Near-field resonances in photon emission via interaction of electrons with coupled nanoparticles

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In this work we construct the first-principles theory for the near-field resonances arising in the system of two different coupled nanoparticles excited by the external field. The obtained expressions are valid for an arbitrary external field; the detailed analysis is performed for the case of a fast electron passing by. The energy of electron is arbitrary, so the results are valid both for relativistic and nonrelativistic electrons. We have obtained rigorous analytical solution for the radiation field and for distribution of radiation over angles and frequencies. The effect of interaction between the single particles manifests itself very distinctly in conditions of resonance, resulting in a spectral shift of the radiation maximum, significant increase of its altitude, and, interestingly, in clear oscillations in radiation intensity not only when the particles are very close to each other, but even when the distance between the particles exceeds considerably their sizes. It attests to the strong influence of interaction on the radiation processes in a system of coupling particles.

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I. INTRODUCTION

Today we observe the burst of interest in the collective effects in the radiation processes of different nature and in different spectral ranges, from submillimeter (terahertz) to infrared, optical, UV, and x-ray ranges. Local field effects [1–5], including giant enhanced surface phenomena [1,6], excitation of plasmons in surface nanostructures [1,7–9], new applications and ways of realization of the Smith-Purcell effect [5,7–22], and many other interesting ideas including the strange, at first glance, ones like search for the new physics with atoms and molecules [23] rather than with collisions of superhigh-energy charged particle beams at modern colliders—-all these appeal to the attention of researchers all over the world. In a way, it is fair to say that today the interest in collective effects in radiation from complex systems, essentially dependent on the effects of coupling between their constituent elements, takes the lead over the existing theoretical background.

Usually, the characteristics of polarization radiation are calculated based on the phenomenological theoretical treatment [10–14,24], while the microscopic calculations remain undeveloped. Of course, this fact has an evident explanation: the number of particles in one cubic centimeter reaches atoms, so it is impossible to perform microscopic consideration. In many problems, however, it is possible to consider macroscopically large single particles (nano- and microparticles, quantum dots, etc.) as initial objects, and to construct the theory based on the exact description of the systems consisting of such objects [6,25–27]. This combined consideration still needs to take into account the properties of single elements and effects caused by their interaction [1–5].

In terms of theoretical background in radiation from the system of coupling particles, we should mention some of the papers which we take to be most relevant. Before shortly discussing the papers we should mention that in literature the research in similar topics is sometimes referred as "gap plasmons" [28], meaning to stress the difference from the Mie plasmons, relating to separate particles rather than to the systems of coupling particles. Besides, almost all research of this kind is totally numerical or seminumerical, while the theory is developed considerably less.

In papers [28,29] the effect of interaction between small nanoparticles is discussed, but only in terms of computer simulations or experiment—-there is no theory of the phenomenon. The review [30] by Losquin and Kociak, showing very interesting comparison between different methods of research (cathodoluminescence, electron-energy-loss spectroscopy, etc.), does contain theoretical description of the problem, but only for noninteracting particles.

In papers [31,32] the very powerful method of T matrix was developed, which makes it possible to explore numerically not only the fundamental systems like 2 interacting particles, but also the systems of 3, 4, and more particles. Despite all its power, the T-matrix method is seminumerical, which hampers qualitative theoretical insight into physics of the phenomena explored. This method, along with seminumerical methods of this kind, was applied also in research by García de Abajo (see Refs. [33,34], and especially his comprehensive review [12]), Zabala *et al.* (see Ref. [35] and a later one [36]), and Chern with coauthors [37]. While in the papers mentioned above theory was also a subject of research, still the numerical calculations dominate.

In this paper we present the fundamental, fully analytical theory describing the process of exciting the radiation from the system of two objects interacting with each other as well as with the field of the external source; as an example of such a source, the charged particle of arbitrary energy is considered.

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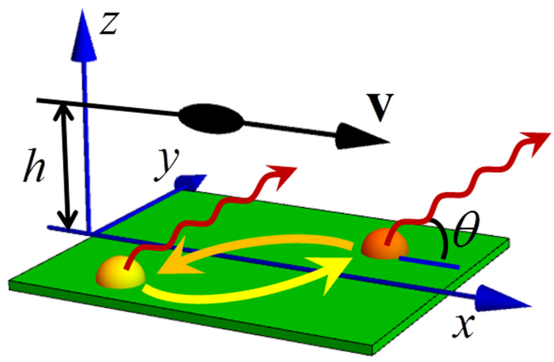

FIG. 1. Passing electron (black) generates radiation (red wavy lines) from two coupled nanoparticles. The particles may be different, which is marked by their different colors.

