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Modification of the EDXD Method for Diagnostics of Polycrystalline and Fine-Grained Media

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Abstract—The possibilities of using broadband X rays for diagnostics of the atomic structure of solids are considered. It is proposed to use virtual photons of the Coulomb field of a relativistic electron beam as primary probe radiation.

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1. The conventional approach to the diagnostics of the atomic structure of matter is based on measurement of the angular distribution of quasi-monochromatic X rays scattered by an object studied. Advances in development of X-ray detectors made it possible to propose a new approach, which is based on measurement of the spectral distribution of broadband X rays scattered at a fixed angle [1]. This approach, known as the energy dispersive X-ray diffraction (EDXD) method, can be modified by replacing the primary flux of free X-ray photons with virtual photons of the Coulomb field of relativistic electrons transmitted through a sample under study. The modified EDXD method has the following advantages: broadband spectrum of virtual photons, possibility of studying small portions using focused electron beams, high sensitivity to interatomic correlations, and possibility of using an X-ray detector in a fixed position.

It is noteworthy that the radiation that is due to the scattering of the Coulomb field of a fast electron during its collision with an atom is well known as polarization bremsstrahlung [2–4]. Polarization bremsstrahlung has been investigated in detail for the collision of fast particle with an isolated atom [4]. At the same time, study of the collective contribution of the atoms of a medium to the formation of the polarization bremsstrahlung yield has only begun [5]. The purpose of this study is to theoretically and experimentally analyze the possibility of using the coherent component of the polarization bremsstrahlung of relativistic electrons in diagnostics of the structure of polycrystalline and fine-grained media.

2. The Maxwell equations yield the following relation for the Fourier components of an electromagnetic field:

$$(k^2 - \omega^2)\mathbf{E}_{\omega\mathbf{k}} - \mathbf{k}(\mathbf{k}\cdot\mathbf{E}_{\omega k}) = 4\pi i\omega\mathbf{J}_{e\omega\mathbf{k}} + \frac{i\omega e}{2\pi^2}\mathbf{V}\delta(\omega - \mathbf{kV}). \quad (1)$$

Here, $\mathbf{V}$ is the velocity of a fast electron and $\mathbf{J}_{e\omega\mathbf{k}}$ is the Fourier transform of the induced current density of the electrons of the medium, defined as the mean of the conventional quantum-mechanical operator of the current density. Generally, the expression for $\mathbf{J}_{e\omega\mathbf{k}}$ has the form

$$J_{e\omega\mathbf{k},l} = \sum_p \int d^3k' G_{lp}(\mathbf{k},\mathbf{k}')E_{\omega\mathbf{k}'p}, \quad (2)$$

where the tensor G_{lp} is generally cumbersome [6]. In the case under consideration (motion of a fast electron through a polycrystal), the main mechanism of Coulomb field scattering is the Bragg scattering from the lattice, which generally occurs in the frequency range $\omega \approx 10$ keV. This value of ω significantly exceeds the electron binding energy in light atoms; therefore, the atomic electrons can be considered as free during the generation of radiation in the atom. Under these conditions, the tensor G_{lp} takes a simple form [7]:

$$4\pi i\omega G_{lp}(\mathbf{k},\mathbf{k}') = -\delta_{lp}\frac{e^2}{2\pi^2 m}\sum_j e^{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_j}F(\mathbf{k}'-\mathbf{k}) \equiv -\delta_{lp}G_{lp}(\mathbf{k}'-\mathbf{k}). \quad (3)$$

Here, $F(\mathbf{k}' - \mathbf{k})$ is the atomic form factor and $\mathbf{r}_j$ is the coordinate of the jth atom.

