Local electric and magnetic fields in semicontinuous metal films: Beyond the quasistatic approximation

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A theory of optical, infrared, and microwave response of metal-dielectric inhomogeneous films is developed. The generalized Ohm's law is formulated for the important case, when the inhomogeneity length scale is comparable with or larger than the skin (penetration) depth in metal grains. In this approach electric and magnetic fields *outside* a film can be related to the currents *inside* the film. Our computer simulations, with the use of the generalized Ohm's law approximation, reproduce the experimentally observed prominent absorption band near the percolation threshold. Calculations show that the local electric and magnetic fields experience giant spatial fluctuations. The fields are localized in small spatially separated peaks: electric and magnetic hot spots. In these hot spots the local fields (both electric and magnetic) exceed the applied field by several orders of magnitude. It is also shown that transmittance of a regular array of small holes in a metal film is strongly enhanced when the incident wave is in resonance with surface polaritons in the film. In addition, there is a skin resonance in transmission, which is of a purely geometrical nature.

I. INTRODUCTION

Random metal-dielectric films, also known as semicontinuous metal films, are usually produced by thermal evaporation or spattering of metal onto an insulating substrate. In the growing process, first, small clusters of metal grains are formed and eventually, at a percolation threshold, a continuous conducting path appears between the ends of the sample, indicating a metal-insulator transition in the system. At high-surface coverage, the film is mostly metallic with voids of irregular shape, and finally the film becomes a uniform metal film. Over the past three decades, the electric-transport properties of semicontinuous metal films have been a topic of active experimental and theoretical study. The classical percolation theory had been employed to describe the anomalous behavior of the conductivity and other transport properties near the percolation threshold.[1–4] Recently it was shown that quantum effects, such as tunneling between metal clusters and electron localization, become important at the percolation even at room temperature (see Refs. 5–8 and references there in). The low-frequency divergence of the dielectric-constant was predicted theoretically[7,8] and obtained then experimentally.[9]

In this paper we will consider the optical response of metal-insulator thin films. Although these films have been intensively studied both experimentally and theoretically (see e.g., Refs. 3,4,10–22), the important role of giant local-field fluctuations was not considered in these earlier studies.

A two-dimensional inhomogeneous film is a thin layer within which the local physical properties are not uniform. The response of such a layer to an incident wave depends crucially on the inhomogeneity length scale compared to the wavelength and also on the angle of incidence. Usually, when the wavelength is smaller than the inhomogeneity scale, the incident wave is scattered in a various directions. The total field that is scattered in certain direction is the sum of the elementary waves scattered in that direction by each elementary scatterer on the surface. As each elementary wave is given not only by its amplitude, but also by its phase, this sum will be a vector sum. The scattered wave is then distributed in various directions, though certain privileged directions may receive more energy than others. By contrast, when the inhomogeneity length scale is much smaller than the wavelength, the resolution of the wave is too small to ''see'' the irregularities, therefore the wave is then reflected specularly and transmitted in well-defined direction, as if the film were a homogeneous layer with bulk effective physical properties (conductivity, permittivity, and permeability) that are uniform. The wave is coupled to the inhomogeneities in such a way that irregular currents are excited on the surface of the layer. Strong distortions of the field then appear near the surface; however, they decay exponentially so that far enough from the surface the wave resumes its plane-wave character.

The problem of scattering from inhomogeneous surfaces has attracted attention since the time of Lord Rayleigh.[23] Due to the wide range of potential application in, e.g., radiowave and radar techniques, most efforts have been concentrated in the regime where the scale of inhomogeneity is larger than the wavelength.[24] In the last decade, a problem of localization of surface polaritons[25] and other ''internal modes'' due to their interaction with surface roughness, attracted a lot of attention. This localization is found to manifest strongly in the angular dependence of the intensity of nonspecularly reflected light, leading to the peak in the antispecular direction[26] and other ''resonance directions.''[27,28] The development of near-field scanning optical microscopy has opened the way to probe the surface polariton field above the surface and visualize its distribution (for review, see, for example, see Ref. 29.) In this paper we consider another limiting case, when the inhomogeneity length scale is much smaller than the wavelength, but can be of the order or even larger than the skin depth. In other words the coupling of a metal grain

with an *electromagnetic* field is supposed to be strong in spite of their subwavelength size. In particular, we focus on the high-frequency response (optical, infrared, and microwave) of thin metal-dielectric random films.

The optical properties of metal-dielectric films show anomalous phenomena that are absent for bulk metal and dielectric components. For example, the anomalous absorption in the near-infrared spectral range leads to unusual behavior of transmittance and reflectance. Typically, the transmittance is much higher than that of continuous metal films, whereas the reflectance is much lower (see, e.g., Refs. 3,4,10,11,16–18). Near and well-below the percolation threshold, the anomalous absorbance can be as high as 50%.[12–16,20] A number of theories were proposed for calculation of the optical properties of semicontinuous random films, including the effective-medium approaches,[30,31] their various modifications,[3,16,17,32–36] and the renormalization-group method (see, e.g. Refs. 4,37,38). In most of these theories the semicontinuous metal-dielectric film is considered as a fully two-dimensional system and a quasistatic approximation is invoked. However, usage of this approximation implies that both the electric and magnetic fields in the film are assumed to be two dimensional and curl free. That assumption ceases to be valid when the fields are changed considerably within the film and in its close neighborhood, which is usually the case for a semicontinuous metal thin film, especially in the regime of strong skin effect.

In the attempt to expand a theoretical treatment beyond the quasistatic approximation, a new approach has recently been proposed that is based on the full set of Maxwell's equations.[18–20] This approach does not use the quasistatic approximation because the fields are not assumed to be curl free inside the physical film. Although the theory was proposed with metal-insulator thin films in mind, it is, in fact, quite general and can be applied to any kind of inhomogeneous film under appropriate conditions. For the reason that will be explained below, that theory is referred to as the ''generalized Ohm's law.'' We use this new theory to find optical properties and distribution of the local fields in a semicontinuous metal film and in a metal film with regular array of holes in it.

Below we restrict ourselves to the case where all the external fields are parallel to the plane of the film. This means that an incident wave, as well as the reflected and transmitted waves, are traveling in the direction perpendicular to the film plane. We focus our consideration on the electric and magnetic-field magnitudes at certain distances *away* from the film and relate them to the currents *inside* the film. We assume that inhomogeneities on a film are much smaller in size than the wavelength λ (but not necessarily smaller than the skin depth δ), so that the fields away from the film are curl free and can be expressed as gradients of potential fields. The electric and magnetic induction currents averaged over the film thickness obey the usual two-dimensional continuity equations. Therefore the equations for the fields (e.g., $\nabla\times\mathbf{E}=0$) and the equations for the currents (e.g., $\nabla\times\mathbf{j}=0$) are the *same* as in the quasistatic case. The only difference, though important, is that the fields and the averaged currents are now related by new constitutive equations and that there are magnetic currents as well as electric currents.

To determine these constitutive equations, we find the

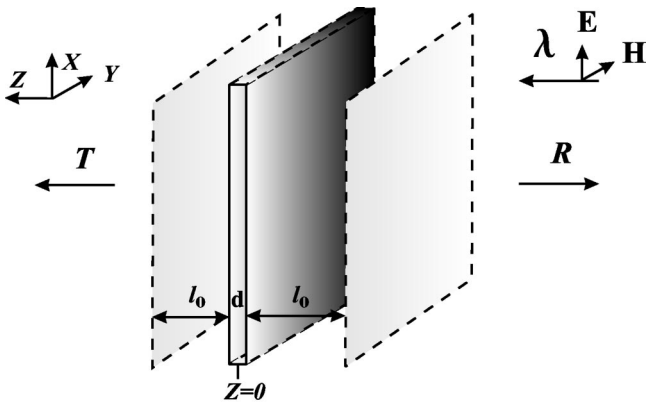

FIG. 1. The scheme used in a theoretical model. Electromagnetic wave of wavelength λ is incident on a thin metal-insulator film with thickness d. It is partially reflected and absorbed, and the remainder is transmitted through the film. The amplitudes of the electric and magnetic fields (averaged over the plane $z=-d/2-l_0$ behind the film) are equal to each other.

electric and magnetic-field distributions inside the conductive and dielectric regions of the film. The boundary conditions completely determine solutions of Maxwell's equations for the fields inside a grain when the frequency is fixed. Therefore the internal fields, which change very rapidly with position in the direction perpendicular to the film, depend linearly on the electric and magnetic field away from the film. The currents inside the film are linear functions of the local internal fields given by the usual local constitutive equations. Therefore the currents flowing *inside* the film also depend linearly on the electric and magnetic fields *outside* the film. However, the electric current averaged over the film thickness now depends not only on the external electric field, but also on the external magnetic field. The same is true for the average magnetic induction current. Thus we have two linear equations that connect the two types of the average internal currents to the external fields. These equations can be considered as a generalization of the Ohm's law to the nonquasistatic case and referred to as generalized Ohm's law (GOL).[19,20] The GOL forms the basis of a new approach to calculating the electromagnetic properties of inhomogeneous films.

It is instructive to consider first the electric and magnetic fields on both sides of the film.[19,20] Namely, the electric and magnetic fields are considered at the distance l_0 behind the film $\mathbf{E}_1(\mathbf{r})=\mathbf{E}(\mathbf{r},-d/2-l_0)$, $\mathbf{H}_1(\mathbf{r})=\mathbf{H}(\mathbf{r},-d/2-l_0)$, and at the distance l_0 in front of the film $\mathbf{E}_2(\mathbf{r})=\mathbf{E}(\mathbf{r},d/2+l_0)$, $\mathbf{H}_2(\mathbf{r})=\mathbf{H}(\mathbf{r},d/2+l_0)$ as shown in Fig. 1. All the fields and currents considered in this paper are monochromatic fields with the usual $\exp(-i\omega t)$ time dependance. The vector $\mathbf{r}=\{x,y\}$ in the above equations is a two-dimensional vector in the film plane perpendicular to the ''z'' axis where d is the thickness of the film. In the case of laterally inhomogeneous films electric current

$$\mathbf{j}_E=\int_{-d/2-l_0}^{d/2+l_0}\mathbf{j}(\mathbf{r},z)\,dz$$

and current of the magnetic induction

$$\mathbf{j}_H=(i\omega/4\pi)\int_{-d/2-l_0}^{d/2+l_0}\mathbf{B}(\mathbf{r},z)dz$$

