Extraordinary optical transmission through a random array of subwavelength holes

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It has been experimentally shown that in near- and mid-IR-wave bands, the transmittance of 17 nm silver layer deposited onto a lavsan film with an irregular system of small holes is two to six times higher than that of the same layer deposited onto a uniform lavsan film. The effect is of a broadband type covering the wave range of 1500–5000 nm. The study of system's microstructure with the help of AFM and electronic microscope shows that the silver film is corrugated with the same system of holes as the lavsan substrate is. The evaluation made within the frames of physical optics approximation has shown that due to smallness of holes' diameter (200 nm), the additional transmittance through the holes is much less than the observed effect. A supposed mechanism of extraordinary light transmittance is discussed.

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I. INTRODUCTION

The recent development of nanotechnologies has stimulated both experimental and theoretical investigations in near field optics and plasmonics.[1–3] A specific interest is drawn to phenomena that can efficiently transform near fields into far fields.

That kind of transformation is a basis of overcoming the diffraction limit and reaching the super-resolution with the help of the Pendry and Veselago lenses.[4–6] It is the resonance of surface plasmons that determines these phenomena.[4] Along with plasmonic amplification of near fields, an important role is played by the phenomenon referred to as filtering, which results in efficient reflection (nontransmission) of traveling waves and in transmission of near fields (evanescent waves).[7]

It is the surface-plasmon excitation that is usually connected with recently discovered extraordinary transparency of thin metal films perforated with a regular array of subwavelength holes.[8–12] The extraordinary transmission was also predicted by computer simulation for regular aperiodic systems with structures that provide spatial resonance of surface waves.[13–20] It is noteworthy that for both periodic and aperiodic systems, these effects are of a pronounced resonance nature.

The present communication is devoted to an experimental observation of extraordinary light transmission through a metal film with a random system of subwavelength holes. This phenomenon is of a broadband type. Opposite to the above-mentioned cases, the phenomenon of near fields' filtering is supposed to be responsible for the observed extraordinary transparency.

II. SAMPLE FABRICATION AND EXPERIMENTAL TECHNIQUE

Track membrane films (TMF) have been used as a basic component of the samples. A TMF is a polymer film (PF) with track holes that appear after heavy-ion bombarding.[21] The TMF without holes (i.e., the TMF edge that was not bombarded) serves as a reference system. Below it is referred to as PF. The AFM study has shown that an average diameter of pores is about 200 nm [Fig. 1(a)]. The holes are randomly distributed over the membrane surface (Fig. 1). Measuring of the membrane thickness by a micrometer gave the value of 10^4 nm.

The deposition of a silver layer on the uniform PF and TMF was performed by thermal evaporation of metal by electron beam at a residual pressure of 0.1 mPa. The layer thickness was controlled by the transparency of the system in visible light (at wavelength of 650 nm). The electron microscope investigation of a chip of a TMF sample shows [Fig. 1(b)] that after silver deposition, the holes remain open, metallization of the internal surface of the holes is insignificant and should not exceed the depth of their diameter value. The thickness of a metal layer inside the hole decreases smoothly as the distance from the surface increases.

The transmission spectra of the TMF were analyzed by IR Fourier spectrometer "Philips PU 9804" with the spectral resolution of 4 cm^{-1}, that made it possible to resolve phonon modes of the condensed media. The membrane material was identified as polyethylene terephthalate of "lavsan" trade mark.

The sample was placed in the flask chamber of spectrometer, in the focal plane of a naturally polarized beam that converges with a cone angle of 13°. Thus, the illuminated area on the sample has the diameter of 10 mm. The wave range of 6000–1900 cm^{-1} (1666–5263 nm) is used. The high-frequency limit of the range was determined by the device characteristics, whereas at the low frequencies, the range was limited by a strong absorption in the membrane.

Primarily, we have determined the electromagnetic properties of the substrate (lavsan). We have focused on the difference in the transmission spectra of PF and TMF (Fig. 2).

