TRANSITION RADIATION AND COLLECTIVE OSCILLATIONS IN METALLIC FILMS

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The radiation produced upon passage of fast electrons through a metallic plate is studied. It is shown that under the conditions of the experiments hitherto carried out an account of the possibility of excitation and propagation of longitudinal waves is not important. A detailed comparison of the theoretical and experimental results shows that the radiation observed in the experiments can be sufficiently well described by the theory of transition radiation.

1. When fast electrons pass through metallic films, discrete energy losses of the electrons are observed. It is clear now that these losses are connected with the excitation of collective oscillations, which correspond in an isotropic medium to longitudinal field oscillations. The spectrum of such oscillations is determined by the zeroes of the dielectric constant [1)]. The longitudinal oscillations cannot propagate in vacuum. However, owing to the presence of the boundary of the medium (surface of the film), it becomes possible to transform the oscillations excited by the fast electrons in the film into transverse electromagnetic radiation which emerges from the film and which can be detected, for example, by optical means.

In 1960 there were published interesting experimental papers[1,2] reporting the observation of electromagnetic radiation occurring when fast electrons pass through thin silver films. In the first of these investigations, the silver films were bombarded with a beam of 25-keV electrons. The expected radiation was observed by locating to the side of the beam a small quartz spectrograph with resolution ~ 10 mμ. No absolute calibration was made, and optical filters were used to fix the point on the spectrum. The radiation was recorded on photographic film. Radiation characterized by a rather sharp peak at $\lambda = 330 \pm 10$ mμ was observed, along with a continuous-spectrum radiation which terminated at $\lambda \lesssim 320$ mμ but extended far in the long-wave region. The intensity and the form of the radiation spectrum varied periodically with the thickness, which assumed in the experiment values of 45, 85, and 150 mμ. The period was ~ 100 mμ. The angle at which the radiation was observed is not indicated.

In the second paper, published in the same issue[2], silver films ~ 500 Å thick and 22-keV electrons normally incident on the film were used. The radiation was registered with a photomultiplier, using an amplifier and a phase analyzer. This experiment also disclosed radiation in the spectral interval indicated above. The angular distribution of the radiation investigated in this experiment has a maximum at $\theta \sim 40°$ (the angle was measured from the normal to the film), falling off as $\theta \to 0°$ and 90°. It was found that the spectral distribution depended essentially on θ. At $\theta = 30°$ the form of the spectrum agreed with that in the first paper, while the maximum was at $\lambda = 340$ mμ. At large angles ($\theta = 75°$) there is no sharp peak on the curve. It was impossible to observe the radiation polarization.

The investigations referred to [1,2] were stimulated by the work of Farrell[3], who made an attempt to determine theoretically the probability of transformation of longitudinal waves into transverse ones from the point of view of the notion of the existence of longitudinal oscillations (plasmons). Without touching on the inconsistency of Farrell's paper[3] (see, for example, the report by Lippmann[4]), it must be stated that no account was taken in it of many specific features of the electrodynamics of media in which longitudinal waves are possible. However, the most important aspect is that the results of the investigations of Steinmann[1] and Brown et al[2] actually correspond to the predictions of the theory of transition radiation[5-7]. We shall show this in detail in what follows. In addition, we shall disclose the specific features due to the possibility of propagation of longitudinal waves (plasmons) in the medium. (A preliminary report on some of the results of this investigation was published by us earlier[8].)

[1)]We shall not discuss here the question of surface oscillations.

2. The interaction between fast electrons and a metal, particularly a metallic film, leads to the occurrence of various types of radiation: x-rays, luminescence, Cerenkov effect, and transition radiation. Let us turn first to an analysis of transition radiation, since it will be shown that this was precisely what was observed in the cited experiments.

