

$$\Psi(\mathbf{r})=e^{ikz}\Psi_0(\boldsymbol{\rho},z)+e^{i(k+g)z}\Psi_1(\boldsymbol{\rho},z). \tag{20}$$

As can be seen from Fig. 1b, at $\vartheta\sim\vartheta_r$ the elastic scattering is characterized by a relatively large momentum transfers $\Delta p\sim(2pg)^{1/2}$ which correspond to small Fourier components of potential (10). For this reason, incorporating the next [in terms of $\exp(igz)$] harmonics in expansion (19) leads to the appearance in the wave function of only small terms on the order of $|m\Lambda/Gp|$, which can be ignored for fast particles according to condition (6).

Substituting (19) and (20) into (5), we find a system of two coupled equations for the functions $\Psi_0(\boldsymbol{\rho},z)$ and $\Psi_1(\boldsymbol{\rho},z)$:

$$\frac{k^2}{2m}\Psi_0-i\frac{k}{m}\frac{\partial}{\partial z}\Psi_0-\frac{1}{2m}\frac{\partial^2}{\partial\boldsymbol{\rho}^2}\Psi_0+U_0(\boldsymbol{\rho})\Psi_0$$
$$+U_{-1}(\boldsymbol{\rho})\Psi_1=\varepsilon_p\Psi_0,$$
$$\frac{(k+g)^2}{2m}\Psi_1-i\frac{(k+g)}{m}\frac{\partial}{\partial z}\Psi_1-\frac{1}{2m}\frac{\partial^2}{\partial\boldsymbol{\rho}^2}\Psi_1 \tag{21}$$
$$+U_0(\boldsymbol{\rho})\Psi_1+U_1(\boldsymbol{\rho})\Psi_0=\varepsilon_p\Psi_1,$$

with the boundary condition

$$\Psi_0(\boldsymbol{\rho},0)=\exp(i\mathbf{q}\boldsymbol{\rho}),\qquad \Psi_1(\boldsymbol{\rho},0)=0.$$

The quantity $U_0(\boldsymbol{\rho})$ in (21) describes a diffraction by a 2*D* potential of a crystal, averaged along the z axis. The terms $U_{\pm1}(\boldsymbol{\rho})$ give rise to components of wave function (5) which oscillate rapidly in the longitudinal direction. It can be seen from Fig. 1b that the diffraction reflections fixed by condition (18) are the ones of primary interest in Eqs. (21). In other words, in solving problem (21) in the limit $\vartheta\gg\vartheta_L$ it is usually legitimate to ignore the effect of the potential $U_0(\boldsymbol{\rho})$ on the first component of wave function (20) (Peng[12] pointed out that this approximation could be used). Using the equality $q^2+k^2=2m\varepsilon_p$ and the condition $g/k\ll1$, we find from (21)

$$i\frac{k}{m}\frac{\partial}{\partial z}\Psi_0=-\frac{\mathbf{q}^2}{2m}\Psi_0-\frac{1}{2m}\frac{\partial^2}{\partial\boldsymbol{\rho}^2}\Psi_0+U_{-1}(\boldsymbol{\rho})\Psi_1,$$
$$i\frac{k}{m}\frac{\partial}{\partial z}\Psi_1=\left\{\frac{(k+g)^2}{2m}-\frac{k^2}{2m}-\frac{\mathbf{q}^2}{2m}\right\}\Psi_1 \tag{22}$$
$$-\frac{1}{2m}\frac{\partial^2}{\partial\boldsymbol{\rho}^2}\Psi_1+U_0(\boldsymbol{\rho})\Psi_1+U_1(\boldsymbol{\rho})\Psi_0.$$

Before we solve Eqs. (22), we would like to point out that the second component $\Psi_1(\boldsymbol{\rho},z)$ of wave function (20) becomes comparable to the first near the point $\mathbf{q}_0$, for which

$$(k+g)^2=k^2+\mathbf{q}_0^2. \tag{23}$$

Comparing (23) with (2), we see that (23) is simply the condition for a tangency of the Ewald sphere and the lower of the planes of reciprocal-lattice vectors of the crystal shown in Fig. 1 (in the terminology of Ref. 4, this is the plane of vectors of the minus-first Laue zone). By analogy with (8)–(15), we seek a solution of (22) in the form