First of all, in Fig. 2 we can observe oscillations in transmission spectra of PF and TMF. These oscillations appear due to interference of the waves, exhibiting multiple reflections on film surface and traversing the film several times.[22] Maxima of the transmittance appear at half-wavelength condition. Our experiment shows a phase shift between these interference oscillations in the transmission spectra of TMF and PF (Fig. 2). At low frequencies of $d/\lambda \sim 4$, the shift reaches $\pi/2$ (the maximum of TMF transmittance coincide with the minimum of PF transmittance). It is obvious that this shift is related to the difference in the refractive indices

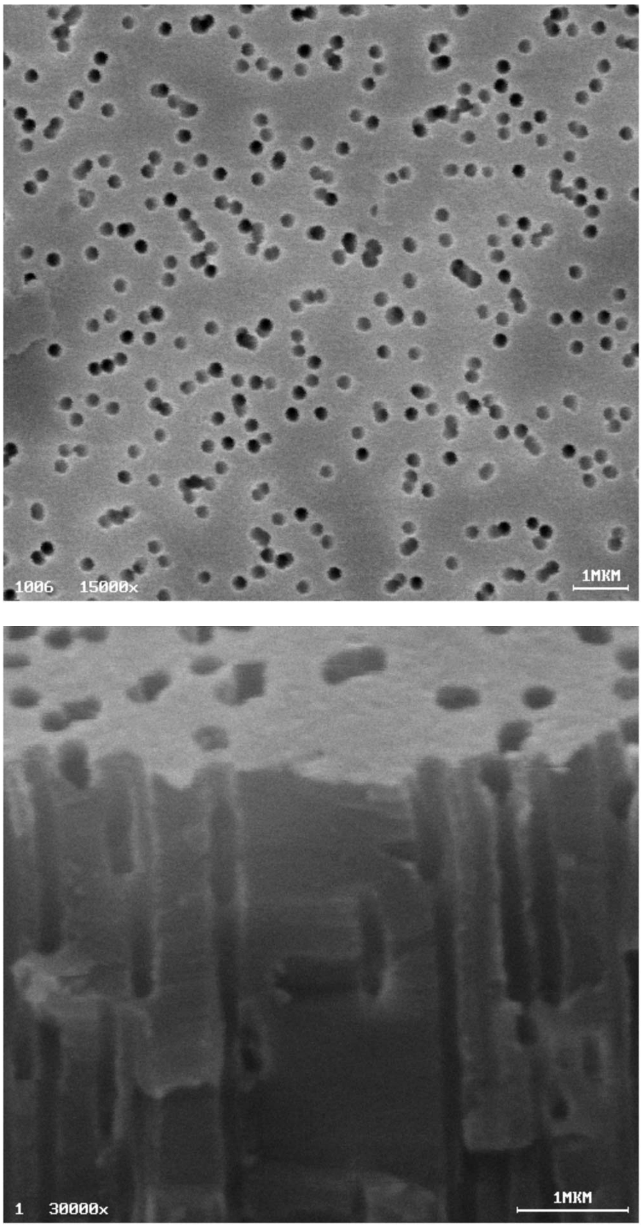

FIG. 1. Picture of a track membrane taken with an electron microscope: (a) top view, (b) chip

caused by a relatively lower density of TMF. Taking into account that the optical path $n_{\mathrm{PF}}k_0d$ of PF is about 12π, where $n_{\mathrm{PF}}\approx 1.5$ is a refractive index of the PF, it is easy to evaluate the refractive index n_{TMF} of TMF as $0.96n_{\mathrm{PF}}$. On the other hand, the results obtained by means of AFM show that the relative area of the holes is about 10%.[23] It is reasonable to expect the same reduction of the specific polarization $\varepsilon-1$ of TMF. Thus, the refraction coefficient should change by 4%, which coincides with the previous estimation.

