MODEL OF INELASTIC SCATTERING OF AN ATOM BY A CRYSTAL SURFACE IN THE IMPULSE APPROXIMATION

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§1. The aim of the work reported in the present communication was to obtain the differential cross section for single scattering of an atom by a simple crystal. The interaction in the system is determined in the form of a sum of two-body potentials characterizing the interaction of the particle with the atoms of the crystal lattice. The processes leading to scattering are assumed to be weakly dependent on the processes responsible for binding and thermal equalization within the crystal. The Hamiltonian of the crystal is thus described in the harmonic approximation [1] and the scattering, in the impulse approximation with a "quasiclassical binding" [2, 3]. We assume that the velocities of the particle are large compared with the thermal velocities of the atoms of the lattice and that interference effects are insignificant. Under these conditions the differential cross section for scattering in the range of angles $d\Omega$ and energies dE_2, referred to a single scattering center, has the form (h = 1) [2]

$$\frac{d^2\sigma}{d\Omega dE_2} = \frac{k_2}{k_1}(1+\mu)^2|F|^2\int_{-\infty}^{\infty} d\tau' e^{-iE\tau'}\langle e^{-i\mathbf{k}\mathbf{u}(0)}e^{i\mathbf{k}\mathbf{u}(\tau')}\rangle_T, \tag{1.1}$$

where $\mu = m/M$ is the ratio of the mass of the particle to the mass of the atom of the lattice; $\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2$, $E = E_1 - E_2$ are, respectively, the change in the momentum and energy of the particle in a single collision; $\mathbf{u}$ is the displacement of the center about the site position; $F = F(\varkappa', \varkappa)$, the single-particle scattering amplitude in a center-of-mass system (incident particle, scattering center), is taken outside the energy surface

$$\varkappa = \mathbf{k}_1/(1+\mu),\ \varkappa' = (\mathbf{k}_2 - \mu\mathbf{k}_1)/(1+\mu);$$

<...> denotes an average over the initial state of the crystal and an average over the canonical distribution for the given crystal temperature T. The conservation laws for the entire particle–crystal system are taken into account in the derivation of (1.1) [2]. Assuming that single collisions are due solely to collisions with topmost atoms, we separate out from the summation over all centers the summation over topmost atoms alone.

§2. In (1.1) the summation is over all final states. It is known [3], however, that in the impulse approximation the cross section for each scattering direction $\mathbf{k}_2/k_2$ has the form of sharp peaks only near states corresponding to recoil of the center as if it were free. Only transitions into such states will be taken into account; their excitation energies we define as the real roots of the equations $a_1(E) = 0$ or $a_1^2(E) = a_2(E)$, which are found in the following manner.

From (1.1) we write down the expression for the autocorrelator in the phonon representation [2]

$$\exp\left\{\sum_{j,\mathbf{q}}\frac{(\mathbf{k}e_j(q))^2}{2NM\omega_j(\mathbf{q})}\{\langle 2n_j(\mathbf{q})+1\rangle[\cos(\omega_j(\mathbf{q})\tau')-1]+i\sin(\omega_j(\mathbf{q})\tau')\}\right\}, \tag{2.1}$$

where $\mathbf{q}$, $\omega_j(\mathbf{q})$, $\mathbf{e}_j(\mathbf{q})$ are, respectively, the phonon wave vector, frequency, and polarization