The theory of transition radiation, the existence of which was predicted theoretically some time ago by Ginzburg and Frank[5], has by now been sufficiently well developed. In particular, transition radiation from a plate was considered by Pafomov [6] and by Garibyan and Chalikyan[7]. They assumed, however, that only transverse electromagnetic waves can propagate in the medium. With an aim at investigating the role of plasmons in radiation from a metallic plate, we shall consider the excitation and propagation in a medium of not only transverse but also longitudinal electromagnetic waves; we shall also consider the question of their transformation into transverse electromagnetic waves which emerge to the vacuum.

The possibility of propagation of longitudinal waves in a medium is accounted for by the spatial dispersion of the dielectric constant[9] which determines, in particular, the group velocity of the collective plasma excitations.

In the general case the connection between the induction D and the electric field E is functional, and the order of the equations is infinite. In the experiments of [1,2] the investigated region of the radiation spectrum corresponds to values of the frequency and wave vectors (k—wave vector of radiation in the medium) such that the parameter kv_0/ω is small (v_0 is the characteristic velocity of the electrons of the medium, with order of magnitude 10^8 cm/sec). This is precisely the parameter that characterizes the role of the spatial dispersion. The smallness of kv_0/ω allows us to speak of weak spatial dispersion[9,10].

Weak dispersion corresponds to retaining in the expansion of the Fourier transform of the dielectric constant only terms quadratic in k. The dispersion of the transverse dielectric constant can be neglected, owing to its smallness, and the longitudinal dielectric constant can be taken in the form

$$\varepsilon^l(\omega, k) = \varepsilon(\omega) - \alpha k^2.$$

The quantity α, for example, in the model of free electrons, is of the form

$$\alpha = \tfrac{3}{5} v_0^2 \omega_{Le}^2/\omega^4,$$

where v_0 is the velocity of the electrons on the Fermi surface and ω_{Le} is the Langmuir frequency of the electrons.

The existence of waves of three polarizations, corresponding to an increase in the order of differential equations of the field, leads to the need for formulating an additional boundary condition or imposing some physical requirements which are equivalent to this condition[9-13]. In a macroscopic approach we can only postulate arbitrarily some boundary conditions, as was done in [11] in an analysis of radiation from a semi-infinite medium. The meaning of the corresponding conditions was not explained there.

A microscopic analysis shifts the emphasis to the model of the boundary. We can, for example, consider a surface from which the conduction electrons are reflected specularly (or diffusely). In such a case the problem is completely determined, and no conditions need be imposed (it must be noted that the condition of specular or diffuse reflection must be understood in the sense that the corresponding condition is formulated for the electron distribution function). Such a microscopic consideration of the problem of oblique incidence of an electromagnetic wave on a semi-infinite medium under conditions of specular reflection of the medium electrons from the surface was carried out in an earlier paper by the author [12] (Kaner and Yakovenko[13] considered later, by an analogous method, the transition radiation from a half space).

An analysis of the longitudinal and transverse fields excited in a medium has made it possible to formulate the following boundary conditions in the region of weak spatial dispersion[12]: the normal component of the radiation field, traveling inside the metal, is equal to zero on the surface. This boundary condition will be used from now on. For known ϵ and for known boundary conditions, the transition radiation is obtained as a result of solving the simple electrodynamic problem. In this case the difference in the derivation of the corresponding formulas is not the main point, so that we shall present only the final result.

We confine ourselves henceforth to the case of nonrelativistic particles and to a plate situated in a vacuum 2). Then we obtain for the intensity of radiation at an angle θ in the angle interval $d\theta$ and frequency interval $d\omega$ the formula

$$dW = (2e^2v^2/\pi c^3)\, d\omega d\theta \sin^3\theta \cos^2\theta\, |A(\omega, \theta)|^2, \tag{1}$$

2)The general case of different media and arbitrary velocities is considered in the appendix of the present article.