At a frequency higher than 3300 cm^{-1} (wavelength of 3000 nm), the reduction of TMF transmittance is observed (Fig. 2). The TMF transmittance decreases with frequency and might be related to the appearance of a diffusion scattering of light. As a consequence, a part of light misses the receiving plate of the spectrometer's receiver. The scattering might be due to bulk and surface irregularities caused by ion

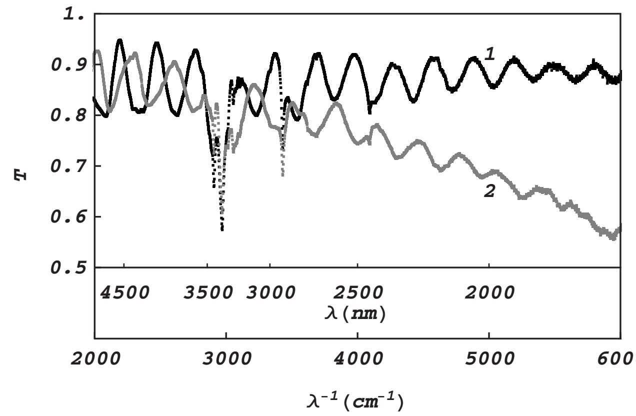

FIG. 2. Experimental data of the light transmittance through a TMF (curve 2) and uniform lavsan film (curve 1)

bombardment. The characteristic size of those irregularities estimated with the help of transmittance spectra is about 1000 nm, which is a typical distance between the holes. (see Fig. 1)

To evaluate the value of PF permittivity, we employ the transmittance and reflection coefficient measured at different angles of incidence.

All the theoretical evaluations are made with assumption that we deal with plane waves and that the permittivity of PF is described by the Lorentz dispersion with four resonances. By making theory fit the experiment, we match film thickness and parameters of dispersion. The PF permittivity was assumed to be anisotropic, which is quite common for polymer films and is caused by mechanical stress. The introduction of anisotropy both improves the level of agreement with the experimental data (some of these results are shown in Figs. 3 and 4) and explains the observed forked maxima of the transmittance coefficient of PF with a deposited silver layer (see Fig. 4).

The experimental data on transmittance of PF and PF with a deposited silver layer are satisfactory described by

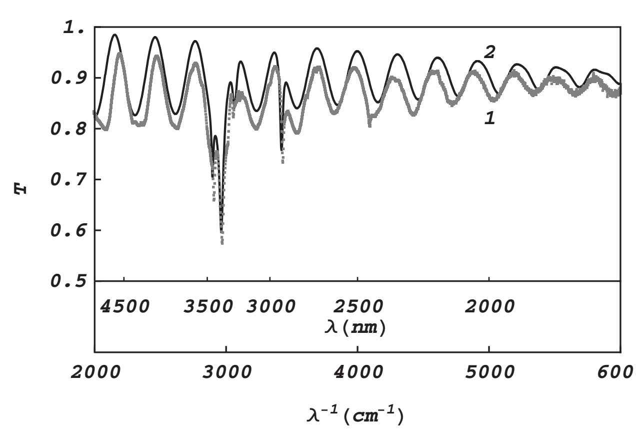

FIG. 3. Coefficient of normal transmission through homogeneous lavsan film: experiment (dots, pointed by 1) and calculation (solid line, pointed by 2).

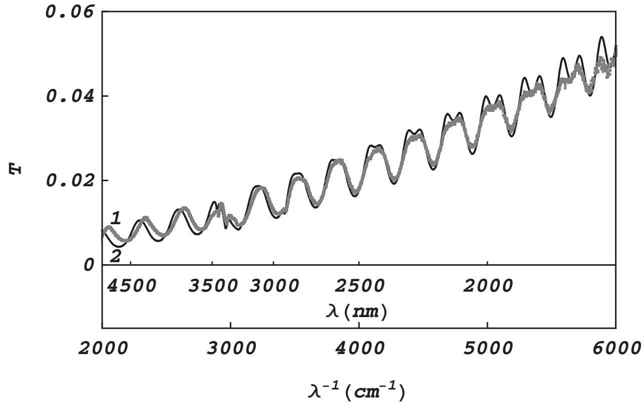

FIG. 4. Coefficient of normal transmission through lavsan film with deposited silver layer: experiment (dots, pointed by 1) and calculation (solid line, pointed by 2).

$$\varepsilon_L = \varepsilon_0 + \sum_{i=1}^{4} \frac{f_i}{\Lambda_{0i}^2 - \Lambda^2 - 2i\gamma_i\Lambda}, \quad (1)$$

where Λ is inverse wavelength in cm^{-1} and ε_0 takes the values of 2.45 and 2.57 along and across the anisotropy axis. The other parameters are considered to be isotropic.[24] The Lorentz-shaped terms are included in Eq. (1) to describe the stretching mode of ($>CH_2$) group at 3000 cm^{-1} and of overtone of ($C=O$) group at 3400 cm^{-1}. The silver permittivity values were taken from Ref. 25. For the sake of convenience, those data were interpolated by a dispersion law of the Drude type (Fig. 5):

$$\varepsilon_{Ag} = 4.06 - \frac{5.52 \cdot 10^9}{\Lambda^2 + 180 \cdot i\Lambda}. \quad (2)$$

It has been found that the thickness of the silver layer is equal to 17.2 nm and that of the PF is $1.03 \cdot 10^4$ nm.