$$\Psi_0(\boldsymbol{\rho},z)=\alpha(z)e^{i\mathbf{q}\boldsymbol{\rho}}, \tag{24}$$
$$\Psi_1(\boldsymbol{\rho},z)=\sum_{\varkappa,j}\beta_{\varkappa j}(z)b_j(\boldsymbol{\varkappa},\boldsymbol{\rho}),$$

where the summation over $\varkappa$ and j is over the quasimomenta and indices of the energy bands in the spectrum of transverse-motion states in (12). Substituting (24) into (22), we find the following expression for the slowly varying coefficients $\alpha(z)$ and $\beta_{\varkappa j}(z)$:

$$i\frac{k}{m}\frac{\partial}{\partial z}\alpha=\sum_{\varkappa,j}\beta_{\varkappa j}(z)\left\{\int d^2\rho\, e^{-i\mathbf{q}\boldsymbol{\rho}}U_{-1}(\boldsymbol{\rho})b_j(\boldsymbol{\varkappa},\boldsymbol{\rho})\right\},$$
$$i\frac{k}{m}\frac{\partial}{\partial z}\beta_{\varkappa j}=[E_j(\boldsymbol{\varkappa})-D(\mathbf{q})]\beta_{\varkappa j} \tag{25}$$
$$+\alpha(z)\sum_{\varkappa,j}\left\{\int d^2\rho\, b_j^*(\boldsymbol{\varkappa},\boldsymbol{\rho})U_1(\boldsymbol{\rho})e^{i\mathbf{q}\boldsymbol{\rho}}\right\},$$

where

$$D(\mathbf{q})=\frac{\mathbf{q}^2}{2m}+\frac{k^2}{2m}-\frac{(k+g)^2}{2m}.$$

To evaluate the matrix elements on the right side of (25), we expand the potential $U_1(\boldsymbol{\rho})$ in reciprocal-lattice vectors:

$$U_1(\boldsymbol{\rho})=\sum_l U_1(\mathbf{G}_l)\exp(i\boldsymbol{\Delta}\boldsymbol{\rho}+i\mathbf{G}_l\boldsymbol{\rho}). \qquad (26)$$

We wish to stress that the series in (26) differs from the usual expansion (9), since the vector $g\mathbf{e}_z$ generally does not belong to the set of reciprocal-lattice vectors of the crystal (except, perhaps, in the case of a simple cubic lattice). For this reason we have had to introduce an additional phase factor $\exp(i\boldsymbol{\Delta}\boldsymbol{\rho})$ in (26), where $\boldsymbol{\Delta}$ is chosen such that the sum $g\mathbf{e}_z+\boldsymbol{\Delta}$ is equal to one of the reciprocal-lattice vectors. Using (13) and (26), we find an explicit expression for the matrix element on the right side of (25):

$$\int d^2\rho\, b_j^*(\boldsymbol{\varkappa},\boldsymbol{\rho})U_1(\boldsymbol{\rho})e^{i\mathbf{q}\boldsymbol{\rho}}$$
$$=(2\pi)^2\sum_{h,l}C_{hj}^*(\boldsymbol{\varkappa})U_1(\mathbf{G}_l)\delta(\boldsymbol{\Delta}+\mathbf{G}_l-\boldsymbol{\varkappa}-\mathbf{G}_h+\mathbf{q}), \qquad (27)$$

where the arbitrariness in the choice of $\mathbf{G}_l$ and $\mathbf{G}_h$ has been eliminated by the condition that the quasimomentum $\boldsymbol{\varkappa}$ is referred to one of the Brillouin zones. When we choose $\boldsymbol{\varkappa}=\mathbf{q}+\boldsymbol{\Delta}$, for example, we obtain $\mathbf{G}_l=\mathbf{G}_h$. Denoting the coefficient of the δ-function in (27) by $U_1(j)$, and omitting the first subscript in $\beta_{\varkappa j}$ for simplicity, we find from (25)

$$i\frac{k}{m}\frac{\partial}{\partial z}\alpha=\sum_j U_1^*(j)\beta_j,$$
$$i\frac{k}{m}\frac{\partial}{\partial z}\beta_j=[E_j(\mathbf{q}+\boldsymbol{\Delta})-D(\mathbf{q})]\beta_j+\alpha\sum_j U_1(j). \qquad (28)$$

The boundary conditions on (28) are $\alpha(0)=1$, and $\beta_j(0)=0$ for all j.