II. RADIATION FIELD AND INTENSITY

Let us consider the electron with charge and velocity, which passes near two nanoparticles; see Fig. 1. The shortest distance between the electron and the plane in which the particles lay is h. The role of substrate (green) is not a subject of our analysis, and is depicted only to show the plane in which the particles lay.

Without loss of generality we shall consider the particles of spherical form having different sizes r_α, r_β, and polarizabilities $\alpha(\omega)$, $\beta(\omega)$. The form does not play an essential role in the long-wave approximation

$$r_\alpha, r_\beta \ll \lambda, \tag{1}$$

which we assume to be true hereinafter. It is important that the particles interact with each other. The positions of the particles are defined by the radius vectors $\mathbf{R}_a$ and $\mathbf{R}_b$, correspondingly.

All the variety of electromagnetic phenomena, which can happen in this system, is described by the system of Maxwell's equations

$$\begin{aligned} curl\mathbf{H}(\mathbf{r},t) &= \frac{4\pi}{c}\mathbf{j}(\mathbf{r},t) + \frac{1}{c}\frac{\partial}{\partial t}\mathbf{E}(\mathbf{r},t)\\ curl\mathbf{E}(\mathbf{r},t) &= -\frac{1}{c}\frac{\partial}{\partial t}\mathbf{H}(\mathbf{r},t)\\ div\mathbf{E}(\mathbf{r},t) &= 4\pi\rho(\mathbf{r},t)\\ div\mathbf{H}(\mathbf{r},t) &= 0. \end{aligned} \tag{2}$$

In our case the current density is

$$\mathbf{j}(\mathbf{r},t) = \mathbf{j}^0(\mathbf{r},t) + \mathbf{j}_a(\mathbf{r},t) + \mathbf{j}_b(\mathbf{r},t), \tag{3}$$

where $\mathbf{j}^0$ describes a moving electron, and $\mathbf{j}_a$, $\mathbf{j}_b$ describe the currents induced in the particles by that electron.

Considering the Fourier-image of Eq. (2) over all the variables, i.e., in the variables $(\mathbf{q}, \omega)$, we can express the ith component of the field $\mathbf{E}$ through the current density $\mathbf{j}$:

$$E_i(\mathbf{q},\omega) = \frac{4\pi i}{\omega}\frac{k^2\delta_{is} - q_iq_s}{q^2 - k^2 - i0}\, j_s(\mathbf{q},\omega), \tag{4}$$

where $k = \omega/c$. The infinitely small imaginary term in the denominator means the usual rule of treatment when the poles $q = \pm k$ are dealt with.

The nearest consequence of Eq. (4) is asymptotic solution for the radiation at $kr \gg 1$:

$$E_i^{\mathrm{rad}}(\mathbf{r},\omega) = \frac{i(2\pi)^3}{\omega}\frac{e^{ikr}}{r}(k^2\delta_{is} - k_ik_s)\, j_s'(\mathbf{k},\omega), \tag{5}$$

where $\mathbf{k} = \mathbf{n}\omega/c$, and one should keep only $\mathbf{j}'(\mathbf{k},\omega) = \mathbf{j}_a(\mathbf{k},\omega) + \mathbf{j}_b(\mathbf{k},\omega)$, since contribution of the electron's own field can be neglected at $kr \gg 1$. Then, the distribution of the radiation over the angles and frequencies is found as

$$\frac{d^2W(\mathbf{n},\omega)}{d\omega d\Omega} = cr^2|\mathbf{E}^{\mathrm{rad}}(\mathbf{r},\omega)|^2, \tag{6}$$

and the only thing left to be done is to express the Fourier images $\mathbf{j}'(\mathbf{k},\omega)$ through the external field $\mathbf{E}^0(\mathbf{r},t)$ defined by the corresponding current density $\mathbf{j}^0(\mathbf{r},t)$.

