

## Percolation-enhanced nonlinear scattering from metal-dielectric composites

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It is shown that large percolation-enhanced nonlinear scattering occurs in metal-dielectric random composites near the percolation threshold. The enhancement is due to giant local electric field fluctuations that are extremely inhomogeneous and consist of spatially separated sharp peaks, “hot” spots, where the local field is greater by many orders of magnitude than the applied field. [S1063-651X(99)00106-3]

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The local fields can experience strong enhancement in the visible and infrared spectral ranges, for metal-dielectric composites comprising metal particles that are characterized by dielectric constant  $\varepsilon_m = \varepsilon'_m + i\varepsilon''_m$ , with the negative real part,  $\varepsilon'_m < 0$ , and the small imaginary part,  $\varepsilon''_m/|\varepsilon'_m| \ll 1$ . In a simplest case of a spherical metal particle, the local electric field  $\propto (2\varepsilon_d + \varepsilon_m)^{-1}$ , and it is strongly enhanced at the plasmon resonance, when  $\varepsilon'_m(\omega) \cong -2\varepsilon_d$  ( $\varepsilon_d$  is the permittivity of a dielectric substrate) [1]. In general, the local field  $\mathbf{E}(\mathbf{r})$  enhancement is due to the collective plasmon resonances in ensembles of metal particles [1–3]. Nonlinear optical processes of the  $n$ th order are proportional to  $E^n(\mathbf{r})$  and, therefore, the enhancement can be especially large.

In this paper we consider percolation-enhanced nonlinear scattering (PENS) from a random metal-dielectric film (also referred to as a semicontinuous metal film) at the metal filling factor  $p$  close to the percolation threshold  $p_c$ . Specifically, we study the enhanced nonlinear scattering resulting in a field oscillating at frequency  $n\omega$ , when a percolating metal-dielectric film is illuminated by an electromagnetic wave of frequency  $\omega$ . At the percolation, an infinite metal cluster spans over the entire sample and the metal-dielectric transition occurs in a semicontinuous metal film [1]. Optical excitations of the self-similar fractal clusters formed by metal particles near  $p_c$  result in giant, scale-invariant, local-field fluctuations that make possible PENS. Linear Rayleigh scattering from semicontinuous metal films has been considered in a recent paper [4]. It was shown that while Rayleigh scattering is strongly enhanced, it is still only a small correction to the specular reflection and transmission. In contrast, below we show that PENS with a broad angular distribution can be a leading optical process.

For simplicity, we assume that a semicontinuous film is illuminated by the light propagating normal to the film, with the wavelength  $\lambda$  larger than any intrinsic spatial scale in the film, including the skin depth,  $\lambda \gg a_0 \sqrt{|\varepsilon_m|}$ , where  $a_0$  is the grain size. (All distances hereafter are given in units  $a_0 = 1$ .) The gaps between metal grains are filled by the dielectric substrate so that a semicontinuous metal film can be thought of as a two-dimensional array of metal and dielectric grains that are randomly distributed over the plane. For an incident

wave of frequency  $\omega$ , we consider  $n$ th harmonic generation ( $n$ HG) in a percolating film. We assume that a semicontinuous metal film is covered by a layer possessing the nonlinear conductivity  $\sigma^{(n)}$  that results in  $n$ HG. The local electric field  $\mathbf{E}_\omega(\mathbf{r})$  induced in the film by the external field  $\mathbf{E}_\omega^{(0)}$  generates in the layer the  $n\omega$  current  $\sigma^{(n)}\mathbf{E}_\omega E_\omega^{n-1}$  [5]. This nonlinear current, in turn, interacts with the film and generates “seed”  $n\omega$  electric field, with the amplitude  $\mathbf{E}^{(n)} = \sigma^{(n)}E_\omega^{n-1}\mathbf{E}_\omega/\sigma^{(1)}$ , where  $\sigma^{(1)}$  is the linear conductivity of the nonlinear layer at frequency  $n\omega$ . The electric field  $\mathbf{E}^{(n)}$  can be thought of as an inhomogeneous external field excit-