Equations of the type in (28) are well known in the theory of resonant scattering.[17,18] Their solution depends on the parameters expressing the detuning from the resonances:

$$\delta_j(\mathbf{q})=E_j(\mathbf{q}+\boldsymbol{\Delta})-D(\mathbf{q}). \qquad (29)$$

Let us consider the behavior of the quantities in (29) as a function of the angle between the electron momentum and the crystal axis. Here we make use of symmetry relations for Bloch wave functions,[19,20]

$$E_j(\mathbf{q})=E_j(\mathbf{q}+\mathbf{G}_l),$$
$$C_{hj}(\mathbf{q}+\mathbf{G}_l)=C_{h+l,j}(\mathbf{q}), \qquad (30)$$

which are valid for all reciprocal-lattice vectors which lie in the xy plane.

Figure 2 shows a cross section of the multisheet $E_j(\mathbf{q})$ surface in the first Brillouin zone, which we have calculated as an example of a $2D$ potential field of atomic chains which run along the ⟨100⟩ axis of a molybdenum crystal. We see in this figure a narrow band of states localized near the axes of the atomic chains, along with a system of high-lying levels which correspond to a nearly free transverse motion of the electrons above the atomic planes of the crystal. The band structure shown in the nonrelativistic region of electron energies in Fig. 2 is fairly typical of most elements in the periodic table. The corresponding curves for the crystals of various elements differ for the most part only in the nature of the dispersion of the states in the lower band, $E_0(\mathbf{q})$, which weakens with increasing atomic number of the target atoms. In the case at hand (the Mo crystal), the dispersion does not exceed 4%, and $E_0(\mathbf{q})$ lies in the interval -85 eV $\leqslant E_0(\mathbf{q})\leqslant -82$ eV. The states in the lower band are separated from the spectrum of quasifree motion by an energy interval on the order of 100 eV.

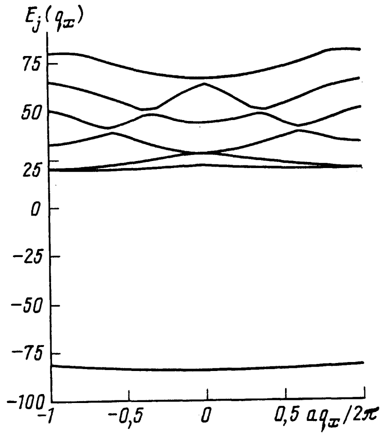

FIG. 2. Intersection of the multisheet $E_j(\mathbf{q})$ surface calculated for the ⟨100⟩ axis of a molybdenum crystal with a (100) plane. The calculation was carried out with 29 reciprocal-lattice vectors orthogonal to ⟨100⟩; the Fourier components of the potential were taken from Ref. 23.

The dependence in Fig. 2 determines the nature of the solutions of Eqs. (28). Specifically, the value of $D(\mathbf{q})$ from (28) is negative at $\mathbf{q}=0$, and at $\varepsilon_p=100$ keV it is given in order of magnitude by $D(0)\approx -pg/m\approx -2.5\cdot 10^3$ eV. With increasing $|\mathbf{q}|$, the function $D(\mathbf{q})$ increases rapidly. As a result, the coefficients $\delta_0(\mathbf{q})$, $\delta_1(\mathbf{q})$, etc., vanish in succession. Of particular interest, as we will see below, is an analysis of the behavior of solutions (28) in the region with $|\delta_0(\mathbf{q})|\lesssim|U_i(j=0)|$. According to the $E_j(\mathbf{q})$ plot shown in Fig. 2, the following inequalities hold in this interval of the vector $\mathbf{q}$:

$$|\delta_0(\mathbf{q})|\ll|\delta_1(\mathbf{q})|,\quad |\delta_0(\mathbf{q})|\ll|\delta_2(\mathbf{q})|,\ \ldots. \qquad (31)$$

Because of these inequalities, we can use the approximation of an isolated resonance[17] in analyzing (28). A further increase in $|\mathbf{q}|$ (i.e., in the angle ϑ between the electron momentum and the crystal axis) results in the simultaneous vanishing of several of the parameters $\delta_j(\mathbf{q})$ in (29) [e.g., $\delta_1(\mathbf{q})$, $\delta_2(\mathbf{q})$ and $\delta_3(\mathbf{q})$] at closely spaced points, and a transition occurs to a resonance regime with a large number of overlapping levels.[17]