Within the dipole approximation we have

$$\mathbf{j}_a(\mathbf{r},\omega) = -i\omega\alpha(\omega)\mathbf{E}^{\mathrm{act}}(\mathbf{R}_a,\omega)\delta(\mathbf{r} - \mathbf{R}_a), \tag{7}$$

and the same for the second particle, meaning only the replacement $\alpha(\omega) \to \beta(\omega)$ and $a \to b$. The fields $\mathbf{E}^{\mathrm{act}}(\mathbf{R}_a,\omega)$, $\mathbf{E}^{\mathrm{act}}(\mathbf{R}_b,\omega)$ acting on the ath and bth particles, correspondingly, are to be found. As the acting field is defined by both the field of external sources $\mathbf{E}^0$ and the field from the neighboring particle, i.e.,

$$\begin{aligned} \mathbf{E}^{\mathrm{act}}(\mathbf{R}_a,\omega) &= \mathbf{E}^0(\mathbf{R}_a,\omega) + \mathbf{E}_b^{\mathrm{act}}(\mathbf{R}_a,\omega)\\ \mathbf{E}^{\mathrm{act}}(\mathbf{R}_b,\omega) &= \mathbf{E}^0(\mathbf{R}_b,\omega) + \mathbf{E}_a^{\mathrm{act}}(\mathbf{R}_b,\omega), \end{aligned} \tag{8}$$

we need to solve the self-consistent system of equations. Here $\mathbf{E}_b^{\mathrm{act}}(\mathbf{R}_a,\omega)$ is the field of the bth particle acting at the point $\mathbf{R}_a$, and $\mathbf{E}_a^{\mathrm{act}}(\mathbf{R}_b,\omega)$ is the field from the ath particle acting at the point $\mathbf{R}_b$.

Mathematically, which opens the opportunity to find the solution analytically, is the fact that the Fourier-image of Eq. (7) in variables $(\mathbf{q},\omega)$

$$\mathbf{j}_a(\mathbf{q},\omega) = -\frac{i\omega}{(2\pi)^3}\alpha(\omega)\mathbf{E}^{\mathrm{act}}(\mathbf{R}_a,\omega)e^{-i\mathbf{q}\mathbf{R}_a} \tag{9}$$

(and the same for the bth particle) is expressed through the field $\mathbf{E}^{\mathrm{act}}$ in variables $(\mathbf{r},\omega)$. Combining expressions (8) and (9) with the Fourier-image of Eq. (4) taken in variables $(\mathbf{r},\omega)$, after some calculations one can obtain the radiation field:

$$\begin{aligned} E_i^{\mathrm{rad}}(\mathbf{r},\omega) = \frac{e^{ikr}}{r}(k^2\delta_{is} - k_ik_s)\big\{&\alpha(\omega)e^{-i\mathbf{k}\mathbf{R}_a}\big(E_s^0(\mathbf{R}_a,\omega)\\ &+ E_s^{(1)}(\mathbf{R}_a,\omega)\big) + \beta(\omega)e^{-i\mathbf{k}\mathbf{R}_b}\big(E_s^0(\mathbf{R}_b,\omega)\\ &+ E_s^{(2)}(\mathbf{R}_b,\omega)\big)\big\}, \end{aligned} \tag{10}$$

where

$$\begin{aligned} E_s^{(1)}(\mathbf{R}_a,\omega) &= -e^{iRk}\frac{\beta(\omega)}{V}t_{sj}^{(1)}E_j^0(\mathbf{R}_b,\omega)\\ &\quad + \beta(\omega)e^{2iRk}\frac{\alpha(\omega)}{V}t_{sj}^{(2)}E_j^0(\mathbf{R}_a,\omega),\\ E_s^{(2)}(\mathbf{R}_b,\omega) &= -e^{iRk}\frac{\alpha(\omega)}{V}t_{sj}^{(1)}E_j^0(\mathbf{R}_a,\omega)\\ &\quad + \beta(\omega)e^{2iRk}\frac{\alpha(\omega)}{V}t_{sj}^{(2)}E_j^0(\mathbf{R}_b,\omega),\\ t_{sj}^{(1)} &= B\delta_{sj} - \frac{\alpha(\omega)\beta(\omega)e^{2iRk}AB(A+B) + A}{W}\frac{R_sR_j}{R^2},\\ t_{sj}^{(2)} &= B^2\delta_{sj} - \frac{A(A+2B)}{W}\frac{R_sR_j}{R^2}, \end{aligned} \tag{11}$$