The obtained parameters of PF permittivity are in a good agreement with known values of the lavsan permittivity. Thus, all the observed specific features of the transmission spectrum, but for the "blooming" effect, can be explained within the frames of continuous medium model. The successful application of the model was expected because the

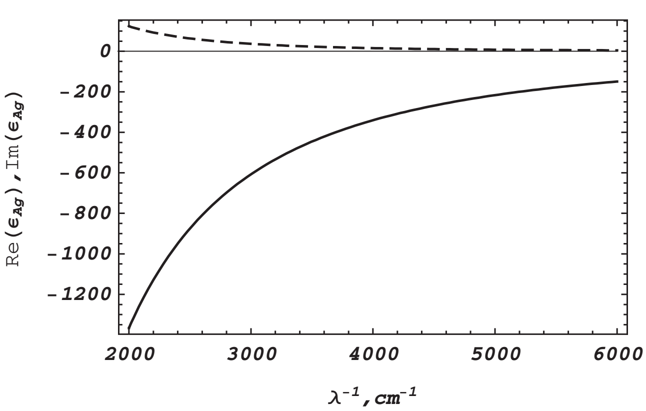

FIG. 5. Dispersion of real (solid line) and imaginary (dashed line) parts of permittivity of silver.

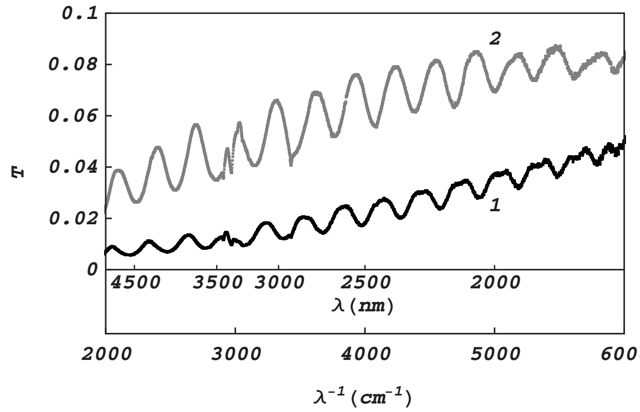

FIG. 6. Experimental data of the light transmittance through a silver film deposited onto a TMF (curve 2) and onto a PF (curve 1). The observed oscillations are caused by the interference in the PF and TMF, respectively.

diameter of holes and the average distance between the holes were much less than the wavelength of the incident radiation.

III. EFFECT OF BLOOMING OF PF WITH SILVER LAYER BY A RANDOM ARRAY OF SUBWAVELENGTH HOLES

The experiment showed that the silver film is "bloomed" by the system of holes. In other words, the transmittance of the silver film deposited on TMF is two to six times higher than that of the silver film deposited on PF (Fig. 6). It is worth noting that without a silver film, we have opposite situation (Fig. 2). The effect is observed within the whole mid-IR range of the spectrum.