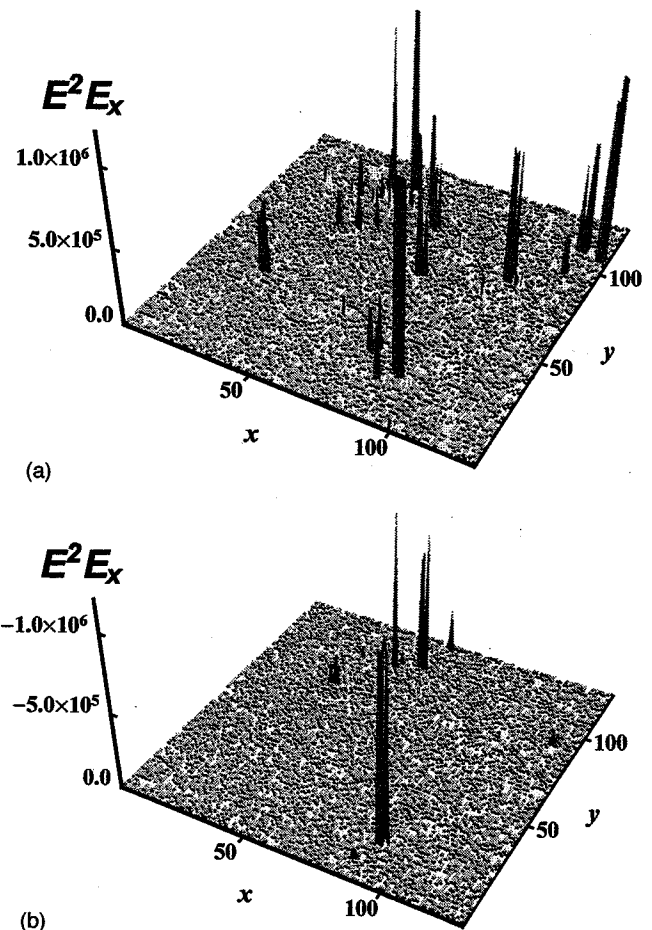


FIG. 1. Distribution of the  $x$  component of the “nonlinear” local field  $\text{Re}[E^2(\mathbf{r})E_x(\mathbf{r})]$ . The applied field  $E_a^{(0)} = 1$ ,  $E_y^{(0)} = 0$ .

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ing the film at  $n\omega$  frequency. The  $n$ HG current  $\mathbf{j}^{(n)}$  induced in the film by the "seed" field  $\mathbf{E}^{(n)}$  can be found in terms of the nonlocal conductivity tensor  $\hat{\Sigma}(\mathbf{r}, \mathbf{r}')$  introduced in [3] that relates the applied (external) field at point  $\mathbf{r}'$  to the current at point  $\mathbf{r}$ ,

$$\mathbf{j}_\beta^{(n)}(\mathbf{r}) = \int \Sigma_{\beta\alpha}^{(n)}(\mathbf{r}, \mathbf{r}') E_\alpha^{(n)}(\mathbf{r}') d\mathbf{r}', \quad (1)$$

where  $\Sigma_{\beta\alpha}^{(n)}$  is the conductivity tensor at frequency  $n\omega$  and the integration is over the entire film area [3]. The Greek indices take values  $\{x, y\}$  and summation over repeated indices is implied. It is the current  $\mathbf{j}^{(n)}$  that eventually generates the nonlinear scattered field at the frequency  $n\omega$ .