$$A(\omega,\theta)=\frac{\varepsilon}{\varepsilon-\alpha/v^2}\left((\varepsilon-1-\frac{\alpha}{v^2}\right)$$
$$\times\{(x+y)\,e^{-i\omega xd/c}+(x-y)\,e^{i\omega xd/c}$$
$$-2xe^{i\omega d/v}+z^{-1}\sin^2\theta\,[e^{-i\omega xd/c}-e^{i\omega d/v}+e^{i\omega zd/c}$$
$$-e^{i\omega d/v+i\omega zd/c-i\omega xd/c}]\}\{(x+y)^2e^{-i\omega xd/c}-(x-y)^2e^{i\omega xd/c}$$
$$+2z^{-1}\sin^2\theta\,[(y-x)\,e^{i\omega zd/c}+(x+y)\,e^{-i\omega xd/c}]$$
$$-z^{-2}\sin^4\theta\,[e^{2i\omega zd/c-i\omega xd/c}-e^{-i\omega xd/c}]\}^{-1},\qquad(2)$$

where

$$z=[\varepsilon(c^2/\alpha)-\varkappa^2]^{1/2},\qquad x=[\varepsilon-\varkappa^2]^{1/2},$$
$$\varkappa^2=\varepsilon_1\sin^2\theta\ \text{ and }\ \varepsilon_1=1,\ y=\varepsilon\cos\theta.$$

In comparing the results of (1) and (2) with the known formulas for transition radiation[6,7], obtained by neglecting spatial dispersion, the following features appear. First, a factor $\epsilon/(\epsilon-\alpha/v^2)$ appears, which can be rewritten in the form

$$\varepsilon/[\varepsilon-\alpha(k/\omega)^2],\qquad k^2=\omega^2\varkappa^2/c^2+\omega^2/v^2.\qquad(3)$$

This factor is due to the change in the Coulomb field of the charge. Since α is equal, roughly speaking, to the square of the Fermi velocity of the electrons ($v_0\sim10^8$ cm/sec), the order of magnitude of α/v^2 for 25-keV incident electrons is 10^{-4}. Since the imaginary part ϵ_2 of the dielectric constant of silver is of the order of 0.2—0.3 near λ = 3300 Å[14], it is obvious that any difference between (3) and unity could not be observed in the experiments of [1,2]. The effect can be made more noticeable by using beams of electrons of lower energy and using targets with a smaller imaginary part, for example alkali metals.

Another feature of (2) is the appearance of terms proportional to z^{-1} and z^{-2}. They turn out to be dependent on the boundary conditions. The boundary condition assumed above corresponds to a definite model of the boundary. In addition, it is obtained from an analysis of a problem with a single boundary, and is then applied to a plate. Therefore, for comparison, formulas analogous to (1) and (2) were obtained also by using other boundary conditions. A corresponding analysis shows that arbitrary variations in the boundary conditions lead to changes in the coefficients preceding the terms z^{-1} and z^{-2}, but the order of magnitude does not change thereby[3)]. On the other hand, under real conditions[14], z turns out to be quite large (namely, $z\sim10^2$), so that to disclose the role of terms proportional to z^{-1} the experimental accuracy must be of the order of 1%.

[3)]Compare, for example, the derivation obtained in [11] in an analysis of transition radiation from one boundary.

The use of alkali metals as targets is desirable in this case, too.

Summarizing the examination of transition radiation in a plate with account of spatial dispersion, we can state that both the specific features of the propagation of longitudinal plasma waves in matter and the specific boundary conditions relating them with the transverse electromagnetic waves are not realizable under the conditions of the experiment of [1,2]. More precise experiments are necessary to observe these features. In the analysis that follows we neglect the spatial dispersion, for which purpose it is sufficient to put α = 0 (then $|z|\to\infty$). In this case (1) and (2) go over into the well known formulas of transition radiation for a plate[6,7], which are free of any model assumptions whatever.