Separating $G(\mathbf{k}' - \mathbf{k})$ into the mean and random components, $G = \overline{G} + \tilde{G}$, $\overline{G} = \langle G(\mathbf{k}' - \mathbf{k}) \rangle_{r_j} = \omega_p^2 \delta(\mathbf{k}' - \mathbf{k})$, where ω_p is the plasma frequency of the medium, we obtain the equation

$$(k^2 - \omega^2 \varepsilon(\omega))\mathbf{E}_{\omega\mathbf{k}} + \int d^3k' \tilde{G}(\mathbf{k}' - \mathbf{k}) \left(\mathbf{E}_{\omega\mathbf{k}'} - \mathbf{k} \frac{\mathbf{k}\mathbf{E}_{\omega\mathbf{k}'}}{\omega^2 \varepsilon(\omega)} \right) = \frac{i\omega e}{2\pi^2} \left(\mathbf{V} - \frac{\mathbf{k}}{\omega\varepsilon(\omega)} \right) \delta(\omega - \mathbf{k}\mathbf{V}), \quad (4)$$

which is convenient to solve by iterations (this approach is admissible in view of the small size of the microcrystallites forming a polycrystal). Here, $\varepsilon(\omega) = 1 - \omega_p^2/\omega^2$ is the conventional permittivity in the X-ray region. In the zero-order approximation with respect to $\tilde{G}$, Eq. (4) yields an expression describing the Coulomb field, which is screened due to the polarization of the electrons of the medium. In the first-order approximation with respect to $\tilde{G}$, we obtain the desired radiation field (generated as a result of scattering of Coulomb virtual photons):

$$\mathbf{E}^S_{\omega k} = -\frac{i\omega e}{2\pi^2} \frac{1}{k^2 - \omega^2\varepsilon} \int \frac{d^3k'}{k'^2 - \omega^2\varepsilon} \tilde{G}(\mathbf{k}' - \mathbf{k}) \left(\mathbf{V} - \frac{\mathbf{k}'}{\omega\varepsilon} - \mathbf{k} \frac{\mathbf{k}\mathbf{V} - \mathbf{k}\mathbf{k}'/\omega\varepsilon}{\omega^2\varepsilon} \right) \delta(\omega - \mathbf{k}''\mathbf{V}). \quad (5)$$

The radiation amplitude $\mathbf{A_n}$ and the spectral angular distribution of the radiation intensity are derived from (5) by the standard methods

$$\omega \frac{dN}{dtd\omega d\Omega} = \langle |\mathbf{A_n}|^2 \rangle,$$
$$\mathbf{A_n} = -i\omega e \int \frac{d^3k}{k^2 - \omega^2\varepsilon(\omega)} \tilde{G}(\mathbf{k} - \omega\sqrt{\varepsilon}\mathbf{n}) \left(\mathbf{V} - \frac{\mathbf{k}}{\omega\varepsilon} - \mathbf{n}\left(\mathbf{n}\mathbf{V} - \frac{\mathbf{n}\mathbf{k}}{\omega\varepsilon} \right) \right) \delta(\omega - \mathbf{k}\mathbf{V}), \quad (6)$$

where $\mathbf{n}$ is the unit vector in the radiation direction and the angular brackets $\langle\ \rangle$ mean averaging over the coordinates of target atoms.

3. As a result of averaging of the squared modulus of the amplitude over the atomic coordinates in a microcrystallite and over the random orientations of microcrystallites, we obtain from (6) the final formula for the coherent component of the radiation intensity,

$$\omega \frac{dN}{dtd\omega d\Omega} = \sum_{\mathbf{g}} \omega \frac{dN_{\mathbf{g}}}{dtd\omega d\Omega},$$
$$\omega \frac{dN_{\mathbf{g}}}{dtd\omega d\Omega} = A_{\mathbf{g}} \Phi_{\mathbf{g}}(\omega, \theta), \quad (7)$$
$$A_{\mathbf{g}} = \frac{\pi e^6 n_a^2}{m^2 g^3} |S(\mathbf{g})|^2 F^2(g) e^{-g^2 u_T^2},$$

where $\mathbf{n}\mathbf{V} = V\cos\theta$, $S(\mathbf{g})$ is the unit-cell structure factor, $e^{-g^2 u_T^2}$ is the Debye–Waller factor, g is the reciprocal lattice vector, and n_a is the atomic density. The contribution of the incoherent component of radiation is suppressed and does not exceed several percent (7). The function $\Phi_{\mathbf{g}}(\omega, \theta)$ in (7) has a cumbersome form and is omitted here. In the case of relativistic electron energies, the spectrum of scattered photons contains sharp peaks with amplitudes

$$\Phi_{\mathbf{g}\,max} \approx \frac{2\gamma \tan(\theta/2)}{\sqrt{1 + \gamma^2 \omega_p^2/\omega^2}} \quad (8)$$

and relative width

$$\frac{\Delta\omega}{\omega} \approx \frac{\sqrt{1 + \gamma^2 \omega_p^2/\omega^2}}{2\gamma \tan(\theta/2)}. \quad (9)$$

