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Superfocusing of surface polaritons in the conical structure

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It is shown that at the propagation of a surface polariton through a pointed cone its wavelength decreases to zero as it approaches the edge of the cone. As a result, the polariton is focused in a very small region and the strengths of the wave fields anomalously increase. The phenomenon could be used for creation of a scanning near-field optical microscope to investigate the nanometer-scale surface areas. © 2000 American Institute of Physics. [S0021-8979(00)09805-4]

I. INTRODUCTION

Extraordinary electromagnetic processes taking place on a metal surface are still in the focus of attention of many researchers. In particular, there exist grating anomalies for electromagnetic waves incident on a dielectric/metal grating interface, usually referred to as the Wood anomalies, and at certain wavelengths and angles of incidence these may lead to the total absorption of the plane wave by the metal grating.^{1,2} Furthermore, an anomalous enhancement of the Raman scattering signal has been observed^{3,4} in experiments on Raman scattering from a molecule adsorbed on the rough surface of metal. When the metal surface is rough on the nanometric scale, an enhancement of the intensity of the Raman signal up to 10⁶ times can be observed. And, finally, a signal enhancement due to the roughness of the metal surface⁵ during the process of surface generation of the second harmonic has also been observed. A detailed study of the conditions of emergence of these phenomena has led to a conclusion that these are caused by excitation of surface electromagnetic waves. One can conclude that the formation of electromagnetic waves on the rough surface sometimes may acquire specific features. In particular, the enhancement of nonlinear response mentioned above is a result of an anomalous increase of the electric field of surface-plasmon polaritons.

Recently, Garcia-Vidal and Pendry⁶ studied the interaction of light with a rough surface of silver. They modeled the rough surface as an array of hemicylinders embedded in the silver surface and found anomalous enhancement of the electric field at the tip of grooves between the hemicylinders. Sambles et al. 7 continued the investigations of grating grooves and disclosed a family of strong standing-wave surface-plasmon resonances. These resonances may lead to a very high absorption of the incident radiation and a strong local field enhancement. However, the methods of numerical simulation used in these papers neither reveal the physical nature of the anomalous increase of wave fields, nor describe the behavior of ongoing processes. In particular, it remains unclear why the diffraction processes do not hinder the localization of the wave in very small space regions. Meanwhile, it seems doubtful to us that this is a collective effect⁶

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and cannot be observed at the tip of an isolated groove.

The problem of creation of gigantic wave fields can be solved only under specific conditions, when a substantial shortening of the radiation wavelength is possible. The surface polariton appears to be of the supposed kind, because its dispersion relation allows the wave number to vary in a rather wide range in the small vicinity of the resonance frequency.⁸ In particular, one can conclude from the analysis of dispersion relations of the surface polaritons in a layer^{9,10} that their wave number is inversely proportional to the layer thickness. In our work, 11 the possibility of superfocusing of the surface polariton was studied analytically. It was shown that during the propagation of surface polaritons through a wedge-like structure, the necessary conditions are realized for the localization of the wave in a very small space region and for anomalous amplification of the electric field. The obtained results are valid both for the wedge-like metal structure and wedge-like groove in the metal. It is easy to see that the analytical results obtained in the work¹¹ agree with numerical ones obtained in Refs. 6 and 7. Therefore, in the wedge-like structure the superfocusing of surface polariton plasmons takes place due to the shortening of their wavelengths as they approach the tip of the wedge. This effect is not of a collective nature and can be observed in an isolated wedge-like structure as well.

First, it is necessary to resolve the problem of universality of this effect in order to define the possibility of detailed experimental investigations. Obviously, the possibility of the superfocusing effect is stipulated by the geometric features of an object under consideration. In the present article, we consider the features of propagation of the surface-plasmon polariton through a pointed metal cone. In our opinion, the suggested structure is the best fit for investigation of the superfocusing effect that can have different applications in the field of scanning near-field optical microscopy. 12–17

II. THEORETICAL MODEL

We choose the origin of a spherical system of coordinates at the apex and direct the polar axis along the axis of the conical structure. The apex angle of the cone is $2\alpha(\alpha \ll 1)$ and the region outside the cone corresponds to values of polar angle $\alpha < \vartheta < \pi$. We assume that the magnetic field is directed along the φ axis and depends only on coordinates

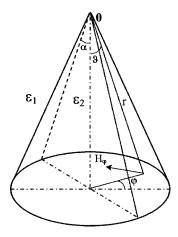


FIG. 1. Geometry of the cone structure where the superfocusing of the surface polaritons takes place.