are functions of the vector $\mathbf{r}$. The metal islands in semicontinuous films typically have an oblate shape so that the grain diameter D can be much larger than the film thickness d (see e.g., Refs. 11). When the thickness of a conducting grain d (or skin depth δ) and distance l_0 are much smaller than the grain diameter D, the relation of the fields $\mathbf{E}_1$ and $\mathbf{H}_1$ (or $\mathbf{E}_2$ and $\mathbf{H}_2$) to the currents becomes fully local. The electric $\mathbf{j}_E$ and magnetic $\mathbf{j}_H$ currents lie in between the planes $z=-d/2-l_0$ and $z=d/2+l_0$. These currents satisfy to the two-dimensional continuity equations

$$\nabla\cdot\mathbf{j}_E(\mathbf{r})=0,\quad \nabla\cdot\mathbf{j}_H(\mathbf{r})=0, \tag{1}$$

which follow from the three-dimensional continuity equations when the z components of $\mathbf{E}_1,\mathbf{H}_1$ and $\mathbf{E}_2,\mathbf{H}_2$ are neglected at the planes $z=-(d/2+l_0)$ and $z=d/2+l_0$, respectively. This is possible because these components are small, in accordance with the fact that the average fields $\langle\mathbf{E}_1\rangle$, $\langle\mathbf{H}_1\rangle$, $\langle\mathbf{E}_2\rangle$, and $\langle\mathbf{H}_2\rangle$ are parallel to the film plane. Moreover, it is supposed in the GOL approximation that ''z'' components of the local fields vanish on average, so that the integrals

$$\int_{-d/2-l_0}^{d/2+l_0}E_z dz=0$$

and

$$\int_{-d/2-l_0}^{d/2+l_0}H_z dz=0$$

are equal to zero. Then, the second Maxwell's equation $\mathrm{curl}\mathbf{H}=(4\pi/c)\mathbf{j}_E$ can be written as

$$\oint\mathbf{H}\cdot d\mathbf{l}=(4\pi/c)(\mathbf{n}_1\cdot\mathbf{j}_E)\Delta, \tag{2}$$

where the integration is over the rectangular contour, which has sides $d+2l_0$ and Δ so that the sides $d+2l_0$ are perpendicular to the film and the sides Δ are in the planes $z=\pm(d/2+l_0)$; the vector $\mathbf{n}_1$ is perpendicular to the contour, i.e., parallel to the film. When $\Delta\to 0$ this equation takes the following form

$$\mathbf{H}_2-\mathbf{H}_1=-\frac{4\pi}{c}[\mathbf{n}\times\mathbf{j}_E], \tag{3}$$

where the vector $\mathbf{n}$ is perpendicular to the film. The first Maxwell equation $\mathrm{curl}\mathbf{E}=ik\mathbf{H}$ can be rewritten as

$$\oint\mathbf{E}\cdot d\mathbf{l}=(4\pi/c)(\mathbf{n}_1\cdot\mathbf{j}_H)\Delta, \tag{4}$$

where the integration contour is the same as in Eq. (2) and $\mathbf{j}_H$ is the current of the magnetic induction, defined earlier in Eq. (1). Thus we obtain the equation

$$\mathbf{E}_2-\mathbf{E}_1=-\frac{4\pi}{c}[\mathbf{n}\times\mathbf{j}_H] \tag{5}$$

relating the electric fields in front of and behind the film. The electric current $\mathbf{j}_E$ and current of the magnetic induction $\mathbf{j}_H$ obey GOL (see Refs. 19,20), namely,

$$\mathbf{j}_E(\mathbf{r})=u(\mathbf{r})\mathbf{E}(\mathbf{r}),\quad \mathbf{j}_H(\mathbf{r})=w(\mathbf{r})\mathbf{H}(\mathbf{r}), \tag{6}$$

where $\mathbf{E}=(\mathbf{E}_1+\mathbf{E}_2)/2$, $\mathbf{H}=(\mathbf{H}_1+\mathbf{H}_2)/2$ and Ohmic parameters u and w are expressed in terms of the local refractive index $n=\sqrt{\varepsilon(\mathbf{r})}$ as

$$u=-i\frac{c}{2\pi}\frac{\tan(Dk/4)+n\tan(dkn/2)}{1-n\tan(Dk/4)\tan(dkn/2)}, \tag{7}$$

$$w=i\frac{c}{2\pi}\frac{n\tan(Dk/4)+\tan(dkn/2)}{n-\tan(Dk/4)\tan(dkn/2)}, \tag{8}$$

where the refractive index n takes values $n_m=\sqrt{\varepsilon_m}$ and $n_d=\sqrt{\varepsilon_d}$ for metal and dielectric regions of the film; the distance to the reference plane is set as $l_0=D/4$.[18,19] The essence of the GOL can be summarized as follows: The entire physics of a three-dimensional inhomogeneous layer, which is described by the full set of Maxwell's equations, has been reduced to a set of two-dimensional Eqs. (1) and (6).

In the quasistatic limit, when the optical thickness of metal grains is small $dk|n_m|\ll 1$, while the metal-dielectric constant is large in magnitude, $|\varepsilon_m|\gg 1$, the following estimates hold for the Ohmic parameters of the metal grains

$$u_m\simeq -i\frac{\omega\varepsilon_m}{4\pi}d,\quad w_m\simeq i\frac{\omega}{4\pi}(d+D/2),\quad (d/\delta\ll 1). \tag{9}$$

In the opposite case of a strong skin effect, when the skin depth (penetration depth) $\delta=1/k\,\mathrm{Im}\,n_m$ is much smaller than the grain thickness d and the electromagnetic field does not penetrate into metal grains, the parameters u_m and w_m take values

$$u_m=i\frac{2c^2}{\pi D\omega},\quad w_m=i\frac{\omega D}{8\pi},\quad (d/\delta\gg 1). \tag{10}$$

For the dielectric region, when the film is thin enough so that $dkn_d\ll 1$ and $\varepsilon_d\sim 1$, Eqs. (7) and (8) give

$$u_d=-i\frac{\omega\varepsilon_d'}{8\pi}D,\quad w_d=i\frac{\omega}{4\pi}(d+D/2), \tag{11}$$

where the reduced dielectric constant $\varepsilon_d'=1+2\varepsilon_d d/D$ is introduced. Note that in the limit of the strong skin effect the Ohmic parameters u_m and w_m are purely imaginary and the parameter u_m is of the inductive character, i.e., it has the sign opposite to the dielectric parameter u_d. In contrast, the Ohmic parameter w remains essentially the same $w\sim iD\omega/8\pi$ for the dielectric and metal regions, regardless of the magnitude of the skin effect.

The rest of this paper is organized as follows: In Sec. II we briefly outline the basic optical properties: reflectance, transmittance, and absorbance, found in the GOL approximation; we also present here the self-consistent approach denoted as the dynamic effective-medium theory. Our numerical method and results of calculations for the local electric and magnetic field are described in Sec. III. In Sec. IV a theory is developed for the giant local field fluctuations and

the spatial high-order moments of the local fields. In Sec. V we consider the optical properties of a metal film which is perforated with an array of small subwavelength holes; we show that the transmittance through such a film can be strongly enhanced in accord with recent experimental observations. Section VI summarizes and concludes the paper.

II. OPTICAL PROPERTIES OF SEMICONTINUOUS FILMS

Since we consider semicontinuous films with an inhomogeneity scale much smaller than the wavelength λ, the fields $\mathbf{E}_1(\mathbf{r})$ and $\mathbf{H}_1(\mathbf{r})$ behind the film are simply the gradients of potential fields, when considered as functions of x and y in the fixed reference plane $z=-d/2-l_0$. Similarly, the potentials for the fields $\mathbf{E}_2(\mathbf{r})$ and $\mathbf{H}_2(\mathbf{r})$ in front of the film can be introduced. Therefore the fields $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$ in Eq. (6) can, in turn, be represented as gradients of some potentials:

$$\mathbf{E}=-\nabla',\quad \mathbf{H}=-\nabla\psi'. \tag{12}$$

By substituting these expressions in Eqs. (6) and then in the continuity Eq. (1), we obtain the following equations

$$\nabla\cdot[u(\mathbf{r})\nabla\varphi'(\mathbf{r})]=0, \tag{13}$$

$$\nabla\cdot[w(\mathbf{r})\nabla\psi'(\mathbf{r})]=0, \tag{14}$$

that can be solved independently for the potentials φ' and ψ'. These equations are solved under the following conditions

$$\langle\nabla\varphi_1'\rangle=\langle\mathbf{E}_1\rangle\equiv\mathbf{E}_0,\quad \langle\nabla\psi_1'\rangle=\langle\mathbf{H}_1\rangle\equiv\mathbf{H}_0, \tag{15}$$

where the constant fields $\mathbf{E}_0$ and $\mathbf{H}_0$ are the external (given) fields that are determined by the incident wave. When the fields $\mathbf{E}$, $\mathbf{H}$ and currents $\mathbf{j}_E$, $\mathbf{j}_H$ are found by solving Eqs. (13), (14), and (15), the local electric and magnetic fields in the plane $z=-l_0-d/2$ are given by

$$\mathbf{E}_1=\mathbf{E}+\frac{2\pi}{c}[\mathbf{n}\times\mathbf{j}_H],\quad \mathbf{H}_1=\mathbf{H}+\frac{2\pi}{c}[\mathbf{n}\times\mathbf{j}_E], \tag{16}$$

as follows from Eqs. (3) and (5) and the definitions of the fields $\mathbf{E}$ and $\mathbf{H}$. Note that the field $\mathbf{E}_1(\mathbf{r})$ can be measured in near-field experiments (see, e.g., Ref. 29). The effective Ohmic parameters u_e and w_e are defined in a usual way

$$\langle\mathbf{j}_E\rangle=u_e\mathbf{E}_0\equiv u_e(\langle\mathbf{E}_1\rangle+\langle\mathbf{E}_2\rangle)/2, \tag{17}$$

$$\langle\mathbf{j}_H\rangle=w_e\mathbf{H}_0\equiv w_e(\langle\mathbf{H}_1\rangle+\langle\mathbf{H}_2\rangle)/2. \tag{18}$$

When these expressions are substituted in Eqs. (3) and (5), which are averaged over the film plane (coordinates $\{x,y\}$), we obtain the equations

$$[\mathbf{n}\times(\langle\mathbf{H}_2\rangle-\langle\mathbf{H}_1\rangle)]=\frac{2\pi}{c}u_e(\langle\mathbf{E}_1\rangle+\langle\mathbf{E}_2\rangle), \tag{19}$$

$$[\mathbf{n}\times(\langle\mathbf{E}_2\rangle-\langle\mathbf{E}_1\rangle)]=\frac{2\pi}{c}w_e(\langle\mathbf{H}_1\rangle+\langle\mathbf{H}_2\rangle), \tag{20}$$

that connect the average fields in front of the film and behind it, i.e., we obtain the equations that determine the optical response of an inhomogeneous film.

