Theory of spontaneous and stimulated emission of radiation by local plasmons on a surface with regularly distributed microirregularities

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It is shown that a fast charged particle moving above the surface of a substance covered by a two-dimensional grid of microirregularities emits intense coherent radiation in the form of photons generated as a result of resonant excitation of local surface plasmons. A theory of spontaneous and stimulated emission of radiation is developed for the case when a dipole resonance of plasmon excitation is important. It is also shown that under certain conditions relativistic particles can exchange energy effectively with an external electromagnetic wave via local surface plasmons.

1. INTRODUCTION

Collective oscillations of an electron plasma in material microparticles, excited by an electromagnetic wave or fast electrons, are the origin of many interesting effects which are currently under close scrutiny (see, for example, the reviews in Refs. 2 and 5). It is shown in Ref. 1 that a local electromagnetic field near the surface of a particle of submicron size may be several orders of magnitude greater than the field of the incident wave if the wave frequency is in resonance with one of the natural frequencies of oscillations of an electron plasma in microparticles. Enhancement of this local field results in turn in a considerable amplification of a number of electro-magnetic processes that occur on metal surfaces or in metal colloids.[2–6] In particular, the cross sections representing the Raman scattering of light and luminescence of adsorbed molecules rise by several orders of magnitude.[2,5] Observations have also been reported of enhancement of nonlinear optical phenomena such as the two-photon luminescence of molecules and the generation of higher harmonics when laser light is reflected by irregularities of metal surfaces.[2,3]

The discovery of considerable enhancement of several electromagnetic processes on metal surfaces has increased the interest in other phenomena involving local surface plasmons. They include the appearance of a "dip" in the coefficient representing reflection of light by metal surfaces when the frequency of light is close to the frequency of local plasmons,[4,6] radiative damping of local plasmons excited by fast electrons,[8–12] and evidently also Wood's anomalies,[7] i.e., the appearance of dips in the case of diffraction by gratings with sufficiently deep grooves, analogous to dips in the specular reflection of light. Some of the observed effects[11,12] have not been explained convincingly for a long time, but they were reviewed in Ref. 10 allowing for the role of local surface plasmons. It was shown there that the unusually high intensity and other properties of optical radiation discovered when metal surfaces were bombarded with fast electrons at grazing angles can be explained fully by radiative decay of local plasmons excited by electrons located in surface irregularities.

Such microirregularities of random surfaces represent particles which differ greatly in size and shape.[8] Since it is these parameters of the particles that determine, in conjunction with the permittivity of the material, the resonance frequencies of local surface plasmons, the plasmon excitation spectrum is characterized by a relatively large inhomogeneous width. On the other hand, microlithography of surfaces can produce[2,5,13–15] regular two-dimensional grids of submicron particles which are of the same size, shape, and orientation. In such cases it is possible, firstly, to minimize the inhomogeneous broadening of plasmon resonances and thus enhance greatly the local field effects. Secondly, the additional effects associated with the translation symmetry of the surface are also observed. In particular, it is reported in Ref. 15 that a grid of ellipsoidal projections can be used to generate the second harmonic of laser light not only (as usual) at an angle close to the reflection angle, but also at an angle corresponding to the diffraction of the second harmonic by a grating. Another experiment[13] has shown how a two-dimensional array of disk-shaped AlGaAs particles on the surface of GaAs affects the *hf* conductivity in a magnetic field, associated with the damping of surface plasmons.

The regular nature of such microirregularities should be manifested particularly in the excitation of local plasmons by a fast charged particle and their subsequent radiative decay, since phase relationships between the individual radiators are important in this process. We shall develop a theory of coherent emission of radiation from local surface plasmons excited by a relativistic electron and we shall formulate the conditions under which considerable enhancement of the emission is possible compared with incoherent radiation emitted by random surfaces.[8–10] We shall discuss the processes of stimulated coherent emission and absorption of photons when, in addition to an electron beam, an electromagnetic wave with the frequency of a plasmon resonance is traveling at an angle to the surface. We shall show that in this case there is an exchange of energy between the electron beam and the electromagnetic wave because of plasmon resonances.

2. FREQUENCY–ANGULAR DISTRIBUTION OF RADIATION EMITTED BY AN ARRAY OF PLASMONS EXCITED BY A FAST ELECTRON

We assume that a relatively flat surface carries a two-dimensional array or grid in the form of separate microparticles with specific shapes, dimensions, and orientations relative to the normal to the surface. We also assume that a fast relativistic electron is traveling in vacuum at a velocity $\mathbf{v}$ and at a distance $\mathbf{x}$ from the surface. The fast electron excites oscillations of the electron density in the particles and these

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in turn become sources of electromagnetic radiation in the form of transverse waves.