r and ϑ (see Fig. 1). The wave equation for the magnetic field H_{φ} in the cone matter having dielectric permittivity ϵ_2 and in the surrounding medium with dielectric permittivity ϵ_1 has the form

$$\frac{1}{r}\frac{\partial^2}{\partial r^2}rH_{\varphi} + \frac{1}{r^2}\frac{\partial}{\partial \vartheta \sin \vartheta}\frac{\partial}{\partial \vartheta}H_{\varphi}\sin \vartheta + \frac{\epsilon_j\omega^2}{c^2}H_{\varphi} = 0,$$

$$j = 1, 2.$$
 (1)

To separate the variables we write the field in the form

$$H_{\alpha} = R(r)\Psi(\vartheta)\exp(i\omega t).$$
 (2)

Then

$$\frac{d}{d\vartheta} \frac{1}{\sin\vartheta} \frac{d}{d\vartheta} \Psi(\vartheta) \sin\vartheta = \eta^2 \Psi(\vartheta), \tag{3}$$

$$\frac{d^{2}R(r)}{dr^{2}} + \frac{2}{r}\frac{dR(r)}{dr} + \left(\frac{\eta^{2}}{r^{2}} + \frac{\epsilon_{j}\omega^{2}}{c^{2}}\right)R(r) = 0.$$
 (4)

Here, η is the separation constant to be determined from the boundary conditions. The existence of the surface polariton is stipulated by the negative dielectric permittivity of one of the bounding media in a definite range of frequencies. In the case under consideration, the solution with the surface polariton properties can be obtained for positive values of η^2 . In the case of $\eta \gg 1$, the wave fields are essentially nonzero in the region $\vartheta \ll 1$, where one can write Eq. (3) as

$$x^{2}\frac{d^{2}\Psi}{dx^{2}} + x\frac{d\Psi}{dx} - (x^{2} + 1)\Psi = 0, \quad x = \eta\vartheta.$$
 (5)

The solution of Eq. (5) that provides the localization of the wave field near the conical structure can be written in the form:

$$\Psi(\vartheta) = AI_1(\eta\vartheta), \quad \text{if } \vartheta \leq \alpha, \tag{6}$$

$$\Psi(\vartheta) = BK_1(\eta\vartheta), \text{ if } \vartheta \geqslant \alpha.$$
 (7)

Here, I_1 and K_1 are the modified Bessel and Hankel functions.¹⁸ Note that $I_1(0) = 0$ and the function $K_1(x)$ exponentially decreases as its argument grows. Therefore, the wave field is localized near the angel $\vartheta = \alpha$.

The relationship between the parameters A and B has to be determined from the boundary conditions. Near the top of the cone, where the condition

$$\frac{\eta^2}{r^2} \gg |\epsilon_j| \frac{\omega^2}{c^2},\tag{8}$$

is satisfied, one can easily derive for the radial part of the field

$$R(r) = \frac{1}{r^{1/2}} \exp\left\{i \int_{r_0}^r k(r') dr'\right\},$$

$$k(r') = \frac{\sqrt{\eta^2 - 1/4}}{r'} \approx \frac{\eta}{r'}.$$
(9)

Here, the value of the constant r_0 is determined by the manner of excitation of surface waves. From Eq. (9) it follows that the wave number k increases as the end of cone tip $(r \rightarrow 0)$ is approached, and correspondingly, the wavelength decreases. As a consequence, when $r \rightarrow 0$ the value of the function R(r) also grows anomalously. From the Maxwell equations we get

$$\frac{1}{r\sin\vartheta}\frac{\partial}{\partial\vartheta}H_{\varphi}\sin\vartheta = \frac{i\omega\epsilon_{1}}{c}E_{r},$$
(10)

$$-\frac{1}{r}\frac{\partial}{\partial r}(rH_{\varphi}) = \frac{i\omega\epsilon_1}{c}E_{\vartheta}. \tag{11}$$

Now it is easy to determine the values of the electric-field components

$$E_r = -\frac{ic}{\omega \epsilon_2} \frac{\eta A}{r^{3/2}} I_0(\eta \vartheta) \exp\left\{-i\omega t + i\eta \ln \frac{r}{r_0}\right\}, \quad \vartheta < \alpha,$$
(12)

$$E_{\vartheta} = \frac{c}{\omega \epsilon_2} \left[\eta + \frac{i}{2} \right] \frac{A}{r^{3/2}} I_1(\eta \vartheta) \exp \left\{ -i\omega t + i \eta \ln \frac{r}{r_0} \right\},$$

$$\vartheta < \alpha, \tag{13}$$

$$E_{r} = \frac{ic}{\omega \epsilon_{1}} \frac{\eta B}{r^{3/2}} K_{0}(\eta \vartheta) \exp\left\{-i\omega t + i\eta \ln \frac{r}{r_{0}}\right\}, \quad \vartheta > \alpha,$$
(14)

$$E_{\vartheta} = \frac{c}{\omega \epsilon_1} \frac{B}{r^{3/2}} \left[\eta + \frac{i}{2} \right] K_1(\eta \vartheta)$$

$$\times \exp\left\{-i\omega t + i\eta \ln \frac{r}{r_0}\right\}, \quad \vartheta > \alpha.$$
 (15)