Let us suppose that the wave enters the film from the right half-space (see Fig. 1), so that its amplitude is proportional to e^{-ikz}. The incident wave is partially reflected and partially transmitted through the film. The electric-field amplitude in the right half-space, away from the film, can be written as $\tilde{\mathbf{E}}_2(z)=\mathbf{e}[e^{-ikz}+re^{ikz}]$, where r is the reflection coefficient and $\mathbf{e}$ is the polarization vector. Well behind the film, the electric component of the electromagnetic wave acquires the form $\tilde{\mathbf{E}}_1(z)=\mathbf{e}te^{-ikz}$, where t is the transmission coefficient. It is supposed for simplicity that the film has no optical activity, so that the wave polarization $\mathbf{e}$ remains the same after passing through the film. At the planes $z=d/2+l_0$ and $z=-d/2-l_0$ the average electric field equals $\langle\mathbf{E}_2\rangle$ and $\langle\mathbf{E}_1\rangle$, respectively. The electric field in the wave is matched with the average fields in the planes $z=d/2+l_0$ and $z=-d/2-l_0$, i.e., $\langle\mathbf{E}_2\rangle=\tilde{\mathbf{E}}_2(d/2+l_0)=\mathbf{e}[e^{-ik(d/2+l_0)}+re^{ik(d/2+l_0)}]$ and $\langle\mathbf{E}_1\rangle=\tilde{\mathbf{E}}_1(-d/2-l_0)=\mathbf{e}te^{ik(d/2+l_0)}$. The same matching with the magnetic fields gives $\langle\mathbf{H}_2\rangle=[\mathbf{n}\times\mathbf{e}][-e^{-ik(d/2+l_0)}+re^{ik(d/2+l_0)}]$ and $\langle\mathbf{H}_1\rangle=-[\mathbf{n}\times\mathbf{e}]te^{ik(d/2+l_0)}$ in the planes $z=d/2+l_0$ and $z=-d/2-l_0$ respectively. Substitution of these expressions for the fields $\langle\mathbf{E}_1\rangle$, $\langle\mathbf{E}_2\rangle$, $\langle\mathbf{H}_1\rangle$, and $\langle\mathbf{H}_2\rangle$ in Eqs. (19) and (20) gives the two linear (scalar) equations for the reflection r and transmission t coefficients. By solving these equations we obtain the reflectance,

$$R\equiv|r|^2=\left|\frac{(2\pi/c)(u_e+w_e)}{(1+(2\pi/c)u_e)(1-(2\pi/c)w_e)}\right|^2, \tag{21}$$

transmittance

$$T\equiv|t|^2=\left|\frac{1+((2\pi/c))^2u_ew_e}{(1+(2\pi/c)u_e)(1-(2\pi/c)w_e)}\right|^2, \tag{22}$$

and absorbance

$$A=1-T-R \tag{23}$$

of the film. Thus, the effective Ohmic parameters u_e and w_e completely determine the optical properties of an inhomogeneous films.

We see that the problem of the field distribution and optical response of the metal-dielectric films reduces to solving the decoupled quasistatic conductivity problems [Eqs. (13) and (14)], for which a number of theoretical approaches are available. Thus efficient analytical and numerical methods (see Sec. III) developed in the frame of the percolation theory can be used to find the effective parameters u_e and w_e of the film.

We consider now the case of the strong skin effect in metal grains and trace the evolution of the optical properties of a semicontinuous metal film with the increase of the metal surface density p. When $p=0$ the film is purely dielectric and the effective parameters u_e and w_e coincide with the dielectric Ohmic parameters given by Eq. (11). By substituting $u_e=u_d$ and $w_e=w_d$ in Eqs. (21), (22), and (23), and assuming that the dielectric film has no losses and is optically thin ($dk\varepsilon_d\ll1$), we obtain that the reflectance $R=d^2(\varepsilon_d-1)^2k^2/4$, transmittance $T=1-d^2(\varepsilon_d-1)^2k^2/4$, and the absorbance $A=0$ in accord with the well-known results for a thin dielectric film.[39,40]

It is not surprising that the film without losses has zero absorbance. When the ratio of the penetration length (skin

depth) $\delta = 1/k\mathrm{Im}n_m$ is negligible in comparison with the film thickness d and $|n_m| \gg 1$ the losses are also absent in the limit of full coverage, when the metal concentration $p=1$. In this case the film is a perfect metal mirror. Indeed substituting the Ohmic parameters $u_e = u_m$ and $w_e = w_m$ from Eq. (10) in Eqs. (21), (22), and (23), we obtain for the reflectance $R=1$, while the transmittance T and absorbance A are both equal to zero. Note that the optical properties of the film do not depend on the particle size D for the metal concentration $p=0$ and $p=1$ since properties of the dielectric and continuous metal films do not depend on the shape of the metal grains.

We consider now the film at the percolation threshold $p=p_c$ with $p_c=1/2$ for a self-dual system.[3,4] A semicontinuous metal film may be thought of as a mirror, which is broken into small pieces with typical size D much smaller than the wavelength λ. At the percolation threshold the exact Dykhne formulas $u_e = \sqrt{u_d u_m}$ and $w_e = \sqrt{w_d w_m}$ hold.[41] Thus following equations for the effective Ohmic parameters are obtained from Eqs. (11) and (10)

$$\frac{2\pi}{c} u_e(p_c) = \sqrt{\varepsilon_d'}, \quad \frac{2\pi}{c} w_e(p_c) = i\frac{Dk}{4}\sqrt{1+\frac{2d}{D}}. \quad (24)$$

From this equation it follows that $|w_e/u_e| \sim Dk \ll 1$ so that the effective Ohmic parameter w_e can be neglected in comparison with u_e. By substituting the effective Ohmic parameter $u_e(p_c)$ given by Eq. (24) in Eqs. (21), (22), and (23), the optical properties at the percolation can be obtained as

$$R(p_c) = \frac{\varepsilon_d'}{(1+\sqrt{\varepsilon_d'})^2}, \quad (25)$$

$$T(p_c) = \frac{1}{(1+\sqrt{\varepsilon_d'})^2}, \quad (26)$$

$$A(p_c) = \frac{2\sqrt{\varepsilon_d'}}{(1+\sqrt{\varepsilon_d'})^2}, \quad (27)$$

where $\varepsilon_d' = 1 + 2\varepsilon_d d/D$. When metal grains are oblate enough so that $\varepsilon_d d/D \ll 1$ and $\varepsilon_d' \to 1$ the above expressions simplify to the universal result

$$R = T = 1/4, \quad A = 1/2 \quad (28)$$

first obtained in Ref. 18. Thus, there is the effective absorption in semicontinuous metal films, even in the case when neither dielectric nor metal grains absorb the light energy, i.e., the mirror broken into small pieces effectively absorbs energy from the electromagnetic field. The effective absorption in a loss-free film means that the electromagnetic energy is stored in the system and that the amplitudes of the local electromagnetic field can increase, in principle, up to infinity. In any real semicontinuous metal film the local field is finite, of course, because of nonzero losses; still, the field fluctuations over the film can be very large provided that losses are small, as discussed below.

To find the optical properties of semicontinuous films for arbitrary metal concentration p, the effective medium theory can be implemented that was originally developed to provide a semiquantitative description of the transport properties of percolation composites.[3] The effective-medium theory being applied to Eqs. (13) and (17) and (14) and (18) results in the following equations for the effective parameters

$$u_e^2 - \Delta p u_e(u_m - u_d) - u_d u_m = 0, \quad (29)$$

$$w_e^2 - \Delta p w_e(w_m - w_d) - w_d w_m = 0, \quad (30)$$

where the ''reduced'' concentration $\Delta p = (p-p_c)/p_c$, ($p_c = 1/2$) is introduced. It follows from Eq. (30) that for the case of strong skin effect, when the Ohmic parameters $w_m \ll c$ and $w_d \ll c$ [see Eqs. (10)–(11)], the effective Ohmic parameter $|w_e| \ll c$, for all metal concentrations p. Therefore the parameter w_e is negligible in Eqs. (21) and (22). For further simplification the Ohmic parameter u_d can be neglected in comparison with u_m in the second term of Eq. (29) [cf. Eqs. (10) and (11)]. Then, by introducing the dimensionless Ohmic parameter $u_e' = (2\pi/c)u_e$, we rewrite Eq. (29) as

$$u_e'^2 - 2i\frac{\lambda \Delta p}{\pi D} u_e' - \varepsilon_d' = 0. \quad (31)$$

Right at the percolation threshold $p = p_c = 1/2$, when the reduced concentration $\Delta p = 0$, Eq. (31) gives the effective Ohmic parameter $u_e'(p_c) = \sqrt{\varepsilon_d'}$ that coincides with the exact Eq. (24) and results in reflectance, transmittance, and absorbance given by Eqs. (25), (26), and (27), respectively. For concentrations different from p_c, Eq. (31) gives

$$u_e' = i\frac{\lambda \Delta p}{\pi D} + \sqrt{-(\lambda \Delta p/\pi D)^2 + \varepsilon_d'}, \quad (32)$$

that becomes purely imaginary for $|\Delta p| > \pi D\sqrt{\varepsilon_d'}/\lambda$. For the imaginary effective Ohmic parameter u_e', Eqs. (25), (26), and (27) result in the zero absorbance: $A = 1 - R - T = 1 - |u_e'|^2/(1+|u_e'|^2) - 1/(1+|u_e'|^2) = 0$ (recall that the effective Ohmic parameter w_e is neglected). In the vicinity of a percolation threshold, namely, for

$$|\Delta p| < \frac{\pi D}{\lambda}\sqrt{\varepsilon_d'} \quad (33)$$

the effective Ohmic parameter u_e' has a nonvanishing real part and, therefore, the absorbance

$$A = \frac{2\sqrt{-(\lambda \Delta p/\pi D)^2 + \varepsilon_d'}}{1 + \varepsilon_d' + 2\sqrt{-(\lambda \Delta p/\pi D)^2 + \varepsilon_d'}} \quad (34)$$

is nonzero and has a well-defined maximum at the percolation threshold; the width of the maximum is inversely proportional to the wavelength. The effective absorption in almost loss-free semicontinuous metal film means that the local electromagnetic fields strongly fluctuate in the system as was speculated above. The concentration width for the strong fluctuations should be the same as the width of the absorption maximum, i.e., it is given by Eq. (33).

