Pis'ma v ZhETF, vol. 89, iss. 2, pp. 75 – 79 © 2009 January 25

Physical nature of anomalous optical transmission of thin absorptive corrugated films

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Submitted 7 November 2008
Resubmitted 3 December 2008

The enhancement of p-polarized light transmittance through periodically relief thin absorptive film on dielectric or semiconductor substrate was considered theoretically. The calculation of transmittance/reflectance was performed in the framework of modified differential formalism method. Numerical results obtained for noble metals (Au, Ag and Cu) under arbitrary relief correlation between two sides of film allow ascertaining physical nature of surface electromagnetic fields responsible for the enhancement transmittance effect. These are Fano, Zenneck-Sommerfeld and Brewster modes on the interfaces with absorbing medium. For anticorrelated reliefs of opposite film surfaces the bound of their modes was found especially effective.

PACS: 42.79.Dj, 71.36.+c, 73.20.Mf, 78.66.Bz

About ten years ago Ebbesen et al. [1] reported on optical transmission enhancement in the case of periodic arrays of subwavelength holes in a thin metallic film, what has been explained by excitation of surface plasmon polaritons (SPP). Simultaneous satisfaction of energy and momentum conservation laws is ensured through the presence of a grating on surface/interface.

Recently, the enhancement of light transmittance through periodically relief thin absorptive (metal) film has been predicted, especially for so-called anti-correlated reliefs of opposite surfaces of film [2]. It was shown that the correlation or anti-correlation between these interfaces leads to un-coupling or coupling between SPP excited on both interfaces. However the concrete nature of the SPP modes responsible for the light transmittance enhancement remains uncertained. Here it should be noted that recently it have been ascertained by us [3] that in the case of interface between the strong absorptive and transparent media a tangential (planar) component of Poynting's vector (in relation to the interface) in an absorptive medium changes its direction in relation to the planar component of Poynting's vector of the incident TM-wave, that corresponds to the non-resonant excitation of surface waves. Usually the excitation of surface electromagnetic waves was considered (see, for example, [4, 5, 6]) under condition of weak absorption, $|\mathrm{Re}\,\varepsilon| \gg \mathrm{Im}\,\varepsilon$, that is conditioned by the convenience of consideration and application. In the case of arbitrary relation between $|\mathrm{Re}\,\varepsilon|$ and $\mathrm{Im}\,\varepsilon$ in addition to the normal component of Poynting's vector the tangential component is present. Their coexistence can be explained by the simultaneous existence of bulk and surface electromagnetic waves (surface polaritons) and by the non-resonant excitation of surface non-radiative waves. Moreover one is discussed last time [7] the problem of existence of strongly damping surface electromagnetic waves of Zenneck's type [8] although experimentally these waves have been observed [9] on the quartz surface near the transversal vibrational frequency, where $\mathrm{Im}\varepsilon$ is maximal and $\mathrm{Re}\varepsilon$ changes its sign. Besides, under strong damping in dissipative medium even at $\mathrm{Re}\varepsilon > 0$ so-called Brewster radiative modes [10] turn out are weakly bounded with surface, $\mathrm{Im}k^{\perp} \ll \mathrm{Re}\,k^{\perp}$, where $k^{\perp}$ is the wave vector component normal to interface.

Surface electromagnetic waves and anomalous transparency of thin dissipative (metal) films in the region, where they are excited, are of large interest for practical application in photodetectors and solar cells, because it increases their photosensivity [11].

Therefore this letter is devoted to calculation of transmittance/reflectance spectra of thin noble metal films with arbitrary correlation between periodical reliefs of their opposite surfaces. The analysis of obtained spectral and angular dependences of transmittance allows us to determine the physical nature of participating surface electromagnetic waves (Fano, Zenneck, Brewster modes) for thin metal films on substrates (absorptive or transparent).

