Resonant Diffusive Radiation in Random Multilayered Systems

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We have theoretically shown that the yield of diffuse radiation generated by relativistic electrons passing random multilayered systems can be increased when a resonant condition is met. Resonant condition can be satisfied for the wavelength region representing visible light as well as soft X-rays. The intensity of diffusive soft X-rays for specific multilayered systems consisting of two components is compared with the intensity of Cherenkov radiation. For radiation at photon energy of 99.4 eV, the intensity of Resonant Diffusive Radiation (RDR) generated by 5 MeV electrons passing a Be/Si multilayer exceeds the intensity of Cherenkov radiation by a factor of \approx 60 for electrons with the same energy passing a Si foil. For a photon energy of 453 eV and 13 MeV electrons passing Be/Ti multilayer generate RDR exceeding Cherenkov radiation generated by electrons passing a Ti foil by a factor \approx 130.

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I. INTRODUCTION

Recently radiation generated by relativistic electrons passing through periodic multilayer structures have received increased attention\textsuperscript{1,2} This has particularly been enabled by the availability of multilayers with periodicities in the nm scale. Yamada and Hosokava\textsuperscript{2} demonstrated Resonant Transition Radiation RTR\textsuperscript{4-6} to be applicable as a radiation source in the wavelength region \( \lambda < 1 \text{nm} \). This was achieved by 15 MeV electrons passing a periodic multilayer with 170 nm thick nickel layers as radiator and 221 nm thick carbon layers as a spacer. As the RTR intensity is proportional to \([\varepsilon_1(\omega) - \varepsilon_2(\omega)]^2\), where \(\varepsilon_1\) and \(\varepsilon_2\) are the dielectric constants of the multilayer components, a sharp increase can be expected around inner-shell absorption edges\textsuperscript{1}.

In the X-ray region, the dielectric constant can be described by the plasma formula and usually is less than unity. However, in narrow region around absorption edges of some materials the dielectric constant can exceed unity. This means that the condition for Cherenkov radiation (CR) can be fulfilled even in the X-ray region. The intensity of CR is proportional to \(\varepsilon(\omega) - 1\).

Bazylev and et al\textsuperscript{7} demonstrated the generation of CR around the K absorption edge of C from 1.2 GeV electrons passing a carbon containing foil. More recently Knulst et al\textsuperscript{8} used 5 MeV electrons to generate CR around the Si L absorption edge \(\approx 99.4\text{MeV}\) and 10 MeV electrons to generate CR around L edges of V (512 eV) and Ti (453 eV).

Previously, we have shown that Diffusive Radiation (DR) generated by relativistic electrons passing a randomly distributed multilayer system can provide an alternative for a source with a high radiation yield\textsuperscript{9}. Experimental evidence in favor of this mechanism was provided by L.C. Yuan and et al\textsuperscript{10} who conducted experiments on radiation generated by relativistic electrons passing a system of microspheres, distributed randomly in a dielectric material. The observed strong dependence of the radiation intensity as a function of the particle energy could only be explained by the DR mechanism\textsuperscript{11}. In this paper we discuss the influence of a resonance effect on the yield of Diffusive Radiation (RDR). As the resonant condition requires that \(n > 1\), this effect can for radiation in the soft x-ray region only be achieved around selected absorption edges representing an atomic inner-shell of an element. Our paper explicitly deals with the generation of Resonant Diffuse Radiation around these inner-shell absorption edges. A complicating factor for this short wavelength region is that DR requires a low absorption to enable multiple scattering. Nevertheless we have shown that when a resonance condition is met related to the anomalous behavior of dielectric constant around an adsorption edge, the yield resulting from RDR exceeds the yield of CR in this soft x-ray region.