Note that the effective parameters u_e and w_e can be determined experimentally by measuring the amplitude and phase of the transmitted and reflected waves or by measuring the film reflectance as a function of the fields $\mathbf{E}_1$ and $\mathbf{H}_1$. In

the latter case, a metal screen placed behind the film can be used to control the values of these fields.[42,43]

III. COMPUTER SIMULATIONS OF LOCAL ELECTRIC AND MAGNETIC FIELDS

To find the local electric $\mathbf{E}(\mathbf{r})$ and magnetic $\mathbf{H}(\mathbf{r})$ fields, Eqs. (13) and (14) should be solved. Consider the first equation (13), which is convenient to rewrite in terms of the renormalized dielectric constant

$$\tilde{\varepsilon} = i\frac{4\pi u(\mathbf{r})}{\omega d} \tag{35}$$

as follows

$$\nabla\cdot[\tilde{\varepsilon}(\mathbf{r})\nabla\phi(\mathbf{r})] = \mathcal{E}, \tag{36}$$

where $\phi(\mathbf{r})$ is the fluctuating part of the potential $\phi'(\mathbf{r})$ so that $\nabla\phi'(\mathbf{r}) = \nabla\phi(\mathbf{r}) - \mathbf{E}_0$, $\langle\phi(\mathbf{r})\rangle = 0$, and $\mathcal{E} = \nabla\cdot[\tilde{\varepsilon}(\mathbf{r})\mathbf{E}_0]$. We recall that the ''external'' field $\mathbf{E}_0$ is defined by Eq. (15). For the metal-dielectric films considered here local dielectric constant $\tilde{\varepsilon}(\mathbf{r})$ equals to $\tilde{\varepsilon}_m = 4\pi i u_m/\omega d$ and $\tilde{\varepsilon}_d = \varepsilon_d' D/2d$, for the metal and dielectric regions, respectively. The external field $\mathbf{E}_0$ in Eq. (36) can be chosen real, while the local potential $\phi(\mathbf{r})$ takes complex values since the dielectric constant $\tilde{\varepsilon}_m$ is complex $\tilde{\varepsilon}_m = \tilde{\varepsilon}_m' + i\tilde{\varepsilon}_m''$.

In the quasistatic limit, when the skin depth δ is much larger than the film thickness d, the dielectric constant $\tilde{\varepsilon}_m$ coincides with the metal-dielectric constant ε_m as follows from Eq. (9). In the optical and infrared spectral ranges, a simple model of the Drude metal can be used to reproduce semiquantitatively the basic optical properties of a metal. In this approach, the dielectric constant of metal grains can be approximated by the Drude formula

$$\varepsilon_m(\omega) = \varepsilon_b - (\omega_p/\omega)^2/[1 + i\omega_\tau/\omega], \tag{37}$$

where ε_b is contribution to ε_m due to the interband transitions, ω_p is the plasma frequency, and $\omega_\tau = 1/\tau \ll \omega_p$ is the relaxation rate. In the high-frequency range, losses in metal grains are relatively small, $\omega_\tau \ll \omega$. Therefore, the real part ε_m' of the metal-dielectric function ε_m is much larger (in modulus) than the imaginary part ε_m'' ($|\varepsilon_m'|/\varepsilon_m'' \cong \omega/\omega_\tau \gg 1$), and ε_m' is negative for the frequencies ω less than the renormalized plasma frequency,

$$\tilde{\omega}_p = \omega_p/\sqrt{\varepsilon_b}. \tag{38}$$

Thus, the metal conductivity $\sigma_m = -i\omega\varepsilon_m/4\pi \cong (\varepsilon_b\tilde{\omega}_p^2/4\pi\omega)[i(1-\omega^2/\tilde{\omega}_p^2) + \omega_\tau/\omega]$ is characterized by positive imaginary part for $\tilde{\omega}_p > \omega \gg \omega_\tau$, i.e., it is of the inductive character. This allows us to model the metal grains as inductances L for the frequencies $\tilde{\omega}_p > \omega \gg \omega_\tau$ while the dielectric gaps can be represented by capacitances C. In the opposite case of the strong skin effect, the Ohmic parameter u_m is inductive according to Eq. (10), for all spectral ranges regardless of the metal properties. Then, the percolation metal-dielectric film represents a set of randomly distributed L and C elements for *all* spectral ranges.

Note that the Ohmic parameter w takes the same sign and is close in magnitude for both metal and dielectric grains, according to Eqs. (9), (10), and (11). A film can be thought of as a collection of C elements in ''w'' space. Therefore, there are no resonances in the solution to Eq. (14). The fluctuations of the potential ψ' can be neglected in comparison with the φ' fluctuations. For this reason we concentrate our attention on the properties of the ''electric'' field $\mathbf{E}(\mathbf{r}) = -\nabla\phi'(\mathbf{r}) = -\nabla\phi(\mathbf{r}) + \mathbf{E}_0$ when considering the fluctuations of the local fields. The field $\mathbf{E}(\mathbf{r})$ can be found by solving Eq. (36).

Equation (36) has the same form as usual quasistatic continuity equation, $\nabla(\varepsilon\nabla\phi) = 0$. Therefore Eq. (36) being discretized on, say, square lattice acquires the form of Kirchhoff's equations. We employ here the efficient real-space renormalization method suggested in Ref. 44 to solve the discretized Eq. (36). In our previous works[21,44–55] the same method was used to find local electric fields for the quasistatic case. Solution of Eq. (36) gives the potential ϕ in the film, the local field $\mathbf{E}(\mathbf{r})$ and the electric current $\mathbf{j}_E(\mathbf{r})$ in terms of the average field $\mathbf{E}_0$. The effective Ohmic parameter u_e is determined by Eq. (17) that can be written as $\langle\mathbf{j}_E\rangle = u_e\mathbf{E}_0$. The effective dielectric constant $\tilde{\varepsilon}_e$ equals $4\pi i u_e/\omega d$. In the same manner the field $\mathbf{H}(\mathbf{r})$, the magnetic current $\mathbf{j}_H(\mathbf{r})$, and the effective parameter w_e can be found from Eq. (14) and its lattice discretization. Note that the *same* lattice should be used to determined the fields $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$.[55] The directions of the external fields $\mathbf{E}_0$ and $\mathbf{H}_0$ may be chosen arbitrary when the effective parameters u_e and w_e are calculated since the effective parameters do not depend on the direction of the field, for an isotropic, on average, film.

Although the effective parameters do not depend on the external field, the local electric $\mathbf{E}_1(\mathbf{r})$ and magnetic and $\mathbf{H}_1(\mathbf{r})$ fields do depend on the incident wave. The local fields $\mathbf{E}_1(\mathbf{r})$ and $\mathbf{H}_1(\mathbf{r})$ are defined in the reference plane $z = -d/2 - l_0$ (see Fig. 1). For calculations below, the electric and magnetic fields of the electromagnetic wave are chosen in the form $\langle\mathbf{E}_1\rangle = \{1,0,0\}$ and $\langle\mathbf{H}_1\rangle = \{0,-1,0\}$ in the plane $z = -l_0 - d/2$. This choice corresponds to the wave vector of the incident wave as $\mathbf{k} = (0,0,-k)$, i.e., there is only a transmitted wave behind the film (see Fig. 1). It follows from the averaging of Eq. (16) (which can be written as $\langle\mathbf{E}_1\rangle = \mathbf{E}_0 + (2\pi/c)w_e[\mathbf{n}\times\mathbf{H}_0]$ and $\langle\mathbf{H}_1\rangle = \mathbf{H}_0 + (2\pi/c)u_e[\mathbf{n}\times\mathbf{E}_0]$) that the fields $\mathbf{E}_0$ and $\mathbf{H}_0$ are given by

$$\mathbf{E}_0 = \frac{\langle\mathbf{E}_1\rangle - (2\pi/c)w_e[\mathbf{n}\times\langle\mathbf{H}_1\rangle]}{1 + (2\pi/c)^2 u_e w_e},$$

$$\mathbf{H}_0 = \frac{\langle\mathbf{H}_1\rangle - (2\pi/c)u_e[\mathbf{n}\times\langle\mathbf{E}_1\rangle]}{1 + (2\pi/c)^2 u_e w_e}. \tag{39}$$

These fields $\mathbf{E}_0$ and $\mathbf{H}_0$ are used to calculate the local fields $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$. The local electric $\mathbf{E}_1(\mathbf{r})$ and magnetic $\mathbf{H}_1(\mathbf{r})$ fields are restored then from the fields $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$ by using Eqs. (16).

The local electric and magnetic fields are calculated for silver-on-glass semicontinuous films, as functions of the surface concentration p of silver grains. The dielectric constant for glass is given by $\varepsilon_d = 2.2$. The dielectric function for silver is chosen in the Drude form (37); the following parameters are used in Eq. (37): the interband-transition contribu-

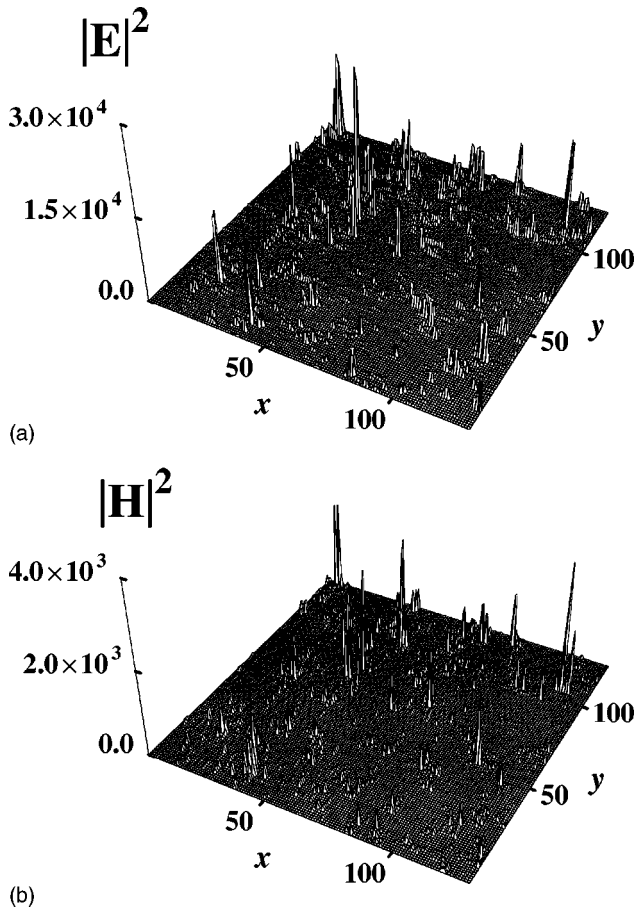

FIG. 2. Distribution of local em field intensities in a semicontinuous silver film at small skin effect $d/\delta=0.2$, where δ is the skin depth and d is the thickness of the film ($\lambda=1$ μm; $p=p_c$).

