Quantum Fluctuations of the Refractive Index near the Interface Between a Metal and a Nonlinear Dielectric

Igor I. Smolyaninov

Department of Electrical and Computer Engineering, University of Maryland, College Park, Maryland 20742, USA

(Received 16 September 2004; published 7 February 2005; publisher error corrected 11 February 2005)

Zero-point fluctuations of surface plasmon modes near the interface between a metal and a nonlinear dielectric are shown to produce a thin layer of shifted fluctuating dielectric constant near the interface. The shift of the dielectric constant in this layer may be sufficiently large to produce multiple metastable states of the surface plasmon vacuum.

DOI: 10.1103/PhysRevLett.94.057403 PACS numbers: 78.67.–n, 42.50.–p, 42.65.–k

Nonlinear optical effects at low light intensities is a topic of great current interest because of their potential implementations in quantum computing and communication [1]. The use of surface electromagnetic waves such as surface plasmon-polaritons (SP) [2,3] is known to reduce considerably the power levels that are necessary to achieve nonlinear optical effects [4,5]. Such a power reduction is possible because of considerable enhancement of the optical field near metal interfaces. Very recently a possible observation of the optical bistability effect at single-photon levels due to localized plasmon excitation in nonlinear nanoholes in gold films has been reported [6]. The experimental findings were confirmed by theoretical calculations in [6,7]. This development together with other photon-blockade effects [8] observed in nonlinear optical cavities at single-photon levels may open new ways of designing optical quantum computing and communication devices. It justifies the continued interest in nonlinear optical effects at very low light levels.

As we continue to reduce the number of energy quanta in a SP system, and still observe nonlinear optical effects, we may ask ourselves if the properties of the nonlinear dielectric near the metal interface are somehow altered due to zero-point fluctuations in the vacuum state of the surface plasmon system. In other words, can we observe nonlinear optical behavior even without a single plasmon quantum? It appears that the answer to this question should be yes. Zero-point fluctuations of the SP field E_{vac} must produce some average $E^2_{\mathrm{vac}}(z)$, which is very weak at large distances z from the metal surface, but must grow exponentially towards the surface (similar to the behavior of the E field in the SP wave) [9]. Let us assume that the dielectric near the metal surface exhibits third-order nonlinearity:

$$\epsilon_d = \epsilon^{(1)} + 4\pi\chi^{(3)}E^2, \tag{1}$$

where $\epsilon^{(1)}$ and $\chi^{(3)}$ are the linear dielectric constant and the third-order nonlinear susceptibility, respectively. Far from the metal surface $\epsilon = \epsilon^{(1)}$. However, near the metal we should see an altered layer of the material with the dielectric constant, which fluctuates around $\epsilon = \epsilon^{(1)} + \Delta\epsilon = \epsilon^{(1)} + 4\pi\chi^{(3)}E^2_{\mathrm{vac}}$. But how large and noticeable is this effect?

Our results indicate that this effect may be surprisingly large even in materials with modest $\chi^{(3)}$ nonlinearities. It appears that the dielectric constant shift may reach values of the order of $\Delta\epsilon \sim 1$. Thus, a strongly fluctuating layer of altered dielectric constant must exist near the metal-dielectric interface, which should appear in experiments as a quantum mechanical analogue of the well-known highway mirage. In this analogy the role of heated highway surface is played by the surface of metal, which supports SP modes and, hence, zero-point fluctuations of these modes. It appears that the average shift of the dielectric constant near the interface is sufficiently large to produce multiple metastable states of the metal-dielectric system, so one can talk about optical bistability of the SP vacuum state itself, regardless of the number of real photons in the system. Even though surprising, this effect is not unique or extremely unusual. Metastable quantum vacuums do occur in many field theories such as quantum chromodynamics, quantum gravity, etc.