The energy of such radiation traveling in vacuum, measured within a frequency interval $d\omega$ and a solid angle $d\Omega$, can be found using the familiar expression taken from classical electrodynamics:

$$\frac{d^2W}{d\omega d\Omega}=\frac{\omega^2}{4\pi^2c^3}\,|\,[\mathbf{n}\mathbf{j}(\mathbf{k},\omega)]\,|^2. \tag{1}$$

Here $\mathbf{n}$ is a unit vector along the direction of the emitted radiation, $\mathbf{k}=\mathbf{n}(\omega/c)$ is the wave vector, and $\mathbf{j}\,(\mathbf{k},\omega)$ is the space–time component of the Fourier current induced by a relativistic electron in the particles on the surface. We assume that the dimensions of these particles are small compared with the radiation wavelength $\lambda=2\pi c/\omega$ and the main periods of the two-dimensional array are comparable with λ, so that the distance between the neighboring particles is much greater than their dimensions. In this case the radiation of the dipole nature and the interaction between neighboring dipoles can be ignored.[2] Consequently, the Fourier component of the induced current can be represented in the form

$$\mathbf{j}(\mathbf{k},\omega)=i\omega\sum_R \mathbf{d}(\omega;x,\mathbf{R})\,e^{-i\mathbf{kR}}, \tag{2}$$

where $\mathbf{d}(\omega;x,\mathbf{R})$ is the dipole moment induced in a single particle and dependent on the distance x between the fast electron and the surface and on the coordinate $\mathbf{R}$ of the "site" in the two-dimensional array (lattice) on the surface.

The dipole moment $\mathbf{d}(\omega;x,\mathbf{R})$ can be expressed in terms of the corresponding Fourier time components $\mathbf{E}(\omega,\mathbf{r})$ of the electron field. We choose a cylindrical coordinate system with the z axis along the electron velocity $\mathbf{v}$ and use $\boldsymbol{\rho}$ to denote the radius vector perpendicular to $\mathbf{v}$. Then the components of the electron field of frequency ω can be described (see, for example, Ref. 10)

$$\begin{aligned} E_z(\omega,\mathbf{r})&=-\frac{2ie\omega}{v^2\gamma^2}K_0\left(\frac{\rho\omega}{v\gamma}\right)e^{i\omega z/v},\\ \mathbf{E}_\perp(\omega,\mathbf{r})&=\frac{2e\omega}{v^2\gamma}K_1\left(\frac{\rho\omega}{v\gamma}\right)e^{i\omega z/v}\frac{\boldsymbol{\rho}}{\rho}. \end{aligned} \tag{3}$$

Here, $\mathbf{r}=\{\boldsymbol{\rho},z\}$; $\gamma=(1-v^2/c^2)^{-1/2}$ is the Lorentz factor of a fast electron; $K_0(\xi)$ and $K_1(\xi)$ are modified Bessel functions of the second kind: E_z and $\mathbf{E}_\perp$ are the parallel and perpendicular (relative to the velocity) Cartesian resolvents of the Fourier component of the field; $\rho(x^2+y^2)^{1/2}$. In the case of sufficiently small islands $\xi=\rho\omega/v\gamma\ll 1$, the behavior of the modified Bessel functions is described by the familiar expressions

$$K_0(\xi)\approx-\ln(\xi/2),\quad K_1(\xi)\approx 1/\xi.$$

An estimate of the spatial derivatives of the field (3), based on these expressions, show that the variation of the field of the electron within a particle can be ignored if the longitudinal a_z and transverse $a_\perp$ dimensions of the particle satisfy the conditions

$$a_z\ll v/\omega,\quad a_\perp\ll v\gamma/\omega.$$

Under these conditions we can assume that a single particle experiences the electric field of the electron of Eq. (3) which alternates in time and is practically constant in space. We assume that the particle material is characterized by a frequency-dependent permittivity $\varepsilon(\omega)$ and the particles are not too small ($a>v_F/\omega\sim 10$ Å), so that we can ignore the influence of the particle dimensions on their dielectric properties. Consequently, the dipole moment induced by the field of the electron within the particle can be calculated by analogy with the corresponding problem in electrostatics. If, for the sake of simplicity, we assume that the particle has three symmetry axes, which coincide with the axes of the selected coordinate system, we can represent the components of the dipole moment in the form

$$\begin{aligned} d_x&=V\alpha_x(\omega)E_\perp(\omega,r)\,(x/\rho),\\ d_y&=V\alpha_y(\omega)E_\perp(\omega,r)\,(y/\rho),\\ d_z&=V\alpha_z(\omega)E_z(\omega,r). \end{aligned} \tag{4}$$

The following notation is introduced above: V is the particle volume; α_x, α_y, and α_z are the components of the polarizability tensor of the particle at a frequency ω, corresponding to the excitation of dipole oscillations along the coordinate axes. The components of the polarizability tensor of the particle can be expressed as follows in terms of electric susceptibility $\chi(\omega)=\varepsilon(\omega)-1$ and in terms of the depolarization coefficients n_1, n_2, and n_3 depending only on the particle shape:

$$\begin{aligned} \alpha_x&=\frac{1}{4\pi}\frac{\chi(\omega)}{1+n_1\chi(\omega)},\\ \alpha_y&=\frac{1}{4\pi}\frac{\chi(\omega)}{1+n_2\chi(\omega)},\\ \alpha_z&=\frac{1}{4\pi}\frac{\chi(\omega)}{1+n_3\chi(\omega)}. \end{aligned} \tag{5}$$