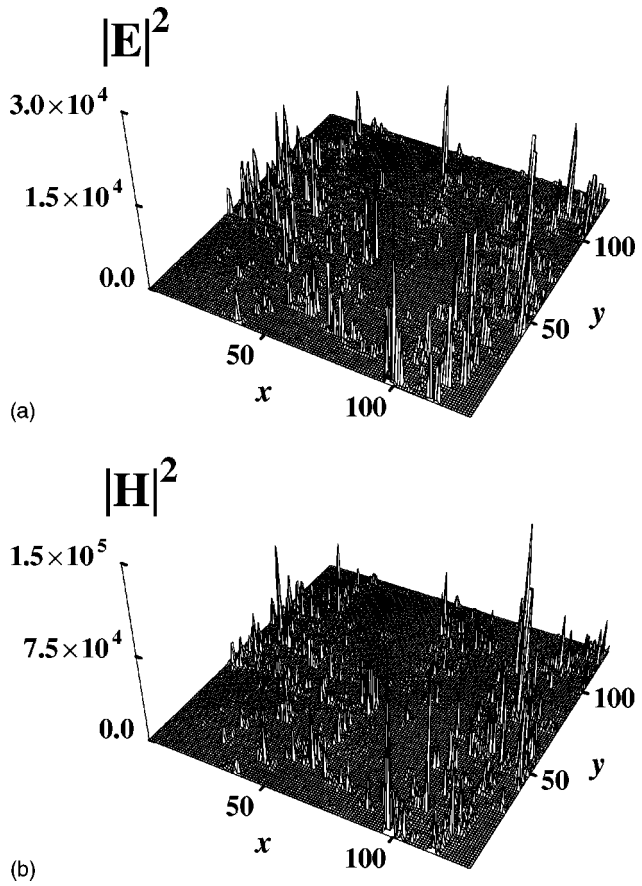

FIG. 3. Distribution of the local-field intensities in a semicontinuous silver film at strong skin effect $d/\delta=2.2$ ($\lambda=1$ μm; $p=p_c$).

tion $\varepsilon_b=5$, the plasma frequency $\omega_p=9.1$ eV, and the relaxation frequency $\omega_\tau=0.021$ eV.[56] The metal grains are supposed to be oblate in shape. The ratio of the grain thickness d (film thickness) to the grain diameter D has been chosen as $D/d=3$, the same as the one used in Ref. 18. To consider the skin effect of different strength (i.e., different interaction between the electric and magnetic fields, occurring through the skin effect), we vary the size d of silver particles in a wide range, $d=1\div100$ nm. The size of metal grains in semicontinuous metal films is usually of the order of few nanometers but it can be increased significantly by using a proper method of preparation.[57] In the microwave experiments of Ref. 20, for example, the films were lithographically prepared, so that the size of a metal particle could vary in a large range.

The space distribution of the electric and magnetic fields is calculated for two sets of parameters as illustrated in Figs. 2 and 3. In Figs. 2 and 3 we show the electric and magnetic-field distributions for $\lambda=1$ μm and two different thicknesses d of the film, $d=5$ nm and $d=50$ nm. The first thickness (Fig. 2) corresponds to a weak skin effect since the dimensionless thickness is small, $\Delta\equiv d/\delta=0.2$ [where $\delta=1/(k\,\mathrm{Im}\,n_m)$ is the skin depth]. In this case we observe the giant field fluctuations of the local electric field; the magnetic field, although it strongly fluctuates over the film, is much smaller in magnitude compared with the electric field. This is because the film itself is not magnetic, $\mu_d=\mu_m=1$, and the interaction of the magnetic field with the electric field through the skin effect is relatively small. For comparison, Fig. 3 shows the fields in the case of a significant skin effect, when the film thickness $d=50$ nm and the dimensionless thickness exceeds one, $\Delta=2.2$. It is interesting to note that the amplitude of the electric field is roughly the same as in Fig. 2(a), despite the fact that the parameter Δ is increased by one order of magnitude. In contrast, the local magnetic field in Fig. 3(b) is strongly increased in this case so that the amplitude of magnetic field in peaks is of the same order of magnitude as the electric field maxima. This behavior can be understood by considering the spatial moments of the local magnetic field as shown in the next section.

Being given the local fields, the effective parameters u_e and w_e can be found and thus the effective optical properties of the film. In Figs. 4 and 5 we show the reflectance, transmittance, and absorbance as functions of silver concentration p, for wavelengths $\lambda=1$ μm and $\lambda=10$ μm, respectively. The absorbance in these figures has an anomalous maximum in the vicinity of the percolation threshold that corresponds to the behavior predicted by Eq. (34). This maximum was detected first in the experiments.[13–16] The maximum in the absorption corresponds to strong fluctuations of the local fields. We estimated in Eq. (33) the concentration range Δp around p_c, where the giant local-field fluctuations occur, as $\Delta p\propto 1/\lambda$. Indeed, the absorbance shrinks at the transition from Fig. 4 to Fig. 5, when wavelength λ increases by a factor of 10.

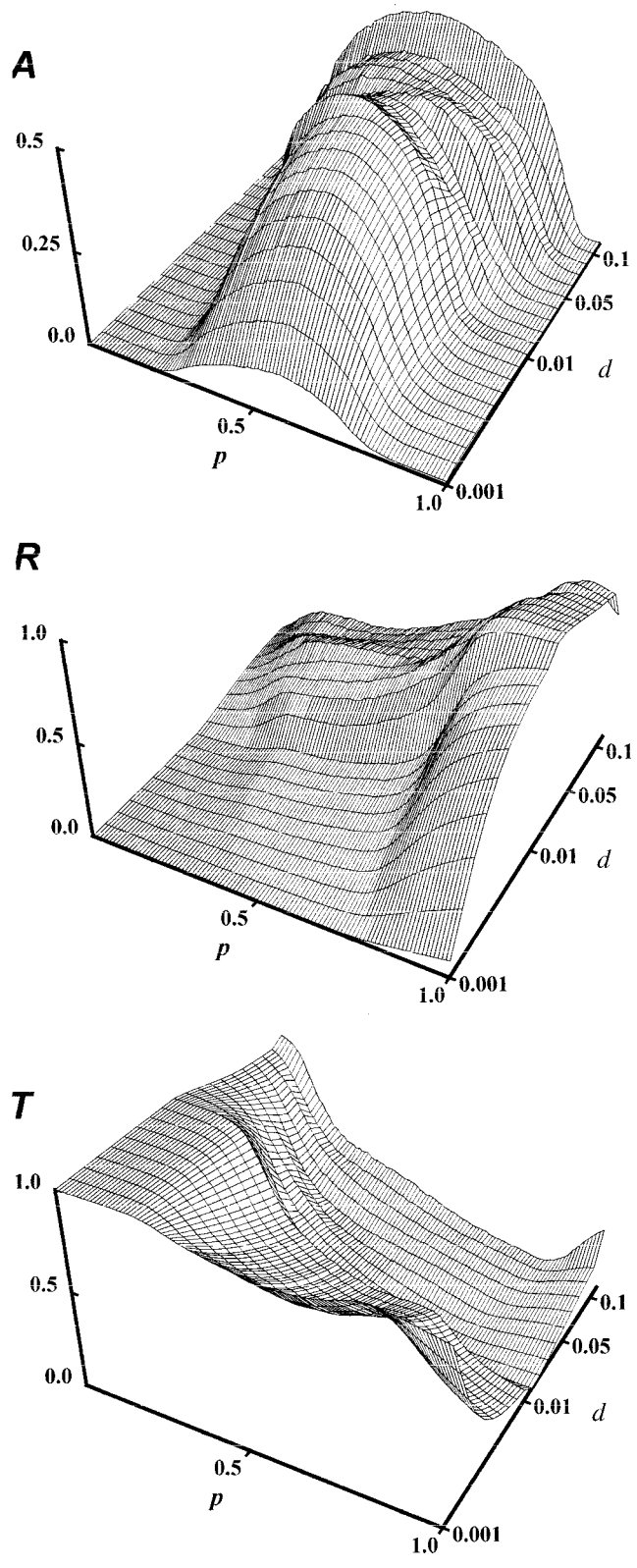

FIG. 4. Calculated absorptance *A*, reflectance *R*, and transmittance *T* for a silver-glass film as functions of metal concentration *p* and film thickness *d* at $\lambda=1$ μm.

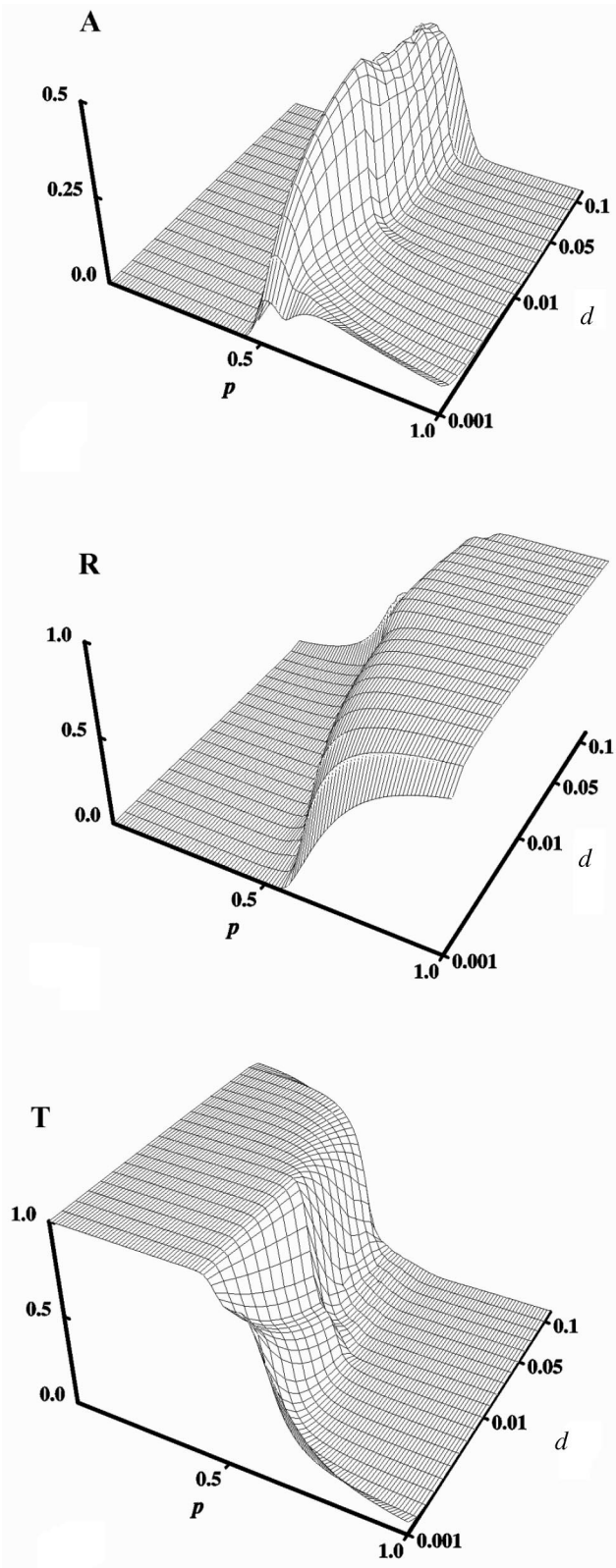

FIG. 5. Calculated absorptance *A*, reflectance *R*, and transmittance *T* for a silver-glass film as functions of metal concentration p and film thickness *d* at $\lambda=10$ μm.