Using the numerical technique described in detail in [3], we calculate the local-field spatial distribution. For example, in Fig. 1 we show the normalized real part of the  $3\omega$  local field  $\text{Re}[E^2(\mathbf{r})E_x(\mathbf{r})]/|E^{(0)}|^3$  in a  $2d$  silver-on-glass film at  $p=p_c$  and  $\lambda=1.5 \mu\text{m}$ . For the silver dielectric constant we use the Drude formula  $\epsilon_m(\omega) = \epsilon_b - (\omega_p/\omega)^2/[1+i\omega\tau/\omega]$ , where the interband-transition contribution  $\epsilon_b=5.0$ , the plasma frequency  $\omega_p=9.1 \text{ eV}$ , and the relaxation frequency

$\omega_\tau=0.021 \text{ eV}$  [6]. As seen in Fig. 1, the fluctuating  $3\omega$  fields form a set of sharp peaks, looking up and down, and having the magnitudes  $\sim 10^6$ . Such huge fluctuations of the local fields are anticipated to trigger the PENS at the frequency  $3\omega$ . The larger the number  $n$  of the harmonic, the stronger the corresponding  $n\omega$  local field fluctuates. Therefore we speculate that the enhancement factor for PENS becomes progressively larger for higher harmonics.

By using the standard approach of the scattering theory adopted for semicontinuous metal films in [4] and assuming that the incident light is unpolarized, we obtain that the integral scattering in all directions but the specular one, i.e., the diffusive scattering, is

$$S = (4k^2/\beta c) \int [\langle j_\alpha^{(n)}(\mathbf{r}_1) j_\alpha^{(n)*}(\mathbf{r}_2) \rangle - |\langle \mathbf{j}^{(n)} \rangle|^2] d\mathbf{r}_1 d\mathbf{r}_2, \quad (2)$$

where the integration is over the entire area  $A$  of the film,  $k=\omega/c$ , and the angular brackets stand for the ensemble average. As in [4], we assume that the integrand vanishes in distances  $r \ll \lambda$ , where  $\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1$ ; therefore, we omit the term  $\sim \exp(i\mathbf{k} \cdot \mathbf{r})$ . By substituting Eq. (1) in Eq. (2), we obtain

$$\int \langle j_\alpha^{(n)}(\mathbf{r}_1) j_\alpha^{(n)*}(\mathbf{r}_2) \rangle d\mathbf{r}_1 d\mathbf{r}_2 = \int \langle \Sigma_{\gamma\beta}^{(n)}(\mathbf{r}_1, \mathbf{r}_3) \Sigma_{\delta\alpha}^{(n)*}(\mathbf{r}_2, \mathbf{r}_4) \delta_{\gamma\delta} \{ E_\beta^{(n)}(\mathbf{r}_3) E_\alpha^{(n)*}(\mathbf{r}_4) \} \rangle \prod_{i=1}^4 d\mathbf{r}_i, \quad (3)$$

where  $\{\dots\}$  denotes the averaging over the light polarizations. For the unpolarized light we have the expression  $\delta_{\gamma\delta} = 2\{E_{n\omega, \gamma}^{(0)*} E_{n\omega, \delta}^{(0)}\}/|E_{n\omega}^{(0)}|^2$ , where  $\mathbf{E}_{n\omega}^{(0)}$  is the amplitude of the uniform "probe" field at the frequency  $n\omega$ . We substitute this expression for  $\delta_{\gamma\delta}$  in Eq. (3), integrate over the coordinates  $\mathbf{r}_1, \mathbf{r}_2$ , and average over independent polarizations of fields  $E_{n\omega}^{(0)}$  and  $E_\omega^{(0)}$ . Thus we obtain

$$\int \langle j_\alpha^{(n)}(\mathbf{r}_1) j_\alpha^{(n)*}(\mathbf{r}_2) \rangle d\mathbf{r}_1 d\mathbf{r}_2 = \frac{1}{|E_{n\omega}^{(0)}|^2} \int \langle \sigma_{n\omega}(\mathbf{r}_3) \sigma_{n\omega}^*(\mathbf{r}_4) [\mathbf{E}_{n\omega}(\mathbf{r}_3) \cdot \mathbf{E}_{n\omega}^*(\mathbf{r}_4)] [\mathbf{E}^{(n)}(\mathbf{r}_3) \cdot \mathbf{E}^{(n)*}(\mathbf{r}_4)] \rangle d\mathbf{r}_3 d\mathbf{r}_4, \quad (4)$$