3. We shall show now that a complete interpretation of the experiments of [1,2] is contained in the usual theory of transition radiation. As was already noted, these experiments have been carried out in a spectral region close to the point of Wood transparency for silver. If we confine ourselves to not too large angles θ and to thicknesses ~ 500 Å (which corresponds, for example, to conditions of the second of the considered experimental investigations[2]), then it is easy to see that the parameter $xd\omega/c$, where $x=\sqrt{\epsilon-\sin^2\theta}$, is small. In such a case formula (1) simplifies still further and can be written in the form

$$dW=(2e^2v^2/\pi c^3)\,d\omega d\theta\sin^3\theta\cos^2\theta\sin^2(\omega d/2v)$$
$$\times\left|\frac{\varepsilon-1}{\varepsilon\cos\theta-i(\omega d/2v)(\varepsilon-\sin^2\theta+\varepsilon^2\cos^2\theta)}\right|^2.\qquad(4)$$

Inasmuch as the imaginary part of the dielectric constant of silver at $\lambda\approx3300$ Å is 0.2—0.3, it is easy to see that (4) gives the angular distribution of the experimentally observed radiation (Fig. 2 of the paper of Brown et al[2]), its spectral distribution and also the oscillations of the intensity of radiation as a function of the thickness of the film with period $\omega d/v$ (Steinmann's investigation [1]). The latter property has an interference character and does not depend on the specific radiation mechanism[4)].

[4)]We note that the sharp peak in the transition radiation in the plate is not the only effect near the transparency region. An analogous phenomenon arises when light is incident on a thin plate. Indeed, in the case of light which is polarized perpendicular to the plane of incidence (p — polarization), the energy coefficient of reflection for a thin film [15-16] in the region of small ϵ assumes the form

$$R_p=\frac{\omega_{Le}^2c^{-2}\omega^2d^2\sin^4\theta/4\cos^2\theta}{4(\omega-\omega_{Le})^2+(\omega_{Le}\varepsilon_2+\omega_{Le}\omega d\sin^2\theta/2c\cos\theta)^2}.$$

Returning to (4), it must be said that the approximation used in this derivation (thin film) is far from applicable for all film thicknesses and in the entire range of frequencies investigated in the experiment. Consequently, we plotted by numerical calculation the angular and spectral dependence, and also curves for the variation of the radiation intensity as a function of the film thickness, on the basis of the exact formulas of transition radiation [6,7], corresponding to formulas (A.1—A.3) of the appendix and neglecting spatial dispersion and for $\epsilon_1 = \epsilon_3 = 1$. The values of the optical constants were taken from experiment [14].

No absolute measurements of the intensity were made in [1,2], but the relations shown in Figs. 1—4 are in very good agreement with the experimental curves. To illustrate the dependence on the optical constants, the curves of Fig. 4 were plotted using data by Minor [18]. All this allows us to conclude that the experimentally observed electromagnetic radiation is the transition radiation which was predicted as long ago as in 1946 [5].

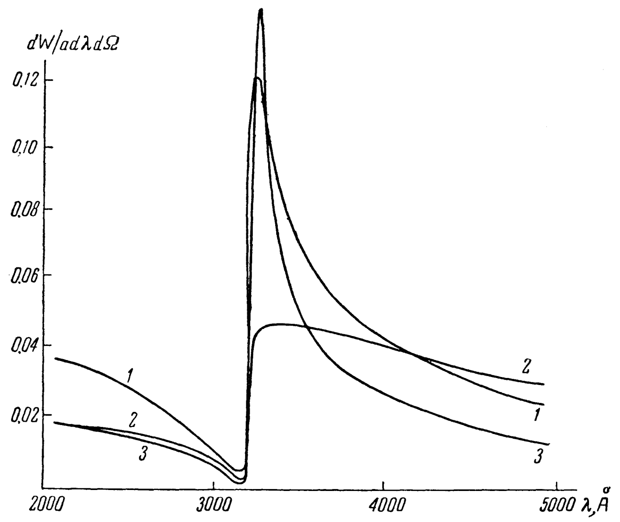

FIG. 1. Radiation intensity dW/Ad λdΩ against the wavelength λ, calculated for several values of the angle θ (curve 1—45°, curve 2—70°, curve 3—30°) for the following values of the parameters: d = 500 Å, v/c = 0.285 (coefficient a = $2e^2v^2/\pi c^3$).