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vector; j is the polarization index; and $\langle n_j(\mathbf{q})\rangle$ is the Planck distribution. Expanding the exponential in the integrand in (1.1) as series in τ', we obtain, using (2.1),

$$e^{-iE\tau'}\langle\ldots\rangle_T = \exp\left\{-ia_1\tau' - a_2\frac{\tau'^2}{2} + \ldots\right\}; \tag{2.2}$$

$$a_1 = E - \sum_{j,\mathbf{q}}\frac{(\mathbf{k}e_j(\mathbf{q}))^2}{2NM}, \quad a_2 = \frac{\langle(\mathbf{k}\mathbf{p}_\nu)^2\rangle}{M^2} = \sum_{j,\mathbf{q}}\frac{(\mathbf{k}e_j(\mathbf{q}))^2\,\omega_j(\mathbf{q})}{2NM}\langle 2n_j(\mathbf{q})+1\rangle, \tag{2.3}$$

where $\mathbf{p}_\nu$ is the specific momentum of the center before scattering. It is clear from the form of expansion (2.2) that the summation in a_1 has the significance of recoil energy, equal to $k^2/2M$ only for an isotropic crystal, and is consequently dependent on the dynamics of the surface. Energy conservation for three bodies (the particle, a fixed center, and an infinite mass) we assume to be expressed by the equation $a_1(E) = 0$. In the case of a mobile (in the site system) center, in the equation $E - k^2/2M = (\mathbf{k}\mathbf{p}_\nu)/M$ we replace the left side by a_1 and perform an ensemble average of the squares of both sides. The net result is $a_1^2(E) = a_2(E)$.

The above method of describing an inelastic process enables us to obviate a problem of the utmost complexity — the direct description of total multiphonon scattering.

§3. We now construct a model of the dispersion of the scattered energy allowing for certain basic properties of the reflection of phonons from two-dimensional defects [1]: for any vector $\mathbf{q}$ the energies $\hbar\omega_j(\mathbf{q})$ of excited phonons polarized along and perpendicular to the surface take on, respectively, values in the ranges from 0 to Θ_n and from 0 to $\varkappa\Theta_n$, where $\varkappa \ll 1$; Θ_n is the Debye temperature for the surface. The dispersion law is linear.

The condition $\varkappa \ll 1$ comes about because the reflecting power of two-dimensional defects is only weakly dependent on the phonon wavelength λ down to $\lambda > L$, where L is the linear dimension of the distortion [1]. For $\varkappa \ll 1$ ($\lambda \gg a$, where a is the lattice constant) normally polarized phonons are only weakly scattered by the defect and pass into the crystal. In this model conservation of periodicity along the surface leads to the removal and redistribution of a large part of $E = E_1 - E_2$ along the surface (with the dynamics of the harmonic approximation). We introduce the dimensionless quantities

$$\varepsilon = E/\Theta_n,\ \varepsilon_1 = E_1/\Theta_n,\ \varepsilon_2 = E_2/\Theta_n,\ t = T/\Theta_n,\ \beta = k_2/k_1,\ \tau = \tau'\Theta_n$$

and go in (2.1) and (2.3) from a summation over $\mathbf{q}$ to an integral. With the aid of the $\varkappa$-model we obtain the following expressions for the integral in (1.1):

$$S = \int_{-\infty}^{\infty} d\tau e^{-i\varepsilon\tau}\langle e^{-i\mathbf{k}\mathbf{u}(0)}e^{i\mathbf{k}\mathbf{u}(\tau)}\rangle_T = \int_{-\infty}^{\infty} d\tau \exp\{iA_1 + A_2\} \tag{3.1}$$

and for $a_1(\varkappa) = a_1/\Theta_n$, $a_2(\varkappa) = a_2/\Theta_n^2$:

$$A_1 = -\tau\varepsilon + \frac{3}{\tau^2}\{e_t(\sin\tau - \tau\cos\tau) + e_n[\sin(\varkappa\tau) - \varkappa\tau\cos(\varkappa\tau)]\}; \tag{3.2}$$

$$A_2 = -\frac{3}{2}(e_t + \varkappa^2 e_n) + \frac{3}{\tau^2}\{e_t(\cos\tau + \tau\sin\tau - 1) + e_n[\cos(\varkappa\tau) +$$

$$+ \varkappa\tau\sin(\varkappa\tau) - 1]\} - 12\left(e_t\int_0^1 + e_n\int_0^\varkappa\right)\frac{dx\,x\sin^2(\tau x/2)}{\exp\{x/t\} - 1}; \tag{3.3}$$