and

$$
\begin{aligned}
V &= 1 - \alpha(\omega)\beta(\omega)e^{2iRk}B^2, \\
W &= \alpha(\omega)\beta(\omega)e^{2iRk}(A+B)^2 - 1, \\
A &= \frac{k^2R^2 + 3ikR - 3}{R^3}, \\
B &= -\frac{k^2R^2 + ikR - 1}{R^3}, \\
R &= |\mathbf{R}_a - \mathbf{R}_b|.
\end{aligned} \tag{12}
$$

Note that in Eq. (11) the $E_s^{(1)}(\mathbf{R}_b, \omega) \rightarrow E_s^{(2)}(\mathbf{R}_a, \omega)$ at $a \rightleftarrows b$ and $\alpha(\omega) \rightleftarrows \beta(\omega)$, which is expectable for symmetry reasons. It is easy to see that the terms $E_s^{(1)}(\mathbf{R}_a, \omega)$ and $E_s^{(2)}(\mathbf{R}_b, \omega)$ describe the resonant summands resulting in difference of the obtained radiation field from a mere sum of radiation fields from two independent, noninteracting particles.

Equation (10) describes the total radiation field from two coupled nanoparticles, excited by the field of external sources. Note that we have not supposed any concrete form of the field $\mathbf{E}^0$ so far. So, the solution has been obtained in the most general form, being restricted only by the inequalities from Eq. (1) (providing the correctness of the dipole approximation) and $kr \gg 1$, meaning that we look for the emission in the wave zone, which is the conventional case for most experiments. In other words, the solution in form Eq. (10) describes the field emitted by the system of two coupled nanoparticles interacting with each other and excited by the external field $\mathbf{E}^0$ of arbitrary nature, be it the field of the plane wave, the wave packet, or the field of single electron or any other charged particles, or the beams of them.

In the case considered, using the current density

$$
\mathbf{j}^0(\mathbf{r}, t) = e\mathbf{v}\delta(\mathbf{r} - \mathbf{v}t - h\mathbf{e}_z), \tag{13}
$$

one can easily find the field $\mathbf{E}^0(\mathbf{r}, \omega)$ from Eq. (4):

$$
\mathbf{E}^0(\mathbf{r}, \omega) = \frac{e\omega}{\pi v^2\gamma} e^{i(\omega/v)x} \left[\frac{\boldsymbol{\rho}}{\rho} K_1\left(\frac{\omega\rho}{v\gamma}\right) - \frac{i\mathbf{e}_x}{\gamma} K_0\left(\frac{\omega\rho}{v\gamma}\right) \right], \tag{14}
$$

where is assumed that $\mathbf{r} = (x\mathbf{e}_x, \boldsymbol{\rho})$ with $\boldsymbol{\rho} = y\mathbf{e}_y + (z-h)\mathbf{e}_z$, and γ is the Lorentz-factor of the electron; K_0, K_1 are the Macdonald functions. So, Eqs. (6), (10), and (14) give us the solution of the problem.

III. NUMERICAL ANALYSIS AND DISCUSSION

To analyze numerically the results, let us choose the frequency range corresponding to visible light. To be more specific, we assume that the particles are aligned along the electron's trajectory, although the resulting expressions work for arbitrary orientation of the particles and the electron's trajectory.

Also, for the sake of simplicity we shall consider two identical particles, i.e., $\alpha(\omega) = \beta(\omega)$. The polarizability can be written in a rather general form:

$$
\alpha(\omega) = r_0^3 \frac{\omega_0^2}{\omega_0^2 - \omega^2 - i\Gamma\omega}, \tag{15}
$$

