Polarization bremsstrahlung of fast electrons on metal nanospheres in a dielectric matrix in view of plasmon interference effects

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Abstract. The paper is devoted to the theoretical analysis of polarization bremsstrahlung (PB) due to scattering of fast electrons by a metal nanosphere embedded in a dielectric matrix in a frequency region in the vicinity of dipole and quadrupole plasmon resonances. Here we take into account plasmon interference effects that arise in the frequency-angular distribution of PB. These effects are a consequence of interrelation between contributions to PBs due to a plasmon on the sphere surface of different multipolarity. Our approach is based on the Fermi method of equivalent photons [2] and the Mie theory of radiation scattering by small metal particles [3]. It is shown that taking into account plasmon interference in the PB differential cross-section results in specific features in the spectral distribution of an emitted photon that depend strongly on the radiation angle and nanosphere radius.

1. Introduction
There are two components in bremsstrahlung (B) of a charged particle on a metal nanosphere in a dielectric matrix. One component is static bremsstrahlung (SB) of a projectile on a Coulomb force center [4]. The second component is radiation caused by the fact that a projectile polarizes electron shells (surface plasmons) of the nanosphere, and this polarization is also a source of radiation. This second component is called, for any targets, polarization bremsstrahlung (PB) [5].

2. Cross-section of virtual photon scattering by a metal nanosphere in a dielectric matrix
In case of metal nanospheres, the spectral-angular cross-section of scattering of monochromatic linearly polarized radiation is represented as

\[ \sigma_{sc}(\omega, \theta, \varphi) = \pi r_s^2 Q_{sc}(\omega, \theta, \varphi), \]  

where \( r_s \) is the nanosphere radius, \( Q_{sc}(\omega, \theta, \varphi) \) is the spectral-angular efficiency of scattering that is expressed in terms of the scattering function \( F_{sc}(\omega, \theta, \varphi) \) as follows [6]:

\[ Q_{sc}(\omega, \theta, \varphi) = \frac{4}{x^2} F_{sc}(\omega, \theta, \varphi), \]
where $x = kr$, $k = \sqrt{\varepsilon_m \omega / c}$ is the propagation vector of radiation in a matrix with a dielectric permittivity $\varepsilon_m$, in which nanospheres with a dielectric permittivity $\varepsilon_m(\omega)$ are placed. The geometry of the process is shown in figure 1, where $\theta, \varphi$ are the polar and azimuth angles of the propagation vector of scattered radiation $\mathbf{k}$.

\[ F_{sc}(\omega, \theta, \varphi) = |S_1(\omega, \theta)|^2 \sin^2 \varphi + |S_2(\omega, \theta)|^2 \cos^2 \varphi, \]

where $S_1(\theta, \omega) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[ a_n(\omega) \pi_n(\theta) + b_n(\omega) \tau_n(\theta) \right]$,  

\[ S_2(\theta, \omega) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[ a_n(\omega) \tau_n(\theta) + b_n(\omega) \pi_n(\theta) \right], \]

\[ \pi_n(\theta) = \frac{P_n'(\cos \theta)}{\sin \theta}, \tau_n(\theta) = \frac{dP_n'(\cos \theta)}{d\theta}. \]

In our case of cylindrical symmetry, the cross section should be averaged over the azimuth angle $\varphi$. Then the scattering function loses the dependence on the azimuth angle, and the scattering cross-section depends only on the frequency and the polar angle.

The multipole expansion coefficients $a_n, b_n$ are given within the framework of the Mie theory [3]:

\[ a_n(x, y, m) = \frac{\psi_n'(y)\psi_n(x) - m\psi_n'(x)\psi_n(y)}{\psi_n'(y)\zeta_n(x) - m\zeta_n'(x)\psi_n(y)}, \]

\[ b_n(x, y, m) = \frac{m\psi_n'(y)\psi_n(x) - \psi_n'(y)\psi_n(x)}{m\psi_n'(y)\zeta_n(x) - \zeta_n'(x)\psi_n(y)}, \]

where $m = \sqrt{\varepsilon_m(\omega)/\varepsilon_m}.$

The coefficients $\psi_n(z) = z j_n(z)$ and $\zeta_n(z) = z\chi_n(z)$ are the functions introduced by Debye, where $j_n(z), \chi_n(z)$ are the spherical Bessel and Hankel functions.