$$a_1(\varkappa) = \varepsilon - e_t - \varkappa^3 e_n,$$

$$a_2(\varkappa) = \frac{3}{4}(e_t + \varkappa^4 e_n) + 6\left(e_t\int_0^1 + e_n\int_0^\varkappa\right)\frac{x^3 dx}{\exp(x/t) - 1}; \tag{3.4}$$

$$e_t = \varepsilon_1\mu(\sin^2\theta_1 + \beta^2\sin^2\theta_2 - 2\beta\sin\theta_1\sin\theta_2\cos(\varphi_1 - \varphi_2)];$$

$$e_n = \varepsilon_1\mu(\cos\theta_1 + \beta\cos\theta_2)^2, \quad \varepsilon = \varepsilon_1(1 - \beta^2),$$

where $e_n + e_t = e_0$ is the reduced recoil energy of the free center; e_t and e_n are the parts of e_0 corresponding to the tangential and normal changes of momentum of the particle; θ_1', φ_1 and θ_2, φ_2 are the directions of $\mathbf{k}_1$ and $\mathbf{k}_2$ in a spherical system of coordinates whose z axis coincides with the external normal to the topmost plane of sites (z = 0); and $\theta_1 = \pi - \theta_1'$ is the angle of incidence.

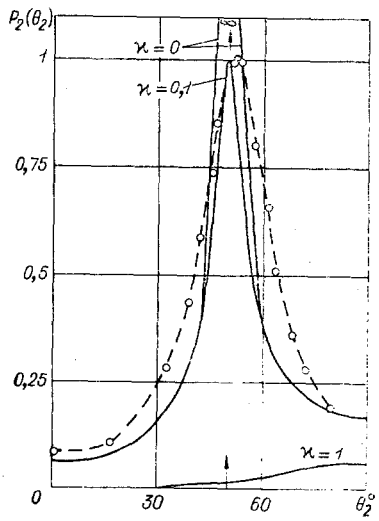

Fig. 1

The integrals in (3.3) and (3.4) vanish for t = 0, while for t > 1 we have that

$$a_2(\varkappa) = 2t(e_t + \varkappa^3 e_n) + 0((20t^2)^{-1}),$$
$$A_2 \approx -6t\left[e_t\left(1 - \frac{\sin\tau}{\tau}\right) + \varkappa e_n\left(1 - \frac{\sin(\varkappa\tau)}{\varkappa\tau}\right)\right]. \tag{3.5}$$

Utilizing expansion (2.2), we obtain the following approximate expressions for S:

$$S = \begin{cases} \sqrt{2\pi/a_2(\varkappa)} & \text{for } a_1(\varkappa) = 0, \\ \sqrt{2\pi/2.718a_1^2(\varkappa)} & \text{for } a_1^2(\varkappa) = a_2(\varkappa). \end{cases} \tag{3.6}$$

The transition from the case with a mobile center to the case with a fixed center occurs when $e_t + \varkappa^3 e_n \gg t$. The expressions for S diverge only for $\varkappa = 0$; $\theta_2 = \theta_1$, $\pi - \theta_1$; $\varphi_1 = \varphi_2$ (see Fig. 1).

In final form the results of the present work can be expressed as follows:

$$\frac{d\sigma}{d\Omega_2} = (1+\mu)^2 \sum_{\beta_i > 0} \beta_i |F(\varkappa'(\beta_i); \varkappa)|^2 S(\varepsilon(\beta_i); e_t(\beta_i); e_n(\beta_i); \varkappa), \tag{3.7}$$

where $d\Omega_2 = \sin\theta_2 d\theta_2 d\varphi_2$; S is given by expressions (3.1)-(3.3), (3.5), (3.6); and β_i are the roots of the equation

$$[\varepsilon(\beta) - e_t(\beta) - \varkappa^3 e_n(\beta)]^2 = \frac{3}{4}[e_t(\beta) + \varkappa^3 e_n(\beta)] + 6\left(e_t\int_0^1 + e_n\int_0^\varkappa\right)\frac{x^3 dx}{e^{xt} - 1}. \tag{3.8}$$

In this manner, the scattering of an atom by a crystal is reduced to a single-particle scattering problem. After normalizing in accordance with the optical theorem, the expression for the density of scattered particles $P_2(\Omega_2) \sim \Sigma|F|^2 S$ can be used to obtain the various characteristics of single scattering.