The depolarization coefficients satisfy the relationship

$$n_1^2+n_2^2+n_3^2=1$$

and can be represented analytically in the form of isolated particles with simple shapes[2,4] and in the case of microirregularities of more complex shape (for example, in the case of hemispherical projections on a flat surface), the value of $\alpha(\omega)$ can be calculated by numerical methods.[16] It follows from Eqs. (4) and (5) that the dipole moment of a particle considered as a function of the frequency is characterized by resonance at frequencies ω_r for which the real parts of the denominators in the system (5) vanishes. The amplitudes and widths of these resonances are then governed by the ratio χ'/χ'' of the real and imaginary parts of the electric susceptibility $\chi(\omega)$ at the resonance frequencies. It therefore follows that strong excitation of local surface plasmons can be expected if the damping at the resonance frequencies is sufficiently weak: $\chi''\ll|\chi'|$.

In the case of a rectangular array of microirregularities with the periods d_1 and d_2 the components of the induced current can be written in the form

$$\begin{aligned} j_x&=\frac{2ie\omega^2}{v^2\gamma}V\alpha_x(\omega)\sum_{\mu=-\infty}^{\infty}\sum_{\nu=0}^{N-1}K_1\left(\frac{\rho_{\mu\nu}}{\rho_0}\right)\frac{x}{\rho_{\mu\nu}}\exp(i\Phi_{\mu\nu}),\\ j_y&=\frac{2ie\omega^2}{v^2\gamma}V\alpha_y(\omega)\sum_{\mu=-\infty}^{\infty}\sum_{\nu=0}^{N-1}K_1\left(\frac{\rho_{\mu\nu}}{\rho_0}\right)\frac{y}{\rho_{\mu\nu}}\exp(i\Phi_{\mu\nu}),\\ j_z&=\frac{2e\omega^2}{v^2\gamma^2}V\alpha_z(\omega)\sum_{\mu=-\infty}^{\infty}\sum_{\nu=0}^{N-1}K_0\left(\frac{\rho_{\mu\nu}}{\rho_0}\right)\exp(i\Phi_{\mu\nu}). \end{aligned} \tag{6}$$

where $\rho_0 = v\gamma/\omega$,

$$y_{\mu\nu}=\nu d_1 \sin\psi+\mu d_2 \cos\psi, \qquad z_{\mu\nu}=\nu d_1 \cos\psi+\mu d_2 \sin\psi$$

are the coordinates of the sites in this array or lattice,

$$\rho_{\mu\nu}=(x^2+y_{\mu\nu}^2)^{1/2}, \qquad \Phi_{\mu\nu}=\omega c^{-1}[(\beta^{-1}-n_z)z_{\mu\nu}-n_y y_{\mu\nu}],$$

ψ is the angle between the electron velocity and one of the sides of the array (along which the period is d_1),

$$n_y=\sin\theta\sin\varphi, \qquad n_z=\cos\theta$$

are the projections of a unit vector oriented parallel to the direction of emission and resolved along the coordinate axes, and N is the number of sites in the array or lattice along the direction of the electron velocity. The quantity $\Phi_{\mu\nu}$ represents the phase of the radiation field in the case of a single dipole and the other factors in the sums represent its amplitude. the substitution of Eq. (6) into Eq. (1) yields the spectra and angular distribution of the radiation.

We now consider some cases of practical importance in which the analysis of the general result can be simplified greatly. We assumed that the direction of motion of an electron is close to the direction of one of the sides of the array ($\psi \ll \beta^{-1} - n_z$). In this case we can assume that $y_{\mu\nu} \approx \mu d_2$, $z_{\mu\nu} \approx \nu d_1$. Consequently, we can sum over the index ν in Eq. (6), using the identity

$$\sum_{\nu=0}^{N-1} e^{i\nu\xi}=(1-e^{iN\xi})/(1-e^{i\xi}),$$

$$\xi=(\omega d_1/c)(\beta^{-1}-\cos\theta), \qquad \beta=v/c. \tag{7}$$

Further simplification is possible in the ultrarelativistic limit $\gamma \gg 1$. In this limit the amplitude of the radiation field includes a comparable contribution of the fairly large number ($\sim\gamma$) of sites with $y_{\mu\nu} \lesssim \lambdabar\gamma$, so that the summation in the system (6) over the index μ can be replaced by integration with respect to the quasicontinuous quantity $y_{\mu\nu} d_2$. In this integration we can use the relationships[17]

$$\int_{-\infty}^{\infty} K_1(\alpha\rho)e^{i\delta y}\frac{x}{\rho}\,dy=\frac{\pi}{\alpha}\exp[-x(\alpha^2+\delta^2)^{1/2}],$$

$$\int_{-\infty}^{\infty} K_1(\alpha\rho)e^{i\delta y}\frac{y}{\rho}\,dy=\frac{i\pi}{\alpha}\frac{\delta}{(\alpha^2+\delta^2)^{1/2}}\exp[-x(\alpha^2+\delta^2)^{1/2}],$$

$$\rho=(x^2+y^2)^{1/2}, \qquad \alpha=1/\rho_0, \qquad \delta=-(\omega/c)n_y. \tag{8}$$