The resonance character of this expression is obvious. An even sharper peak is possessed by the absorption coefficient $A_p = 1 - (R_p + T_p)$ where T_p is the transmission coefficient. This resonance effect was observed experimentally [17]. Finally, we note that the increase in the intensity of the transition radiation in the vicinity of the Wood transparency occurs also for a semi-infinite metal, although it is less sharply pronounced (a small parameter $\omega x d/c$ appears also for a film).

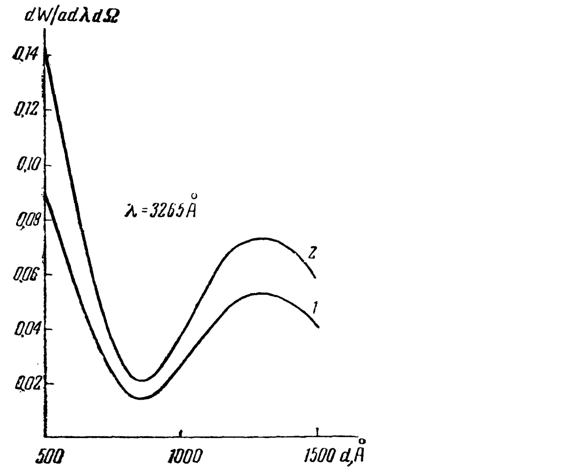

FIG. 2. Dependence of the radiation intensity dW/adλdΩ on the film thickness, calculated for incidence angles θ = 20° (curve 1) and θ = 30° (curve 2) (v/c and a are the same as in Fig. 1).

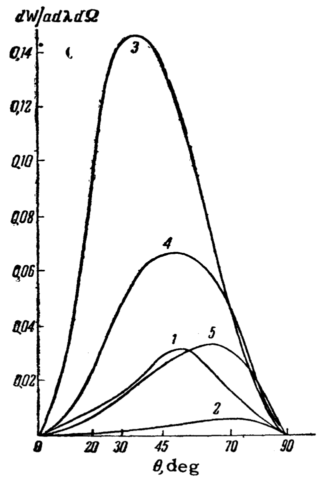

FIG. 3. Angular distribution of the radiation, calculated for several wavelengths λ: curve 1—2,390 Å, curve 2—3,150 Å, curve 3—3,265 Å, curve 4—3,540 Å, curve 5—4,965 Å (d,v/c, and a are the same as in Fig. 1).

Strictly speaking, however, we must estimate the contribution that can be made by other types of radiation. This includes, first, the ever present x-rays, and also luminescence (the conditions for the appearance of Cerenkov radiation for the considered energies of the electrons and for the spectral region are not satisfied). A characteristic feature of the transition radiation is total polarization in a plane passing through the trajectory of the incident particle in the radiation direction. This enables us to separate cathode luminescence and the glow of the gases adsorbed on the surface of the film, since these forms of radiation are not polarized. In addition, they should disclose a strong temperature dependence, whereas transi-

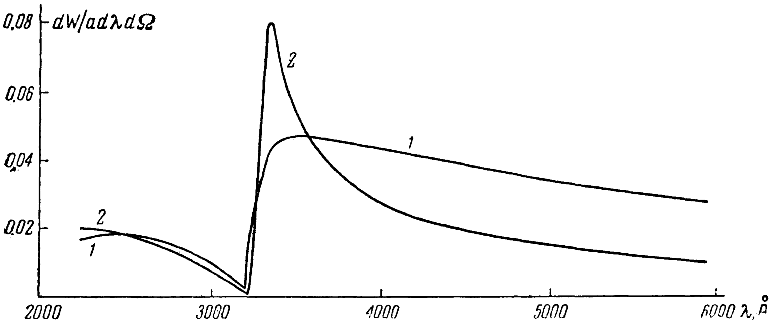

FIG. 4. Spectral dependence of the radiation, obtained using Minor's experimental data[18], Curve 1—70°, curve 2—30° (d, v/c, and a are the same as in Fig. 1).

tion radiation depends on the temperature only via the optical constants, i.e., quite weakly.