Under conditions (31), Eqs. (28) reduce to a system of two equations:

$$i\frac{k}{m}\frac{\partial}{\partial z}\alpha=U_1^*(0)\beta_0,$$
$$i\frac{k}{m}\frac{\partial}{\partial z}\beta_0=[E_0(\mathbf{q}+\boldsymbol{\Delta})-D(\mathbf{q})]\beta_0+\alpha U_1(0), \qquad (32)$$

with the boundary conditions $\alpha(0) = 1, \beta_0(0) = 0$. The solution of (32) is simple in form:

$$\alpha(z)=\exp\left(-\frac{iz\varepsilon}{2v\cos\vartheta}\right)\left\{\cos\left[\frac{z}{2v\cos\vartheta}(\varepsilon^2+4|U|^2)^{1/2}\right]\right.$$
$$\left.+\frac{i\varepsilon}{(\varepsilon^2+4|U|^2)^{1/2}}\sin\left[\frac{z}{2v\cos\vartheta}(\varepsilon^2+4|U|^2)^{1/2}\right]\right\},$$
$$\beta_0(z)=-\frac{2iU}{(\varepsilon^2+4|U|^2)^{1/2}}\exp\left(-\frac{iz\varepsilon}{2v\cos\vartheta}\right)$$
$$\times\sin\left[\frac{z}{2v\cos\vartheta}(\varepsilon^2+4|U|^2)^{1/2}\right], \tag{33}$$

where we have used $\varepsilon = E_0(\mathbf{q}+\Delta) - D(\mathbf{q})$ and $U = U_1(0)$. The unusual nature of this solution becomes particularly noticeable when we use the strong-coupling approximation to calculate the wave functions of the Bloch states in the lower band.[7] Substituting (33) into (20), we find

$$\Psi(\mathbf{r})=\alpha(z)\exp(i\mathbf{pr})$$
$$+\beta_0(z)\exp[i(k+g)z]S_1^{1/2}\sum_A\exp[i(\mathbf{q}+\Delta)\boldsymbol{\rho}_A]\Phi(\boldsymbol{\rho}-\boldsymbol{\rho}_A), \tag{34}$$

where the localized orbital $\Phi(\boldsymbol{\rho})$ describes a $2D$ bound state in the potential of an isolated chain. The summation over A in (34) is over all atomic chains with centers at $\boldsymbol{\rho}_A$ which run along the z axis; and $S_1 = 1/\nu$, where ν is the number of chains per unit surface area.

Solution (34) describes a periodic restructuring of plane wave (5a) into a highly localized state with a minimal transverse-motion energy, and vice versa. By way of comparison, we note that a solution oscillating as in (33) for two amplitudes is well known in the theory of dynamic diffraction.[21,22] In the two-wave approximation of the dynamic theory according to (33), however, the amplitudes of the transmitted and diffracted plane waves oscillate, in contrast with the situation in (34). In the case discussed above, a resonant coupling arises between a plane wave and a state in a narrow band consisting of a coherent superposition of tens of diffraction reflections. One might say that resonance (34) represents physically an acceleration of the electron along the axes of atomic chains, accompanied by a simultaneous and sharp decrease in the energy of the transverse motion of the electron in the xy plane.

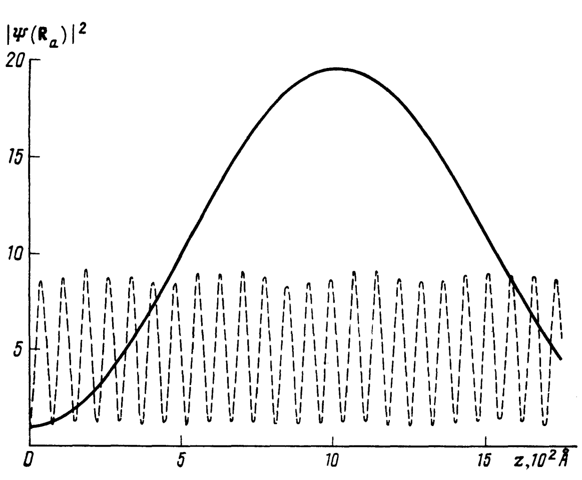

FIG. 3. Squared wave function of a fast electron at lattice sites versus the depth to which the particle penetrates into the crystal. Solid line—exact resonance, $\varepsilon = 0$; dashed line—calculated from (17) for motion along the ⟨100⟩ axis of a Mo crystal.