In the limit $\gamma \gg 1$ the component z of the induced dipole moment is γ times less than its transverse components d_x and d_y, so that we can ignore the excitation of the dipole oscillations along the direction of the electron velocity. It should be noted also that according to Eq. (8) the phases of the x and y components of the current described by Eqs. (6) are shifted relative to one another by $\pi/2$ and, therefore, the cross terms in Eq. (1) proportional to $\mathrm{Re}\, j_x j_y^*$ vanish. Consequently, the frequency and angular distribution of the radiation energy in a distance $L = Nd_1$ traveled by the electron above the surface becomes quite simple:

$$\frac{d^2W}{d\omega d\Omega}=\frac{e^2\omega^4V^2}{c^5d_2^2}\exp\left[-\frac{2x}{\lambdabar}(n_y^2+\gamma^{-2})^{1/2}\right]$$
$$\times\left[|\alpha_x(\omega)|^2(1-n_x^2)+|\alpha_y(\omega)|^2\frac{n_y^2(1-n_y^2)}{n_y^2+\gamma^{-2}}\right]S(\omega,\theta), \tag{9}$$

where

$$S(\omega,\theta)=\sin^2(N\xi/2)/\sin^2(\xi/2)$$

is the interference factor for the radiation emitted by a chain of N dipoles located along the electron path. If the number of dipoles is large, $N \gg 1$, we can present the above results in the approximation form

$$S(\omega,\theta)\approx\sum_{n=1}^{\infty}\frac{\sin^2(N\xi_n)}{\xi_n^2},$$
$$\xi_n=\frac{\omega d_1}{2c}(\beta^{-1}-\cos\theta)-\pi n. \tag{10}$$

We also note that

$$\lim_{N\to\infty}\frac{\sin^2(N\xi_n)}{N\xi_n^2}=\pi\delta(\xi_n), \tag{10'}$$

where $\delta(\xi_n)$ is the Dirac function.

The frequency and angular distribution of the radiation energy given by Eq. (9) as a function of the polar angle θ at a fixed frequency ω or, conversely, as a function of the frequency at a fixed angle θ has sharp maxima when the condition $\xi_n(\omega,\theta) = 0$ is satisfied, which is a consequence of the coherence of the radiators. The width of this maxima $\Delta\xi_n$ on the ξ_n scale is governed by the number of periods N along the electron path: $\Delta\xi_n \approx \pi/2N$. The magnitude of the frequency–angular density of the radiation energy at the maxima is proportional to the square of the number of periods, to the square of the polarizability of the microparticles, and (subject to the condition $d_2 \sim \lambdabar^2$) to the square of the ratio of the microparticle volume V to the quantity $\lambdabar^3$.

As pointed out above, the polarizabilities $\alpha_x(\omega)$ and $\alpha_y(\omega)$ have resonances at frequencies ω_r where the real part of the susceptibility satisfies the conditions $\xi'(\omega_r^{(1)}) = -n_1^{-1}$ or $\chi'(\omega_r^{(2)}) = -n_2^{-1}$, so that the radiation is emitted mainly at angles θ_r corresponding to such resonances: $\xi_n(\omega_r,\theta_r) = 0$. If the susceptibility in Eq. (5) is expanded in powers of the small parameter $\Delta\omega = \omega - \omega_r$, we obtain expressions for $|\alpha_x(\omega)|^2$, $|\alpha_y(\omega)|^2$ near the resonance frequencies which yield

$$|\alpha(\omega)|^2\approx\frac{\chi'^4}{(4\pi)^2}\left[\left(\frac{d\chi'}{d\omega}\right)^2(\Delta\omega)^2+\chi''^2\right]^{-1}$$

where χ', χ'', and $d\chi'/d\omega$ are taken at the resonance frequency ω_r. It is assumed here that the absorption is relatively weak: $\chi'' \ll |\chi'|$. Therefore, to lowest order the line profile representing excitation of a local plasmon is Lorentzian with the following width at a half-amplitude:

$$\Gamma=2\chi''/(d\chi'/d\omega).$$

Although this width is small compared with the resonance frequency ω_r, if the structure is sufficiently long ($N \gtrsim 10^2$) it exceeds the width determined by the interference between dipoles ($\Delta\omega/\omega \approx 1/2Nn$). Therefore, the total width of the distribution of the radiation in the polar angle around θ_r is governed by the spectral width Γ of the plasmon resonance.

