

POLES OF STRUCTURAL FACTOR AND SECONDARY EMISSION FROM A SEMI-INFINITE CRYSTAL

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Secondary emission from a semi-infinite crystal are considered. Contribution to the emission amplitude originating from structural factor singularities are analyzed. Poles of structural factor determine the main contribution to the emission amplitude. The energy and angular distributions of the emission depends on the crystallographic orientation of the crystal surface and is characterized by a set of permitted vectors of reciprocal lattice. It is shown, that emission anisotropy related to the crystallographic directions is due to the coherent character of photoabsorption process and diffraction scattering leads only to relative change of the emission intensity in permitted low-index directions. The concrete calculations are performed for (100) and (111) surfaces of the diamond like lattice.

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Photo-, Auger- and fluorescence emissions from X-ray irradiated crystals are intensively used for structure reconstruction on a crystal surface. Development of experimental facilities in this field allows to obtain information concerning surface structure of irradiated crystals [1–5]. Application of these emissions for crystal holography is also widely discussed [6–11]. Characteristic features of angular distribution of the emission investigated in the papers cited above are interpreted as a result of diffraction scattering. A few authors try to analyze interference between direct and scattered emissions from individual atoms (see [12] and ref. there). Mathematical methods of structure reconstruction are also developed. Parallel with new holographic schemes some refined methods such as a Patterson-like scheme are proposed [13]. Discussed schemes of structure reconstruction based on measuring of angular and energy distribution of the emission take into account scattering by individual atoms as by independent scatterers and do not take into account periodic nature of scattering potential. Correct determination of positions of displacement atoms near a crystal surface make it desirable to have for comparison emission scattered by periodic potential of a semi-infinite perfect crystal. In this paper analysis of anisotropy of the secondary emission from a single semi-infinite crystal under X-ray irradiation is proposed.

During emission registration experiments it is not possible to distinguish an explicit atom-emitter or an atom-scatterer. Therefore in the case of identical atoms it is convenient to consider an amplitude of the emission probability, which depends only on the emission wave vector \mathbf{k} , which determines energy and direction of the measured emission. It is known, that the scattering potential acting on the electrons in a crystal is a sum of all atomic potentials. A Fourier transform of this potential $U(\mathbf{q})$, which determines the scattering with transmitted momentum \mathbf{q} , is given by

$$U(\mathbf{q}) = V(\mathbf{q})S(\mathbf{q}). \quad (1)$$

Here $V(\mathbf{q})$ is the atomic factor, $S(\mathbf{q}) = \sum_n \exp(i\mathbf{q}\mathbf{r}_n)$ is the structural factor (SF) of a semi-infinite crystal, \mathbf{r}_n is an atomic position and the summation is taken over all

atomic positions with $z_n \leq 0$, where the z -axis of a Cartesian coordination system is directed along the external normal to the irradiated surface. The eq.(1) does not take into account thermal atomic motion, which leads to the Debye - Waller factor in this equation. The finite summation limit over $z_n \leq 0$ in SF determines influence of crystallographic orientation of the surface. The explicit form of the \mathbf{q} -dependence of SF can be obtained layer by layer summation for investigated lattice and given surface orientation. It is known that $V(\mathbf{q})$ does not have singularities and change of the wave vector \mathbf{k}' during the scattering is determined by the poles \mathbf{q}_{jlm} of $S(\mathbf{q})$, which are a subset of the total set of vectors of reciprocal lattice. This set depends strongly on the crystallographic orientation of crystal surface. Below on the example of the diamond like lattice we demonstrate dependence of the set on surface crystallography.

The concrete calculations, fulfilled for the important case of (100)-surface of diamond like lattice, allow to obtain the explicit form of SF. It has the following form

$$S_{100}(\mathbf{q}) = \left(4 \cos \frac{q_x a}{4} \cos \frac{q_y a}{4} \cos \frac{q_z a}{4} \right)^{-1}. \quad (2)$$

