

ON 'EXTRAORDINARY OPTICAL TRANSMISSION' FROM PERIODIC AND RANDOM NANOSTRUCTURES

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Recently, an enhanced optical transmission (EOT) through 2D periodic arrays of sub-wavelength holes in a metal layer was found. It generated intense theoretical and experimental research, but its physical nature is still not elucidated. In this communication, we report experimental investigations on 1D relief gratings, and numerical analysis of the composite structure of randomly distributed dielectric nanoparticles embedded in a thin metallic matrix. We show that EOT is also present in such structures.

(Received December 9, 2004; accepted January 26, 2005)

Keywords: Gratings, Composite thin films, Nanoparticles

1. Introduction

The so-called 'extraordinary optical transmission' (EOT) is a phenomenon related to an enhancement of optical transmission through metal films in presence of periodic structures. EOT was first reported in 1998 [1] for a metal layer with 2D periodic sub-wavelength holes. Later, it was shown [2] that EOT does not require the existence of holes or other apertures. Continuous thin metal films with a constant thickness give a similar enhancement of transmission, provided they are 1D or 2D modulated. The effect of EOT has drawn considerable theoretical and experimental interest, due to its potential applications in modern optics. Still, there is no detailed understanding of its physical nature. EOT could be explained by excitation of surface plasmon polaritons (SPP); another hypothesis suggests that it occurs from waveguide mode resonance [3].

In this communication, we report investigations of simple 1D periodic structures that can support EOT. Next, we analyse the optical response of a thin composite metallic film. We show that neither holes nor relief gratings are necessary in order to observe the EOT effect.

2. CD-stampers for measuring techniques, based on EOT

We here studied EOT with 1D periodic metalized relief structures. The purpose of these investigations was to demonstrate the use of the position of the resonance peak in transmission as a very sensitive probe of the optical properties of symmetricized structures: a superstrate with refractive index N_0 ; a continuous metal film with N_m ; a relief grating on a substrate with N_s . As a model system, we used compact-disc stampers (CD-S) which are pregrooved polycarbonate, $N_s = 1.58$. In fact, CD-S are relief diffraction gratings with periodicity in the radial direction of the

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disc. AFM analysis was performed with a DS 40-45 (Danish Micro Engineering) on the CD-S. It has revealed that the grating period is 800 nm and the groove depth is 170 nm; the edges are rounded out and the walls are not vertical. CD-S structures have the advantage of being cheap, commercially available and highly standardized (within a specific series). On the relief structure, an Al film, 20-25 nm thick, was deposited in a 2700 P2 (Leybold Heraeus) vacuum installation, by dc magnetron sputtering. It is known that an oxide film a few nanometers thick grows on the surface of freshly deposited Al films, but we consider the groove structure to be covered with one equivalent continuous film of effective complex refractive index N_m .

The reflectance (R) and transmittance (T) were measured in the visible (VIS) and near infrared (NIR) regions, with a Cary 5E high precision spectrophotometer. Light polarization for the spectra in Fig. 1 was parallel to the grating vector. We used different contact liquids (all grade pro analysis, transparent in the region of the resonance peak) with $N_0 = 1.333$; 1.361; 1.474; and 1.515. For $N_0 = 1$, the resonance peak at $\lambda = 1130$ nm is not fully revealed. With increasing N_0 , it is red shifted – curve (b), Fig. 1. Thus, a calibration curve for the quantitative determination of a compound's refractive index is build up.

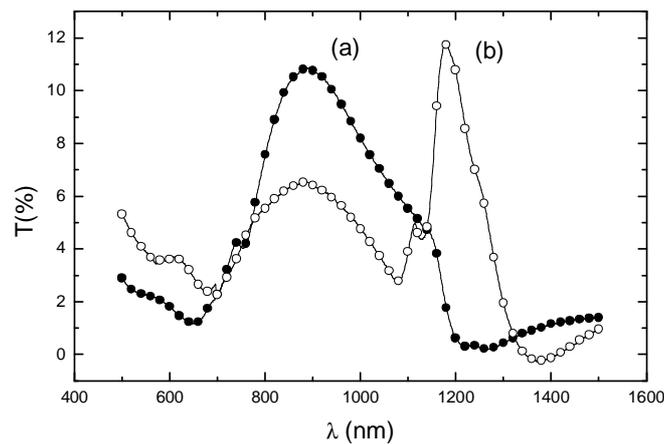


Fig. 1. Transmittance of CD-S metalized structures: (a) grating with a superstrate of $N_0 = 1$; (b) the same grating with a superstrate of $N_0 = 1.515$.