The calculations of transmittance/reflectance spectra for arbitrary relief correlation of both sides of metal film were performed in the framework of differential formalism. Periodic profile functions have been taken in the form of 1D grating $\xi(x, \phi)$ with the same periods

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l_x, where angle ϕ defines the in-plane reliefs' shift (see inset in Fig.1). We initialize Cartesian coordinate z

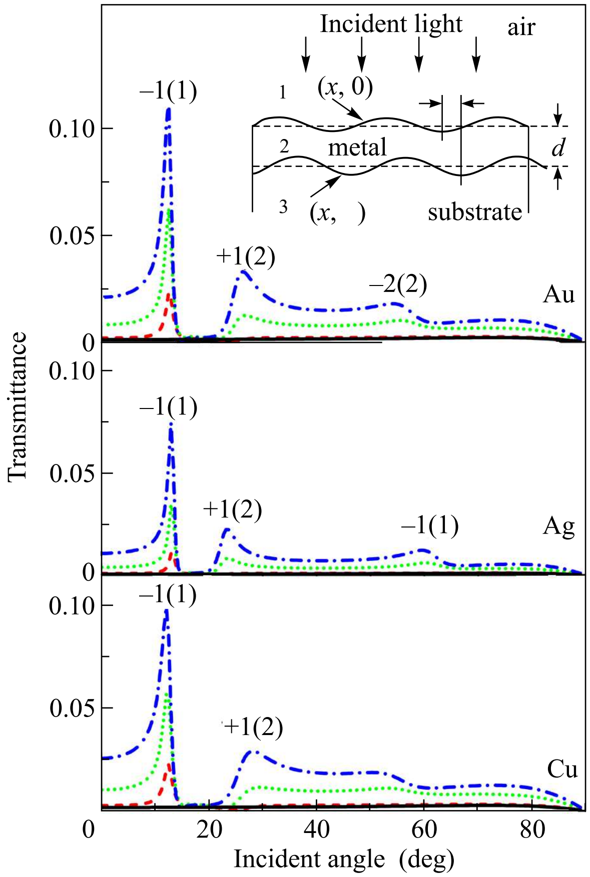

Fig.1. Angular for 632.8 nm incident wave dependencies of full transmittance through metal film with 100 nm thickness on the glass substrate for the cases: $\phi = 0$ – correlated (dashed), $\phi = \pi/4$ (dotted), $\phi = \pi/2$ – anticorrelated (dash-dotted) and flat (solid) film interfaces with the 50 nm relief depth and 500 nm relief period. All peaks are denoted by diffraction order and mode type (in parentheses). In inset, schematic intersection of considered structure is presented

downwards along the structure growth axis (normal to interface), so the numbers z_n and ε_n are denoted averaged positions and permittivity of layers respectively, where $n = 1, 2, 3$ being the medium number. Thin film boundaries are defined in the form $z = z_1 + \xi(x, 0)$ and $z = z_2 + \xi(x, \phi)$. The profile of each corrugated interfaces can be transformed to flat profile by introducing of two coordinate systems, where first and second coordinates are coincided with corresponding Cartesian ones but third coordinate can be written as follows $x^3 = z + \xi(x, \phi)$.

Following calculation of transmittance/reflectance was performed in the framework of modified differential formalism method [12]. The solution for Maxwell's equations can be found in the form of the partial flat waves superposition defined by wave vectors with longitudinal, $\boldsymbol{\kappa}$, and transverse, $k_n^{\perp}(\kappa) \equiv \sqrt{(\omega/c)^2\varepsilon_n - \kappa^2}$, components. Taking into account relief function periodicity and performing transformation to curvilinear coordinates, the in-plane components of electromagnetic field takes the following form

$$\hat{\boldsymbol{\Psi}}_n(\boldsymbol{\rho}, x^3, \phi) = \\ = \sum_{\sigma,m,m'} e^{i(\boldsymbol{\kappa}_i + \mathbf{G}_m)\cdot\boldsymbol{\rho}} M_{n\,\boldsymbol{\kappa}_i+\mathbf{G}_{m'},x^3}^{\,m-m',\sigma}(\phi)\hat{c}_{n,m'}^{\sigma}, \qquad (1)$$