The dielectric permittivity of a metal sphere can be expressed in terms of the real $n_s(\omega)$ and imaginary $\kappa_s(\omega)$ parts of the refractive index of a metal:
$$\varepsilon_r(\omega) = \varepsilon_i(\omega) + i \varepsilon_2(\omega) = \left[ n_i(\omega) \right]^2 - \left[ \kappa_i(\omega) \right]^2 + 2 i n_i(\omega) \kappa_i(\omega).$$  \hspace{1cm} (9)

For the functions $n_i(\omega)$ and $\kappa_i(\omega)$, we use experimental data obtained in the work [7].

We assume the dielectric permittivity of the matrix to be real and independent of radiation frequency:

$$\varepsilon_m = \text{const.}$$

Following the Fermi method of equivalent photons [2], the general formula for the PBs differential cross-section used in our calculations looks like

$$\frac{d\sigma^{(pb)}}{d\omega d\Omega} = \frac{c}{(2\pi)^2} \int_0^\infty \sigma_{sc}^{(pb)}(\omega, \theta) F^2 \left( \frac{\sqrt{\omega^2 + \rho^2}}{\omega} \right) \frac{|E(\omega, \rho, \nu)|^2}{h \omega} \rho d\rho,$$  \hspace{1cm} (10)

where $\rho$ is the impact parameter, $\omega$ is the electron velocity, $\Omega$, $\theta$ are the solid and polar angles of radiation, $\omega$ is the PBs frequency, $\sigma_{sc}^{(pb)}(\omega, \theta)$ is the cross-section of radiation scattering by a metal nanosphere, $E(\omega, \rho, \nu)$ is the Fourier transform of the scattered electron electric field strength for the specified impact parameter, $F(q)$ is the nanosphere form factor, $F(q) = 3 j_1(qr) / qr$ and $j_1(x) = \frac{\sin(x)}{x^2} - \frac{\cos(x)}{x}$. In the approximation of straight trajectories, for the normal component of the Fourier transform of the electric field strength in medium with the impact parameter $\rho$ we have

$$E_n = \frac{2 e \omega}{v^2} K_1 \left( \frac{\omega \rho}{\sqrt{\gamma}} \right),$$

where $K_1(z)$ is the Macdonald function of the first order and $\gamma = \left( 1 - \varepsilon_m v^2 / c^2 \right)^{-1/2}$ is relativistic factor in medium with the dielectric permittivity $\varepsilon_m$. For parallel electric field strength we have:

$$E_z = \frac{2 e \omega}{\sqrt{\gamma} \gamma} K_0 \left( \frac{\omega \rho}{v \sqrt{\gamma}} \right),$$

where $K_0(z)$ is the Macdonald function of the zero order.

In the following calculations we consider the electron velocity $v = 90$ at.u. For this velocity and glass dielectric permittivity $\varepsilon_m = 2.25$ we have $\gamma \approx 5.87$. Numerical analysis shows that for such parameters the contribution of parallel electric field into PB cross section is negligible. Thus we can substitute $E_n$ in expression (10) instead of $E$ in our calculations.

3. Results of calculations

In this section we present the results of calculations of electron PBs on silver and gold nanospheres in a glass matrix in different regions of PB frequencies in the vicinity of plasmon resonances for different radiation angles and nanosphere radii.

For metal nanospheres, values of radii about 50 nm were chosen, which corresponds to optical frequencies of corresponding plasmon resonances. The PB directional pattern was studied mainly for forward and backward directions; besides, integration and averaging over the PB solid angle were carried out. All calculations were done in the MathCAD system with the use of experimental data on dielectric permittivities of gold and silver in corresponding frequency ranges [7].