II. INITIAL RELATIONS

Let us consider radiation from a charged particle uniformly moving in a homogeneous medium with randomly embedded parallel foils in it. The origin of the radiation can be explained as follows. Each charged particle creates an electromagnetic field around it which is not yet photon but a pseudophoton. These pseudophotons are scattered on the inhomogeneities of dielectric constant and convert into real photons. We consider the radiation due to scattering of pseudophotons which includes also the conventional transition radiation (TR). We do not consider the bremstrahlung and Cherenkov radiation which have different origin. We have shown\textsuperscript{9} that the radiation intensity can be represented as a sum of two contributions. One is the contribution by single scattering and the another is the contribution caused by multiple scattering of pseudophotons. The single scattering contribution actually is TR from randomly spaced interfaces. However here we are interested in the contribution by multiple scattering\textsuperscript{9}, as it was shown that it leads to
the diffusion of pseudophotons. As demonstrated in a simplified expression for the yield for DR can be achieved by introducing a random multilayer system with Gaussian distribution of distances between the foils. It should be emphasised that other random systems are not excluded to be applicable. The statistical properties of the Gaussian distribution enters to the solution through the elastic mean free path of the photons.

That means that in a real system the randomness created by the parallel foils should fulfill the requirement to treat the dielectric constant as Gaussian distributed random function. For a large number of foils \( N \gg 1 \), in the wavelength range \( \lambda \ll l \) (where \( l \) is the photon mean free path in the normal to the foils direction, for more details see [2]) diffusion contribution to the radiation intensity is the main one and given by the formula

\[
I(\omega, \theta) = \frac{5\varepsilon^2\gamma^2(\omega) L_z L^2 \sin^2 \theta}{2\varepsilon(\omega)c^2|\cos \theta|}
\]

where \( \varepsilon \) is the average dielectric constant of the system

\[
\varepsilon = \varepsilon_0 + na(\varepsilon_f - \varepsilon_0)
\]

Here \( \varepsilon_0 \) and \( \varepsilon_f \) are dielectric constants of the medium and the plate, respectively, \( \alpha \) is the thickness of the foils, \( \gamma_m = (1 - \varepsilon^2/\varepsilon_f^2)^{-1/2} \) is the Lorentz factor of the particle in the medium, \( L_z \) is the system size in the z direction (we assume that the particle is moving in this direction) and \( L \) is the characteristic size of the system. The formula [1] has a clear physical meaning [2]. The quantity \( \varepsilon^2\gamma^2 L_z/c \) is the total number of pseudophotons in the medium, \( 1/l \) is the probability of the photon scattering and \( L^2/\lambda^2 \) is the average number of pseudophoton scatterings in the medium. The Eq.[1] is correct provided that \( \gamma^2_m \gg ak \), where \( k = \omega/c \) is the photon wave number. Taking into account that \( \gamma^2_m/k \) is the coherence length (radiation formation zone) [12] this condition means that many foils can be placed in a coherence length. Note that at the resonance point where \( \gamma_m \rightarrow \infty \), the condition \( \gamma^2_m \gg ak \) is automatically fulfilled. This regime differs from those one which is usually explored in the periodical multilayered systems [1, 2] where an enhancement of photon yield appear as a result of constructive interference.

Note that Eq.[1] is correct provided that \( |\cos \theta| \gg (\lambda/l)^{1/3} \). This condition is caused by the fact that parallel to foils pseudophotons are not diffusing so why the theory is not applicable for very large angles. For the derivation of Eq.[1] we have neglected the absorption of the electromagnetic field. However a weak absorption \( l_{in} \gg l \) (where \( l_{in} \) is the photon absorption length) can be taken into account in the following way [13] and also [11]. When \( L > \sqrt{l_{in}/m} \) the length \( \sqrt{l_{in}/m} \) becomes an effective size of the system and one should substitute \( L^2 \) by \( l_{in} \) in Eq.[1], to obtain

\[
I(\omega, \theta) = \frac{5\varepsilon^2\gamma^2(\omega) L_z l_{in}(\omega) \sin^2 \theta}{2\varepsilon(\omega)c^2|\cos \theta|}
\]

Note that absorption introduces an extra frequency dependence in radiation intensity. In section IV we will investigate the photon mean free paths in the medium more detail.