If the resonance frequency ω_r of a local plasmon is sufficiently low compared with the frequencies of interband transitions, it follows that the electric susceptibility of the metal can be described by the Drude expression

$$\chi(\omega)=-\frac{\omega_p^2}{\omega^2}+i\frac{\omega_p^2\tau}{\omega},$$

where ω_p and τ represent respectively the plasma frequency and the relaxation time of carriers. In this approximation the resonance frequencies and the widths of the resonances are given by the simple relationships:

$$\omega_r^{(i)}=\omega_p n_i^{1/2},\quad \Gamma_i=\omega_p^2\tau n_i.$$

The distribution of the radiation in the azimuthal angle φ depends on the distance between the moving electron and the plane of the surface and on the polar angle of the emission of radiation. If the distance x is large compared with λ and the polar angle is not too close to zero or to π ($\sin\theta\gg\gamma^{-1}$), the effective values of φ are small: $\varphi_{\rm eff}\sim\gamma^{-1}$. Subject to these restrictions we can now integrate Eq. (9) with respect to the solid angle by an analytic procedure. We represent $n_y=\sin\theta\sin\varphi$ in its approximate form so that $\sin\varphi\approx\varphi$ and extend the limits of integration with respect to φ to infinity. We can then use the relationship

$$\int_0^\infty \exp[-X(t^2+1)^{1/2}]dt=K_1(X),\tag{11}$$

where $t=\gamma\varphi|\sin\theta|$ and $X=2x/\lambda\gamma$, which is the familiar[17,18] integral representation of the modified Bessel function. The other integral which is obtained can be represented in the form

$$\int_0^\infty \exp[-X(t^2+1)^{1/2}]\frac{dt}{t^2+1}=Ki_1(X),$$
$$Ki_1(X)=\int_X^\infty K_0(p)\,dp.\tag{12}$$

The relationship given by Eqs. (12) is obtained by integrating Eq. (11) twice with respect to the parameter X. The functions $K_1(X)$ and $Ki_1(X)$ are tabulated in, for example, Ref. 18. In the case of large parameters $X\gtrsim 1$ they decrease exponentially.[18] Further integration with respect to the polar angle θ carried out using Eq. (10) gives

$$\frac{dW}{d\omega}=\frac{2\pi e^2\omega^3V^2L}{c^4S^2\gamma}\sum_{n=1}^{[2d_1/\lambda]}\left[1-\left(1-\frac{\lambda n}{d_1}\right)^2\right]^{-1/2}$$
$$\times\left\{|\alpha_x(\omega)|^2\left(1-\frac{\lambda n}{d_1}\right)^2K_1(X)+|\alpha_y(\omega)|^2[K_1(X)-Ki_1(X)]\right\},\tag{13}$$

where $[x]$ is the integral part of x; $S=d_1d_2$ is the area of the surface per one microirregularity.

The spectral density of the radiation energy given by Eq. (13) for the motion of an electron above a regular structure on the surface of a metal is on the order of the value obtained in Ref. 10 for a random distribution of surface microirregularities with an appropriate distribution density. On the other hand, along certain directions the coherence of the radiation may enhance the spectral and angular density of the radiation emitted by the regular structure [Eq. (9)] by several orders of magnitude.

The relatively high spectral and angular density and the monochromaticity of the coherent radiation emitted by local plasmons is of interest in connection with the possibility of laser acceleration of particles and generation of laser radiation, which will be discussed below.

3. INTERACTION BETWEEN AN ELECTROMAGNETIC WAVE AND AN ELECTRON BEAM TRAVELING ABOVE A TWO-DIMENSIONAL ARRAY

We assume that a beam of electrons moving above an array of microirregularities meets an electromagnetic wave of frequency corresponding to resonant excitation of local plasmons at an angle θ to the direction of the electron velocity, and let us assume that this angle is close to the angle of coherent spontaneous emission [see Eq. (10)]. We shall show later that such a wave may be amplified by the process of stimulated emission or it may be damped, transferring its energy to the moving electrons, depending on its relationship to the electron beam.

When the wave loses energy, we have to find the increase ΔE in the energy of an electron after it passes above a structure of length L at a distance x from the surface. In the other case we have to find the gain G, defined as the ratio of the increase in the density of the energy flux carried by the incident wave to its initial energy density. Their are several ways of calculating the gain when the active medium is an electron beam interacting with matter or with an external field [see, for example, Refs. 19 and 20]. The simplest of these is based on allowance for the influence of the quantum recoil effect, which accompanies the emission or absorption of the photon by an electron, on the condition specifying coherence of the radiation. Let us assume that $w^{\rm sp}(\omega,\mathbf{n})$ denotes the frequency–angular probability density of coherent spontaneous emission of a photon by an electron in a distance L. Since the photon energy is considerably less than the electron energy, it follows that the lowest order in the parameter $\hbar\omega/E$, i.e., in the classical approximation, the quantity $w^{\rm sp}(\omega,\mathbf{n})$ is found by the dividing the right-hand side of Eq. (9) by the photon energy $\hbar\omega$. We shall now assume that $j_{\rm ph}(\omega,\mathbf{n})$ denotes the frequency–angular density of the energy flux carried by the external electromagnetic wave. Then, the corresponding density of the probability of simulated emission $w^{\rm em}$ and absorption $w^{\rm ab}$ can be related to $w^{\rm sp}$ by the familiar expressions:

