Bulk resonant scattering of fast electrons in crystals

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A new type of dynamic scattering of fast electrons has been observed in a crystal. This scattering is accompanied by a resonant localization of a particle in a narrow region of the spectrum of transverse-motion states. The appearance of a resonance explains the nature of the annular and parabolic diffraction patterns and resolves the problem of interpreting images in high-resolution electron microscopy.

1. Emslie reported the observation of annular and parabolic diffraction patterns in early 1934.[1] Emslie suggested that the nature of the patterns was related to an inelastic trapping of electrons in states localized near axes or atomic planes of the crystal. Recent experiments by Peng *et al.*[2–4] and some subsequent studies[5,6] have shown that the rings and parabolas of elevated intensity which arise in various scattering configurations result from the "one-dimensional" diffraction of electrons which are localized in the potential of an individual atomic plane or axis. So far, there has been no theoretical description of this effect. Below we find a purely elastic local mechanism which explains the nature of the annular and parabolic diffraction patterns.[1–6]

2. When the angle ϑ made by the momentum vector of the electron, $\vec{p}$, with the crystal axis z is close to $\vartheta_r = \arccos(1 - 2\pi/pa_\parallel)$, where $a_\parallel$ is the period of the atomic chains along the z axis, an elastic interaction with a momentum transfer along the axis plays an important role.[3,4] Restricting the Fourier expansion of the periodic potential to reciprocal-lattice vectors with zero and minimal nonzero projections onto the axis, $U(\vec{r}) = U_0(\vec{\rho}) + U_1(\vec{\rho})\exp(igz) + U_{-1}(\vec{\rho})\exp(-igz)$, where $g = 2\pi/a_\parallel$, we find the wave function of our scattering problem to be

$$\Psi_{\vec{p}}^{(+)}(\vec{r}) = \alpha(z)\exp(i\vec{p}\vec{r}) + \exp(i(p\cos\vartheta + g)z)\sum_j \beta_j(z) b_j(\vec{p}_\perp + \vec{\Delta}, \vec{\rho}), \tag{1}$$

The summation over j is over the zones of the Bloch state spectrum of the transverse motion of the electron in the potential $U_0(\vec{\rho})$, with a dispersion law $E_j(\vec{p}_\perp)$; α and β_j are smooth functions of z, and $\vec{\Delta}$ is the (vector) transverse displacement of the plane of reciprocal-lattice vectors of the minus first Laue zone with respect to the zeroth Laue zone. The amplitudes $\beta_j(z)$ depend on the relations among the parameter expressing the excursion from the mass shell,

$$\delta_j(\vec{p}) = E_j(\vec{p}_\perp + \vec{\Delta}) + \frac{(p\cos\vartheta + g)^2}{2m} - \frac{\vec{p}^2}{2m}, \tag{2}$$

the matrix element of the coupling of channels, $U_1(j) = \int d^2\rho e^{-i\vec{p}_1\vec{\rho}} U_1(\vec{\rho}) b_j(\vec{p}_\perp + \vec{\Delta}, \vec{\rho})$, and the inelastic electron absorption coefficient $\mu = nv\sigma_{\text{inel}}$. Under normal conditions, the inequality $|U_1(j)| > \mu$ is satisfied only for states for the lower zone of the Bloch state spectrum, with $j = 0$. For $j > 0$ we then have $|\beta_j(z)| \lesssim |U_1(j)|/\mu \ll 1$, and a "two-level" resonant behavior of the wave function arises near ϑ_r with

$$|\beta_0(z)|^2 = 1 - |\alpha(z)|^2 = \frac{1}{1+Y^2}\sin^2\left(\frac{m|U_1(0)|}{p} z\sqrt{1+Y^2}\right), \tag{3}$$

where $Y = \delta_0(\vec{p})/2U_1(0)$. At $\vartheta = \vartheta_0 = \arccos\{1 - (2\pi/pa_\parallel) - [mE_0(\vec{p}_\perp + \vec{\Delta})/p^2]\}$, at a depth $z = \pi v/(2|U_1(0)|)$, the electron localizes in the narrow lower zone of the Bloch state spectrum. This localization is accompanied by an acceleration of the electron along the direction of the crystal axis and a decrease in the energy of the motion in the plane $\vec{\rho} = (x,y)$. A behavior similar to that in (1)–(3) is exhibited by the wave function of the problem of resonant surface scattering.[7] Working from this analogy, we might call the motion in (1), (3) a "bulk resonance." The angular width of the resonant region is determined by the condition $|Y| < 1$, i.e., by $|\vartheta - \vartheta_0| < m|U_1(0)/g^{1/2}p^{3/2}$. It is a strong function of the temperature:

$$|U_1(0)| \sim \exp(-pg < u^2 >), \tag{4}$$

where $\sqrt{\langle u^2 \rangle}$ is the linear thermal displacement. A comparison of (4) with the condition for an inelastic disruption of the resonance, $\mu > |U_1(0)|$, shows that the zero-point vibrations of the atomic nuclei limit the region in which the bulk resonance exists to the range of nonrelativistic particle energies, $E_p = p^2/2m < [mg^2(\langle u^2 \rangle)^2]^{-1} \sim 10^4$ eV. The annular and parabolic diffraction patterns have been observed in this range of electron energies.