Let us start by considering the zero-point energy of the SP vacuum of a square $a \times a$ region of a thin metal film with thickness $d \ll a$. Let us assume that the half-spaces above and below the film are filled with the same third-order nonlinear dielectric, and consider the dispersion law of a SP, which propagates along the metal-dielectric interface. In such a case the dispersion law can be written as [10]

$$k^2 = \frac{\omega^2}{c^2}\frac{\epsilon_d\epsilon_m(\omega)}{\epsilon_d + \epsilon_m(\omega) \pm 2\epsilon_d e^{-kd}}, \tag{2}$$

where $\epsilon_m(\omega)$ is the frequency-dependent dielectric constant of the metal. If we assume that, according to the Drude model $\epsilon_m = 1 - \omega_p^2/\omega^2$ (ω_p is the plasma frequency of the lossless metal) and d is large, the dispersion law can be simplified as

$$k^2 = \frac{\omega^2}{c^2}\frac{\epsilon_d\epsilon_m(\omega)}{\epsilon_d + \epsilon_m(\omega)}, \tag{3}$$

where $\epsilon_m(\omega)$ is real. This dispersion law is shown in Fig. 1(a) for the cases of metal-vacuum and metal-dielectric interfaces. It starts as a "light line" in the respective dielectric at low frequencies and approaches asymptotically $\omega_{sp} = \omega_p/(1+\epsilon_d)^{1/2}$ at large wave vectors. The latter frequency corresponds to the so-called surface plasmon resonance. The dispersion law (2) also looks very similar to Fig. 1(a) in the general case of a lossy metal film if the metal film thickness d is small, so that the imaginary part of the term $\pm 2\epsilon_d e^{-kd}$ compensates the imaginary part of ϵ_m. In such cases the SP wave vector k also diverges at some frequency, which is very close to $\omega_{sp} = \omega_p/(1+\epsilon_d)^{1/2}$. Very short wavelength (in the 10–50 nm range) plasmons with the frequencies near the frequency of the SP resonance were observed in a number of near-field optical and low energy electron diffraction experiments [11,12].

The main contribution to the zero-point energy of the SP field comes from the frequency region near the SP resonance ω_{sp} in which ω does not depend on k. Hence, the total zero-point energy of the SP vacuum can be approximated as

$$U_{\rm vac} \approx \tfrac{1}{2}\Sigma_k \hbar\omega_{sp}, \qquad (4)$$

where summation is done over all the plasmon modes k of the square region of the thin metal film. The SP eigenmodes of this square region are defined by the two-component wave vector $(k_x, k_y) = \pi/a \times (n_x, n_y)$, where n_x and n_y are integer. If a of the order of 100–200 nm is selected, the quasistatic approximation used in Eq. (4) is justified. The summation over all possible SP wave vectors has to be cut off at some $k_{\rm max}$, which according to [13] is defined by the Landau damping and equals the electron Fermi momentum $k_{\rm F}$ so that $|k_{\rm max}| \sim k_{\rm F} = (6\pi^2 n_e)^{1/3}$ (where n_e is the electron density). However, if we are less optimistic about the total zero-point energy, we may assume that $k_{\rm max} = 2\pi/\lambda_{\rm min}$ corresponds to the shortest SP wavelength $\lambda_{\rm min}$ observed in the experiment. In the 10–50 nm wavelength range the experimentally observed linewidth of the SP resonances remains quite narrow (~10%) [11,12], which indicates that despite losses these SP modes remain relatively well-defined eigenmodes of the system.

Thus, the total number of SP modes on both the top and the bottom interfaces of the metal film is approximately $4k_{\rm max}^2 a^2/\pi^2$, and

$$U_{\rm vac} = \frac{2}{\pi^2}\hbar\omega_{sp}k_{\rm max}^2 a^2. \qquad (5)$$

On the other hand, the zero-point energy of the plasmon field in this region can be expressed as

$$U_{\rm vac} = \frac{\langle E_{sp}^2\rangle}{4\pi} a^2 \langle 1/k_{sp}\rangle, \qquad (6)$$

where $\langle E_{sp}^2\rangle$ is the average of the square of the electric field of the zero-point fluctuations, and $\langle 1/k_{sp}\rangle = 2/k_{\rm max}$ is the average decay length of the SP field away from the metal surface, which is obtained by averaging $1/k$ over all the SP modes of the square film region. As a result, the electric field of plasmon zero-point fluctuations can be found as

$$\langle E_{sp}^2\rangle = \frac{4}{\pi}\hbar\omega_{sp}k_{\rm max}^3. \qquad (7)$$