These peaks arise at the frequencies

$$\omega = \omega_g = \frac{gV}{\sqrt{2(1 - \sqrt{\varepsilon} V \cos\theta)}} \approx \frac{g}{2\sin(\theta/2)}, \quad (10)$$

which are the Bragg frequencies for a fixed observation angle θ and a given reciprocal lattice vector g (the contribution to the radiation yield in the vicinity of ω_g is formed mainly by the microcrystallites that are in the position of specular reflection of virtual photons of the Coulomb field of fast electrons). Here, γ is the electron Lorentz factor.

Formulas (7)–(10) indicate the possibility of simultaneous determination of many reciprocal vectors of the polycrystal lattice from measurement of the spectrum of the scattered Coulomb field of a fast electron. This possibility was experimentally verified in [5]. Figure 1 shows the measured and calculated curves of the spectral distribution of the polarization bremsstrahlung (collimated at an angle of 90°) of 7-MeV electrons passing through a copper film. Note good agreement between the measurement results and theoretical pre-

dictions, not only in the positions of coherent peaks but also in their amplitudes.

Note also the very strong influence of density (following from (8)), which leads to saturation of the radiation yield at sufficiently high energies of probe electrons: $\gamma > \gamma_* = \omega_g/\omega_p$ (generally, $\omega_g \approx 5$–10 keV and $\omega_p \approx$ 20–50 eV; therefore, $\gamma_* \approx 100$–500). These estimates show that electron beams with energies not higher than 100 MeV are most appropriate for the diagnostic method under consideration.

4. When a fast electron moves through a fine-grained medium, the mechanism of Bragg scattering of the Coulomb field is not implemented due to the small grain size. Under the conditions considered here, the main factor is the coherent scattering from grains, which is most effective in the range of wavelengths exceeding the grain size. In this case, the dipole approximation is more appropriate to calculate the tensor G_{lp} in (3). Within this approximation, expression (2) gives the formula

$$4\pi i\omega G_{lp}(\mathbf{k}, \mathbf{k}') = -\delta_{lp}\frac{\omega^2}{2\pi^2}\alpha(\omega, \mathbf{k}' - \mathbf{k})\sum_j e^{i(\mathbf{k}' - \mathbf{k})\mathbf{r}_j},$$

$$\alpha(\omega, \mathbf{k}' - \mathbf{k}) = \alpha_0 f(\mathbf{k}' - \mathbf{k}), \qquad (11)$$

$$\alpha_0 = \frac{e^2}{m}\sum_n \frac{f_{n0}}{\omega_{n0}^2 - \omega^2}, \quad f(\mathbf{k}' - \mathbf{k}) = \frac{F(\mathbf{k}' - \mathbf{k})}{Z},$$

where $\alpha_0(\omega)$ is the conventional dipole polarizability of an atom, $\omega_{n0} = E_n - E_0$ is the difference of atomic energies in the $|n\rangle$ and $|0\rangle$ states, f_{n0} is the oscillator strength of the $|0\rangle \to |n\rangle$ transition, and Z is the number of electrons in an atom.