$$w^{\rm ab}=w^{\rm em}=\frac{8\pi^3c^2}{\hbar\omega^3}J_{\rm ph}w^{sp}.$$

The energy ΔE acquired by an electron is proportional to the difference $w^{\rm ab}-w^{\rm em}$ and, consequently, a nonvanishing effect ($\Delta E\neq 0$) appears only in the next order in the small parameter $\hbar\omega/E$. Inclusion of the quantum corrections modifies the coherence condition for the emission and absorption in such a way that the quantity ξ_n [see Eq. (10)] experiences different increments: $\delta\xi_n^{(-)}$ in the case of absorption. This splits the spectral line for a fixed angle θ. On the other hand, the influence of the quantum effects on the amplitude of the frequency–angular distribution of the radiation observed for $\xi_n\approx 0$ can be ignored. We shall introduce $\Delta\xi_n=\delta\xi_n^{(+)}-\Delta\xi_n^{(-)}$ as the difference between quantum increments in the quantity ξ_n. Then, the increment in the electron energy due to the absorption of external radiation can be represented in the form

$$\Delta E=2\pi\lambda^2 j_{\rm ph}\frac{\partial w^{sp}}{\partial \xi_n}\Delta\xi_n, \tag{14}$$

where $J_{\rm ph}$ is the density of the energy flux of the external wave which is assumed to have spectral and angular widths small compared with the corresponding quantities in the case of spontaneous emission.

If the right-hand side of Eq. (14) is negative, then the wave is amplified as a result of stimulated emission. The gain G is found from Eq. (14) as follows. Let us assume that $j_e(x)$ is the density of the electron flux which may depend on the distance from the surface of the substance, $\sigma = L_y D$ is the cross section of the photon beam, L_y is the dimension of this beam along the y axis, and D is the other side of the rectangular beam cross section. The optimum conditions for magnification are obtained when the linear dimensions of the electron and photon (electromagnetic) beams coincide along the y axis and the length of the region of overlap of the beams along the z axis is equal to the total length of the structure, i.e., $D = L|\sin\theta|$. Multiplying the right-hand side of Eq. (14), by the electron flux $J_e(x) = j_e(x)\Delta x$, where $\Delta x \ll \lambda\gamma$ is the width of the electron beam, and dividing next by the photon beam cross section, we find the magnitude of the increment in the photon energy flux. Consequently, the gain becomes

$$G=-\frac{\Delta E}{J_{\rm ph}}j_e(x)\frac{\Delta x}{L|\sin\theta|},\qquad D>\Delta x. \tag{14'}$$

It follows from Eq. (9) that the probability of spontaneous emission and, consequently, the gain described by Eq. (14) depend exponentially on the distance x between the electron beam and the surface and the dependence. Therefore, if the dimensions of the electron beam along the x axis are not sufficiently small compared with $\lambda\gamma$, it is necessary to carry out integration in Eq. (14') over the transverse dimensions of the beam.

We can find the quantum shift of ξ_n without solving the problem of emission again. This shift can be deduced quite readily from the conservation laws.

We use δE and $\delta\mathbf{p}$ for the increment in the energy E and in the momentum $\mathbf{p}$ of a fast electron; we use ω and $\mathbf{k}$ to denote the energy and momentum of the photon. The laws of conservation of energy and momentum (accurate to within the reciprocal lattice vector $\mathbf{K}$ parallel to the surface) can be written in the form (on the assumption that $\hbar = m = c = 1$)

$$\delta E=\pm\omega,\ \delta\mathbf{p}=\pm(\mathbf{k}+\mathbf{K}).$$

The upper sign corresponds to the absorption and the lower to the emission of a photon by a fast electron, accompanied by the transfer of momentum $\mp\mathbf{K}$ to the lattice of local plasmons. Since the change in the electron energy is small, it can be represented in the form

$$\delta E=\frac{\partial E}{\partial p_i}\delta p_i+\frac{1}{2}\frac{\partial^2 E}{\partial p_i \partial p_j}\delta p_i\delta p_j,$$

$$\frac{\partial E}{\partial p_i}=v_i,\qquad \frac{\partial^2 E}{\partial p_i\partial p_j}=\frac{E^2\delta_{ij}-p_ip_j}{E^3},$$

where v_i is the Cartesian components of the electron velocity; $i,j = 1,2,3$. Therefore, the condition of coherence in the case of absorption and also emission (subject to quantum corrections) becomes

$$\omega-(\mathbf{k}+\mathbf{K})\mathbf{v}\mp\frac{1}{E}(K^2+2\mathbf{kK})=0.$$

Bearing in mind that the above case of spontaneous emission described by Eq. (9) corresponds to $\mathbf{K}\cdot\mathbf{v} = 0$, where $K = 2\pi n/d_1$, we find that in this case the difference between the shifts of ξ_n is

$$\Delta\xi_n=\pi n\frac{\hbar\omega}{E}\left[\frac{n\lambda}{d_1}+2\cos\theta\right]. \tag{15}$$