III. RESONANT EMISSION

As is seen from Eqs.[1] and [3] the intensity of Diffusive Radiation is proportional to the square of particle Lorentz factor in the medium \( \gamma^2_m \)

\[
\gamma^2_m = 1 - \frac{\omega^2\varepsilon}{c^2} = 1 - \frac{\omega^2\varepsilon}{c^2}[\varepsilon_0 + (\varepsilon_f - \varepsilon_0)na]
\]

It follows from Eqs.[1], [3] and [4] that when \( v \) is such that \( \gamma^2_m = 0 \) (resonant condition) the radiation intensity drastically increases. When \( \gamma_m \rightarrow \infty \) the radiation intensity also becomes infinite. However physically this is impossible. This infinity arises because in theoretical consideration we assumed the system size infinite. However in reality the system size is finite and the intensity even at the resonance point also will be finite. Now consider the resonance condition \( \gamma^2_m = 0 \) in the optical and X-ray regions, respectively. In the optical region taking vacuum as a homogeneous medium, \( \varepsilon_0 = 1 \) and assuming for relativistic electrons that \( v \approx c \), from Eq.[1], one gets

\[
\gamma^2_m = 1 - \delta - \delta
\]

where \( \gamma = (1 - \omega^2/c^2)^{1/2} \) is the Lorentz factor of the particle and \( \delta = na(\varepsilon_f - 1) \). So the resonance condition in this region has the form \( \gamma^2 = \delta \) and weakly depends on the frequency.

Now consider the X-ray region. Assuming that \( \varepsilon_0 = 1 - \delta_s \) and \( \varepsilon_f = 1 - \delta_f \), for relativistic electrons \( v = c \), one has from Eq.[4]

\[
\gamma^2_m = \gamma^2 + \delta_s(na) + \delta_f na
\]

In the X-ray region one usually has \( \delta_s, \delta_f > 0 \) and the resonance condition \( \gamma^2_m = 0 \) can not be fulfilled. However in the atomic inner-shell region the dielectric constant can resonantly increase and exceed unity, [1, 15]. This means that for those frequencies, for example, one can have \( \delta_f(\omega) < 0 \). As is seen from Eq.[7] if \( \delta_f(\omega) < 0 \) then a resonant \( \gamma^2_m \rightarrow \infty \) is possible in the X-ray region too. The corresponding X-ray spectrum will be quite narrow because the region where \( \varepsilon_f(\omega) \) exceeds unity is small.

It should be emphasized that the resonance condition \( \gamma^2_m = 0 \) is the Cherenkov condition for the average dielectric constant. Hence one can say that RDR origins from the interaction of two processes. The Cherenkov condition creates resonantly large number of pseudophotons and multiple scattering converts them into real photons.

Although the resonant condition is the same as for Cherenkov radiation, the essence of RDR completely different. While CR is based on interference of wave fronts
and inhomogeneity is not needed for its generation, DR is caused by multiple scattering of pseudophotons on inhomogeneities. We show below that in some cases the photon yield in RDR can exceed that one in CR.

IV. ELASTIC AND INELASTIC MEAN FREE PATHS

The elastic mean free paths appearing in Eqs. (11) and (14) correspond to the photons falling in under the normal of the foils although those expressions determine the radiation intensity for all angles except for a small region. The photon mean free path in the medium is related to the transmission coefficient $t(\omega)$ through a foil by

$$l(\omega) = \frac{1 - Ret(\omega)}{n}$$  \hfill (7)

where $n$ is the concentration of the foils and $t(\omega)$ is given by the formula

$$t(\omega) = \frac{2i\sqrt{\epsilon_f(\omega)/\epsilon_0} \exp(-ika)}{[\sqrt{\epsilon_f(\omega)/\epsilon_0} + 1]|\sin\sqrt{\epsilon_f(\omega)/\epsilon_0}ka + 2i\sqrt{\epsilon_f(\omega)/\epsilon_0} \cos\sqrt{\epsilon_f(\omega)/\epsilon_0}ka|}$$  \hfill (8)