The shift of the dielectric constant of the nonlinear optical material near the metal surface equals

$$\Delta\epsilon_d = 4\pi\chi^{(3)}\langle E_{sp}^2\rangle = 16\chi^{(3)}\hbar\omega_{sp}k_{\rm max}^3 = A\omega_{sp}. \qquad (8)$$

Assuming a rather cautious $\lambda_{\rm min} = 30$ nm, we obtain $\Delta\epsilon_d \sim 10^9\chi^{(3)}$ esu^{-1}. Taking into account that such nonlinear dielectrics as 3BCMU and 4BCMU polydiacetylenes have $\chi^{(3)}$ as large as 10^{-10} esu [14], very large zero-field induced shifts $\Delta\epsilon_d \sim 0.1$ may be expected near the metal interface. If a less cautious upper limit $|k_{\rm max}| \sim k_{\rm F}$ is assumed, the resulting shift would be much larger ($\Delta\epsilon_d \sim 10^{14}\chi^{(3)}$ esu^{-1}), which would make this effect noticeable even in regular nonlinear optical materials. Very large fluctuations of the dielectric constant near the metal interface make this effect a rather remarkable analogue of the well-known highway mirage. In this analogy the role of the elevated temperature of the highway surface is played by zero-point fluctuations of the SP field near the metal surface. In the experiment this quantum highway mirage may be detected by essentially similar optical means. A focused probe laser beam may be sent almost parallel to the metal interface [Fig. 1(b)], and its scattering by the "quantum turbulence" in the interface layer of the nonlinear dielectric may be studied. Highway mirage

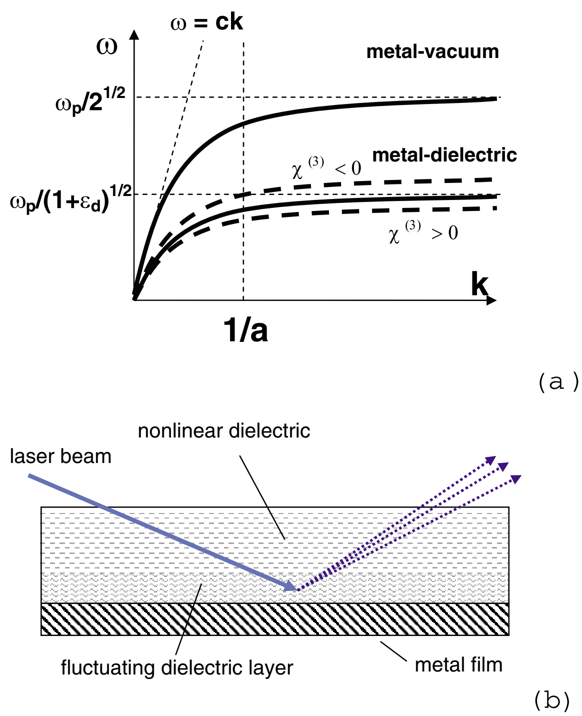

FIG. 1 (color online). (a) SP dispersion law for the cases of metal-vacuum and metal-dielectric interfaces. Nonlinear shifts of the SP resonance are shown for the cases of $\chi^{(3)} > 0$ and $\chi^{(3)} < 0$. (b) Geometry of quantum highway mirage observation.

causes fluctuations of light intensity, propagation direction, and phase of the optical rays upon reflection. Thus, changes in phase coherence of the laser beam transmitted through the surface "turbulent" layer near the metal interface may be measured. The phase fluctuations of the order of $\Delta\epsilon_d \lambda_{\min}/\lambda$ may be expected if the probe beam of wavelength λ is used.