Let us consider in detail the phenomenon of blooming. In the physical optics approximation, the existence of such small holes ($0.04\lambda - 0.13\lambda$) with the surface density of 10% should not cause a considerable additional radiation transmission.[26] Actually, within the frequency range of investigation, the silver permittivity is about $\varepsilon \approx -1000$ and the transmission of a continuous film is several percent.[27] Therefore, to evaluate the contribution of the holes to the transmission we can use Bethe's result,[26] see also Ref. 29. It reveals that at a normal wave incidence onto the perfectly conducting screen with a hole that is much less than the wavelength, the field, which is excited behind the screen, is equivalent to the field of a magnetic dipole with the magnetic moment of $M \sim a^3 H_i$, where a is the size of the hole and H_i is the magnetic field in the incident wave. A system of magnetic dipoles can be considered as the surface magnetic current $j_M = \dot{M} \sim c(ka)Na^2 H_i$, where N is the density of holes with $Na^2 = \alpha \sim 0.1$. Thus, we arrive at the estimation $j_M \sim c(ka)\alpha H_i$. The amplitude of the wave generated by the current is defined by the value $E = H \sim j_M / c$ with the accuracy of up to a numerical factor.[30] The Poynting vector of the transmitted wave is proportional to $EH \sim (ka)^2 \alpha^2 H_i^2$. Thus, the expected transmittance coefficient is $T \sim \alpha^2 (ka)^2 \sim 0.1^2 (200\ \mathrm{nm}/2000\ \mathrm{nm})^2 \sim 10^{-4}$, which is much lower than the observed effects (4%–6%).[31] As we can see in Fig. 1, the holes may form clusters. This is inevitable feature of the technology. The size

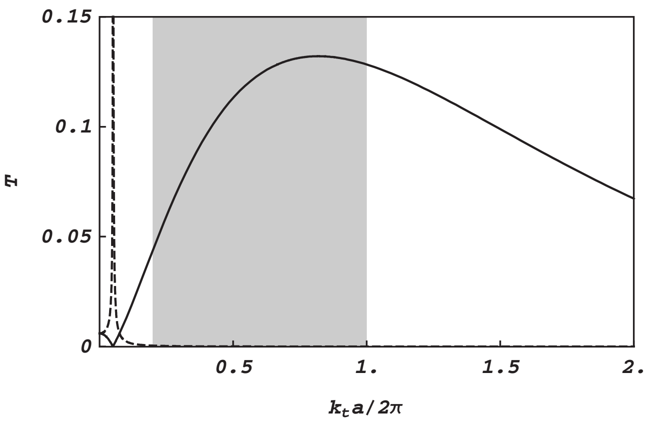

FIG. 7. A relative amplitude of a surface wave at the rear surface of the silver film at fixed frequency $\Lambda=2500$ cm^{-1}, which corresponds to silver permittivity $\varepsilon_{Ag}=-854+63i$. The plate is 17 nm thick. The solid line corresponds to TE polarization, the dashed line corresponds to TM polarization. The waves in vacuum are evanescent ($k_0^2<k_t^2$) to the right from the point $k_t a/2\pi\approx 0.05$

of the clusters does not exceed $5a$; thus, the account of clusters' formation can increase the evaluation by 25 times that is still not enough to explain the observed transmission.

Hence, an increase in the transmission is impossible to explain, if we do not consider the phenomena related to the excitation of near fields that appear when the incident wave is scattered by holes. That is why we refer to this effect as extraordinary transmission.

The near fields that appear at the scattering of a plane wave by a system of holes can be expanded in series of inhomogeneous waves that have the real tangential wave numbers k_t.

The transmittance of TE- and TM-polarized inhomogeneous waves is defined by the longitudinal wave number $k_z=\sqrt{\varepsilon k_0^2-k_t^2}$ and impedance values $Z_{TE}=k_0/\sqrt{\varepsilon k_0^2-k_t^2}$, $Z_{TM}=\sqrt{\varepsilon k_0^2-k_t^2}/(\varepsilon k_0)$, respectively.

At $k_t^2 \gg \varepsilon k_0^2$, the impedance of TE-polarized waves ceases to depend on ε ($Z_{TE}\approx Z_{TEvac}\approx -ik_0/k_t$). Since the longitudinal wave number $k_z\approx ik_t$ also ceases to depend on ε, the transmitted wave behaves as if there is no metal at all: $T\approx\exp(-k_t d)$ (see Fig. 7)[33] and there is no efficient reflection from the silver film. In the other extreme case $k_t\to k_0$, the wave impedance in the film tends to a constant $-i/\sqrt{1-\varepsilon}$, whereas the impedance in vacuum Z_{vacTE} tends to infinity. As a consequence, the wave is totally reflected, and $T\ll\exp(-k_t d)$. The case of $k_t a/2\pi\sim 1$, which we are interested in, corresponds to a transitional mode, where T exceeds 10% (see the shaded area in Fig. 7).