In the Born approximation it holds that $|\sqrt{\epsilon_f/\epsilon_0} - 1|/ka \ll 1$ even though $(ka \gg 1)$. So one obtains from Eqs. (7) and (8)

$$l(\omega) = \frac{2}{n(\sqrt{\epsilon_f(\omega)/\epsilon_0} - 1)^2 ka^2}$$  \hfill (9)

More interesting for us is the geometrical optics region $|\sqrt{\epsilon_f/\epsilon_0} - 1|/ka \gg 1$. Substituting Eq. (8) into Eq. (7) and neglecting strongly oscillating terms in the geometrical optics region, one has $l(\omega) \approx 1/n$. Thus in this region the photon elastic mean free path does not depend on frequency and the radiation intensity is maximal. Usually, in the X-region $|\sqrt{\epsilon_f/\epsilon_0} - 1|/ka \ll 1$ it is therefore difficult to satisfy the condition of applicability of geometrical optics. However, for the atomic inner shell frequencies the dielectric constant some times, even in the X-ray region, can exceed unity and for those frequencies the geometrical optics condition $|\sqrt{\epsilon_f/\epsilon_0} - 1|/ka \gg 1$ is fulfilled. Now about the photon inelastic mean free path. In the frequency region we are mainly interested in the dominant mechanism of inelastic interaction of photon with the medium is the photoabsorption. Therefore the inelastic mean free path can be represented in the form

$$l^{-1}_{in} = N_1\sigma_{1a}(\omega) + N_2\sigma_{2a}(\omega)$$  \hfill (10)

where $N_1, N_2, \sigma_{1a}, \sigma_{2a}$ are the numbers of atoms in an unit volume and photoabsorption cross section for the first and second media, respectively.

V. ABSORPTION OF RDR PHOTONS IN THE MEDIUM

Above we have discussed the absorption of pseudophotons in the random medium. However already formed real photons also will be absorbed in the medium. Therefore to know what part of already created real photons will escape the system one should take into account the absorption of real photons in the medium. Suppose that we are interested in the photon yield from the depth $z$ in the material. Using Eq. (6) and adding an exponential decaying factor which takes into account the difference of paths of photons with different emitted angles, one has

$$\frac{dI}{dz} = \frac{5e^{2\gamma_m^2(\omega)|l_{in}(\omega)\sin^2\theta|}}{2\epsilon(\omega)c^2|\cos\theta|} \exp \left[ \frac{-z}{l_{in}|\cos\theta|} \right]$$  \hfill (11)

Here $\frac{dI}{dz}$ is the spectral-angular radiation intensity per unit length of electron path in the medium. Note the suppression of radiation intensity at very large angles. The real DR photons are formed in the effective size $(ll_{in})^{1/2}$. Therefore when finding the total radiation intensity one should cut the integral in the lower limit on this length. After integration for total spectral angular intensity, we have

$$I = \frac{5e^{2\gamma_m^2(\omega)|l_{in}\sin^2\theta|^2}}{2\epsilon(\omega)c^2} \exp \left[ \left( \frac{l_{in}}{l} \right)^{1/2} \frac{1}{|\cos\theta|} \right]$$  \hfill (12)

So as one could anticipate the absorption of real DR photons leads to the cutoff of radiation intensity at large angles and maximum lies at medium angles.

VI. QUALITATIVE ESTIMATES FOR SPECIFIC ELEMENTS

We have shown that resonant emission of radiation intensity is possible in a system with one-dimensional randomness of the dielectric constant. Resonant emission is possible in the optical as well as in the X-ray regions. The latter case is most interesting because it forms high bright soft X-ray source using MeV electrons. 1, 3.