The average shift of the nonlinear dielectric constant near the metal-dielectric interface predicted by Eq. (8) is so large that the effects of bistability of the metal-dielectric system may be expected. The origin of this effect would be roughly the same as the origin of the bistability effects in the regular SP propagation [4]. Using the constant A from Eq. (8) which is proportional to $\chi^{(3)}$, the frequency of the SP resonance may be written as

$$\omega_{sp} = \frac{\omega_p}{(1 + \epsilon_d)^{1/2}} = \frac{\omega_p}{(1 + \epsilon^{(1)} + A\omega_{sp})^{1/2}}. \qquad (9)$$

Here we assume that the probe optical beam is weak, so the number of real optical quanta is small compared to $4k_{\max}^2 a^2/\pi^2$, and the nonlinear shift of the dielectric constant due to the probe beam itself may be disregarded.

Let us initially assume that $\chi^{(3)}$ and A are constants independent of the optical frequency. Under such an assumption the frequency of the SP resonance is given by the cubic equation

$$A\omega_{sp}^3 + (\epsilon^{(1)} + 1)\omega_{sp}^2 - \omega_p^2 = 0. \qquad (10)$$

The graphic solution of this cubic equation is presented in Fig. 2(a) for the cases of positive and negative $\chi^{(3)}$. Positive $\chi^{(3)}$ increases the dielectric constant in the immediate vicinity of the metal surface and shifts the frequency of the SP resonance downward because of the zero-point fluctuations. Negative $\chi^{(3)}$ has an opposite effect on the SP resonance: the dielectric constant near the metal surface decreases, which leads to an increase of the frequency of the SP resonance. In addition, another meaningful root of Eq. (10) may appear at even higher frequencies, if this root is below the plasma frequency of metal ω_p. The analytical solution of the cubic Eq. (10) indicates that this interesting behavior occurs near $A^2 = 4(\epsilon^{(1)} + 1)^3/27\omega_p^2$ and the two roots are located about

$$\omega_{2,3} = \frac{3^{1/2}\omega_p}{(\epsilon^{(1)} + 1)^{1/2}}. \qquad (11)$$

If $\epsilon^{(1)} > 2$, these two roots are, indeed, located below ω_p. Thus, two states are possible for the metal-dielectric interface (and hence for the SP vacuum) when $\chi^{(3)} = \text{const} < 0$. The state which corresponds to the larger SP frequency is metastable, since the total energy of the system is higher in this state. Once created, it should eventually spontaneously decay into the lower-energy state. Since $\chi^{(3)}$ of such materials as 4BCMU polidyacetylene is not actually constant as a function of frequency, but rather quickly oscillates in magnitude and sign in the

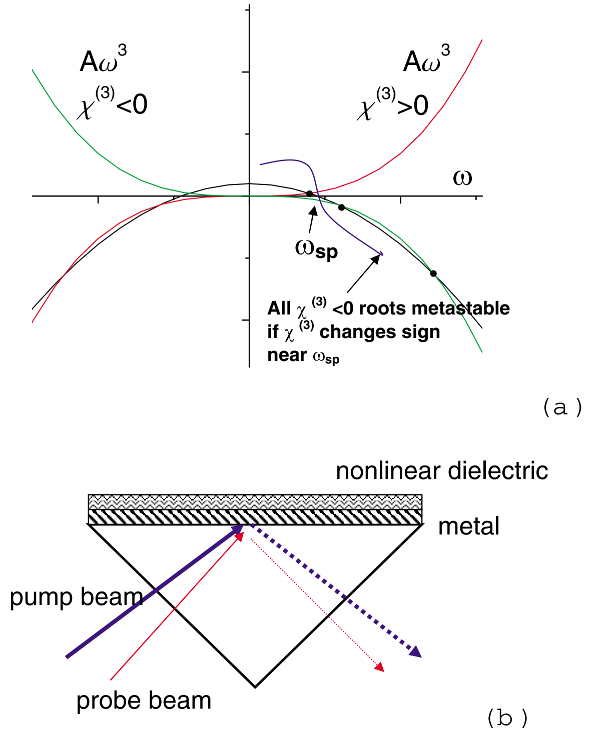

FIG. 2 (color online). (a) Graphic solution of Eq. (10) which defines the frequency of the SP resonance for the cases of positive and negative $\chi^{(3)}$ of the nonlinear optical material. The roots are shown by black dots. If $\chi^{(3)}$ changes sign near ω_{sp}, all $\chi^{(3)} < 0$ roots become metastable states of SP vacuum. (b) Vacuum bistability may be observed in a SP resonance pump-probe experiment in the Kretschman geometry. The average dielectric constant of the fluctuating interface layer can be measured by measuring the angle of the SP resonance with a weak probe beam.

