Surface plasmons at nanoscale relief gratings between a metal and a dielectric medium with optical gain

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Surface plasmons at the interface between metal and a dielectric with strong optical amplification are analyzed theoretically. It is shown that proper choice of optical indices of the dielectric medium results in an infinitely large effective refractive index of surface waves. Such resonant plasmons have extremely low group velocity and are localized within a vanishingly small distance near the interface. The plasmon-related anomalies in the UV reflection spectra are predicted for nanoscale gratings on a surface of a silver film covered by a concentrated dye solution with high optical amplification.

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Surface plasmons and polaritons are electromagnetic oscillation localized at the interface between materials with positive and negative dielectric permittivities. When the negative permittivity results from the plasma effect of free electrons in metals, such surface oscillations are called plasmons. In polar dielectrics, the permittivity becomes negative in a relatively narrow spectral range in the middle infrared between the frequencies of transverse and longitudinal optical phonons. Corresponding surface modes are called polaritons. Once the dielectric permittivity is specified, the electrodynamic description of surface plasmons and polaritons becomes essentially identical: it is nothing more than a solution of Maxwell equations that exponentially decays in both directions away from the interface. In this article, we refer to the material with negative permittivity $\varepsilon_m=\varepsilon_m'+i\varepsilon_m''$ $(\varepsilon_m'<0)$ as metal, and the other material with positive permittivity $\varepsilon_d=\varepsilon_d'+i\varepsilon_d''$ $(\varepsilon_d'>0)$, as dielectric. We show that at certain conditions the surface plasmons (polaritons) can be localized within the distance from the interface that can be orders of magnitude less than the light wavelength in vacuum. For visible and near ultraviolet light, this means that plasmons' energy is localized within few nanometers at the surface. The group index approaches infinity, which results in extremely slow group velocity. The plasmons' effective refractive index becomes very large, which allows to suggest that such surface modes can efficiently interact with nanoscale objects. We show by simulations that plasmon-related anomalies in reflection and transmission can exist for near UV wavelengths when the grating has a period as short as 10 nm and a half-depth as small as 0.1 nm.

Surface plasmons have been studied comprehensively.[1–5] The plasmons' effective refractive index, $n_p=\sqrt{\varepsilon_d\varepsilon_m/(\varepsilon_d+\varepsilon_m)}$, in a typical case of an interface between vacuum $(\varepsilon_d=1)$ and metal with low losses $(\varepsilon_m''\ll|\varepsilon_m'|)$ such that $|\varepsilon_m'|\gg1$, is reduced to

$$n_p\approx 1+\frac{1}{2|\varepsilon_m'|}+\frac{i}{2}\frac{\varepsilon_m''}{(\varepsilon_m')^2}. \tag{1}$$

The real part of the effective index is slightly larger than the refractive index of the dielectric medium at the interface, and the plasmons' optical loss at wavelength λ,

$$\alpha_p=\frac{4\pi}{\lambda}\,\mathrm{Im}(n_p)\approx\frac{2\pi}{\lambda}\frac{\varepsilon_m''}{(\varepsilon_m')^2}, \tag{2}$$

is small enough to allow for propagation length of dozens of wavelengths and larger. The strength of the plasmons' electric field decays exponentially, so that the field penetration depth t at $1/e$ level is

$$t_{d(m)}=\frac{\lambda}{2\pi}\,\mathrm{Re}\sqrt{\frac{\varepsilon_d+\varepsilon_m}{-\varepsilon_{d(m)}^2}}, \tag{3}$$

where subscripts d and m correspond to dielectric and metal, respectively. Thus, in the case of a "good" metal with low losses and large negative dielectric permittivity, the plasmons' field penetration into the dielectric medium $t_d\approx(\lambda/2\pi)\sqrt{-\varepsilon_m'}$ is comparable or larger than the vacuum wavelength, while penetration into metal $t_m\approx\lambda/2\pi\sqrt{-\varepsilon_m'}$ is much smaller than the wavelength. The maximum of the plasmons' electromagnetic field is located exactly at the surface, which accounts for practical applications of surface plasmons in optical sensors capable of detecting a monolayer of molecules adsorbed by the surface.[6] Surface plasmons are also responsible for gigantic enhancement of Raman scattering.[7] Strong localization of plasmons' field offers the potential for developing miniaturized photonic circuits.[8] Surface plasmons allow for a new form of scanning-probe microscopy and spectroscopy.[9] Recently, extraordinary high light transmission through subwavelength periodic hole arrays in metallic films has been found to result from resonant excitation of surface plasmons.[10] Due to the strong localization of the plasmons' field at the metal interface, the resonant transmission through the periodically modulated thin metal film is expected to be highly nonlinear.[11] Surface magneto-optical interaction[12] and dipole-dipole interactions between nanoparticles[13] can be enhanced using surface plasmons.