The question of separating the bremsstrahlung is more subtle, since, like the transition radiation, it is linearly polarized and one can speak only of quantitative differences in the degree of polarization. But the bremsstrahlung intensity, like that of luminescence, decreases with increasing energy of the particle beam, whereas for the transition radiation there is a linear increase. Further, in bremsstrahlung there is no angular dependence, but there is a characteristic dependence on the atomic number of the element.

New experiments[19-20] have greatly clarified and supplemented the previously obtained picture [1,2]. In the first investigation[19], targets of bulk Ag, Al, and Ni were used; in the second[2]—bulk samples of W, Ta, Mo, Ti, Pt, Ni, Cu, Al, Ag, and Cs, and also plates thicker than 1 micron. These experiments have established the existence of transition radiation, and have splendidly confirmed the main properties of this radiation, noted above, including the presence of high polarization ($\gtrsim$ 90%). The absolute measurements of the intensity[20] yielded for most metals good agreement with the theoretical quantities.

The experiments considered pertain directly to bulk samples, but they enable us to estimate the influence of extraneous effects. First, the surface conditions are important[19], for a poorly finished surface gives an appreciable luminescence glow. Clean well prepared samples, placed in vacuum, are almost free of luminescence. They are characterized by the absence of any noticeable temperature dependence in the region from 300 to 2000—3000°K[20]. The radiation intensity in the high-energy region (10—60 keV) increases linearly with the electron energy. The polarization for most metals is almost complete and is much higher than expected for bremsstrahlung[19,20].

With decreasing electron energy, deviations arise in the linear dependence on the energy, which reach an order of 30% at $E \cong 6$ keV[19]. The deviation from the transition-radiation theory at low energies is also manifest in the decrease of degree of polarization, the absolute intensity, and the angular distribution[19-21].

All this points to an increasing role of bremsstrahlung at low energies of the electron beam. An exact separation is difficult, since, for example, unpolarized radiation is partially due to depolarization of the transition radiation itself inside the metal, and is also produced on the irregularities on the sample surface.

We note that the presence of absorption singles out on the target surface an effective layer of ~ 100 Å, which can be the only source of bremsstrahlung. Therefore the laws and estimates obtained above remain in force also for thin metallic films.

Recently new experiments were performed[21] with silver films 500 Å thick, bombarded with 25-keV electrons. The radiation was highly polarized—on the order of 80—90%. The spectral distribution of the radiation at different angles duplicates with high accuracy the curves of Fig. 1.

Thus, we can state that the radiation observed in the experiment[1,2,21], which occurs when fast electrons pass through metallic films, can be sufficiently adequately explained by the theory of transition radiation.

We must dwell, finally, on one other question. Namely, in some investigations the maximum of the transition-radiation intensity near the region of Wood transparency of metals, as well as the absorption of light in thin films, is interpreted as the appearance of longitudinal field oscillations (plasmons), the frequency of which corresponds precisely to the occurrence of transparency of the film. This point of view is confirmed by the quantum theory of the interaction between an electron and radiation, a theory in which account is taken of plasma oscillations in thin films (see the papers by Kanazawa[22] and Matsudaira[23]). The overall result of such quantum analyses reduces actually to a microscopic derivation of results of the macroscopic theory. Namely, for example, a consistent microscopic analysis of transition radiation[5)] leads to results which are obtained from the macroscopic electrodynamics, the only difference being that the results of the microscopic theory are limited to a specific model, and are therefore narrower and naturally agree less with experiment.