In Fig. 6(a) and 6(b) we compare results of numerical simulations for the optical properties of silver semicontinuous films with the calculations based on our generalized effective-medium approach [Eqs. (29) and (30)], with the new Ohm's parameters u and v. Results of such a ''dynamic'' effective-medium theory are in accord with our numerical simulations, for arbitrary-strength skin effect. We can see that the theory reproduces well the maximum of absorptance in the vicinity of the percolation threshold.

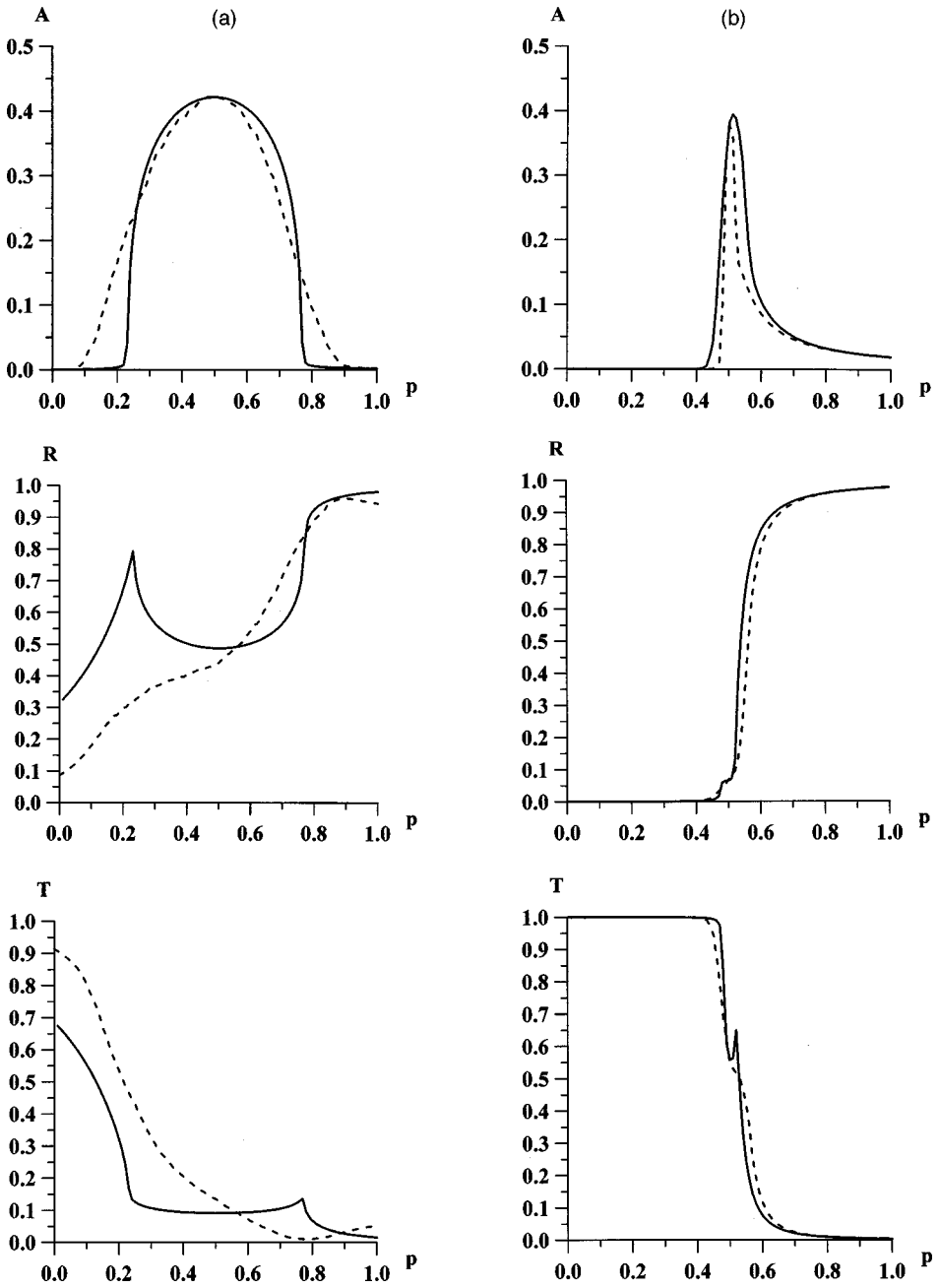

FIG. 6. Results of numerical simulations (dashed line) and the dynamic effective-medium theory (solid line) for absorptance A, reflectance R, and transmittance T of silver-on-glass semicontinuous film as function of the metal concentration p at the following wavelength λ and film thickness d: (1) $\lambda=1.0$ μm and $d=50$ nm; (b) $\lambda=10.0$ μm and $d=5$ nm.

IV. SPATIAL MOMENTS OF THE LOCAL ELECTRIC AND MAGNETIC FIELDS

Below we find the spatial high-order moments for the local electric $\mathbf{E}_1$ distribution in the reference plane $z=-d/2-l_0$ (see Fig. 1) back of the film. The electric field $\mathbf{E}_1$ is expressed in terms of fields $\mathbf{E}$ and $\mathbf{H}$ by means of Eq. (16). The fluctuations of the local magnetic current $\mathbf{j}_H(\mathbf{r})=w(\mathbf{r})\mathbf{H}(\mathbf{r})$ can be neglected in the first equation in Eq. (16), as discussed below Eq. (37). Therefore, the moment $M_n^E=\langle|\mathbf{E}_1(\mathbf{r})|^n\rangle/|\langle\mathbf{E}_1\rangle|^n$ equals approximately the moment $M_n=\langle|\mathbf{E}(\mathbf{r})|^n\rangle/|\langle\mathbf{E}\rangle|^n$ for the field $\mathbf{E}(\mathbf{r})$.

We consider now the moments M_n^E in the optical and infrared spectral ranges for arbitrarily strong skin effect. We assume ε_m is almost negative and large in the absolute value. Since Eq. (36) has the same form as the quasistatic equation $\nabla(\varepsilon\nabla\phi)=0$ (investigated in detail in our previous works) we use the estimate for the moments of the local-electric field obtained in Refs. 52 and 53

$$M_n^E\sim\left(\frac{|\tilde{\varepsilon}_m|^{3/2}}{\sqrt{\varepsilon_d}\tilde{\varepsilon}_m''}\right)^{n-1}, \tag{40}$$

where the metal permittivity is replaced by the renormalized dielectric constant $\tilde{\varepsilon}_m$ given by Eq. (35). The Drude formula Eq. (37) is substituted in Eq. (7) to obtain the Ohmic parameter u_m in the limit $\omega_p\gg\omega\gg\omega_\tau$. Then the Ohmic parameter u_m is substituted in Eq. (35) to obtain $\tilde{\varepsilon}_m$. Finally, the moment M_n^E is obtained from Eq. (40) as

$$M_n^E\sim\rho\left[\frac{\omega_p}{\omega_\tau}\sqrt{f_0(x)}\right]^{n-1}, \tag{41}$$

$$f_0(x)=\frac{4\tanh^3(x)[1+D/(4\delta)\tanh(x)]}{x\{\tanh(x)+x[1-\tanh^2(x)]\}^2}, \qquad (42)$$

where $x=d/2\delta\simeq d\omega_p/2c$ is the ratio of the film thickness d and the skin depth $\delta\approx c/\omega_p$. It follows from these equations that the moments of the local-electric field are independent of the frequency in the wide frequency band $\omega_p\gg\omega>\omega_\tau$, which typically includes the optical and infrared spectral ranges. When the skin effect increases, the function f_0 in Eq. (41) also increases monotonically from $f_0(0)=1$ to $f_0(\infty)=D/(2d)$. Provided that the shape of metal grains is fixed and they are very oblate, i.e., $D/d\gg1$, the moments M_n^E increase significantly with increasing the parameter x.

Let us consider now the far-infrared, microwave, and radio frequency ranges, where the metal conductivity σ_m acquires its static value, i.e., it is positive and does not depend on frequency. Then it follows from Eqs. (10), (11), and (35) that Eq. (40) for the field moments acquires the following form

$$M_n^E\sim\left(\frac{2\pi\sigma_m}{\omega}\right)^{(n-1)/2}, \qquad (43)$$

in the limit of strong skin effect. Since metal conductivity is typically much larger than frequency ω in the microwave and radio bands, the moments remain large at these frequencies.

We proceed now with fluctuations for the local magnetic field $\mathbf{H}_1(\mathbf{r})$ in the reference plane $z=-d/2-l_0$. The fluctuations of the field $\mathbf{H}(\mathbf{r})=[\mathbf{H}_1(\mathbf{r})+\mathbf{H}_2(\mathbf{r})]/2$ defined in Eq. (6) can be neglected. Then as follows from the second Eq. (16), the moments $M_n^H=\langle|\mathbf{H}_1(\mathbf{r})|^n\rangle/|\langle\mathbf{H}_1(\mathbf{r})\rangle|^n$ of the local magnetic field can be estimated as

$$M_n^H\simeq(2\pi/c)^n\langle|\mathbf{j}_E(\mathbf{r})|^n\rangle/|\langle\mathbf{E}_1\rangle|^n, \qquad (44)$$

where we used the conditions $|\langle\mathbf{E}_1\rangle|=|\langle\mathbf{H}_1\rangle|$ that correspond to the wave incident onto the film from the right (see Fig. 1). Thus the external electric field induces the electric currents in a semicontinuous metal film and these currents, in turn, generate the strongly fluctuating local magnetic field.

To estimate the moments $\langle|\mathbf{j}_E(\mathbf{r})|^n\rangle$ of the electric current density in semicontinuous metal films we generalize, to include the nonlinear case, the approach suggested earlier by Dykhne[41] (see discussion in Ref. 58). Since in the considered case the electric current $\mathbf{j}_E$ is related to the local field $\mathbf{E}$ via the first equation in Eqs. (6), the following equation $\langle|\mathbf{j}_E|^n\rangle=\alpha(u_m,u_d)\langle|\mathbf{E}(\mathbf{r})|^n\rangle$ is valid [where the coefficient $\alpha(u_m,u_d)$ is a function of variables u_m and u_d].