Therefore, when the tip of the cone is approached, the values of the wave fields increase anomalously. From the continuity conditions of tangential components of the fields on the interface $\vartheta = \alpha$, one can easily obtain the following dispersion relation:

$$\epsilon_2 \frac{I_1(\eta \alpha)}{I_0(\eta \alpha)} = -\epsilon_1 \frac{K_1(\eta \alpha)}{K_0(\eta \alpha)}. \tag{16}$$

Equation (16) determines the value of η uniquely. As for the modified cylindrical functions, the following inequalities

hold true when $I_0(\eta\alpha) > I_1(\eta\alpha)$ and $K_1(\eta\alpha) > K_0(\eta\alpha)$, ¹⁸ that is, the wave of the type under investigation can exist if $|\epsilon_2| > |\epsilon_1|$.

One can take into account the absorption effect when the dielectric permittivity is written in the form $\epsilon_2 = \epsilon_2' + i \epsilon_2''$. Then, according to Eq. (16), the parameter η also becomes complex: $\eta = \eta' + i \eta''$. Because of the complexity of dielectric permittivity in Eqs. (12)–(15), there arises an additional factor $r^{\eta''}$. As the intensity of propagating radiation is determined by the product of electric and magnetic fields, the superfocusing can be observed when $\eta'' < 1$. In the case of interest $|\epsilon_2'| \gg \epsilon_2''$. One can derive the values of η' and $\eta'' (\eta' \gg \eta'')$ from Eq. (16) by using the modified Bessel function and taking only the first-order term. Then, the equations determining the values of η' and η'' have the form

$$\epsilon_{2}^{\prime} \frac{I_{1}(\eta^{\prime}\alpha)}{I_{0}(\eta^{\prime}\alpha)} = -\epsilon_{1} \frac{K_{1}(\eta^{\prime}\alpha)}{K_{0}(\eta^{\prime}\alpha)},\tag{17}$$

$$\eta''\alpha = \frac{\epsilon_2'' I_1(\eta'\alpha) K_0(\eta'\alpha)}{(\epsilon_1 - \epsilon_2') [I_0(\eta'\alpha) K_0(\eta'\alpha) - I_1(\eta'\alpha) K_1(\eta'\alpha)]}.$$
(18)

In the case of a silver cone in vacuum, $\epsilon_2 = -18 + i0.6$ (for the wavelength of exciting radiation $\lambda_0 = 0.633 \,\mu\text{m}$) (Ref. 1) and $\epsilon_1 = 1$. Then, for the apex angle of the cone $2\alpha = 0.1$ rad, one can easily obtain from Eqs. (17) and (18) the values $\eta' = 5.2$ and $\eta'' = 0.12$. This case is within the framework of our approximation ($\alpha < 1, \eta' > 1, \eta'' < 1$) and reveals the superfocusing effect. It is noteworthy that at $|\epsilon_2'| > \epsilon_1$, which is the case rather often in the experiment, formulas (17) and (18) can be written in a simpler form. Analyzing the features of the modified Bessel and Hankel functions, one can notice that in this case relationship (17) is fulfilled for values of the argument $\eta' \alpha < 1$. Then, for modified Bessel and Hankel functions, the following asymptotic representations 18 take place:

$$\begin{split} I_0(\eta'\alpha) \approx 1, \quad I_1(\eta'\alpha) \approx \frac{\eta'\alpha}{2}, \quad K_0(\eta'\alpha) \approx \ln\frac{2}{\gamma\eta'\alpha}, \\ K_1(\eta'\alpha) \approx \frac{1}{\eta'\alpha} \quad (\gamma = 1.78). \end{split}$$

As a result, in the limit $|\epsilon_2'| \gg \epsilon_1$, formulas (17) and (18) take on the form

$$\frac{1}{2}(\eta'\alpha)\ln\frac{\gamma\eta'\alpha}{2} = \frac{\epsilon_1}{\epsilon_2'},\tag{20}$$

$$\eta''\alpha = \frac{\epsilon_2'' \frac{\eta' \alpha}{2} \ln \frac{\gamma \eta' \alpha}{2}}{(\epsilon_1 - \epsilon_2') \left(\ln \frac{\gamma \eta' \alpha}{2} + \frac{1}{2} \right)}.$$
 (21)

Formulas (17) and (18) may be used in studying the propagation of the surface-plasmon polariton through a silver conical structure.