where  $\mathbf{E}_{n\omega}(\mathbf{r})$  is the local  $n\omega$  field excited in the film by "probe" field  $\mathbf{E}_{n\omega}^{(0)}$ , and  $\sigma_{n\omega}(\mathbf{r})$  is the film conductivity at frequency  $n\omega$ . In the macroscopically homogeneous and isotropic film considered here, the current correlator given by Eq. (4) does not depend on the direction of the probe field  $\mathbf{E}_{n\omega}^{(0)}$ . Therefore, we can choose now the field  $\mathbf{E}_{n\omega}^{(0)}$  to be collinear with external field,  $\mathbf{E}_{n\omega}^{(0)} \parallel \mathbf{E}_\omega^{(0)}$ . The average nonlinear current  $\langle \mathbf{j}^{(n)} \rangle$  is aligned with  $\mathbf{E}_\omega^{(0)}$  and, therefore, the square of the nonlinear current can be written as  $|\langle \mathbf{j}^{(n)} \rangle|^2 = |\langle \mathbf{E}_{n\omega}^{(0)} \cdot \mathbf{j}^{(n)} \rangle|^2 / |E_{n\omega}^{(0)}|^2$ . Using Eq. (1) for  $\mathbf{j}^{(n)}$ , we find

$$|\langle \mathbf{j}^{(n)} \rangle|^2 = \left| A^{-1} \int E_{n\omega, \beta}^{(0)} \Sigma_{\beta\alpha}^{(n)}(\mathbf{r}_1, \mathbf{r}_2) E_\alpha^{(n)}(\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2 \right|^2 / |E_{n\omega}^{(0)}|^2 = |\langle \sigma_{n\omega}(\mathbf{E}_{n\omega} \cdot \mathbf{E}^{(n)}) \rangle|^2 / |E_{n\omega}^{(0)}|^2. \quad (5)$$

By substituting Eqs. (4) and (5) in Eq. (2), we obtain

$$S = \frac{8\pi k^2}{3c |E_{n\omega}^{(0)}|^2} \left| \frac{\sigma^{(n)}}{\sigma^{(1)}} \right|^2 A \langle |\sigma_{n\omega} \mathbf{E}_{n\omega}|^2 |\mathbf{E}_\omega|^2 |E_\omega|^{2(n-1)} \rangle \int_0^\infty g^{(n)}(r) r dr, \quad (6)$$

where  $g^{(n)}(r)$  is the nonlinear correlation function defined as

$$g^{(n)}(r) = \frac{\langle \sigma_{n\omega}(\mathbf{r}_1) \sigma_{n\omega}^*(\mathbf{r}_2) (\mathbf{E}_{n\omega}(\mathbf{r}_1) \cdot \mathbf{E}_{n\omega}^*(\mathbf{r}_2)) (\mathbf{E}^{(n)}(\mathbf{r}_1) \cdot \mathbf{E}^{(n)*}(\mathbf{r}_2)) \rangle - |\langle \sigma_{n\omega}(\mathbf{E}^{(n)} \cdot \mathbf{E}_{n\omega}) \rangle|^2}{\langle |\sigma_{n\omega} \mathbf{E}_{n\omega}|^2 |\mathbf{E}^{(n)}|^2 \rangle}, \quad (7)$$

which depends only on the distance  $r = |\mathbf{r}_1 - \mathbf{r}_2|$  between points  $\mathbf{r}_1$  and  $\mathbf{r}_2$  for macroscopically homogeneous and isotropic films. We compare this PENS with the  $n\omega$  signal  $I_{n\omega}$  from a nonlinear layer on a dielectric film with no metal grains on it,  $I_{n\omega} = (c\varepsilon_d^2/2\pi)A|\sigma^{(n)}/\sigma^{(1)}|^2|E_\omega^{(0)}|^2|E_\omega^{(0)}|^{2(n-1)}$ . By expressing the enhancement factor for PENS,  $G^{(n)} = S/I_{n\omega}$ , in terms of the local dielectric constant  $\varepsilon_{n\omega}$  at the frequency  $n\omega$ , we obtain

$$G^{(n)} = \frac{(ka_0)^4}{3} \frac{\langle |\varepsilon_{n\omega} \mathbf{E}_{n\omega}|^2 |E_\omega|^{2(n-1)} \rangle n^2}{\varepsilon_d^2 |E_{n\omega}^{(0)}|^2 |E_\omega^{(0)}|^{2(n-1)} a_0^2} \times \int_0^\infty g^{(n)}(r) r dr. \quad (8)$$