The effective refractive index and the penetration depth are expected to change dramatically when $\varepsilon_d+\varepsilon_m\to0$. This is known as a resonance condition for a plasmon at a planar interface: $\varepsilon_m=-1$, assuming that the dielectric medium is a vacuum. The plasmon waves in this case are called surface resonant plasmons. It does not seem to be easy to satisfy the

resonance condition due to substantial optical losses in metals. The dielectric function of metals can actually be approximated by the Drude-Sommerfeld formula

$$\varepsilon_m = 1 - \frac{\omega_p^2}{\omega(\omega + i\Gamma)}, \tag{4}$$

where $\omega = 2\pi c/\lambda$ is the radian frequency, c is the speed of light in vacuum, ω_p is the Drude plasma frequency of free electrons, and Γ is the phenomenological relaxation constant. By setting, in Eq. (4), $\mathrm{Re}(\varepsilon_m) = \varepsilon_m' = -1$, one gets the resonance frequency $\omega_R = \sqrt{(\omega_p^2 - 2\Gamma^2)/2} \approx \omega_p/\sqrt{2}$ and inevitably a nonzero imaginary part $\mathrm{Im}(\varepsilon_m) = \varepsilon_m'' = 2\Gamma/\omega_R \approx 2\sqrt{2}\Gamma/\omega_p$. The optical loss of the metal at the resonant frequency is rather high, which greatly reduces resonant phenomena. In addition to the plasma oscillation relaxation, there are electronic interband transitions, which contribute to the optical loss at high frequencies.

For a practical estimation we took optical constants of silver from[14] and, using spline interpolation, found that $\varepsilon_m' = -1$ at $\lambda \approx 337$ nm. The imaginary part of the dielectric function at this wavelength is $\varepsilon_m'' \approx 0.582$. It is easy to check that effective refractive index of the plasmon at this wavelength becomes $\mathrm{Re}(n_p) \approx 1.22$, only slightly higher than unity rather than being infinitely large, as one may expect at $\varepsilon_m + 1 \to 0$. The propagation loss is as high as $26\mu^{-1}$, so that the plasmon is strongly overdamped. One can hardly expect any resonant phenomena with such strong plasmon losses. When dielectric material at the interface has high refractive index (e.g., $n_d = 3$, $\varepsilon_d = n_d^2 = 9$) the resonance is shifted to a longer wavelength. For silver, $\varepsilon_m' = -9$ at $\lambda = 509$ nm. The effective refractive index becomes $\mathrm{Re}(n_p) \approx 7.5$, more than twice the index of the dielectric. The optical propagation loss is still very large: $170\mu^{-1}$.

The singularity of plasmons' effective index at $\varepsilon_d + \varepsilon_m \to 0$ is quite evident so it has been briefly discussed in early works on surface plasmons (see, e.g., Ref. 3). An experimental observation has been reported very recently by Smolyaninov.[15] He used 488 nm laser light in the Kretschman geometry (illumination of a thin film through a glass prism) to excite surface plasmons at the interface between gold and glycerin and observed whispering gallery modes of surface plasmons in liquid microdroplets on a gold film. The high effective refractive index of plasmons allows for the whispering modes to exist in micrometer-scale droplets. The excitation of the whispering modes has been confirmed by observation of enhanced light intensity in the vicinity of the droplets' boundaries using the near-field measurements. No quantitative studies have been made to characterize the effective refractive index and optical loss of surface plasmons. Using interpolated optical data[14] for gold, one can estimate $\varepsilon_m \approx -2.213 + i3.831$ at $\lambda = 488$ nm. At the interface with glycerin (refractive index $n_d = 1.473$, $\varepsilon_d = n_d^2 = 2.170$), the plasmons' effective index becomes $n_p = 1.531 + i0.401$, and the optical propagation loss is $10.3\ \mu\mathrm{m}^{-1}$. Remarkably, qualitative observations are possible at such a high level of optical loss.

The plasmons' properties change dramatically when the dielectric at the interface has a strong optical gain. In this case the imaginary part of dielectric function is negative ($\varepsilon_d'' < 0$), so that the resonance condition $\varepsilon_d + \varepsilon_m = 0$ may indeed be satisfied, resulting in an infinitely large effective index of the plasmon $n_p \to \infty$, which corresponds to the infinitely small effective plasmon wavelength $\lambda_p = \lambda/n_p \to 0$. Furthermore, the optical gain in the dielectric may compensate for losses in the metal, resulting in a longer propagation length of the plasmon. Finally, the penetration depth also becomes much smaller: $t_{d(m)} = \lambda/(2\pi\,\mathrm{Re}\sqrt{n_p^2 - \varepsilon_{d(m)}})$. In other words, at a frequency in the visible or near ultraviolet spectral range the surface resonant plasmon can be localized within a vanishing thin slab near the interface, its effective wavelength can be orders of magnitude smaller than the vacuum wavelength, and the plasmon can propagate for a macroscopic distance. A practical limit will be set by the feasibility of obtaining necessary optical gain in the dielectric and, more fundamentally, by the fact that materials have atomic structure and that the concept of a uniform medium with local polarizability will eventually become inappropriate.