Here a is the size of a primary cell of the diamond like lattice. This equality shows the presence of poles at

$$\mathbf{q}_{jlm} = (2j + 1, 2l + 1, 2m + 1)2\pi/a, \quad (3)$$

where j, l, m are integers. In the case of a weakly scattering potential, $U_s \ll E$, where E is the electron energy, the pole singularities of SF (2) allow to calculate the emission amplitude accounting elastic scattering on the periodic potential in the Born approximation (BA) [14]. Let $F(\mathbf{k})$ be the probability amplitude of unscattered emission. Then account of scattering leads to the following form of the emission amplitude

$$G(\mathbf{k}) = F(\mathbf{k}) + W(\mathbf{k})/E, \quad (4)$$

where

$$W(\mathbf{k}) = \sum_{jlm} b_{jlm}^{(1)} F(\mathbf{k} - \mathbf{q}_{jlm}) + \sum_{jlm} b_{jlm}^{(2)} F(\mathbf{k} - 2\mathbf{q}_{jlm}) + \dots$$

has meaning of the effective scattering potential of the semi-infinite crystal, which takes into account the momentum transfer multiple to all permitted \mathbf{q}_{jlm} . Calculation of the first two coefficients in the BA gives

$$b_{jlm}^{(1)} = \frac{32\sqrt{\pi}}{ea^3} (-1)^{j+l+m} V(\mathbf{q}_{jlm}), \quad (5)$$

$$b_{jlm}^{(2)} = i \frac{512\sqrt{\pi}}{3ea^6} \frac{V^2(\mathbf{q}_{jlm})}{E_{jlm}}.$$

Here

$$E_{j,l,m} = \frac{\hbar^2 \mathbf{q}_{jlm}^2}{2m_e} = 2E_s^e [(2j + 1)^2 + (2l + 1)^2 + (2m + 1)^2], \quad (6)$$

$E_s^e = \text{Ry} (\pi a_B/a)^2$, Ry is the Ridberg's energy and a_B is the Bohr radius, and m_e is the electron mass. The second term in the right hand side of eq.(4) describe multiple scattering as a series decomposition with respect to the scattering potential. The summing in $W(\mathbf{k})$ is fulfilled over all poles of SF, which satisfy the following conditions $(\mathbf{k} - \mathbf{q}_{jlm})^2 = \mathbf{k}^2$ and $(\mathbf{k} - 2\mathbf{q}_{jlm})^2 = \mathbf{k}^2$ in the first and second orders of BA, respectively. In particular it

means that the emission amplitude with the wave vectors $\mathbf{k} = \mathbf{q}_{jlm}$ are preserved during scattering in the first order of BA. Eqs.(4) and (5) show that in the case of isotropic form of the amplitude $F(\mathbf{k})$ elastic scattering preserves the isotropy in the emission intensity $|G(\mathbf{k})|^2$. This leads to the conclusion, that anisotropy, appearing in experimental data and related to the crystal structure [15], is formed in the process of photoabsorption and not during the diffraction scattering. There are two possible causes of this anisotropy. The first one is coherent character of the photoabsorption related to the crystalline atomic structure. The second cause of the anisotropy of $F(\mathbf{k})$ is due to anisotropy of photoprocess on an individual atom and can be due to polarization of the X-ray beam and to initial electron state in an individual atom. This second cause in no way is related to the crystalline structure. Thus the experimental data of Ref. [15] can be considered as an indication on the coherent character of the photoabsorption process. It is possible to show that in the case of coherent photoabsorption, when $F(\mathbf{k}) \sim S(\mathbf{k})$ and all Fourier transforms of the amplitude $F(\mathbf{k})$ correspond to the poles of SF, the first order of BA does not contribute to the elastic scattering and the second order of BA leads only to relative change of emission intensity in permitted low-index directions.

The positions of the poles of SF in the \mathbf{k} -space are determined by the crystal symmetry and surface orientation. Let $[2j + 1, 2l + 1, 2m + 1]$ designates a multiplet of reciprocal vectors corresponding to equal energy (6). In a general case it corresponds to 48 different poles in the \mathbf{k} -space. These poles form a 48-plet with 6 different wave vectors in each octant of the \mathbf{k} -space. In the case when, for example, $q_\alpha = q_\beta \neq q_\gamma$ the poles form a 24-plet with 3 different wave vectors in each octant of the \mathbf{k} -space. The spatial diagonals of the octants are 3-fold axes for this wave vector system. In the case when $q_\alpha = q_\beta = q_\gamma$ the poles form an octet with wave vectors located along the spatial diagonals. The lowest energy octet of such states has energy $E_{[1,1,1]} = 6E_s^e$ (~ 18 eV in the case of Si). The next energy state is a 24-plet and has the energy $E_{[1,1,3]} = 22E_s^e$ (~ 66 eV in the case of Si). The lowest energy 48-plet has the energy $E_{[1,3,5]} = 70E_s^e$ (~ 210 eV in the case of Si).