Note that for a homogeneous ( $p=0$  and  $p=1$ ) surface  $g^{(n)}(r)=0$  and, therefore,  $G^{(n)}=0$ , so that the scattering occurs in the reflected direction only. Besides the small factor  $(ka_0)^4$ , which is similar to that in the standard Rayleigh scattering, the enhancement  $G^{(n)}$  for PENS is proportional to the  $2(n+1)$  power of the local field. For highly fluctuating local fields, this factor can be very large (see Fig. 1). To understand the origin of the strong fluctuations, we consider below the local field distribution in more detail.

Because  $\lambda \gg a_0 \sqrt{|\varepsilon_m|}$ , we can introduce a potential  $\phi(\mathbf{r})$  for the local electric field, and the field distribution problem reduces to solving the equation representing the current conservation law,  $\nabla \cdot (\varepsilon(\mathbf{r})[-\nabla\phi(\mathbf{r}) + \mathbf{E}^{(0)}(\mathbf{r})]) = 0$ . Being discretized, this relation acquires the form of Kirchhoff's equations defined on a square lattice [1]. Kirchhoff's equations, in turn, can be written in the matrix form, with the local dielectric constants as the matrix elements of the Hamiltonian, which we refer to hereafter as Kirchhoff's Hamiltonian (KH). The off-diagonal elements of the KH are  $H_{ij} = -\varepsilon_{ij}$  and the diagonal elements are  $H_{ii} = \sum_j \varepsilon_{ij}$ , where  $j$  refers to nearest neighbors of  $i$ . The dielectric constants  $\varepsilon_{ij}$  take values  $\varepsilon_m$  and  $\varepsilon_d$ , with probabilities  $p_c$  and  $(1-p_c)$ , respec-

tively. In this form, the KH is of Anderson's type, with both on- and off-diagonal correlated disorder [7]. We consider first the case when  $-\varepsilon'_m \approx \varepsilon_d \sim 1$  and the loss factor  $\kappa = \varepsilon''_m/|\varepsilon_m| \ll 1$ . Now we express the fluctuating potential  $\phi(\mathbf{r})$  in terms of the eigenvalues  $\Lambda_n$  and eigenfunctions  $\Psi_n$  of the real part  $H'$  of the non-Hermitian KH,  $H = H' + iH''$ , where  $H'' \propto \kappa$  is small since  $\kappa \ll 1$ . Then, the problem formally maps the Anderson quantum-mechanical problem, and the eigenfunctions  $\Psi_n$  are all localized (see Fig. 1) in the considered  $2d$  case [7]. Then we neglect the overlapping of the eigenstates in calculating the moments  $M_n \equiv \langle |E(\mathbf{r})/E^{(0)}|^n \rangle$ , assuming that  $\langle |\Psi_n| |\Psi_m| \rangle \sim \delta_{nm}$ . Thus we obtain  $M_n \sim \int \rho(\Lambda) / |\Lambda + ib\kappa|^n d\Lambda \sim \kappa^{-n+1}$  ( $\kappa \rightarrow 0$ ), for  $n \geq 2$ , where  $\rho(\Lambda)$  is the density of the eigenstates of KH of  $H'$  and  $ib\kappa$  is a small correction to the eigenvalue  $\Lambda$  due to  $\kappa \neq 0$  ( $b \sim 1$ ). The fluctuations tend to infinity [ $M_n \sim (|\varepsilon_m/\varepsilon''_m|)^{n-1} \rightarrow \infty$ ] when losses vanish in the system. For the large contrast  $|\varepsilon_m/\varepsilon_d| \gg 1$  the moments become even larger,  $M_n \sim (|\varepsilon_m/\varepsilon_d|)^{(n-1)/2} (|\varepsilon_m/\varepsilon''_m|)^{n-1}$  [3].