We illustrate results (3.7) and (3.8) on the example of scattering by rigid spheres. In this case $|F|^2$ = const and the angular dependence $P_2(\Omega_2)$ is determined by the form factor. In Fig. 1 we compare the curve $P_2(\theta_2, \varkappa = 0.1)$ with the experimental (unnormalized) diagram [4] for the scattering of argon from silver, E_1 = 2.56 eV, T = 300°C, $\theta_1 = 50°$, $\varphi_1 = \varphi_2 = 0$. For the silver surface Θ_n = 180°K. The different slopes of the curves near the surface are connected with scattering by a relief of microcrystals.

For $\varkappa = 0$ the energy E is redistributed only along the surface. For $\varkappa = 1$ the form of $P_2(\theta_2)$ corresponds to a freezing-in of the particles [the form of $P_2(\theta_2, T = 0, \varkappa = 1)$ is similar to that of $P_2(\theta_2, T \neq 0, \varkappa = 1)$]. Since $\varkappa = \varkappa(T)$ and since a certain equalization of the bindings in the topmost layer occurs as $T \to 0$, it follows that $\varkappa$ increases.

The value of κ is determined from experimental plots with saturation, for example, from $\Delta\theta_{2m}(E_1) = \theta_1 - \theta_{2m}(E_1)$ for $E_1 > 10$ eV [5], where θ_{2m} is the direction of the peak. For rigid spheres we obtain

$$\varkappa^3 = [(\sin\theta_{2m} - \sin\theta_1)\cos\theta_{2m}]/[\cos\theta_{2m} + \cos\theta_1)\sin\theta_{2m}].$$

For $\varkappa \ll 1$ and any E_1 the quantity $\theta_{2m} \approx \theta_1$ only for rigid spheres; on going over to softer potentials $\Delta\theta_{2m}$ becomes a function of E_1. With increasing E_1 we have the narrowing of $P_2(\theta_2)$ observed in [5].

We note that for $E_1 < 0.1$ eV the problem has been investigated in a quantum formalism for elastic and single-phonon scattering by a one-dimensional potential wall [6]. Quantum model representations for energies $E_1 > 0.1$ eV are few [7]. Classical models are more widely used for these energies. It is considered in these models that for $k_1 a \gg 1$ only multiply scattered particles reproduce the peak reflection by the wall [8].

However, particles experiencing above-barrier scattering in the crystal and multiple mixing in it give rise to a weak dependence of the peak on θ_1 [9]. The formation of such a peak is observed only for E_1 in the range 100-400 eV [9]. Accordingly, for $E < 100$ eV we most probably have a solution of the gas in the crystal, with an accompanying diffusion of the particles to the surface and diffuse desorption of them with $\langle E_2 \rangle \ll E_1$. Scattering by the relief is also diffuse in nature.

It has been shown in a number of experiments over the last 10 years that the reflection of atoms of the noble gases with E_1 up to 100 eV by controlled surfaces of simple single crystals has a single-peak quasispecular character [4, 5, 9]. In the present work the observed form of scattering is obtained for single collisions, and we suggest that quasispecular scattering is connected with the specifics of the recoil of surface centers. The obtained results indicate that the noise [the contribution to $P_2(\Omega_2)$ from multiple, physically unobservable collisions] can be separated from the contribution due to single-reflected particles, which carry information on the dynamics and structure of the surface and on the interaction of the atom with the crystal.

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