here $\hat{c}_{n,m'}^{\sigma}$ is a matrix-column which consists of two unknown polarization amplitudes of partial flat wave, $\boldsymbol{\rho}$ is the in-plane coordinate, $\boldsymbol{\kappa}_i$ is the in-plane component of the incident wave vector and $\mathbf{G}_m \equiv (2\pi/l_x)m\hat{\mathbf{x}}$ is the in-plane inverse vector due to profiles periodicity, $m = 0; \pm1; \pm2; \ldots$. In (1), the in-plane components of polarization unit vectors was introduced in the matrix form that independent of coordinate systems presentation

$$M_{n\,\boldsymbol{\kappa},x^3}^{\,m,\sigma}(\phi) \equiv \\ \equiv \begin{pmatrix} \hat{\mathbf{z}} \times (\boldsymbol{\kappa}/\kappa) & \frac{\kappa \mathbf{G}_m - k_n^{\perp}(\kappa)^2(\boldsymbol{\kappa}/\kappa)}{\sigma k_n^{\perp}(\kappa)} \\ \frac{\kappa \mathbf{G}_m - k_n^{\perp}(\kappa)^2(\boldsymbol{\kappa}/\kappa)}{\sigma k_n^{\perp}(\kappa)} & -\varepsilon_n \hat{\mathbf{z}} \times (\boldsymbol{\kappa}/\kappa) \end{pmatrix} \times \\ \times e^{im\phi}\Lambda_m(\sigma k_n^{\perp}(\kappa)) e^{i\sigma k_n^{\perp}(\kappa)(x^3 - z_{n-1})}, \qquad (2)$$

$$\Lambda_m(\beta) \equiv (1/S_{\text{cell}}) \int_{S_{cell}} e^{-i(\mathbf{G}_m\cdot\boldsymbol{\rho} - \beta\xi(\boldsymbol{\rho}))} d\boldsymbol{\rho}$$

with integration over the low-level cell, S_{cell}, defined by l_x.

The boundary conditions for the system of Maxwell's equations in the case of curvilinear coordinates are reduced to the infinity system of algebraic linear equations

$$\sum_{\sigma,m'} \left[M_{2\,\boldsymbol{\kappa}_i+\mathbf{G}_{m'},z_n}^{\,m-m',\sigma}(0)\hat{c}_{2,m'}^{\sigma} - M_{1\,\boldsymbol{\kappa}_i+\mathbf{G}_{m'},z_n}^{\,m-m',(-)}(0)\hat{c}_{1,m'}^{(-)} \right] \\ = M_{1\,\boldsymbol{\kappa}_i,z_1}^{\,m,(+)} \begin{pmatrix} \cos\theta_\alpha \\ \sin\theta_\alpha \end{pmatrix}, \\ \sum_{\sigma,m'} \left[M_{3\,\boldsymbol{\kappa}_i+\mathbf{G}_{m'},z_n}^{\,m-m',(+)}(\phi)\hat{c}_{3,m'}^{(+)} - M_{2\,\boldsymbol{\kappa}_i+\mathbf{G}_{m'},z_n}^{\,m-m',\sigma}(\phi)\hat{c}_{2,m'}^{\sigma} \right] \\ = 0, \qquad (3)$$

where θ_α is the polarization angle (for s-polarized incident wave θ_α is equal to $0°$ and for p-polarized incident wave θ_α is equal to $90°$). In the case of finite indexes m and m' the system of Eqs. (3) can be solved directly or using the transfer/scatering matrix method [13].