Exactly at resonance, with $\varepsilon = 0$, there is a complete restructuring of the plane wave at a depth $z_0 = \pi v \cos\vartheta/2|U|$ into a state of "channeling" along the axes of the atomic chains [however, the regime of motion in (34) cannot be referred to as "capture into a channel," since the probability for finding an electron in a low-lying narrow band of the energy spectrum vanishes again at a depth $2z_0$]. To compare the qualitative behavior of the solutions of scattering problems under the conditions of the resonance in (34) with the ordinary multiwave diffraction in (16), we show in Fig. 3 the squared electron wave function at the equilibrium points of the crystal nuclei ($\mathbf{R}_a$) versus the depth to which the particles penetrate into the target. As can be seen from Fig. 3, the electron density at the nuclei reaches its maximum not when the particles are incident normally on the crystal surface [(16), (17)] but when there is a finite angle ϑ between the momentum $\mathbf{p}$ and the lattice axis under the conditions of resonance (34). The reason is that when the particles are incident normally on the crystal surface the probability $|C_{0j}(\mathbf{p})|^2$ for the filling of above-barrier states[8] localized in the interatomic regions, which contribute next to nothing to the behavior shown in Fig. 3, is large. At the same time, under the conditions of resonance (32) there may be a complete restructuring of the plane wave into a below-barrier transverse-motion state localized at the nuclei. This intensification of the localization of the wave function of a fast electron at the crystal nuclei could cause pronounced changes in the cross sections for all inelastic processes involving small impact parameters: the excitation and ionization of inner shells, the emission of Auger electrons, the emission of characteristic x radiation, etc.

A parameter characterizing the angular width of the resonance is the potential matrix element $U_1(j)$. We must stress that the value $U_1(j=0)$ in the case of the isolated resonance of (31)–(34) is many times the Fourier component of the potential corresponding to ordinary elastic diffractive reflection through an angle ϑ_r from (2). The reason is that according to (27) the quantity $U_1(j)$ contains a factor not present in $U_1(\mathbf{q})$:

$$|U_1(j)|=\left|\sum_h C_{hj}^*(\mathbf{q}+\Delta)U_1(\mathbf{G}_h)\right|\sim|U_1(\mathbf{q})|\,|b_j(\mathbf{q}+\Delta,\boldsymbol{\rho}_A)|. \tag{35}$$

This factor is proportional to the probability amplitude for the localization of the wave function of the state in band j at the crystal nuclei. A numerical calculation carried out for the ⟨100⟩ axis of a molybdenum crystal with the data of Ref. 23 shows that $U_1(j=0)$ is approximately -2 eV, or about five times the corresponding Fourier component of the potential $U_1(\mathbf{q})$. Estimate (35) leads to an important conclusion: The multiwave resonance is most strongly manifested for just the states of the lower transverse-motion band, since it is for those states, localized at nuclei, that the matrix element $U_1(j)$ of the channel coupling, reaches its maximum value.

It thus follows from this analysis of the elastic-scattering problem that in the diffraction geometry in Fig. 1b the solution is resonant. This resonance may be accompanied by a pronounced restructuring of the electron wave function from a plane wave into a Bloch state localized at the crystal nuclei. In the next section of this paper we take a look at how the resonant elastic restructuring of the wave function alters the differential cross section for inelastic collisions of electrons accompanied by the excitation of internal degrees of freedom of the target.

2. INELASTIC SCATTERING UNDER CONDITIONS OF A MULTIWAVE RESONANCE

The cross section for inelastic scattering of particles in a medium can be expressed in terms of the nonvanishing matrix elements of the interaction potential $U(\mathbf{r})$ with changes of the quantum numbers of the internal degrees of freedom of the target:

$$\langle n'|\delta U(\mathbf{r})|n\rangle$$
$$=\langle n'|U(\mathbf{r})|n\rangle-\delta_{nn'}\sum_{n''}\frac{1}{\mathscr{Z}}\exp\left(-\frac{E_{n''}}{T}\right)\langle n''|U(\mathbf{r})|n''\rangle. \tag{36}$$