The information yielded by EOT would be more difficult to obtain using conventional reflectance (R) / transmittance (T) measurements for inverse optical problem techniques, because only the spectral position of the resonance peak is monitored, not the absolute values of R and T. Thus, the experimental uncertainty with EOT measurements is much lower.

We have also measured what we term the ‘enhanced optical reflection decrease’ (EORD). Due to the specific angular dependence, our results support the interpretation of EOT and EORD as linked to the excitation of surface plasmon polaritons in the grating, rather than the waveguide mode resonance approach.

3. EOT in random structures

Investigations on EOT relate this phenomenon to the presence of periodic structures. We will numerically analyse the optical response of a thin composite film of dielectric particles (spheres of radius R and refractive index N_p) embedded in a metallic host matrix with a complex refractive index N_m . We will show that EOT could be observed in such random structures.

In our model, we have chosen a 150 nm thick Ag film supported on a glass substrate ($N_s = 1.5$). The Ag optical constants are derived from [4] and interpolated with a piecewise cubic Hermite interpolant. On the basis of this data, we have estimated several characteristics of the film. The bulk plasmon frequency is 3.81 eV (325.4 nm), and at the two interfaces planar surface plasmons are supported at 337 nm ($N_0 = 1$) and 358 nm ($N_s = 1.5$). The thickness of the film is greater than the bulk mean free path of the Ag conduction electrons (52 nm); the skin depth is

evaluated as well. The film thickness is large enough to ignore the coupling between surface plasmons. The transmittance for normal incidence of light (rigorous Maxwell calculations [5]) has a maximum of 0.10 at 319 nm and a HBW of 12 nm. The reflectance minimum of 0.03 is at 317.5 nm and the absorption ($A=1-R-T$) maximum of 0.89 is at 312.2 nm.

The optical behaviour of a single dielectric particle in an absorbing matrix is analysed by the help of the Maxwell vector equation of scattering theory [5]. We have calculated the extinction cross section per unit volume C_v , which is a well-defined observable quantity [6]. It depends only on the size of particle, its shape and refractive index and the surrounding medium. In an absorbing host matrix, C_v might have positive or negative values, as follows from the optical theorem [6].

A negative C_v means an increase of the signal of a hypothetical detector, and this is independent of the distance between the particle and the detector. Moreover, in this case C_v cannot be presented as the sum of the absorption and scattering by the particle, due to the absorption by the surrounding medium. In Fig. 2, we show C_v for spheres with $R = 70$ nm; $N_p = 1$ (spherical voids) and $N_p = 1.5$ (glass spheres).

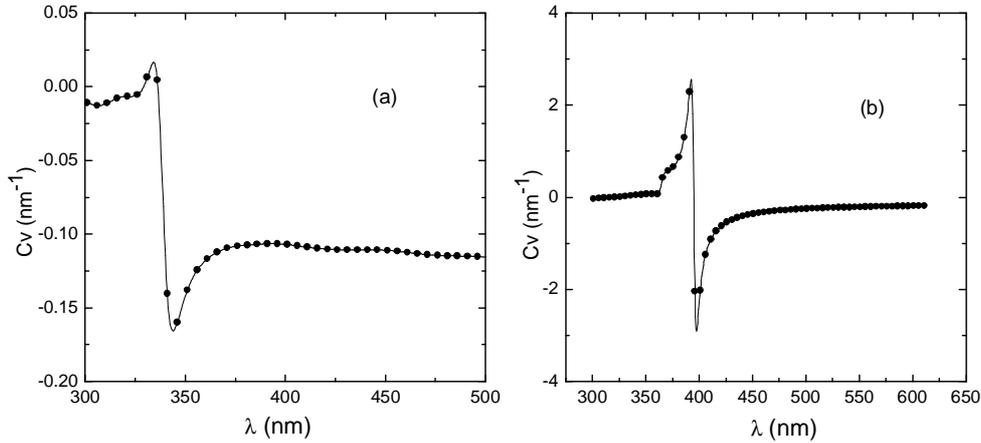


Fig. 2. The volume extinction coefficient C_v for a dielectric sphere with $R = 70$ nm in an Ag matrix: a) $N_p = 1$ (voids); b) $N_p = 1.5$ (glass). The solid lines result from calculations, symbols are to lead the eye.