It follows from the above consideration that the eigenstates  $\Psi_n$  with  $\Lambda_n \approx 0$  are in resonance with external field and are excited indeed. Correspondingly, the local-field fluctuations are of the resonant character, and the field correlation function  $g^{(3)}(r)$ , shown in Fig. 2 for a silver-glass semicontinuous film, drops very rapidly for  $r > 1$  and has a negative minimum. The anticorrelation occurs because the field maxima have different signs, as seen in Fig. 1. The power-law decrease of  $g^{(3)}(r)$ , which is typical for critical phenomena, occurs in the tail only (see the inset in Fig. 2). The correlation function  $g^{(3)}(r)$  departs from the power-law (the straight line in Fig. 2), for  $r$  larger than field correlation length  $\xi_e$ , which was estimated in [3,4] as  $\xi_e \sim |\varepsilon_m|/\sqrt{\varepsilon_d \varepsilon''_m}$ . This estimate gives  $\xi_e \approx 5, 20,$  and  $30$  (in  $a_0$  units), for  $\lambda = 0.34, 0.53$  and  $0.9 \mu\text{m}$ , respectively, which is in agreement with our numerical results (inset in Fig. 2). The integral of  $g^{(3)}(r)$  in Eq. (8) is about unity, for all frequencies. Based on the above consideration we estimate that  $a_0^{-2} \int_0^\infty g^{(n)}(r) r dr \sim 1$  for all  $n$ .

To estimate PENS given by Eq. (8) quantitatively we take into account that the spatial scale  $\xi_e(\omega)$  for the field fluctua-

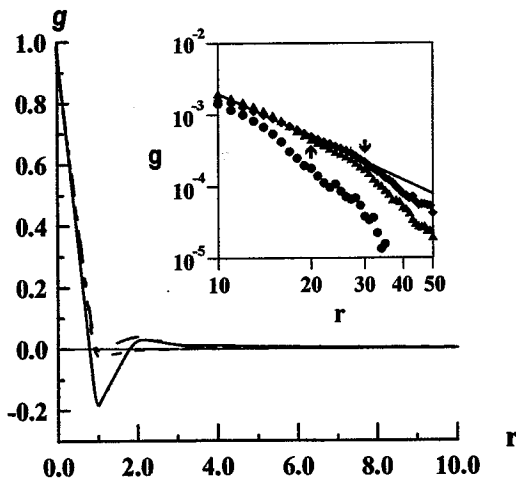


FIG. 2. Correlation function  $g^{(3)}(r)$  for silver on glass semicontinuous film at the percolation threshold  $p = p_c$ . Different curves correspond to  $\lambda_1 = 0.34 \mu\text{m}$  (solid line and circles, in the inset),  $\lambda_2 = 0.53 \mu\text{m}$  (dashed line and triangles), and  $\lambda_3 = 0.9 \mu\text{m}$  (point-dashed line and diamonds); the arrows are theoretical estimates for  $\xi_e(\lambda_2)$  and  $\xi_e(\lambda_3)$ . The straight line illustrates the scaling dependence of  $g^{(3)}(r)$  in the tail. The units  $a_0 = 1$  are used.