To determine the transmittance, we use the energy flux averaged over the periodically translated element of corrugated interface surface

$$T = \int_{S_{\rm cell}} (\mathbf{S}\cdot\mathbf{N})d\boldsymbol{\rho} \Big/ \int_{S_{\rm cell}} (\mathbf{S}_i\cdot\mathbf{N})d\boldsymbol{\rho}\,, \qquad (4)$$

where $\mathbf{S}_i$ is only incident wave Poynting's vector. The electromagnetic energy flux from corrugated surface determined by the covariant coordinate of Poynting's vector placed normal to the surface direction

$$\mathbf{S}\cdot\mathbf{N} \propto \frac{Re\,(E_1^* H_2 - E_2^* H_1)}{\sqrt{1+(\partial\xi(\boldsymbol{\rho})/\partial x)^2+(\partial\xi(\boldsymbol{\rho})/\partial y)^2}}, \qquad (5)$$

where $\mathbf{N}$ is the unit surface normal to corrugation interface.

The present formalism describes multilayer systems with identical relief's periods and depths only and can be extended for the case of different relief's depths. Moreover, there are no limitation on the film's thickness. And the ratio h/l_x is limited by validity of the boundary conditions that are in continuity of the in-plane covariant components of the electromagnetic field.

For elucidation of physical nature of electromagnetic modes determining the light transmittance, we have considered 1D profiled thin metal film, deposited on glass (or semiconductor) substrate and calculated the transmittance/reflectance for ± 1 diffraction orders at the fixed wave length (Fig.1). We see two maxima on angular dependencies $T(\theta)$ which correspond to the $m=-1$ order of surface plasmon polariton (SPP) on the air-metal interface and to the $m=+1$ order of SPP on the metal-glass (for example, BK7 with $\sqrt{\varepsilon_3}\approx 1.5$) interface for three noble metals Ag, Au and Cu. The corresponding $T(\lambda)$ spectra at $\theta = 12^o$ ($m=-1$ diffraction order) are shown in Fig.2 for three relative phases, ϕ, of the profile function in the form $\xi(x,\phi) = h\sin(G_x x + \phi)$ on both sides of metal film, where $2h$ is grating depth, $G_x = 2\pi/l_x$ is reciprocal vector (in calculations the ratio between h and l_x was chosen around 0.1 that is best for clear observation of SPP in the case of sinusoidal grating [14]). The value of $\phi=0$ corresponds to correlated film and $\phi=\pi$ corresponds to anticorrelated one. For each metal there are at least four maxima of different height. Using the dispersion equations for SPPs on the flat metal-dielectric interface and the momentum conservation law for SPP excitation $k_{\rm SPP} = (\omega/c)n\sin\theta + mG_x$, we can determine that maxima 1 and 2 correspond to ± 1 orders of surface Fano modes on the metal-glass and air-metal interface, respectively. For identification of nature of other maxima (3), (4) in the spectra of $T(\lambda)$ the optical constants of metals, $\mathrm{Re}\varepsilon(\omega)$ and $\mathrm{Im}\varepsilon(\omega)$,