In the case of diamond like lattice a normal to a (111)-surface is the symmetry axes of the third order. In this case layer by layer calculation of SF leads to the following expression

$$S_{111}(\mathbf{q}) \sim -i \exp\left(\frac{iq a_0}{2}\right) \left[4 \sin \frac{q a_0}{6} \cos \frac{q a_0}{3}\right]^{-1} \delta\left(\frac{\mathbf{q}}{q} - \mathbf{n}_z\right). \quad (7)$$

Here $a_0 = \sqrt{3}a/4$ is the minimal distance between atoms in the diamond like lattice with the size of primary cell equals a . Note that the summation over any monolayer, which is parallel to a (111)-surface, gives the same result equals 0 for all direction except the normal one. In this case the diffraction scattering changes only z -component of the wave vector.

The presence of this single direction of \mathbf{q} is the result of interference. The poles at

$$\mathbf{q}_j = (0, 0, j) \frac{6\pi}{a_0}, \quad \mathbf{q}_l = (0, 0, 2l + 1) \frac{3\pi}{2a_0} \quad (8)$$

lead to sharp increase of the scattering along the normal direction to the (111)-surface for these wave length. The electrons with energies

$$E_j = 96j^2 E_s^e, \quad E_l = 6(2l + 1)^2 E_s^e, \quad (9)$$

give the main input in the emission. In this case all poles form doublets. Characteristic distance between two lowest levels for the (111)-surface of Si is ~ 36 eV.

In the case of fluorescence emission from the (100)-surface the main input can be expected at energies equal

$$E_{j,l,m}^f = E_s^f [(2j+1)^2 + (2l+1)^2 + (2m+1)^2]^{1/2}, \quad (10)$$

and in the case of the (111)-surface

$$E_j^f = 4\sqrt{3}jE_s^f, \quad E_l^f = \sqrt{3}(2l+1)E_s^f. \quad (11)$$

Here $E_s^f = 2\pi\alpha R_y (a_B/a)$ determines the energy scale for the fluorescence emission, $\alpha = c\hbar/e_0^2 = 137$. Characteristic value of E_s^f equals ~ 2300 eV, while $E_s^e \sim 3$ eV.

Thus we see that the influence of the diffraction scattering on the emission anisotropy depends on the crystallographic orientation of the crystal surface. Eqs.(4), (5) allow to estimate the contribution of the diffraction scattering to the electron emission. Contributions of inelastic processes, and in particularly plasmon excitations, can be easily estimated by experimental data from Ref.[15]. Similar angle dependences of the electron emission measured in [15] for elastic and satellite plasmon peaks indicate that they are related to the same poles of SF. Relatively small energy losses of the electrons due to the excitation of plasma oscillations are considerably smaller than the energy intervals between neighboring multiplets (6) and angular distributions of electrons, both excited and not excited plasmons, are formed by the same multiplet of reciprocal vectors.

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1. K.Robinson and D.J.Tweet, Rep. Prog. Phys. **55**, 599 (1992).
 2. C.S.Fadley, Physica Scripta **T17**, 39 (1987).
 3. M.-L.Xu and M.A.Van Hove, Surface Science **207**, 215 (1989).
 4. I.H.Hong, S.C.Shyu, Y.C.Chou, and C.M.Wei, Phys. Rev. **B52**, 16884 (1995).
 5. P.M.Len, J.D.Denlinger, E.Rotenberg et al., Phys. Rev. **B59**, 5857 (1999).
 6. P.M.Len, S.Thevuthasan, C.S.Fadley et al., Phys. Rev. **B50**, 11275 (1994).
 7. Hua Li, S.Y.Tong, D.Naumovic et al., Phys. Rev. **B47**, 10036 (1993).
 8. S.Thevuthasan, R.X.Ynzunza, E.D.Tober et al., Phys. Rev. Lett. **70**, 595 (1993).
 9. D.K.Saldin, K.Reuter, P.L.De Andres et al., Phys. Rev. **B54**, 8172 (1996).
 10. T.Gog, P.M.Len, G.Materlik et al., Phys. Rev. Lett. **76**, 3132 (1996).
 11. P.M.Len, T.Gog, C.S.Fadley, and G.Materlik, Phys. Rev. **B55**, R3323 (1997).
 12. H.Wu, G.J.Lapeyer, H.Huang, and S.Y. Tong, Phys. Rev. Lett. **71**, 251 (1993).
 13. C.Y.Chang, Z.C.Lin, Y.C.Chou, and C.M.Wei, Phys. Rev. Lett. **73**, 2580 (1999).
 14. L.D.Landau and E.M.Lifshitz, *Quantum mechanics*, Nauka, Moscow, 1974.
 15. E.Puppini, C.Carbhone, and R.Rochow, Phys. Rev. **B46**, 13215 (1992).