The expression for the differential inelastic cross section in a thin crystal ($L<l$), for scattering accompanied by a transition from the state $|n\rangle$ to $|n'\rangle$, is given by [Ref. 24, § 148]

$$d\sigma_{n\to n'}=\frac{2\pi}{v}\left|\int d^3r(\Psi_{\mathbf{p}'}^{(-)}(\mathbf{r}))^*\langle n'|\delta U(\mathbf{r})|n\rangle\Psi_{\mathbf{p}}^{(+)}(\mathbf{r})\right|^2$$
$$\times\delta\left(\frac{\mathbf{p}'^2-\mathbf{p}^2}{2m}+E_{n'}-E_n\right)\frac{d^3p'}{(2\pi)^3}, \tag{37}$$

where the wave functions of the initial and final states, $\Psi_{\mathbf{p}}^{(+)}(\mathbf{r})$ and $\Psi_{\mathbf{p}'}^{(-)}(\mathbf{r})$, constitute the solutions of the problems of elastic scattering in potential (4) which satisfy different boundary conditions at infinity [Ref. 24, § 136]

$$\Psi_{\mathbf{p}}{}^{(+)}(\mathbf{r})\propto\exp(i\mathbf{pr}), \tag{38}$$

$$\Psi_{\mathbf{p}'}^{(-)}(\mathbf{r})\propto\exp(-i\mathbf{p}'\mathbf{r}). \tag{39}$$

Since the asymptotic behavior in (38) is the same as (5a), the wave function found above, in (5) and (34), is actually one of the states $\Psi_{\mathbf{p}}^{(+)}(\mathbf{r})$ which we are seeking. In order to avoid having to solve Eq. (5) with boundary condition (39) repeatedly, we construct the wave function of the final state, $\Psi_{\mathbf{p}'}^{(-)}(\mathbf{r})$, using a corollary of the reciprocity theorem [Ref. 22 and Ref. 24, § 136]

$$(\Psi_{\mathbf{p}'}^{(-)}(\mathbf{r}))^*=\Psi_{-\mathbf{p}'}^{(+)}(\mathbf{r}). \tag{40}$$

After an averaging over the initial states of the target and summing over the final states, this result makes it possible to write the following expression for the differential cross section for the scattering of an electron into a solid-angle element do':

$$\frac{d\sigma}{do'}=\left(\frac{m}{2\pi}\right)^2\int d\omega\frac{v'}{v}\int d^3r\,d^3r'\bar{s}(\mathbf{r}',\mathbf{r};\omega)$$
$$\cdot\Psi_{-\mathbf{p}'}^{(+)}(\mathbf{r})\Psi_{\mathbf{p}}^{(+)}(\mathbf{r})[\Psi_{-\mathbf{p}'}^{(+)}(\mathbf{r}')\Psi_{\mathbf{p}}^{(+)}(\mathbf{r}')]^*, \tag{41}$$

where $v'=(\mathbf{p}^2-2m\omega)^{1/2}/m$, and $\bar{s}(\mathbf{r}',\mathbf{r};\omega)$ is defined as

$$\bar{s}(\mathbf{r}',\mathbf{r};\omega)$$
$$=\frac{1}{\mathscr{Z}}\sum_{n,n'}\langle n|\delta U(\mathbf{r}')|n'\rangle\langle n'|\delta U(\mathbf{r})|n\rangle\exp\left(-\frac{E_n}{T}\right)$$
$$\times\delta(\omega+E_n-E_{n'}). \tag{42}$$

The Fourier transform of (42) is proportional to the dynamic structure factor for inelastic excitations of the crystal:[25,26]

$$s(\mathbf{q},\mathbf{k};\omega)=\frac{q^2k^2}{(4\pi e^2)^2}\int d^3r\,d^3r'\bar{s}(\mathbf{r}',\mathbf{r},\omega)\exp(-i\mathbf{qr}'+i\mathbf{kr}). \tag{43}$$

In the case of the excitation and ionization of inner shells of atoms, or in the case of phonon scattering (Fig. 3), the behavior of cross section (41) as a function of the orientation of the momentum $\mathbf{p}$ of the incident electron is basically determined by the z dependence of the square of the electron wave function, $|\Psi_{\mathbf{p}}^{(+)}(\mathbf{r})|^2$, near the nuclei. We accordingly restrict the analysis below to the effect of the resonant restructuring of the wave functions $\Psi_{-\mathbf{p}'}^{(-)}(\mathbf{r})$ of the final states on the angular distribution of the particles emitted from the crystal.