One of the main conditions for realization of diffusive radiation mechanism is $l_{in} \gg l$. The minimal value of $l$ is reached in the geometrical optics region, $l \sim 1/n$. The geometrical optics approximation for $l$ is justified provided that $|\sqrt{\epsilon_f/\epsilon_0} - 1|/ka \gtrsim 1$. In order to obtain an impression how large the yield of DR photons can be let us make a crude comparison with Cherenkov radiation. As is shown in 12 the ratio of DR and Cherenkov radiation intensities under weak absorption is of order

$$\frac{I_D}{I_C} \sim \frac{1}{kl_{in}^{1/2}} \frac{l_{in}}{l}$$  \hfill (13)

It should be emphasized that this formula represents a leading order comparison for the yield RDR and CR. When the resonance condition $v \rightarrow c/\sqrt{\epsilon}$ is fulfilled, CR
as well as RDR within the same wavelength are generated. While the emission angle of Cherenkov radiation is fixed at $\cos \theta = c/\nu \sqrt{\epsilon}$, RDR is emitted within a wide angular distribution. We omit the angular part in Eq. (10) for simplicity. Consider the conditions of generation of RDR for specific structures using the data in [10].

First consider the structure $Be/Si$ at the photon energy $\omega = 99.4eV$ near the L3 edge of Si. Note that we take the photon energy at the long wavelength side of the Si L3 edge attenuation length at $\omega = 99.8eV$ where the absorption weaker. However for 99.4eV the refractive index $n_{Si}(\omega) = 1 + 11.56 \times 10^{-3}$ still exceeds unity. Using also that $n_{Be} = 1 - 4.27 \times 10^{-3}$ one can be convinced that the geometrical optics approximation is valid provided that $a > 130nm$. The photon attenuation length at $\omega = 99.4eV$ in Si and Be is 0.55$\mu$m and 0.76$\mu$m, respectively [10]. Therefore if one takes the average distance $d$ between Si foils $\sim 130nm$, the elastic and inelastic mean free paths of photon approximately can be estimated as $l \sim d \sim 130nm$ and $l_{in} \sim 0.62\mu$m. As a result the photon multiple scattering condition $l_{in} \gg l$ is fulfilled. Now let us find the resonant electron energy. Using the above mentioned values of refraction index for Be and Si, assuming the filling factor of Si approximately, $na \sim 1/2$, one finds from Eq. (10) that $\gamma_{r} = 11.71$. Correspondingly the resonant electron energy will be $E_{r} = 5.355MeV$. Now estimate the ratio $I^{D}/I^{C}$ using Eq. (10). For a photon energy $\omega = 99.4eV$ for $l = 130nm$, the factor $1/kl \approx 0.01$. As at the resonance point our formulae are not applicable we choose an electron energy of $E = 5.1MeV$ and find $\gamma_{m}^{2} \approx 1471$. Finally, taking into account that $l_{in}/l \sim 4$, one finds a much larger diffusive radiation than Cherenkov radiation $I^{D}/I^{C} \sim 60$. The number of foils and correspondingly the system size can not be too large because of absorption. On the other hands one should have enough number of foils to treat the system as a random. For the average photon attenuation length 0.6$\mu$m a system size of order 10$\mu$m is reasonable. This size means approximately 40 foils. Another interesting possibility is to generate RDR around the L3 edge of Ti. For a photon energy of $\omega = 453eV$ the refractive index of Ti is $n_{Ti} = 1 + 3.0826 \times 10^{-3}$. As a spacer material choose a light element to make reduce the total absorption. Let consider, for example, Be. For Be at 453eV $n_{Be} = 1 - 1.7932 \times 10^{-3}$ and attenuation length is 0.9117$\mu$m. The attenuation length for Ti at 453eV is 0.661$\mu$m. Analogous calculations as for the Be/Si combination show that the geometrical optics condition $ka/n_{Ti}/n_{Be} - 1| \geq 1$ can be satisfied for 100nm thick Ti foils. The resonance electron energy for a Ti filling factor $na \sim 1/2$ is 13.45MeV. Taking for the electron energy $E = 13MeV$ one obtains for the ratio $I^{D}/I^{C} \sim 130$. The number of foils could be 40–50 and the system size 10$\mu$m.