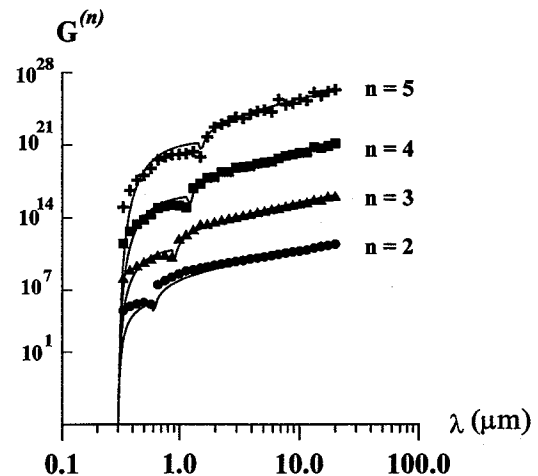


FIG. 3. PENS factor  $G^{(n)}$  for  $n$ -harmonic generation in a silver semicontinuous film at  $p = p_c$ . Numerical calculations for  $n = 2, 3, 4,$  and  $5$  are represented by  $\bullet, \blacktriangle, \blacksquare,$  and  $+$ , respectively. The solid lines describe  $G^{(n)}$  found from Eq. (6).

tions at the fundamental frequency  $\omega$  is significantly larger than  $\xi_e(n\omega)$  at the generated frequency  $n\omega$ . Therefore, we can decouple the average  $\langle |\varepsilon_{n\omega} E_{n\omega}|^2 |E_\omega|^2 |E_\omega|^{2(n-1)} \rangle$  in Eq. (8) and approximate it by  $\langle |\varepsilon_{n\omega} E_{n\omega}|^2 |E_\omega|^{2(n-1)} \rangle \sim \langle |\varepsilon_{n\omega} E_{n\omega}|^2 \rangle |E_\omega|^{2(n-1)}$ . The second moment of the current  $\langle |\varepsilon_{n\omega} E_{n\omega}|^2 \rangle$  we estimated as  $\langle |\varepsilon_{n\omega} E_{n\omega}|^2 \rangle \sim |E_{n\omega}^{(0)}|^2 |\varepsilon_m(n\omega)|^{5/2} \varepsilon_d^{1/2} / \varepsilon_m''(n\omega)$  according to [4]. Then Eq. (8) takes the form

$$\frac{G^{(n)}}{(ka_0)^4} \sim C n^2 \frac{|\varepsilon_m(n\omega)|^{5/2} |\varepsilon_m(\omega)|^{3(2n-1)/2}}{\varepsilon_d^{n+1} \varepsilon_m''(n\omega) \varepsilon_m''(\omega)^{2n-1}}, \quad (9)$$

where  $C$  is an adjustable prefactor. For the Drude metal and  $\omega, n\omega \ll \omega_p$ , we can simplify Eq. (9) as

$$G^{(n)} \sim C (ka_0)^4 \frac{1}{\varepsilon_d^{n+1}} \left( \frac{\omega_p}{\omega_\tau} \right)^{2n} \left( \frac{\omega_p}{\omega} \right)^2, \quad (10)$$

i.e., PENS increases with increasing the order of a nonlinear process and decreases toward the infrared part of the spectrum as  $G^{(n)} \propto \lambda^{-2}$ , in contrast to the well-known law  $\lambda^{-4}$  for Rayleigh scattering. It is interesting to note that PENS is

inversely proportional to the wavelength squared for high-harmonic scattering, independently of the order of optical nonlinearity.

In Fig. 3 we compare the numerical results for the PENS factors  $G^{(n)}$  with the predictions of the scaling formula (9), where we used  $C \approx 10^{-3}$  (note that  $C$  is small because the decoupling used above is, of course, the upper estimate). For a very large spectral interval, there is good agreement between the developed scaling theory and numerical calculations. The PENS effect appears to be really huge, e.g., the enhancement for the fifth harmonic generation is  $G^{(5)}/(ka_0)^4 \sim 10^{21}$ , for  $\lambda = 1.5 \mu\text{m}$ . Note that the diffusive scattering was observed in experiments on second-harmonic generation from semicontinuous silver films [8].

To summarize, large-field fluctuations in random metal-dielectric composites near percolation result in the percolation-enhanced nonlinear scattering (PENS), which is characterized by giant enhancement and a broad-angle distribution.

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$E^{n-1} = (\mathbf{E} \cdot \mathbf{E})^k$ . However, for estimates, the formula can be used in the general case, for arbitrary  $n$ .

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