The comparison of the PB spectra in the vicinity of surface plasmon resonances for different radiation angles $\theta$ on a silver nanosphere in glass is given in figures 2 and 3. Solid line (forward PB) – $\theta = 0.1$, dotted line – the calculation was performed with the scattering cross-section averaged over the solid angle of radiation, dashed line (backward PBs) – $\theta = 9\pi/10$. The ordinate is plotted in nm$^2$ / (eV sr), the abscissa is plotted in eV.
Figure 2. For a silver nanosphere in glass with the radius $r_s = 40$ nm, the electron velocity $v = 90$ atomic units: solid line – forward PB, dotted line – the calculation was performed with the scattering cross-section averaged over the solid angle of radiation, dashed line – backward PB.

Figure 3. For a silver nanosphere in glass with the radius $r_s = 60$ nm, the electron velocity $v = 90$ atomic units: solid line – forward PB, dotted line – the calculation was performed with the scattering cross-section averaged over the solid angle of radiation, dashed line – backward PB.

The dependences of the PB cross-section on the nanosphere radius for two different photon energies are shown in figures 4, 5.
Figure 4. For a silver nanosphere in glass, $\hbar \omega = 3.14$ eV (in the vicinity of the spectral extremum), the electron velocity $v = 90$ atomic units: solid line – forward PB, dotted line – the calculation was performed with the scattering cross-section averaged over the solid angle of radiation, dashed line – backward PB.

Figure 5. For a silver nanosphere in glass, $\hbar \omega = 2.48$ eV (in the vicinity of the dipole maximum), the electron velocity $v = 90$ atomic units: solid line – forward PB, dotted line – the calculation was performed with the scattering cross-section averaged over the solid angle of radiation, dashed line – backward PB.

The comparison of the PB spectra for different radiation angles $\theta$ on a gold nanosphere in glass with the radius $r_s = 40$ nm is given in figure 6.
Figure 6. For a gold nanosphere in glass with the radius $r_s = 40$ nm, the electron velocity $v = 90$ atomic units: solid line – forward PB, dotted line – the calculation was performed with the scattering cross-section averaged over the solid angle of radiation, dashed line – backward PB.

The imaginary parts of dielectric permittivities of silver (solid line) and gold (dotted line) as functions of photon energy are demonstrated in figure 7.

Figure 7. The imaginary parts of dielectric permittivities of silver (solid line) and gold (dotted line) as functions of photon energy are taken from [7].

The normalized angular dependence of PB cross-section for various frequencies is given in figure 8.
Figure 8. For a silver nanosphere in glass with the radius \( r_s = 40 \) nm, the electron velocity \( v = 90 \) atomic units: solid line – \( \hbar \omega = 3.14 \) eV, dotted line – \( \hbar \omega = 3.0 \) eV, dashed line – \( \hbar \omega = 2.48 \) eV.

4. Conclusion
Polarization bremsstrahlung (PB) of fast electron scattered by a metal nanosphere embedded in a dielectric matrix in a frequency region in the vicinity of dipole and quadrupole plasmon resonances was studied theoretically. The numerical analysis of electron PB on a metal nanosphere in a dielectric was carried out in view of surface plasmon interference effects.

It was shown that the spectrum of forward PB on a silver nanosphere in glass has a maximum due to constructive interference between dipole and quadrupole plasmon contributions, while the spectrum of backward PBs has a minimum at the same photon energy.

The sharpness of spectral extrema depends on the nanosphere radius and the nanosphere metal. The angular distribution of PB depends strongly on the photon energy and the nanosphere radius.

The calculation has shown that the role of plasmon interference effects is negligible for PB on a gold nanosphere because of interband transitions in gold, which results in the high value of the imaginary part of the dielectric permittivity of gold in the actual spectral range.

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