Next, using Eqs. (9), (14), and (15), we obtain the increment in the electron energy in the field of the wave because of the interaction with nth harmonic in the form

$$\Delta E=-2^5\pi^6 J_\Phi\frac{e^2}{cE}\left(\frac{V}{S}\right)^2\left(\frac{L}{\lambda}\right)^3\exp\left[-\frac{2x}{\lambda}(n_y^2+\gamma^{-2})^{1/2}\right]$$
$$\times\left[|\alpha_x(\omega)|^2(1-n_x^2)+|\alpha_y(\omega)|^2\frac{n_y^2(1-n_y^2)}{n_y^2+\gamma^{-2}}\right]$$
$$\times\frac{n\lambda}{d_1}\left[\frac{n\lambda}{d_1}+2\cos\theta\right]f(\xi_n). \tag{16}$$

Here the notation is as follows:

$$\xi_n=\frac{\pi Lc}{\lambda}\left(\frac{1}{v}-\frac{1}{v_n}\right),\qquad f(\xi)=\frac{\partial}{\partial\xi}\left(\frac{\sin\xi}{\xi}\right)^2, \tag{17}$$

and $v_n = c(n\lambda/d_1 - \cos\theta)$ is the phase velocity of the surface wave. The relevant gain is found from Eqs. (16) and (14). The electron energy increment and the gain are proportional to the probability of spontaneous emission in the direction of the coherent maximum and to the derivative of the profile of the spontaneous emission line, which is typical of the low-gain approximation. The gain maximum occurs at $\xi_n \approx -0.4\pi$, where $v > v_n$ and $f(\xi_n) \approx 0.54$. The positive value of $\xi_n \approx 0.4\pi$ for $v < v_n$ corresponds to the maximum of the electron energy increment [Eq. (14)].

It is clear that the validity of Eq. (16) requires that the variation of ξ_n due to a change in the parameters occurring in this quantity should not exceed $\approx\pi/2$. Differentiating Eq. (17) with respect to these parameters, we obtain the following restrictions on the monochromaticity and collimation of the electron beam of photons carried by the external wave, and also on possible deviations of the surface structure from strict periodicity:

$$\frac{\Delta\lambda}{\lambda}\leqslant\frac{1}{2nN},\qquad \frac{\Delta d_1}{d_1}\leqslant\frac{1}{2nN},$$
$$\frac{\Delta\gamma}{\gamma}\leqslant\frac{\lambda\gamma^2}{2L},\qquad \Delta\theta\leqslant\frac{\lambda}{2L|\sin\theta|}. \tag{18}$$

In the ultrarelavistic case ($\gamma \gg 1$) the less stringent of these conditions is that imposed on the spread in the electron energies. On the other hand, when the number of the structure periods N is large, it is necessary to ensure a high degree of periodicity of the structure and a high degree of collimation of both beams.

The mean square deviations of the structure period $\langle\Delta d_1^2\rangle$ and of variation of the shape, volume, and orientation of the particles affect the probability of coherent emission [Eq. (9)], but do not alter significantly the coherence condition $\xi_n(\omega,\theta) = 0$ [see Eq. (10)], which is related to the existence of long-range order in the array.

By analogy with the scattering of neutrons or x-ray phonons in crystals, we can show that the mean-square deviations of the array period give rise, in Eq. (9), to a factor e^{-W} of the Debye–Waller type, where $W=(2\pi n/d_1)^2\langle\Delta d_1^2\rangle$ and the quantity $\langle\Delta d_1^2\rangle$ then plays the role of the square of the amplitude of thermal vibrations of the lattice. Variations of the shape, volume, and orientation of the particles relative to the surface also influence the probability of coherent emission, which is analogous to the influence of the isotopic composition of the nuclei on the coherent scattering of neutrons in a crystal. In general, the quantities V, α_x, α_y must be understood to represent average values. The averaging does not result in a significant reduction in the polarizability α if the scatter of the plasmon resonance frequencies caused by variation of the particle phase does not exceed the width of the resonances. Hence, the following restrictions should be imposed on variation of the depolarization coefficients [see Eq. (5)]:

$$\Delta n/n \leqslant \chi''/\chi'$$

In the case of an ellipsoid elongated along the normal to the surface the depolarization coefficient parallel to the longest of the axes of the ellipsoid is[2,5]

$$n_1=\frac{1-e^2}{e^3}\left(\frac{1}{2}\ln\frac{1+e}{1-e}-e\right),\quad e=\left(1-\frac{b^2}{a^2}\right)^{1/2},$$

where $b/a \leqslant 1$ is the ratio of the minor to the major semiaxis. Since in cases of practical importance we can assume[10] that $\chi'/\chi''\sim 10$ applies in the plasmon resonance region, it follows that deviations of the eccentricity e of the ellipsoids from the average value should not exceed a few percent.