The impedance $Z_{TM}=ik_t/\varepsilon k_0$ of TM-polarized wave in the film always considerably differs from the impedance $Z_{vacTM}=ik_t/k_0$ in vacuum (see frequency dispersion of silver permittivity in Fig. 5). However, due to $\varepsilon<0$ the evanescent TM-polarized wave can excite a surface wave.[34] At the exact frequency of plasmon resonance, there is a significant transmission (a peak of the dashed line in Fig. 7). Nevertheless, at spatial frequencies determined by scattering of the incident plane wave on separate hole ($k_t\sim 2\pi/a$), on ensemble of holes ($k_t\sim 2\pi/d$, where $d\sim 1/\sqrt{N}\sim a/\sqrt{\alpha}\sim 3a$ is an average distance among holes) or on a cluster of holes ($k_t\sim 2\pi/d_{cl}, d_{cl}\leq 5a$), the transmittance of the TM-polarized wave is about 10^{-4} (see the shaded area in Fig. 7) and cannot explain the observed effect of transmission.

Thus, we regard a possible mechanism of blooming as follows: though the incident plane wave has a low transmittance coefficient, it produces evanescent waves (near fields) while scattering. For TE-polarized evanescent waves, the transmission coefficient exceeds that of a normally incident plane wave by more than an order. On transmission, the evanescent waves are scattered by the same system of holes[35] and, in accordance with the reciprocity theorem, efficiently form the transmitted wave.

IV. CONCLUSIONS

For the first time, it is proved experimentally that a random system of holes in a metal film can produce an extraordinary transmission in a wide frequency range. This phenomenon is related to the phenomenon of filtration of inhomogeneous TE waves.[7]

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[23] As seen in Fig. 3 the intensity of all characteristic peaks decrease by the same value.

[24] $\Lambda_{01}=2897cm^{-1}$, $\Lambda_{02}=2963cm^{-1}$, $\Lambda_{03}=3065cm^{-1}$, $\Lambda_{04}=3422cm^{-1}$, $\gamma_1=7.96cm^{-1}$, $\gamma_2=15.92cm^{-1}$, $\gamma_3=20.69cm^{-1}$, $\gamma_4=12.73cm^{-1}$, $f_1=648cm^{-2}$, $f_2=2928cm^{-2}$, $f_3=1226cm^{-2}$, $f_4=1340cm^{-2}$.

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[27] Note that interference pulsing of the transmittance can be related only to the imaginary part of permittivity if the real part is negative (Ref. 28). Since the thickness and tangent of losses of the silver film are small we do not observe in the investigated frequency range any influence of transmittance interference.

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[31] From percolation theory it is well known that in a random system a cluster contains on average 2-3 particles (Ref. 32). The diameter of a triple-hole cluster is $a_3=a(1+2/\sqrt{3})\approx 2.1547$. Supposing that all the holes are concentrated in triple-hole clusters we arrive at the following upper estimation of the magnetic current $j_M=\dot{M}\sim c(ka_3)n_3a_3^2H_i$ where n_3 is the density of triple-hole cluster with $n_3a_3^2=\alpha\sim 0.1$. Thus, at a fixed relative area occupied with holes the magnetic current increases linearly with the diameter of a hole whereas the transmittance coefficient does it quadratic. The clusterization of the holes may increase the transmittance coefficient by a factor of 4.6 and cannot explain the experimental results. Computer simulation of transmittance trough a perfectly conducting screen with a single hole and with three-holes cluster shows in the latter case a fourfold rise of the transmittance in agreement with our analytical estimation.

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[33] At a normal incidence impedance is $Z_{TE}^{-1}=i\sqrt{|\varepsilon|}$, which provides the impedance contrast and causes reflection.

[34] At $|\varepsilon|>>1$ from the condition of surface-plasmon excitation $\sqrt{k_t^2-k_0^2}=\sqrt{k_t^2+|\varepsilon|k_0^2}/|\varepsilon|$ one can derive $k_t\approx k_0(1+1/2\varepsilon)$. In Fig. 7 the plasmon resonance is revealed as a peak of *TM*-polarized inhomogeneous wave transmission.

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