3. The nature of the annular and parabolic diffraction patterns observed experimentally[1-6] is related to the appearance of a bulk resonance in the final state $\Psi^{(-)}_{\vec{p}'}(\vec{r})$ of the inelastic scattering of an electron accompanied by the excitation of internal degrees of freedom of the crystal (phonons, electron–hole pairs, and so forth). The final-state wave function $\Psi^{(-)}_{\vec{p}'}(\vec{r})$ is related to (1) by the reciprocity relation $(\Psi^{(-)}_{\vec{p}'}(\vec{r}) = (\Psi^{(+)}_{\vec{p}'}(\vec{r}))^*$. For collisions accompanied by the ionization of atoms in cubic crystals (fcc or bcc), the scattering cross section integrated over energy is

$$\frac{d\sigma}{do'} \sim 1 - b_0(-\vec{p}'_\perp + \vec{\Delta}, 0)\frac{Y'}{1+Y'^2} + \{|b_0(-\vec{p}'_\perp + \vec{\Delta}, 0)|^2 - 1\}\frac{1}{2(1+Y'^2)}, \tag{5}$$

where $Y' = \delta_0(-\vec{p}')/[2U_1(0)]$. The parameter Y' for states in a narrow zone $E_0(-\vec{p}'_\perp + \vec{\Delta}) \approx \text{const}$ is constant in the direction cone $\vartheta' = \text{const}$, and distribution (5) describes an intensity ring with a maximum near $\vartheta'_0 = \arccos\{1 - (2\pi/p'a\|) - [mE_0(-\vec{p}'_\perp + \vec{\Delta})/p'^2]\}$ and with the profile shown in Fig. 1. The value of ϑ_0 and the intensity profile in (5) agree well with the experimental data of Refs. 1–6. The very fact that regular rings of elevated intensity are observed near close-packing directions[1-6] directly proves the existence of a narrow zone in the Bloch state spectrum of the transverse motion of an electron in the 2D potential of the chains of atoms in a crystal, $U_0(\vec{\rho})$. If there are several narrow zones in the Bloch spectrum $E_j(\vec{p}_\perp)$, several concentric rings differing in intensity will arise on the electron diffraction pattern.[1,3,4]

Arguments similar to those above show that in a certain diffraction configuration—in which the Ewald sphere passing through the origin of coordinates in $\vec{p}$ space is tangent to some system of reciprocal-lattice sites which lie on a common straight line and which belong to the plane of the minus first Laue zone—an electron can localize in the lower narrow zone of the Bloch state spectrum of the motion of the particle in the potential of the atomic planes. The "planar" bulk resonances are mani-

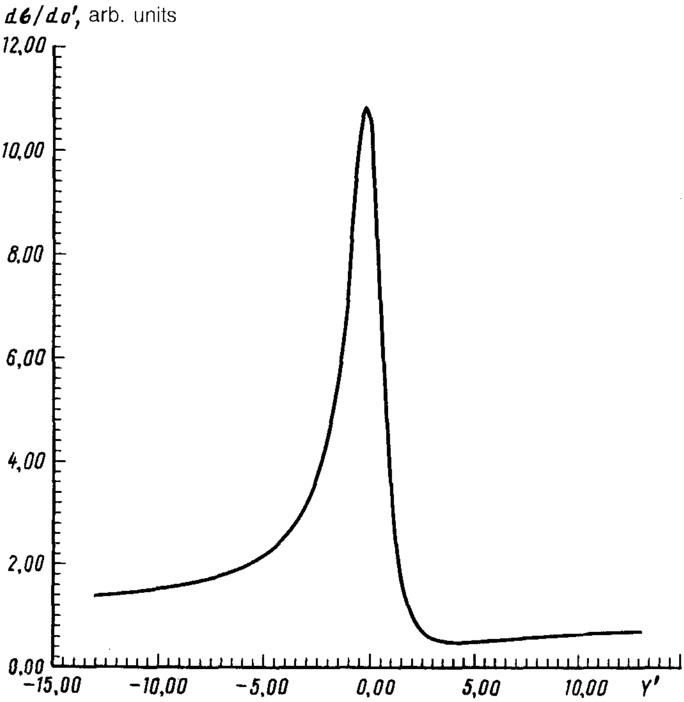

FIG. 1. Intensity profile of an annular diffraction pattern in the "radial" direction. The value $\vartheta' = 0$ corresponds to the $\langle 100 \rangle$ direction in an Mo crystal; $|b_0(-p'_\perp + \Delta, 0)| = 4.43$; the electron energy is 100 keV; the spectrum $E_j(\vec{p}_\perp)$ was calculated on the basis of 29 diffracted waves.

fested in the distribution of inelastically scattered electrons as a system of parabolas with foci toward the center of the diffraction pattern.[2-5] The matrix elements $U_1(0)$ for the planar bulk resonances are smaller than the corresponding quantities for an axial bulk resonance, so the contrast of the parabolas is always weaker than that of the rings. The rapid inelastic disruption of the planar bulk resonances with decreasing electron energy ($\mu \sim E_p^{-1/2}$) explains why there are no parabolic patterns in the photographs in Fig. 6.

4. The possibility of a selective localization of an electron in a low-lying narrow zone of the Bloch state spectrum in bulk-resonance regime (3) may simplify the problem of interpreting images in high-resolution electron microscopy.[8] Under the typical conditions (with $\vec{p}$ parallel to z), the image is formed by several zones of the Bloch state spectrum, and it oscillates rapidly with the thickness of the crystal. In bulk-resonance regime (3), all the nonuniformity of the wave field in the transverse direction, $\vec{\rho}$, stems from the single low-lying state of the Bloch state spectrum. The effect is to stabilize the image and to improve the resolution to the scales of the localization of the function $b_0(\vec{p}_\perp + \vec{\Delta}, \vec{\rho})$ in the potential of an individual atomic chain, i.e., to $\lesssim 0.1$ Å.

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