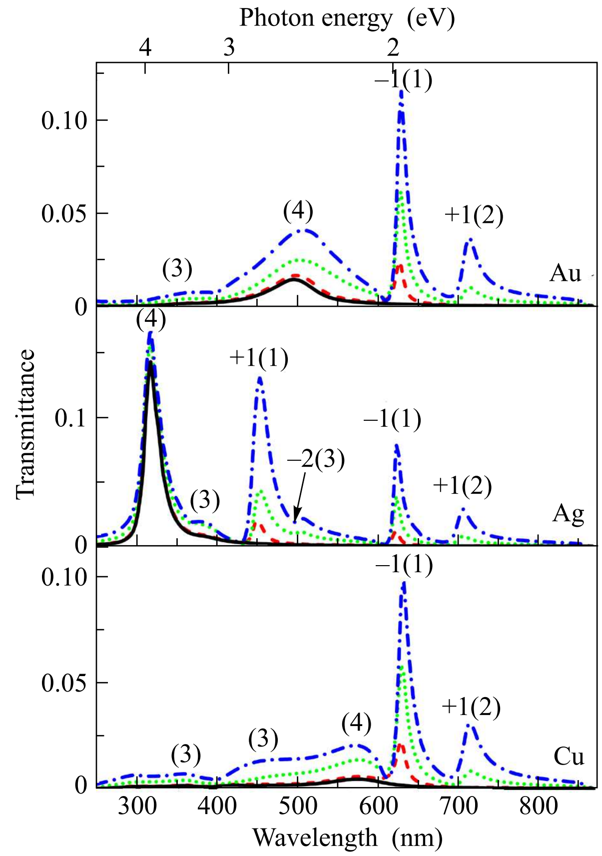

Fig.2. Spectral for 12° incident wave dependencies for transmittance of metal film on a glass substrate for the cases: $\phi = 0$ – correlated (dashed), $\phi = \pi/4$ (dotted), $\phi = \pi/2$ – anticorrelated (dash-dotted) and flat (solid) film interfaces. Other parameters are the same as in Fig.1

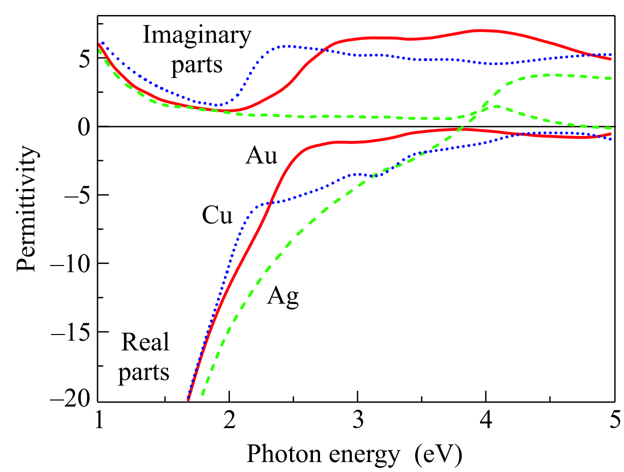

Fig.3. Permittivity of noble metals: Au, Ag and Cu. Solid lines correspond to real and dotted to imaginary parts

are necessary. The permittivity for Au, Ag and Cu was taken from [15] and presented in Fig.3. The spectral region, where $\mathrm{Re}\varepsilon(\omega) < 0$ corresponds to excitation of surface waves, and $\mathrm{Re}\varepsilon(\omega) > 0$ corresponds to bulk waves excitation. There are three ultimate cases: 1) $\mathrm{Re}\varepsilon(\omega) < 0$ and $|\mathrm{Re}\varepsilon(\omega)| \gg \mathrm{Im}\varepsilon(\omega)$, when surface Fano polariton modes are exist on the dispersion curve, $\omega(k)$, at right part from the light line $\omega = kc/\sqrt{\varepsilon_1}$; 2) $\mathrm{Re}\varepsilon(\omega) < 0$ and $|\mathrm{Re}\varepsilon(\omega)| \ll \mathrm{Im}\varepsilon(\omega)$, when so-called Zenneck-Sommerfeld surface modes are exist near by the light line; 3) $\mathrm{Re}\varepsilon(\omega) > 0$, when the Brewster modes are exist at left part from light line, which also are bounded with the interface, but this bound is very weak ($\mathrm{Im}k^{\perp} \ll \mathrm{Re}k^{\perp}$).

Using Fig.3, we can conclude that at $\lambda \lesssim 600$ nm for Cu, Au and $\lambda \lesssim 400$ nm for Ag the surface Zenneck-Sommerfeld modes can be excited on the air-metal or metal-glass interface. However, the maxima of $T(\lambda)$ at $\lambda \approx 570$ nm for Cu, 500 nm for Au, and 320 nm for Ag (Fig.2) are conditioned by existence of bulk modes according to their dispersion equation: $\omega = kc/\sqrt{\varepsilon_2(\omega)}$. Therefore corresponding maxima in transmittance are revealed in the case of flat metal surface too (Fig.2).