visible frequency range [14], other scenarios of bistable behavior are possible. If $\chi^{(3)}(\omega)$ changes sign from positive to negative in the vicinity of $\omega_{sp}^{(1)} = \omega_p/(1 + \epsilon^{(1)})^{1/2}$, the lower-energy state from the pair obtained for $\chi^{(3)} < 0$, which is situated above $\omega_{sp}^{(1)}$, becomes metastable and would eventually decay into a stable state located below $\omega_{sp}^{(1)}$ at $\chi^{(3)} > 0$.

The metastable states of the metal-dielectric interface may be created and observed in the pump-probe optical experiment in the Kretschman configuration [2] [Fig. 2(b)] using the effect of bistability observed experimentally in the excitation of SPs over an interface between metal and nonlinear dielectric [4]. If the light intensity in the pump beam is sufficiently large, SPs of the pump beam excited in the upper energy state would create a necessary seed layer with an altered dielectric constant near the metal interface. After all the real plasmons decay or radiate, the metal-dielectric interface (and the SP vacuum) may stay in the upper metastable state for a while, which may be detected by observing SP resonance with a weak probe beam.

Exactly how long this metastable vacuum state is going to last is difficult to predict because of the following theoretical consideration.

Spatial distribution of the electromagnetic field near the metal-dielectric interface is defined by the spatial distribution of ϵ. On the other hand, due to nonlinearity of the dielectric medium described by Eq. (1), the energy density of the electromagnetic field affects the spatial distribution of ϵ. Considerable self-focusing effects generally appear in nonlinear optics, which in our case are complicated by the fact that the diffraction limit of self-focusing of the SP field is extremely small due to the nanometer range of plasmon wavelengths, and due to the fact that quantum fluctuations of ϵ are strong. Thus, we meet the same type of theoretical problem as in quantum gravity. In order to see and understand this difficulty more clearly, let us recall the well-known analogy between media electrodynamics and gravitation theory.

It is well known that Maxwell equations in a curved space-time background $g_{ij}(x, t)$ are equivalent to the Maxwell equations in the presence of a matter background with nontrivial dielectric and magnetic permeability tensors $\epsilon_{ij}(x, t)$ and $\mu_{ij}(x, t)$ [15]:

$$\epsilon_{ij}(x, t) = \mu_{ij}(x, t) = (-g)^{1/2} \frac{g^{ij}(x, t)}{g_{00}(x, t)}. \quad (12)$$

If we accept this analogy, the nonlinear optical corrections to $\epsilon_{ij}(x, t)$ and $\mu_{ij}(x, t)$ of the form described by Eq. (1) would describe an effective gravitational interaction between the quanta of the electromagnetic field. This fact is very easy to illustrate in situations in which $\epsilon_{ij}(x, t)$ and $\mu_{ij}(x, t)$ depend on only one spatial coordinate z (such as the Rindler geometry). After straightforward calculations one obtains the following expression for the R_{00} component of the Ricci tensor:

$$R_{00} = \frac{1}{\epsilon} \frac{\partial^2}{\partial z^2}\left(\frac{1}{\epsilon}\right) = \frac{16\pi\chi^{(3)}k^2}{\epsilon^{(1)3}} E^2 = \frac{8\pi k_{\text{eff}}}{c^4} T_{00}, \quad (13)$$