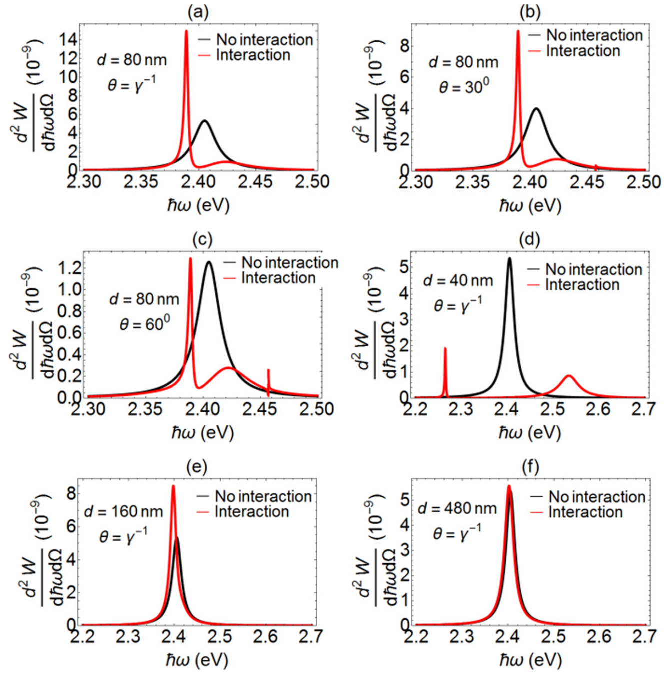

FIG. 2. Spectral distribution of radiation for electrons Lorentz-factor $\gamma = 40$, impact-parameter $h = 10$ μm, size of the particles $r_0 = 20$ nm, and resonant frequency $\omega_0 = 2.4$ eV ($\lambda_0 = 0.5$ μm).

where r_0 is the radius of the particle, and the value of Γ describes the width of the radiation line. The proportionality of $\alpha(\omega)$ to r_0^3 reflects the well-known fact that for little objects with forms close to the ball, the polarizability is directly proportional to the volume of the object. The second factor in Eq. (15) has the so-called Lorentz form, and describes a resonance when $\omega = \omega_0$ and $\Gamma \ll \omega$. Note that in more general form the denominator in Eq. (15) contains the term proportional to ω^3, which can be explained by the effect of radiative friction. This effect was described by Sipe and Kranendonk [38]; see also more recent discussion by Belov *et al.* in Ref. [39], and it seems to be important in systems containing a great deal of interacting particles, which is not the case within this research. Anyway, it affects only the concrete values of $\alpha(\omega)$, while the general expressions obtained above remain unchanged.

Various size of the particles can be chosen, depending on the wavelengths: what is important is only that the inequality $r_0 \ll \lambda$ holds true. For example, to analyze the effect in optical frequency domain, we should consider $r_0 = 20$ nm and less.

Figure 2 shows that the effect of interaction is clearly pronounced when the particles lay close enough to "feel" each other via the electromagnetic fields excited in the near zone. Note that the blue peaks (interaction is switched off) have maxima at $\omega = \omega_0$ corresponding to the simple maxima of $\alpha(\omega)$, while the red peaks (corresponding the interaction) are shifted. This shift, along with increasing the amplitude, is a direct consequence of interaction between particles.

Figures 3(a) and 3(b) show the ratio of the spectral-angular distribution of the radiation from interacting particles to one

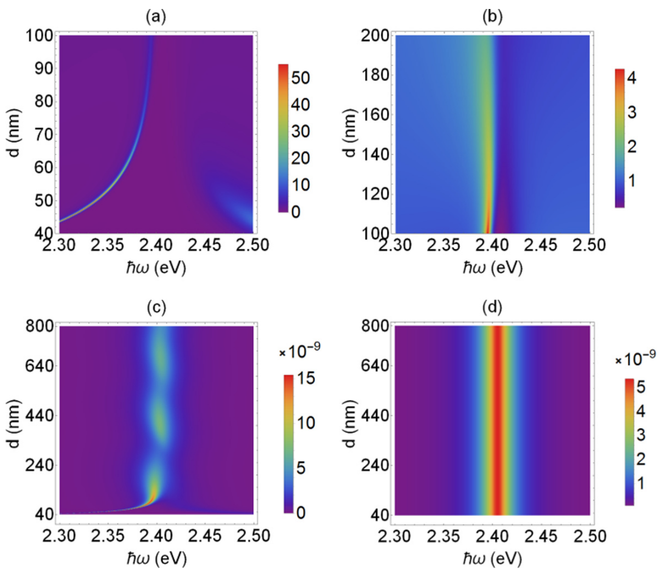

FIG. 3. (a), (b) Dependence of ratio of spectral-angular distribution with and without interaction on the distance between particles and on the photon energy. (c) Dependence of spectral-angular distribution with interaction on the distance between particles and on the photon energy. (d) Same as (c), but with no interaction between the particles. All the parameters are the same as in Fig. 2.