APPENDIX

In many cases the need arises for investigating transition radiation which occurs when a particle passes through plane-parallel layers of matter with different dielectric constants. This must be done, in particular, for the case of a plate with oxidized surface. Then formula (1) of our article becomes unsuitable. The corresponding more general formula for the intensity can be obtained from the following expression for the tangential Fourier component of the radiation field in the space in front of the plate, occupying the region $(-d/2, +d/2)$ and bordering on media characterized by dielectric constants ϵ_1 and ϵ_3:

$$\begin{aligned} \mathbf{E}_t(k) = \frac{ei}{2\pi^2}\frac{\varkappa\omega x_1}{c\varepsilon_1 F}\{(y_2 - y_3)\,\beta e^{i\omega d y_2/c} \\ + \gamma\,(y_2 + y_3 + \varkappa^2/z)\,e^{-i\omega d y_2/c} \\ - 2\varepsilon\delta\,(y_2 + \varkappa^2/z + z^{-1}\varkappa^2 e^{i\omega d\,(z-y_2)/c})\,e^{i\omega d/v} \\ + (v/c)\,\mu z^{-2}\varkappa^4 e^{i\omega d\,(2z-y_2)/c} + z^{-1}\varkappa^2 \\ \times [v\mu\,(y_2 - y_3)/c + \beta]\,e^{i\omega dz/c}\}\,e^{-i\omega d\,(y_1 - c/v)/c}, \end{aligned} \qquad (A.1)$$

where

$$x_i = [\varepsilon_i - \varkappa^2]^{1/2},\quad y_i = x_i\varepsilon_2/\varepsilon_i$$

with i = 1, 2, 3 and

$$\begin{aligned} F = (y_2 - y_1)(y_2 - y_3)\,e^{i\omega d y_2/c} \\ - (y_2 + y_1 + \varkappa^2/z)(y_2 + y_3 + \varkappa^2/z)\,e^{-i\omega d y_2/c} \\ + z^{-1}\varkappa^2\,(2y_2 - y_1 - y_3)\,e^{i\omega dz/c} + z^{-2}\varkappa^4 e^{i\omega d\,(2z-y_2)/c}. \end{aligned} \qquad (A.2)$$

These formulas are similar in structure to those of Garibyan and Chalikyan[7], obtained for a plate in vacuum neglecting spatial dispersion. However (A.1) and (A.2) contain additional terms, which depend on the parameter z. Furthermore, the quantities β, γ, δ, and μ depend on α/c^2 and also on ϵ_1 and ϵ_3, and have the following form:

$$\begin{aligned} \beta &= \frac{\varepsilon + v\varepsilon_1 x_2/c}{(k^2 - \omega^2\varepsilon_1/c^2)\,\varepsilon_1} - \frac{\varepsilon\,(1 - \alpha/c^2) + v x_2 \varepsilon^l/c}{(k^2 - \omega^2\varepsilon/c^2)\,\varepsilon^l}, \\ \delta &= \frac{1 - v x_2/c}{(k^2 - \omega^2\varepsilon_3/c^2)\,\varepsilon_3} + \frac{1 - \alpha/c^2 - v x_3 \varepsilon^l/x_3 c}{(k^2 - \omega^2\varepsilon/c^2)\,\varepsilon^l}, \\ \gamma &= \frac{\varepsilon - v\,(x_2 - \varkappa^2/z)\,\varepsilon_1/c}{(k^2 - \omega^2\varepsilon/c^2)\,\varepsilon_1} - \frac{\varepsilon\,(1 - \alpha/c^2) - v\,(x_2 + \varkappa^2/z)\,\varepsilon^l/c}{(k^2 - \omega^2\varepsilon/c^2)\,\varepsilon^l}, \\ \mu &= \frac{\omega^2(\varepsilon_1 - \varepsilon)/c^2}{(k^2 - \omega^2\varepsilon_1/c^2)(k^2 - \omega^2\varepsilon/c^2)}. \end{aligned} \qquad (A.3)$$

The radiation field behind the plate is obtained from (A.1)–(A.3) by making the substitutions $v \to -v$, $\epsilon \to \epsilon_3$, and $\epsilon_3 \to \epsilon_1$.

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