which reproduces the Einstein equation with the effective gravitational constant k_{eff}, which is proportional to $\chi^{(3)}$. In three-dimensional optics $\epsilon^{(1)}$ and $\chi^{(3)}$ of optical materials are small, and no nontrivial gravitational effects may be emulated. Very recently we have pointed out that this situation changes dramatically in the case of two-dimensional SP optics [16]. It is clear from Eq. (13) that the effective gravitational constant for SPs with large wave vectors becomes very large (since $k_{\text{eff}} \sim k^2$). In order to estimate if we can expect any quantum gravitylike effects in SP optics, let us derive the effective Planck scale $L_{pl}^{\text{eff}} = (\hbar k_{\text{eff}}/c^3)^{1/2}$ of our system. Taking into account the value of the effective gravitational constant k_{eff} from Eq. (13), we find

$$L_{pl}^{\text{eff}} = \left(\frac{\hbar k_{\text{eff}}}{c^3}\right)^{1/2} = \left(\frac{8\pi\hbar c\chi^{(3)}k^2}{\epsilon^{(1)3}}\right)^{1/2}. \quad (14)$$

Assuming $\chi^{(3)} \sim 10^{-10}$ esu, and the plasmon wavelength $\lambda = 10$ nm, which defines $k = 2\pi/\lambda$, we obtain the effective Planck scale as $L_{pl}^{\text{eff}} = 10$ nm. These simple arguments indicate that the effects of effective quantum gravity are very strong in our system. For example, the dephasing effect of the dielectric constant fluctuations described above is a direct analogue of the phase coherence loss in quantum gravity [17]. Since the lifetime of the metastable SP vacuum is determined by the quantum fluctuations of the effective metric ϵ_d of the nonlinear optical material, a theoretical answer to the problem of vacuum lifetime seems to be difficult at the moment. On the other hand, this lifetime may be measured experimentally in the configuration described in Fig. 2(b), which thus becomes a toy ‘‘big-bang’’-like experiment. Such experiments would be very beneficial both from the point of view of SP optics and from the point of view of advancing theoretical models of quantum gravity.

In conclusion, we have considered the effect of zero-point fluctuations of SP modes near the interface between a metal and a nonlinear dielectric on the dielectric constant in the immediate vicinity of the interface. Zero-point fluctuations are shown to produce a thin layer of a strongly fluctuating dielectric constant near the interface. This effect may be considered a quantum mechanical analogue of the well-known highway mirage. The average shift of the dielectric constant in this layer may be sufficiently large to produce multiple metastable states of the metal-dielectric interface, and hence the SP vacuum.

This work has been supported in part by the NSF Grant No. ECS-0304046 and CRDF Grant No. RP1-2332-MO-02.

[1] A. V. Sergienko and G. S. Jaeger, Contemp. Phys. **44**, 341 (2003).
[2] H. Raether, *Surface Plasmons* (Springer, Berlin, 1988).
[3] A. V. Zayats and I. I. Smolyaninov, J. Opt. A **5**, S16 (2003).
[4] P. Martinot *et al.*, J. Phys. (Paris) **45**, 597 (1984).
[5] A. M. Dykhne *et al.*, Phys. Rev. B **67**, 195402 (2003).
[6] I. I. Smolyaninov *et al.*, Phys. Rev. Lett. **88**, 187402 (2002).
[7] J. A. Porto *et al.*, Phys. Rev. B **70**, 081402 (2004).
[8] A. Imamoglu *et al.*, Phys. Rev. Lett. **79**, 1467 (1997).
[9] V. A. Shubin *et al.*, J. Lightwave Technol. **17**, 2183 (1999).
[10] J. J. Burke *et al.*, Phys. Rev. B **33**, 5186 (1986).
[11] I. I. Smolyaninov *et al.*, Phys. Lett. A **158**, 337 (1991).
[12] H.-J. Maas *et al.*, J. Microsc. **209**, 241 (2002).
[13] A. G. Malshukov, Phys. Rep. **194**, 343 (1990).
[14] K. Yang *et al.*, Opt. Commun. **164**, 203 (1999).
[15] B. Reznik, Phys. Rev. D **62**, 044044 (2000).
[16] I. I. Smolyaninov and C. C. Davis, Phys. Rev. B **69**, 205417 (2004).
[17] L. J. Garay, Int. J. Mod. Phys. A **14**, 4079 (1999).