We consider now the percolation threshold $p=p_c$ and set p_c as $p_c=1/2$. It is also supposed that the statistical properties of the system do not change when interreplacing metal and dielectric. If all the conductivities are increased by the factor k then the average nonlinear current $\langle|\mathbf{j}_E|^n\rangle$ also increases, by the factor $|k|^n$; therefore, the coefficient $\alpha(u_m,u_d)$ also increases, by the $|k|^n$. Then the coefficient $\alpha(u_m,u_d)$ has an important scaling property, namely, $\alpha(ku_m,ku_d)=|k|^n\alpha(u_m,u_d)$. By taking $k=1/u_m$ the following equation is obtained

$$\alpha(u_m,u_d)=|u_m|^n\alpha_1(u_m/u_d). \qquad (45)$$

Now we perform the Dykhne transformation

$$\mathbf{j}^*=[\mathbf{n}\times\mathbf{E}],\quad \mathbf{E}^*=[\mathbf{n}\times\mathbf{j}_E]. \qquad (46)$$

It is easy to verify that thus introduced field $\mathbf{E}^*$ is still potential, i.e., $\nabla\times\mathbf{E}^*=0$, and the current $\mathbf{j}^*$ is conserved, i.e., $\nabla\cdot\mathbf{j}^*=0$. The current $\mathbf{j}^*$ is coupled to the field $\mathbf{E}^*$ by the Ohm's formula $\mathbf{j}^*=u^*\mathbf{E}^*$, where the ''conductivity'' u^* takes values $1/u_m$ and $1/u_d$. Therefore, the following equation $\langle|\mathbf{j}^*|^n\rangle=\alpha(1/u_m,1/u_d)\langle|\mathbf{E}^*|^n\rangle$ holds, from which it follows that $\alpha(1/u_m,1/u_d)\alpha(u_m,u_d)=1$. Since we suppose that at the percolation threshold $p_c=1/2$ and the statistical properties of the system do not change when inter-replacing metal and dielectric, the arguments in the first function can be changed to obtain $\alpha(1/u_d,1/u_m)\alpha(u_m,u_d)=1$. This equation, in turn, can be rewritten using Eq. (45) as $|u_m/u_d|^n\alpha_1^2(u_m/u_d)=1$, where the function α_1 is defined in Eq. (45). Thus we find that $\alpha_1(u_m/u_d)=|u_d/u_m|^{n/2}$, and the final result is given by $\alpha(u_m,u_d)=|u_mu_d|^{n/2}$, i.e., the following generalization of the Dykhne's formula is valid

$$\langle|\mathbf{j}_E|^n\rangle=|u_du_m|^{n/2}\langle|\mathbf{E}|^n\rangle. \qquad (47)$$

Now this expression for $\langle|\mathbf{j}_E|^n\rangle$ can be substituted in Eq. (44) to obtain that

$$M_n^H=\left[\left(\frac{2\pi}{c}\right)^2|u_du_m|\right]^{n/2}M_n^E. \qquad (48)$$

In the optical and infrared spectral ranges it is possible to simplify this equation as done for Eq. (44) above. Using again the Drude formula (37) and assuming that $\omega_\tau\ll\omega\ll\omega_p$, the following estimate is obtained

$$M_n^H=\left[\varepsilon_d'\frac{x\tanh x}{(2d/D)+x\tanh x}\right]^{n/2}M_n^E, \qquad (49)$$

where the moment M_n^E is given by Eq. (44) and $x=d/2\delta\simeq d\omega_p/2c$ has the same meaning as in Eq. (44). As follows from Eq. (49), the spatial moments of the local magnetic field M_n^H are of the same order of magnitude as the moments of the local electric field M_n^E in the limit of strong skin effect, i.e., when $x\gg1$.

We can estimate now the moments of the local electric and magnetic fields from Eqs. (41) and (49) for silver-on-glass semicontinuous films, with $\omega_p=9.1$ eV and $\omega_\tau=0.021$ eV. The moments of the local electric field are as $M_n^E\sim(4\times10^2)^{n-1}$, so that the field fluctuations are huge, in agreement with the numerical results shown in Figs. 2 and 3. For sufficiently strong skin effect ($x>1$), the moments of local magnetic field $M_n^H\sim M_n^E$, which is also in accord with our simulations.

As mentioned, at frequencies much smaller than the relaxation rate $\omega_\tau\simeq3.2\times10^{13}\,\mathrm{sec}^{-1}$, the silver conductivity acquires its static value $\omega_p^2/4\pi\omega_\tau\simeq10^{18}\,\mathrm{sec}^{-1}$. In this case, the moments are given by Eq. (43). Thus for wavelength $\lambda=3$ cm ($\omega/2\pi=\nu=10$ GHz) the moments are as $M_n^H\sim M_n^E\sim(10^4)^{n-1}$. We can conclude that the local electric and magnetic field strongly fluctuate in a very large frequency range from the optical down to the microwave and radio spectral ranges. The fluctuations become even stronger for the microwave and radio bands. This is because for the strong skin

effect (when the penetration depth is much smaller than the size of a metal grain), losses are small in comparison with the electromagnetic field energy accumulated around the film. This opens a fascinating possibility to observe the Anderson localization of surface plasmons, predicted in Refs. 52 and 53, in microwave experiments with localization length in the centimeter scale.

V. ENHANCED LIGHT TRANSMISSION THROUGH METAL FILMS WITH ARRAYS OF SMALL HOLES

Light transmission through a tiny (less than the wavelength) hole in an optically thick metal film is known to be small $T\sim(d/\lambda)^4$.[59] However, provided that there is an array of such subwavelength holes in the metal film and special resonance conditions are fulfilled, the optical transmission can be enhanced by several orders of magnitude. For instance, in a silver film of thickness $d=200$ nm with an array of holes of diameter $D=150$ nm and lattice constant $a_0=600$ nm, large transmission peaks have been observed at wavelengths $\lambda\sim 300$, 400, 500, 700, and 950 nm. The maximum transmission can exceed one, when it is normalized to the projected area of the holes. This corresponds to the enhancement of nearly three orders of magnitude, when compared to what one could expect for the same number of single holes.[60–62] Since the skin depth in silver $\delta\sim 20$ nm is much less than the film thickness, these experiments cannot be explained using the quasistatic approximation for the surface conductivity. Note also that the metal films with periodic arrays of nanoholes represent an example of nanoengineered structures known as electromagnetic crystals.[63–66]

A. Skin resonance

We use the GOL approximation to calculate the transmittance of an array of subwavelength holes for the case of strong skin effect. It is assumed, for simplicity, that the surface hole concentration $p_h=1-p=\pi(D/2a_0)^2$ is small, $p_h\ll 1$, which is typical for the experiments mentioned above. To evaluate the effective parameters u_e and w_e in a sparse array of holes, the Maxwell-Garnet (dipole) approximation can be used.[6,30,67] In this approximation the effective Ohmic parameter, say, u_e is related to the ''metal'' (u_m) and ''dielectric'' (u_d) Ohmic parameters by the following equation

$$u_e=u_m\frac{(1+p_h)u_d+(1-p_h)u_m}{(1-p_h)u_d+(1+p_h)u_m}; \tag{50}$$

the same equation connects w_e to w_m and w_d. The substitution of expressions (10) and (11) in the equations above gives

$$u_e=i\frac{8\pi}{c}\frac{16(1-p_h)-D(2d+D)k^2(1+p_h)}{Dk[16(1+p_h)-D(2d+D)k^2(1-p_h)]} \tag{51}$$

and

$$w_e=i\frac{8\pi}{c}\frac{Dk(d+D+dp_h)}{d+D-dp_h}, \tag{52}$$

where $k=\omega/c$ is the wave vector, d is the thickness of the film, and D is the hole diameter. Then, in the considered limit $p_h\ll 1$, we find the transmittance of the hole array from Eq. (22) in the following form

$$T=\frac{1024p_h^2}{[D(2d+D)k^2-16]^2+1024p_h^2}. \tag{53}$$

That is the transmittance has a resonance at the wavelength $\lambda_r=\sqrt{D(2d+D)}$, where $T(\lambda_r)=1$ and the film becomes *transparent*, regardless of the surface concentration of the holes. When the hole surface concentration p_h decreases, the width of the resonance shrinks, but exactly at the resonance, the film remains transparent, even for $p_h\to 0$.

The nature of the described resonance, which can be denoted as ''*skin resonance*,'' is purely geometrical, i.e., the skin resonance does not depend on the metal-dielectric function ε_m. Therefore, this resonance is different from the well-known plasmon resonance of a hole in the metal film. In particular, the skin resonance should be also observable in the microwave range. It is also important to emphasize that the skin resonance does not actually depend on the hole periodicity.

It would be interesting to check out experimentally the position of the resonance as a function of the holes' diameter D, when the hole concentration p_h is fixed. It is possible that the skin resonance described above was observed in the cited experiments[60,61] at wavelength $\lambda\simeq 300$ nm. Unfortunately, the parameters of the film investigated in Ref. 60 and 61 were chosen in such a way that the frequency of the skin resonance is close to the renormalized plasma frequency $\tilde{\omega}_p$ where the real part of the metal-dielectric constant ε_m' vanishes. Therefore, the strong skin effect condition may not hold in the vicinity of the resonance and losses become important. It could be the reason why the transmittance, although increased up to 20%, does not achieve 100%. Provided that the diameter of the holes is increased, the skin resonance can be shifted to larger wavelengths so that its amplitude can be increased, even at much smaller hole concentration p_h.

Equation (53) is valid in the vicinity of the skin resonance. For wavelengths much larger than the resonance λ_r, the equation for the transmittance T can be simplified since the terms $\propto Dk$ can be neglected. This results in the following expression

$$T=\frac{p_h^2k^2D^4}{4(d+D)^2}. \tag{54}$$

The obtained transmittance T is independent of metal properties (e.g., it does not depend on the metal dielectric constant ε_m). It is an anticipated result for the strong skin effect when the penetration of the electromagnetic field in metal can be neglected. The transmittance is proportional to the ratio of the hole size D and the wavelength λ squared. Therefore, the transmittance T is much larger than $T\sim(D/\lambda)^4$ resulting from the Fraunhofer diffraction on single holes in the limit of $D/\lambda\ll 1$. It is the ''background'' transmittance, above which the considered hereafter resonances are imposed.

B. Surface polariton resonance

Consider now surface waves in a metal film. When the real part of the metal permittivity is negative $\varepsilon_m' < 0$ and losses are small, it is well known that surface polaritons can propagate along the surface. On metal surfaces, these waves are referred to as surface plasmon polaritons.