Let us consider the case in which the momentum of the incident electrons is far from a close-packing direction,

$$\Psi_{\mathbf{p}}{}^{(+)}(\mathbf{r})=\exp(i\mathbf{pr}),$$

and the vector $-\mathbf{p}'$ satisfies the condition for an isolated resonance, (31),

$$\Psi_{-\mathbf{p}'}^{(+)}(\mathbf{r})=\alpha(z)\exp(-i\mathbf{p}'\mathbf{r})$$
$$+\beta_0(z)\exp(-ik'z+igz)S_1{}^{1/2}\sum_A\exp(-i\mathbf{q}'\boldsymbol{\rho}_A+i\Delta\boldsymbol{\rho}_A)\Phi(\boldsymbol{\rho}-\boldsymbol{\rho}_A). \tag{44}$$

The angular spread of the fast-particle flux in a crystal is determined primarily by the quasielastic thermal diffuse (phonon) scattering.[3,27] The structure factor for this type of scattering can be written in the Einstein model of thermal motion as a sum of independent contributions representing the scattering by individual crystal atoms:[21]

$$\bar{s}(\mathbf{r}',\mathbf{r};\omega)\approx\delta(\omega)\sum_a\bar{s}_a(\mathbf{r}'-\mathbf{R}_a;\mathbf{r}-\mathbf{R}_a). \tag{45}$$

Estimates show that incorporating the correlations in the thermal displacements of atoms[28] results in this case in only a slight change in the distribution of the diffuse intensity from the results calculated in model (45).

Substituting (4) and (45) into (41), we find the following expression for the angular distribution of electrons scattered quasielastically by a monatomic crystal:

$$\frac{d\sigma}{do'}=\sum_a\{|\alpha(z_a)|^2W_0(\mathbf{p}',\mathbf{p})+2\,\mathrm{Re}\,\beta_0^*(z_a)\alpha(z_a)W_1(\mathbf{p}',\mathbf{p})$$
$$+|\beta(z_a)|^2W_2(\mathbf{p}',\mathbf{p})\}, \tag{46}$$

where

$$W_0(\mathbf{p}',\mathbf{p})=\left(\frac{m}{2\pi}\right)^2\int d^3r\,d^3r'\bar{s}_a(\mathbf{r}',\mathbf{r})\exp[i(\mathbf{p}-\mathbf{p}')(\mathbf{r}-\mathbf{r}')],$$

$$W_1(\mathbf{p}',\mathbf{p})=\left(\frac{m}{2\pi}\right)^2\int d^3r\,d^3r'\bar{s}_a(\mathbf{r}',\mathbf{r})\exp[i(\mathbf{p}-\mathbf{p}')\mathbf{r}]$$
$$\times S_1{}^{1/2}\Phi(\boldsymbol{\rho}')\exp(-i\mathbf{pr}'+ik'z'-igz'),$$

$$W_2(\mathbf{p}',\mathbf{p})=\left(\frac{m}{2\pi}\right)^2\int d^3r\,d^3r'\bar{s}_a(\mathbf{r}',\mathbf{r})\exp[i\mathbf{p}(\mathbf{r}-\mathbf{r}')]$$
$$\times\exp[-i(k'-g)(z-z')]S_1\Phi(\rho)\Phi(\rho').$$

Replacing the summation over the lattice sites $\mathbf{R}_a$ in (46) by an integration,

$$\sum_a\to n\Sigma\int_0^L dz_a,$$

where n is the number of atoms per unit volume, and Σ is the surface area of the crystal, we find from (46) the final result for the angular distribution of the inelastically scattered particles:

$$\frac{1}{nL\Sigma}\left(\frac{d\sigma}{do'}\right)=W_0(\mathbf{p}',\mathbf{p})$$
$$+\{W_2(\mathbf{p}',\mathbf{p})-W_0(\mathbf{p}',\mathbf{p})\}\frac{1}{2(1+Y^2)}\left(1-\frac{\sin[\zeta(1+Y^2)^{1/2}]}{\zeta(1+Y^2)^{1/2}}\right)$$
$$+\mathrm{Re}\left\{\frac{U^*}{|U|}W_1(\mathbf{p}',\mathbf{p})\left[\frac{Y}{1+Y^2}\left(1-\frac{\sin[\zeta(1+Y^2)^{1/2}]}{\zeta(1+Y^2)^{1/2}}\right)\right.\right.$$
$$\left.\left.+\frac{i}{(1+Y^2)^{1/2}}\frac{1-\cos[\zeta(1+Y^2)^{1/2}]}{\zeta(1+Y^2)^{1/2}}\right]\right\}, \tag{47}$$

where

$$\zeta=2L|U|/v\cos\vartheta,\quad Y=[D(-\mathbf{q}')-E_0(-\mathbf{q}'+\Delta)]/2|U|.$$