VII. DISCUSSION

Let us first point out the main differences between random and periodical systems. Gaussian averaging over the randomness enables one to obtain a compact expression for radiation yield in contrary to the sum of complicated terms in periodical case. It is easier to construct a random system than a periodical one therefore random systems are more convenient from practical point of view. Consideration of pseudophoton multiple scattering effects in periodical systems remains as an open problem.

Many approximations are used when we estimate the ratio $I^{D}/I^{C}$. Therefore we emphasize that the calculated values for $I^{D}/I^{C}$ do not represent exact optimized values. The calculations only give an impression about a possible yield. The before mentioned ratios are very sensitive to electron energy, refractive index, filling factor $na$, frequency etc. because the resonant character of radiation. Theoretically for the resonance condition the ratio can be very large. However this requires electrons in the MeV range with an energy accuracy of better than 0.01MeV. As shown before, RDR for the Extreme Ultra Violet and shorter wavelength regions is only possible within a narrow bandwidth for which $n(\omega) > 1$.

VIII. CONCLUSIONS

The possibility to fundamentally generate Resonant Diffusive Radiation for the short wavelength region (EUV and soft x-rays) using a multilayer system with random periodicity has been demonstrated theoretically. For a resonant condition it is required that $\gamma_{m}^{2} \approx 0$. For the wavelength region as considered, this is only possible around absorption edges of some elements where $\varepsilon > 1$.

We demonstrated the feasibility to apply this phenomenon on a practical bright source. Therefore we made some preliminary calculations for specific materials combinations, comparing the RDR yield with the CR yield. Experimental research is justified to further understanding of the phenomenon. Moreover, we expect the new high brightness photon sources can be developed, applicable in x-ray microscopy or EUV lithography.

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[1] A.E.Kaplan, C.T.Law and P.L.Shkolnikov, Phys.Rev.E 52, 6795 (1995).
[2] K.Yamada, and T.Hosokawa, Phys.Rev.A 59, 3673 (1999).
[3] B.Lastgrader, A.Tip and J.Verhoeven, Phys.Rev.E 61, 5767 (2000).
[4] M.L.Ter-Mikaelian, High-Energy Electromagnetic Processes in Condensed Media, John Wiley and Sons Inc., N.Y. (1972).
[5] P.Rullhusen, X.Artru and P.Dhez, Novel Radiation Sources Using Relativistic Electrons, World Scientific, (1998).
[6] M.L.Cherry, G.Hartmann, D.Muller and T.A.Prince, Phys.Rev.D 10, 3594 (1974).
[7] V.A.Bazylev, V.I.Glebov, E.I.Denisov, N.K.Zhevago and A.S.Khlebnikov, JETP Lett., 24, 371 (1976).
[8] W.Knulst, J.Luiten, M.van der Wiel and J.Verhoeven, Appl.Phys.Lett., 79, 2999, (2001); ibid 83, 4050, (2003).
[9] Zh.S.Gevorkian, Phys.Rev.E 57, 2338 (1998); Sov.Phys.JETP 114, 91, (1998).
[10] L.C.Yuan et al., NIM A 441, 479 (2000).
[11] Zh.S.Gevorkian, C.P.Chen and Chin-Kun Hu, Phys.Rev.Lett., 86, 3324, (2001).
[12] Zh.S.Gevorkian, NIM B 145, 185 (1998).
[13] P.W.Anderson, Phil.Mag. B52, 505 (1985).
[14] V.A.Kosobukin, Solid State Physics 32, 227 (1990).
[15] B.L.Henke, E.M.Gullikson and J.C.Davis, At.Data Tables 54, 181, (1993).
[16] see the website, [http://www-cxro.lbl.gov/optical] constants