For $N_p = 1$, there is a sharp negative minimum at 344 nm, preceded by a positive maximum at 334 nm. For $N_p = 1.5$, the corresponding wavelength positions are red shifted at 397 and 392.5 nm. However, the absolute values of C_v are an order of magnitude higher in the latter case. We should expect EOT at the C_v minima.

In order to numerically evaluate the composite film transmittance, we need to fix the number of particles in the metal film and to calculate a formal complex refractive index of the slab, N_{eff} . We do not apply the usual effective medium approximation [5]. Following [7], N_{eff} is:

$$N_{\text{eff}} = N_m \frac{1 + i\pi f S(0)/k^3 v_p}{1 - i\pi f S(0)/k^3 v_p} \quad (1)$$

where $S(0)$ is the forward scattering amplitude by a particle, v_p is the particle (sphere) volume and f is the filling factor [5]. With no particles in the film, $f = 0$; the maximum filling factor for identical closed-packed spheres is $f = 0.74$. Values of $f \sim 0.3-0.4$ are considered as high [5]. A low filling factor means that multiple scattering between particles can be neglected.

In Fig. 3, the transmittance of our model composite film is shown. Fig. 3a describes the case of specific voids ($N_p = 1$) and $f = 0.05$. The transmission maximum at 319 nm is increased by 20%, compared to the case of $f = 0$. A new peak ($T = 0.06$) appears at 341 nm, preceded by a minimum at 334 nm, which has a lower value than that of the 'virgin' film ($f = 0$). The situation for $N_p = 1.5$;

$f = 0.01$ is even more impressive (Fig. 3b). EOT is observed at 397.3 nm, $T_{\max} = 0.9$. There is an additional sharp structure at 393 nm, $T = 0.32$.

Thus, EOT should be observed in random nano-composite structures, although neither plasmons (particles have no free electrons), nor phonons (voids, $N_p = 1$) are available. The transmittance of the metallic film is strongly influenced by the presence of the dielectric spheres, because their presence affects the manner in which light is extinguished in the metal. In our view, it has a resonant nature and is related to collective plasmon-polariton excitations at the metal/dielectric surface boundary.

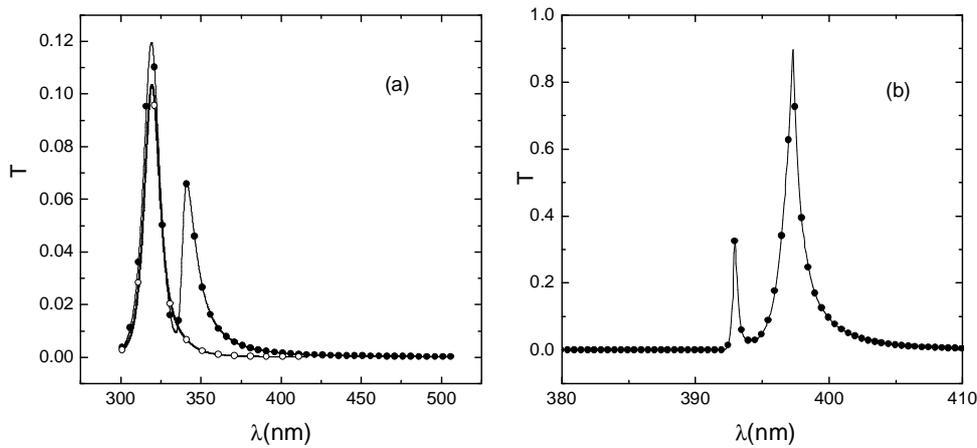


Fig. 3. Transmittance of a 150 nm Ag film: a) Filling factor $f = 0.05$, $N_p = 1$, curve (o) is for a film with no particles; curve (•) is for the composite film; b) Filling factor $f = 0.01$, $N_p = 1.5$. The solid lines result from calculations, symbols are to lead the eye.

4. Conclusions

We have experimentally demonstrated the sensitivity of the wavelength EOT peak position to the superstrate refractive index. The use of simple structures, such as metalized CD stampers, opens the way to a new class of self-built devices for the evaluation of the refractive indices of some relevant compounds. Next, we have shown that EOT is present in composite metallic nanofilms with randomly dispersed dielectric particles. EOT is a phenomenon of a resonant nature, so neither 2D nor 1D gratings are necessary for its observation.

Acknowledgments

Two of the authors (P. Sharlandjiev and D. Nazarova) are much obliged to Dr. L. Tsonev from ISSP (Bulgarian Academy of Sciences) for introducing us to the wonders of EOT. We also thank Dr. E. Popov for many stimulating discussions.

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