For identification of coupled light-matter surface modes their dispersion has been calculated (Fg.4). The

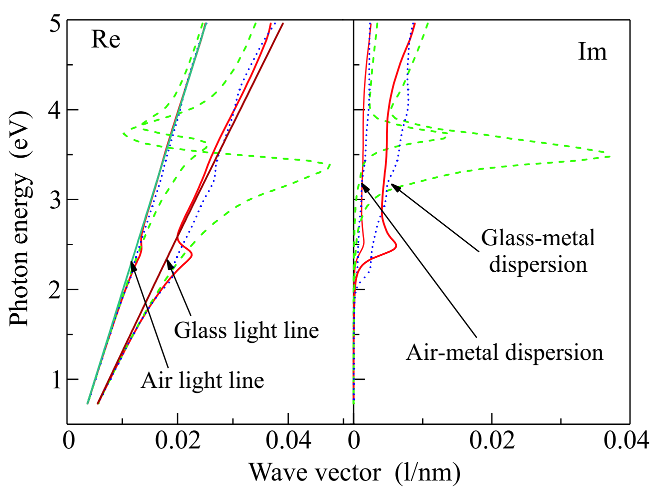

Fig.4. Dispersion (Re) and damping (Im) of SPP excited at "air-metal" interface for Au (solid), Ag (dashed) and Cu (dotted)

dispersion curves are obtained from the zeros of Fresnel coefficients denominator for stratified structures at fixed real photon energy, $\hbar\omega$, and complex wave vectors. The real part of obtained complex wave vectors corresponds to dispersion and its imaginary part corresponds to damping of electromagnetic modes of correspondent flat structure.

Thus, on the upper surface of metallic film take place as resonant excitation of surface plasmon polaritons (Fano modes) as non-resonant (or weakly resonant) excitation of strongly damped surface Zenneck-Sommerfeld modes (polaritons). Transition from the Fano modes to Zenneck-Sommerfeld modes proceeds in metals smoothly as the damping, $\mathrm{Im}\varepsilon(\omega)$, increases with the wavelength λ decreases, and at these frequencies the surface modes dissipative attenuation sharply increases. Moreover, in vicinity of the light line, the dissipative damping surface modes transform into bulk Brewster modes, whose large attenuation is due to radiation decay. So, due to absorption the non-resonant surface wave excitation takes place (see, also [3]), i.e. the wave vector of the light-matter interaction has real and imaginary components: $\mathrm{Re}k_{\mathrm{SPP}} \geq \mathrm{Im}\,k_{\mathrm{SPP}}$, where $\mathrm{Im}k_{\mathrm{SPP}}$ determines broadening of its value. So we can identify spectral peaks in Fig.2 as follows: 1,2) air-metal or glass-metal SPP (Fano modes); 3) SPP of Zenneck-Sommerfeld type; 4) surface-enhanced bulk polaritons. On the lower surface of absorptive film near by substrate the opposite process of transformation of surface/bulk modes into light (homogeneous transversal electromagnetic waves) proceeds. And for this process efficiency it is necessary the bound between two (upper and lower) sets of electromagnetic modes. This bound is especially strong in the case of anticorrelated reliefs of both film interfaces (Fig.1, 2), that leads to anomalous light transmittance.

In this letter, the electromagnetic interaction between two 1D periodic reliefs of absorptive thin film has been considered in the framework of differential formalism. For such typical plasmon-carrying media as noble metals (Ag, Au and Cu) has been predicted anomalous enhancement of transmittance through thin film with anticorrelated reliefs of its opposite sides, which is useful for many photonic and optoelectronic devices. Calculated dependencies of this effect of anomalous optical transmission on light polarization, the incident angles and the wave length for several noble metals allow us to ascertain the physical nature of surface electromagnetic modes participating in the process of light-matter interaction. Besides the usual resonant excitation of SPPs (Fano modes), almost non-resonant excitation of strongly dissipative damped Zenneck-Sommerfeld modes takes place in the strong absorption region of plasmon-carrying medium. Under the conditions of anticorrelation of the opposite surfaces of film the bound of upper and lower plasmon polariton modes is especially effective.

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