The obtained results are valid for regions where the concept of dielectric permittivity makes sense. This means that this theory permits the observation of the propagation of the

surface polariton as long as the wavelength of the surface polariton is much greater than the lattice constant. Note that, as a rule, the radius of the circular region of the cone tip actually much exceeds the lattice constant.

III. DISCUSSION

Based on the above theoretical analysis, one can conclude that propagation of surface polaritons through a conical structure has two inherent features. First, when the top of the cone is approached, the wave number of the surface polariton increases according to the law $k=\eta/r$ and, consequently, its wavelength decreases. Besides, in this process the wave fields grow anomalously $(H_{\varphi} \sim r^{-1/2}, E_r, E_{\vartheta} \sim r^{-3/2})$. The combination of these two features makes the essence of the surface polariton superfocusing effect that consists in localization of waves in very small space regions.

To assess the result, it is necessary to point out that being an elementary excitation, the polariton combines the properties of both the photon and plasmon. As a rule, the speed of plasmons is less than that of photons by a few orders of magnitude. When the tip of the cone is approached, the properties of plasmons show more distinctly and, consequently, the speed of the wave decreases. This leads to a decrease of the wavelength of plasmon polaritons, and the diffraction processes do not hinder the localization of the wave in very small regions of space. This results in an anomalous increase of the wave fields.

This effect is analogous to the quantum-mechanical effect of the incidence of particles on the center. ¹⁹

Comparing these results with those for the wedge, one easily sees that the geometry of the cone is more suitable for observation of the superfocusing effect. Although the process of decrease of the wavelength in both cases is identical, in the case of cones, the wave fields increase more rapidly because of the more rigid space limitation. In the case of wedges, the strength of the electric field grows according to the low $E \sim r^{-1}$ and the magnetic field remains constant, ¹¹ while in case of cones we have $E \sim r^{-3/2}$ and $H \sim r^{-1/2}$.

It has been shown^{12–17} that scanning near-field optical microscopy is a highly effective method of surface investigation. The possibility of strong localization of wave fields given in this article may essentially foster this direction in microscopy.

¹H. Raeter, Surface Plasmons on Smooth and Rough Surfaces and on Gratings (Springer, Berlin, 1988).

² Electromagnetic Surface Model, edited by A. D. Boardman (Wiley, New York, 1982).

³M. Fleischmann, P. J. Hendra, and A. J. McQuillan, Chem. Phys. Lett. **26**, 163 (1974).

⁴M. Moskovits, Rev. Mod. Phys. **57**, 783 (1985).

⁵Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).

⁶F. J. Garsia-Vidal and J. B. Pendry, Phys. Rev. Lett. **77**, 1163 (1996).

⁷M. B. Sobnack, W. C. Tan, N. P. Wanstall, T. W. Preist, and J. R. Sambles, Phys. Rev. Lett. **80**, 5667 (1998).

⁸ Surface Polaritons, edited by V. M. Agranovich and D. L. Mills (North-Holland, Amsterdam, 1982).

⁹K. L. Kliver and R. Fuchs, Phys. Rev. **141**, 495 (1966).

¹⁰D. L. Mills and A. A. Maradudin, Phys. Rev. Lett. **31**, 372 (1973).

¹¹Kh. V. Nerkararyan, Phys. Lett. A 237, 103 (1997).

¹²P. M. Adam, L. Salomon, F. de Fornel, and J. P. Goudonnt, Phys. Rev. B 48, 2680 (1993).

- ¹³ H. H. Marchman and A. E. Nevemre, Appl. Phys. Lett. **66**, 3269 (1995).
- ¹⁴ Y-K. Kim, P. M. Lundquist, J. A. Helfrich, J. H. Mirkut, G. K. Wong, and P. R. Auvil, Appl. Phys. Lett. 66, 3407 (1995).
- ¹⁵S. I. Bozhevolni, I. I. Smolyaninov, and A. V. Zayats, Phys. Rev. B 51, 17916 (1995).
- $^{16}\mathrm{E.}$ Bourillot, F. de Fornel, J. P. Goudonnt, D. Persegol, A. Kevorkian, and
- D. Delacour, J. Opt. Soc. Am. B 12, 95 (1995).
- ¹⁷ A. Jolocha and N. F. Van Hulst, Opt. Commun. **119**, 17 (1995).
- ¹⁸ G. A. Korn and T. M. Korn, *Mathematical Handbook* (McGraw-Hill, New York, 1968).
- ¹⁹L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon, Oxford, 1977).