Further calculations are similar to those reported above. In particular, general formula (6) remains valid, in which one has to make the replacement $\varepsilon(\omega) \to \varepsilon_d(\omega) = 1 + 4\pi n_a \alpha_0(\omega) \equiv 1 + \chi(\omega)$. When necessary averagings are performed in general formula (6), one has to take into account the grain size finiteness. The calculations give the formula

$$\omega\frac{dN}{dtd\omega d\Omega} = \frac{e^2}{4(2\pi)^4 n_a}\omega^4|\chi(\omega)|^2 F_N(\omega), \qquad (12)$$

where the function $F_N(\omega)$ (omitted here because of its complexity) describes the coherent component of the polarization bremsstrahlung from grains (in this study, we considered grains in the form of a cube with N atoms on a side and a distance a between atoms). The function $\omega^4|\chi(\omega)|^2$ can be considered as known, because the dielectric susceptibilities of many materials have been experimentally determined in a wide frequency range. Obviously, this function becomes zero at $\omega \to 0$ and tends to a constant (ω_p^4) with an increase in ω. At the same time, the analysis performed showed that

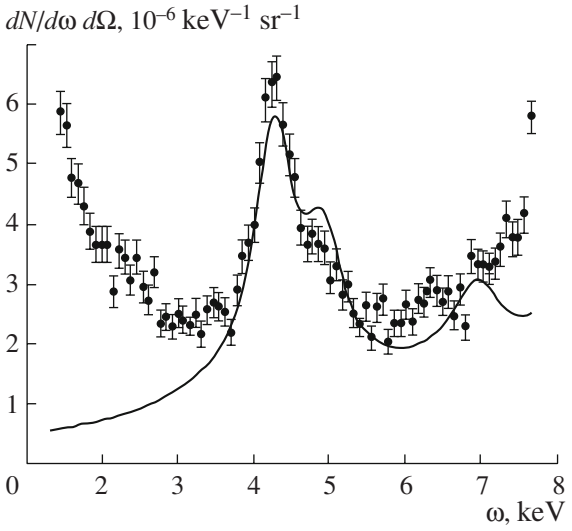

Fig. 1. Spectra of collimated polarization bremsstrahlung from a copper foil, obtained by absolute measurements (circles) and calculated from formula (7) (solid line) with the following parameters: electron energy 7 MeV, emission angle $\theta = 90°$, and foil thickness 15 μm; the electron velocity vector makes an angle of 45° with the foil surface.

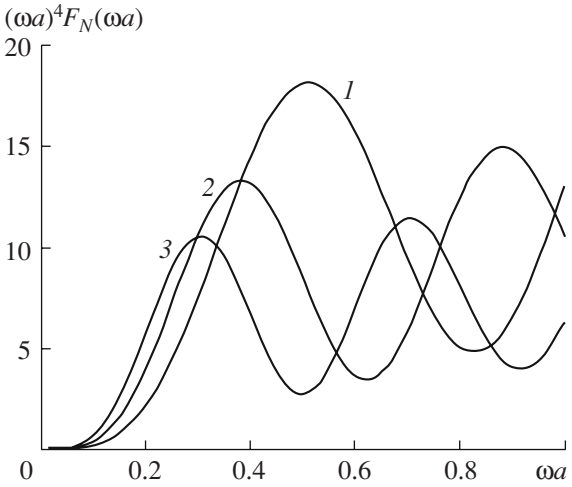

Fig. 2. Model spectrum of collimated polarization bremsstrahlung from a fine-grained medium. The quantity $\omega^4 a^4 F_N(\omega a)$ was calculated at $N = (1)$ 6, (2) 8, and (3) 10 and fixed parameters $\rho = \gamma^{-1} = 0.04$, $R/a = 0/1$, and $\theta = 90°$; the mutual grain screening was disregarded.

$F_N(\omega)$ is a decreasing function of ω; at the same time, this function increases with an increase in N and shifts to small values of ω (this effect is quite natural because the coherent response of a scattering object shifts to lower frequencies with an increase in the object sizes). The most important property of distribution (12) is the sharp dependence of the shape of this distribution on N, i.e., on the grain size. Figure 2 shows as an illustration

the dependences $\omega^4 F_N(\omega)$ for different N, which disregard mutual screening of grains. The plotted curves demonstrate a clear dependence of the spectrum on the grain size.

5. The analysis performed here shows good prospects of using the polarization bremsstrahlung of relativistic electrons in solution of the following diagnostic problems: (i) establishment of the structure of the atomic lattice of microcrystallites in polycrystalline samples and (ii) determination of the grain sizes in fine-grained media.

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