For the sake of simplicity we neglect losses in the further consideration, i.e., we assume that the metal-dielectric constant is negative $\varepsilon_m = -n^2$; we also suppose that $n > 1$. The wave vector k_p of the plasmon polariton in the metal-vacuum interface is given by $k_p = kn/\sqrt{(n^2-1)} > k$ (see e.g., Refs. 28 and 39). There are two kinds of polaritons in the film with a finite thickness, which correspond to symmetric and antisymmetric (with respect to reflection in the plane $z=0$) solutions to the Maxwell equations. The wave vectors k_1 and k_2 of these polaritons are determined by the following equations

$$k_1^2 = (kn)^2 \frac{n^2 + \tanh^2[(d/2)\sqrt{(kn)^2 + k_1^2}]}{n^4 - \tanh^2[(d/2)\sqrt{(kn)^2 + k_1^2}]}, \tag{55}$$

$$k_2^2 = (kn)^2 \frac{1 + n^2 \tanh^2[(d/2)\sqrt{(kn)^2 + k_2^2}]}{n^4 \tanh^2[(d/2)\sqrt{(kn)^2 + k_2^2}] - 1}. \tag{56}$$

In the case of the strong skin effect, when $\exp(dkn) \gg 1$, the wave vectors for the symmetric and antisymmetric polaritons are equal to

$$k_1^2 = (kn)^2 \frac{n^4 - 1 - 4n^2 \exp(dkn^2/\sqrt{n^2-1})}{(n^2-1)(n^4-1)} \tag{57}$$

and

$$k_2^2 = (kn)^2 \frac{n^4 - 1 + 4n^2 \exp(dkn^2/\sqrt{n^2-1})}{(n^2-1)(n^4-1)}, \tag{58}$$

respectively. It is important for the further consideration that both symmetric and antisymmetric polaritons propagate on both interfaces of the film. Moreover, the absolute value of the electric and magnetic fields are the same on the two interfaces. This consideration holds for arbitrarily thick film, although the difference between the two types of polaritons becomes exponentially small for the optically thick films.

Since the polariton wave vectors are larger than the wave vector k of an electromagnetic wave (normal to the film) the wave cannot excite the polaritons in a continuous metal film (see, e.g., Ref. 28). The situation changes dramatically when the film is periodically corrugated, with the same spatial period a_0 on both interfaces. The example of such corrugation is a regular array of holes. When wave vector k of the incident wave is such that one of the polariton wavelengths $\lambda_1(k) = 2\pi/k_1(k)$ or $\lambda_2(k) = 2\pi/k_2(k)$ coincides with a_0, the corresponding polariton is excited on the film. This polariton spreads out onto both sides of the film and interacts with the corrugation. As a result of this interaction, the excited polariton can be transformed back to the plane wave. This can also occur on the backside of the film. Therefore the transmittance has a maximum at the resonance condition $2\pi m/a_0 = k_i \{i = 1,2; m = 1,2, \ldots\}$. Since the polariton amplitude is the same on both sides of the film we speculate that at the resonance the film becomes almost transparent, regardless of its thickness, though the width of the resonance shrinks when the film thickness increases. The set of maxima in transmittance was indeed observed in experiments of Refs. 60 and 61.

For a qualitative analysis of the resonance transmittance we consider a simple model of the periodically inhomogeneous film, with the local dielectric constant varying as $\varepsilon = -n^2[1 - g\cos(qx)]$ in the film plane $\{x,y\}$. Then, the whole spectrum of the spatial harmonics $\sim \cos(q_m x)$ is generated in the film, with the wave vectors $q_m = mq$. We consider, for simplicity, small modulations assuming that parameter $g \ll 1$. Then, we can restrict our consideration to the first two harmonics, i.e., we suggest that the local electric and magnetic fields can be written as

$$\tilde{\mathbf{E}}(x,z) = \mathbf{E}(z) + \mathbf{E}_q(z)\cos(qx), \tag{59}$$

$$\tilde{\mathbf{H}}(x,z) = \mathbf{H}(z) + \mathbf{H}_q(z)\cos(qx), \tag{60}$$

where, as above, the ''z'' axis is perpendicular to the film plane. The wavelength λ of the incident wave is supposed to be larger than the period of the film modulation $\lambda > a_0 = 2\pi/q$. The Maxwell equations in the film can be written as

$$\mathrm{curl}[(1 + g\cos(qx))\mathrm{curl}\ \tilde{\mathbf{H}}] = -(kn)^2 \tilde{\mathbf{H}}. \tag{61}$$

We consider the incident wave with the magnetic and electric fields polarized along the ''y'' and ''x'' axes. Then the magnetic field $\tilde{\mathbf{H}}$ in the film has the ''y'' component only and Eq. (61) takes the following form

$$H''(z) + \frac{g}{2} H_q''(z) - k^2 n^2 H(z) = 0, \tag{62}$$

$$H_q''(z) + gH''(z) - (k^2 n^2 + q^2) H_q(z) = 0, \tag{63}$$

where we neglected the higher spatial harmonics and equate the terms that have the same dependence on the coordinate x.[23] Solutions to Eqs. (62) and (63) give the magnetic field inside the film. The electric fields $\mathbf{E}(z)$ and $\mathbf{E}_q(z)$ are obtained from the second Maxwell equation $\tilde{\mathbf{E}} = i/[kn^2(1 - g\cos(qx))]\mathrm{curl}\ \tilde{\mathbf{H}}$. The internal fields $\mathbf{E}(z)$ and $\mathbf{H}(z)$ are matched with the field of the incident wave at the front interface of the film ($z = d/2$) and these fields, in turn, are matched with the field of the transmitted wave at the back interface ($z = -d/2$). Note that the fields $\mathbf{E}_q(z)$ and $\mathbf{H}_q(z)$ decay exponentially $\sim \exp(-|z|\sqrt{q^2 - k^2})$ outside the film since the spatial period of the corrugation is smaller than the wavelength of the incident wave $\lambda > a_0\ (k < q)$. Thus Eqs. (62) and (63) connect incident and transmitted wave and its solution gives the transmittance T of the film. The transmittance T, as discussed above, has sharp maxima at the plasmon-polariton resonances. At the resonance, T acquires the following form

$$T(k)=\frac{(gkn^3)^4}{(gkn^3)^4+(n^2+1)(n^4-1)^3\{1+4n[2n(n^2-1)+\sqrt{n^2-1}(2n^2-1)]\}\Delta^2(k)}, \tag{64}$$

with the detuning from the resonance Δ given by

$$\Delta(k)=k_i^2(k)+\frac{2\sqrt{n^2-1}(n^6+1)-n(3-n^4+2n^6)}{2n(n^4-1)^2(n^2+1)}\times(gkn^3)^2-q^2, \tag{65}$$

where $k_i(k)=k_1$ or k_2 defined in Eqs. (55)–(58). Thus we obtain the important result that there is a subwavelength resonance in transmittance of a corrugated metal film due to surface polaritons. The resonance has a form of two peaks close to each other since $k_2-k_1\sim\exp(-dkn)\ll 1$. When the skin effect is so strong that $\exp(-dkn)\ll g^2$ these two peaks merge together. Exactly at the resonance $T=1$, that is, the film becomes transparent at arbitrary thickness. In a real film the transmittance is limited by losses. Yet, the losses are rather small for a typical metal in the optical and infrared spectral ranges. Note also that the resonance position is defined by the condition $\Delta=0$ [see Eq. (64)]; it is somewhat shifted with respect to the condition $k_i(k)=q$ since the corrugation of a film affects the propagation conditions for surface polaritons.

Similar resonances take place when $k_i(k)\simeq mq$. We believe that the skin resonance described in Sec. V A is responsible for the sharp short-wavelength peak (near 300 nm) obtained in Ref. 60, whereas long-wavelength resonances of Ref. 60 occur due to surface polaritons considered here. A quantitative comparison with the experiments will be presented elsewhere. Now we only note that the quartz substrate used in the experiment makes the film asymmetric. Therefore, surface plasmon polaritons propagate in a different way on the two sides of the film. This is a possible reason why the observed resonance transmittance, though strongly enhanced, does not exceed 10%.

VI. SUMMARY AND CONCLUSIONS

In this paper a detailed theoretical consideration of the high-frequency response (optical, infrared, and microwave) of thin metal-dielectric films is presented. The generalized GOL approximation was employed. The developed GOL approximation is based on direct solution of Maxwell's equations, without having to invoke the quasistatic approximation. In this approximation the electromagnetic properties of semicontinuous metal films are described in terms of *two* parameters u_e and w_e, in contrast to the usual description with a single complex conductivity. This approach allows us to calculate field distributions and optical properties of semicontinuous metal films in a wide frequency range: from the optical to the microwave and radio frequencies.

It is shown that metal-dielectric films can exhibit especially interesting properties when there is a strong skin effect in metal grains. In particular, the theory predicts that the local magnetic fields, as well as the electric fields, strongly fluctuate within the large optics-to-microwave spectral range. The obtained equations for the field distributions and the high-order field moments for the electric and magnetic fields allow one to describe various optical phenomena in percolation films, both linear and nonlinear. For example, surface enhancement for Raman scattering is proportional to the fourth field moment[46,48] and, therefore, it is strongly enhanced in a wide spectral range. The same is valid for the Kerr nonlinearity, which is also proportional to the fourth moment.[47,52] The giant electric-field fluctuations near the percolation threshold in metal-dielectric films have been already observed in the microwave[20] and optical[21] ranges.

The large electric- and magnetic-field fluctuations explain, in particular, a number of previously obtained phenomena that remained so far unclear. For example, the absorbance in a percolation film calculated using the quasistatic approximation predicts for the maximum $A\simeq 0.2$,[35] which is twice less than the measured value[13–16,20] and the one following from the GOL equations.[18] This can occur because of neglecting the magnetic field in the quasistatic calculations. The energy of the electric field can be converted into the energy of the magnetic field, so that the magnetic component of the em field is also responsible for the absorption. As seen in Figs. 2 and 3, the magnetic and electric fields can be comparable in magnitudes, even at a relatively moderate skin effect. This indicates that the magnetic field can carry out roughly the same amount of energy as the electric field. In accordance with this, Figs. 4–6 show that the absorbance in the GOL beyond-the-quasistatic-approximation reaches the value $A\simeq 0.45$ (in agreement with experiments), which is roughly twice larger than that found in the quasistatic approximation.

We also considered the optical properties of a metal film with an array of subwavelength holes and showed that transmittance of such a film is much larger than the Fraunhofer diffraction predicts. This result is an agreement with recent experiments.[60–62] For such films, a new effect, skin resonance, is predicted; at this resonance the transmittance can increase up to 100%, regardless of the surface concentration of the holes. We also predict series of the surface plasmon-polaritons resonances in the transmittance.

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