The irregularities of the two-dimensional array should also give rise to an incoherent background in the radiation, but this background is of no interest in the problem under discussion.

4. DISCUSSION OF RESULTS

Coherent emission by local plasmons excited by a fast electron is treated theoretically and it is found that the properties of the emitted radiation are closest to those of Cherenkov radiation[21] and Smith–Purcell radiation.[22] It is known that the latter can be regarded also as coherent radiation emitted by dipoles induced by the fast particle. In all such cases spontaneous radiation is emitted when the velocity of the fast particle becomes equal to the phase velocity of electromagnetic excitations in the medium. In the case of the Smith–Purcell radiation a particle moving above the surface of a linear diffraction grating interacts with a surface wave characterized by an electric vector parallel to the particle velocity. When a wave of local plasmons is excited by an electron of higher energy ($\gamma \gg 1$) we have the opposite situation: only those plasma oscillations which have the electric vector perpendicular to the particle velocity are important. The strength of the interaction of such plasmons with a relativistic electron is, according to Eq. (6), approximately γ times greater than in the case of plasmon oscillations parallel to the electron velocity. This in turn makes the preexponential factor in Eq. (16) describing the energy acquired by an electron from an external wave via local plasmons inversely proportional to the first power of the Lorentz factor and not to the third power, as in the case of the Smith–Purcell effect.[23] On the other hand, in both cases it is preferable to use particles with a large value of the Lorentz factor because this relaxes the stringent requirements on the transverse dimensions of the particle beam associated with the exponential decrease of the parameter representing the interaction of electrons with surface plasmons as we move away from the surface.

Another characteristic feature of the radiation discussed above is its strong dependence on the shape of microirregularities of the surface and dielectric properties of matter, related to the resonant behavior of the polarizability $\alpha(\omega)$ as a function of the frequency.

By way of example we consider the case when a two-dimensional array of microinhomogeneities consists of silver ellipsoids with the ratio of the semiaxes $b/a=3.4$ and oriented so that the major axis is perpendicular to the surface.[1)] Calculations (see, for example, Ref. 10) show that a plasma resonance along the major axis occurs at $\lambda \approx 500$ Å and at the resonance we have $|\sigma_x|^2 \approx 880$. We assume that the other parameters of the structure are as follows: $b=\lambda/2$, $d_1=2\lambda$, $d_2=3a$, and $L=0.5$ cm. If the initial energy is $E=0.51$ GeV ($\gamma=10^3$), and if the power density of the external source is $P=10^9$ W/cm^2 we find that the $\varphi=0$ Eq. (16) yields the following estimate of the average rate of laser acceleration of electrons:

$$\Delta E/L \approx 8.5\exp(-0.025x)\ \text{MeV/cm},$$

where the distance of the electron to the surface is measured in microns.

Estimates carried out using Eq. (14) also show that for $\gamma \approx 10^2$ the gain due to stimulated emission at optical frequencies ($\lambda \approx 5000$ Å) reaches a value of order unity when the electron current density is $\sim 10^4$ A/cm^2 and all the electrons in the beam travel no further than $\approx 10\ \mu$m from the surface. A laser operating on this amplification principle may be frequency-tunable within the width of a plasmon resonance by selection of the angle between the direction of the electron beam and the resonator axis. The frequencies of plasma resonances governed by the shape of the microirregularities and the dielectric properties of the medium cover the ultraviolet, optical, and infrared ranges.

The above theory developed for the case of a two-dimensional array can be applied also to the case when the particles form a three-dimensional colloidal crystal.[24,25] This opens up an opportunity for investigating the properties of colloidal crystals using a beam of fast electrons.

We conclude by noting likely trends in the development of the theory of coherent emission by local plasmons. According to these results, the probability of emission is proportional to the square of the volume of the microirregularities and the square of their concentration. Since the dimensions of the particles are assumed to be initially small compared with the wavelength of the radiation (dipole approximation) and the interaction of the induced dipoles has been ignored, the validity of the results obtained is limited to fairly small particles and low concentrations. We escape these restrictions only at the expense of a considerable complication of the theory. An increase in the particle size and their interaction result in deterioration of the plasmon resonances which in the final analysis should balance out the increase in the spectral density of the radiation. Hence, we can assume that the optimal parameters of such a two-dimensional array or structure should be of the order of the radiation wavelength.

[1)] Structures with similar parameters were used in Ref. 15 in nonlinear optics experiments.

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