from noninteracting particles on both the photon energy and the distance between particles for different diapasons of the value d. It is clearly seen that the larger d is, the closer the maximum of radiation intensity to the resonance frequency is and the weaker the effect of interaction is. For relatively small distances between the particles the effect of their interaction is strong enough to shift the maximum of intensity far from the resonant frequency and also to split the single peak into two asymmetrical ones. As, for the parameters taken, the intensity of radiation from noninteracting particles is practically independent of d, the vertical pattern is strongly influenced by the effect of interaction, which is demonstrated in Fig. 3(c).

Interestingly, the oscillations with d in Fig. 3(c) manifest themselves even for the case when the distance between the particles exceeds considerably the size of the particles. It attests to the strong influence of interaction on the radiation intensity not only when the particles are very close to each other, i.e., in the near-field zone relatively to each other (which is expected intuitively), but when they are distant, separated by distances exceeding considerably their sizes. The question can arise: may it be so that the oscillations in Fig. 3(c) are merely due to conventional interference? To answer this question, one should switch the coupling effect off, which has been done in Fig. 3(d). So, we can see that "switching off" the effect of coupling in the system results in dying out of the oscillations. Note that of course "natural" oscillations due to the conventional interference of radiation from two distant particles take place as well, but for much larger values of d. Thus, even relatively remote (although being in the near-field zone still) particles interact with each other through the electromagnetic fields, and this interaction accounts for the oscillations which we see in Fig. 3(c).

Now let us discuss the limiting assumptions made to deduce the results. The most important of those used above is a long-wave approximation (sometimes it is called quasistatic limit as well). Usually, by force of its name, it is associated with phenomena in electrostatics, radio frequencies (rf), and so on. Yet, this impression is deceptive: what is really important is the inequality Eq. (1) only. For example, we in this research apply our analytics for the optical range, which is well far from the rf and electrostatics.

It is worth also mentioning about the place of the results in view of existing powerful numerical methods. For example, the T-matrix method allows describing much more complicated systems than what was considered above. Still, T matrix is a semianalytical approach, and the difference is explicit when expressions like Eq. (21) from Ref. [31] and Eq. (10) from this paper are compared. The former means that the evaluation of the involved Bessel and Hankel functions is too complex to be done analytically and so it requires numerical efforts; the latter lets us trace analytically all the details of physics, including effect of coupling, or relativistic effects of retardation. So, we hope that our simple and explicit analytical results can be of use in terms of developing the theory and be an essential step towards the further rigorous analysis of the radiation processes in the systems with strong interaction and collective effects, such as plasmons and surface plasmon polaritons.

IV. CONCLUSION

In this paper we constructed theory of radiation for the fundamental problem: the system of two interacting particles excited by the field of passing charged particle (electron). Interestingly, the resulting field of radiation differs from the mere sum of the radiation fields from two particles, while the basic equations are linear. It demonstrates that the superposition principle does not hold true always within the linear electrodynamics: the effect of interaction destroys it when the phenomenon of resonance is introduced into consideration.

Another interesting issue is the following. As it is well known, the interactions within many-body systems, starting from the number of objects equaling 3, i.e., the famous "three-body problem," are extremely problematic in terms of obtaining analytical solutions, as this kind of problems easily ends up with unstable solutions like those described by bifurcation points, etc. So, we can only obtain the analytical solution in the case of two interacting particles, and that is what has been done in this research.

On the other hand, even in the most fundamental problems of physics, like the laws of conservation for a system consisting of many objects, all the interactions are considered via two-body interactions, between all possible couples. This way, although being feebly substantiated in its fundamentals (because the contribution of three and more particles should contribute as well, and that is the question when we can neglect it)—-the way consisting of taking into consideration only two-particle interactions is generally accepted. For that reason, the exact theory describing analytically the effects of interaction between two particles can be of primary importance for further rigorous analysis of the radiation processes when it comes to the systems with strong interaction and collective effects, local field effects, including giant surface enhanced phenomena, etc.

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