It can be seen from this expression that the differential cross section for the quasielastic scattering has a sharp anomaly near directions which correspond to values of the parameter Y in the interval $|Y|\lesssim 1$. Since the function $E_0(\mathbf{q})$ depends only weakly on its argument, the equation

$$D(-\mathbf{q}')=E_0(-\mathbf{q}'+\Delta),\text{ i.e., } Y=0 \tag{48}$$

determines a curve which is nearly a circle in the q'_x, q'_y plane. A relation similar to (48) was proposed in Ref. 10 to explain the geometry of patterns observed experimentally (see also Refs. 12 and 13). According to (48), a sharp intensity anomaly arises in the angular distribution (47) of quasielastically scattered electrons. This anomaly takes the form of a ring around a close-packed crystallographic direction. An anomaly of precisely this type was recently observed in some experiments on the transmission of fast electrons through a gallium arsenide crystal.[11] Similar ring patterns had been observed previously by Peng *et al.*[9] in the reflection of fast particles from a crystal surface.

The radial distribution of the intensity near the ⟨100⟩ axis of a molybdenum crystal, calculated from (47), is shown in Fig. 4 (the momentum **p** is directed along the crystal axis). Values $Y<0$ correspond to the inner region (and values $Y>0$ to the outer region) of the ring patterns observed in the experiments of Refs. 9–13. As can be seen from Fig. 4, an increase in the incoherent intensity occurs near resonance (48) during the scattering of the particles, even in relatively thin crystals, in good agreement with the data of Ref. 11.

As the angle ϑ' (or, equivalently, the magnitude of the vector $\mathbf{q}'$) increases in (28), there is a transition to a regime of overlapping resonances involving above-barrier Bloch states. A weakly expressed fine structure appears in angular distribution (41) around ring pattern (47). The inner intensity ring is separated from this fine structure by a finite angular interval,[10,12] whose width is proportional to the size of the first band gap in the energy spectrum of transverse-motion states shown in Fig. 2.

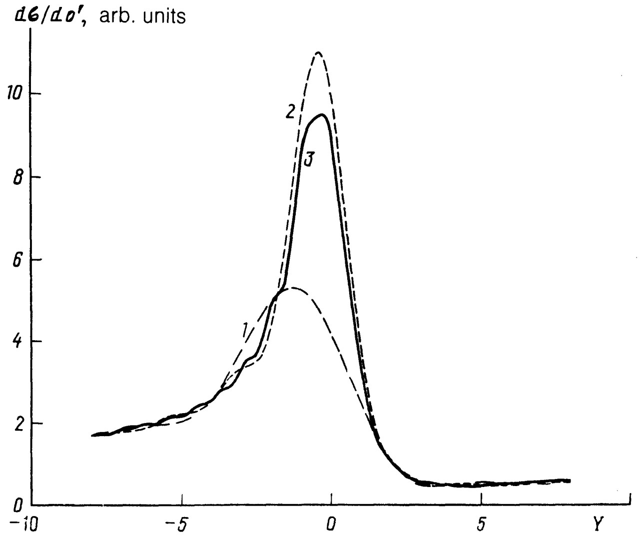

FIG. 4. Intensity distribution in the ring pattern which arises around the ⟨100⟩ axis of molybdenum during phonon scattering [Eq. (47)]. Crystal thickness: 1—L = 500 Å; 2—L = 1000 Å; 3—L = 2500 Å.

3. CONCLUSIONS

It follows from the discussion above that in the geometry shown in Fig. 1b for the diffraction of fast but nonrelativistic electrons in crystals a particle motion of a new type may arise. This new motion is characterized by a resonant coupling of free-motion states with states of the narrow lower band of the electron energy spectrum in the 2*D* effective potential of the crystal. The angular restructuring of the wave functions under multiwave-resonance conditions can substantially alter the angular distribution of the particles scattered inelastically by the crystal and also the extent of all inelastic processes with small collision impact parameters. Comparison of the theoretical results with experimental data on fast-particle scattering in thin crystals leads to the conclusion that the anomalies observed in the differential cross sections in Refs. 11 and 13 may stem from multiwave resonant elastic scattering on the motion of an electron in a crystal after an inelastic collision in the interior of the target. The structural features observed[9,12] in the angular distribution of the quasielastic-scattering intensity in experiments on electron reflection from crystal surfaces appear to be of a similar physical nature.

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