To illustrate these conclusions, we perform following numerical simulations. First, we note that the condition $\varepsilon_d = -\varepsilon_m$ in terms of optical indices $n + ik = \sqrt{\varepsilon}$ means $n_d = k_m$ and $k_d = -n_m$. Assuming that the dielectric is a concentrated dye solution with strong optical gain, we take $n_d = 1.34$, close to the refractive index of methanol at UV wavelengths. Using optical data for silver, we find that $k_m = 1.34$ at a wavelength of $\lambda \approx 349$ nm, and then $n_m \approx 0.224$ at this wavelength. Thus, to achieve the resonance, the dielectric medium must have $k_d \approx -0.224$, which corresponds to an optical gain of $g = -4\pi k_d/\lambda \approx 8.07\ \mu\mathrm{m}^{-1} = 8.07 \times 10^4\ \mathrm{cm}^{-1}$. By adding high-index oil to the solution one can shift the resonance to longer wavelengths where the losses in metal are smaller, so that a smaller optical gain will be required.

Commercially available laser dyes (e.g., Exciton, Inc.) densely cover the wavelength range from 311 to 1530 nm. Several groups of chemicals provide optical gain and lasing around the wavelength of interest: ~350 nm (known as Terphenyl, TMQ, BPBD, PBD, etc.). For application in lasers, the optical gain should be in the range from a single to hundreds of cm^{-1}, and these numbers are often reported in the literature. Larger gain is possible with a higher concentration of dyes. To quantify the optical gain of highly concentrated dye, one should work with extremely short optical path. With a cavity length of 10 μm, an optical gain in thousands of cm^{-1} can be measured. For example, in solid thin films of low-molar-mass dyes, such as thiophrene-based oligomers,[16] an optical gain cross section of $6 \times 10^{-16}\ \mathrm{cm}^2$ has been estimated from pump-probe experiments, which corresponds to optical gain of the order of $2.2 \times 10^3\ \mathrm{cm}^{-1}$. These experiments were done at visible wavelength. It has been pointed out, though, that the reported value of the gain cross section is underestimated. We would like to emphasize that the theoretical limit for the gain cross section set by quantum mechanics, $\omega = 3\lambda^2/2\pi$,[17] is many orders of magnitude higher ($5.8 \times 10^{-10}\ \mathrm{cm}^2$ at $\lambda = 349$ nm), so that achieving larger optical gain must be feasible. Such large figures of optical gain are not reported mainly for two reasons. First, for practical application in dye lasers so large a gain is not needed and the

dyes are usually strongly diluted to bring the optical gain to a technologically comfortable range usually below 100 cm^{-1}. Second, even if that strong optical gain is achieved, measuring it would require a very short cavity. For example, if the cavity mirrors have antireflectance coatings with a reflection as small as 10^{-5}, the cavity should be only $\ln(10^5)/g \approx 1.4$ μm long to avoid lasing of medium with $g=8$ μm^{-1}. Another difficulty in quantifying a large gain is the gain saturation at a high level of optical power. Short cavities and low input optical signals would be required to measure high optical gain and to avoid saturation. As of now, there are no studies of optical gain of highly concentrated dyes in ~ 1 μm thick flow cells with $<10^{-5}$ residual reflectance, but this certainly does not mean that high optical gain is not achievable. Another candidate for high-gain dielectric near the metal interface is a semiconductor quantum well or an array of quantum dots. With a confinement factor of 10^{-2}, optical gain for guided modes in semiconductor lasers easily reaches $100-300$ cm^{-1},[18] which corresponds to the optical gain in the quantum well of $(1-3)\times 10^4$ cm^{-1}. In addition, due to a much larger refractive index of semiconductors, the resonance will be shifted to the visible spectral range, or, with appropriate choice of metal, even to the near infrared.

In a close proximity of the metal interface, typically below 5 nm, there could be nonradiative processes that may result in direct transfer of energy from a dye molecule to an atom of the metal (Forster's resonant dipole coupling[19]). Fortunately, such direct interactions are strong only in a resonant case, when the energy of an electron in an excited state in the dye molecule is close to the energy of the electron at the metal interface. Moreover, studies of molecular fluorescence in close proximity of metallic surface[20,21] suggest that the observed quenching of the emission is produced mainly by transferring energy to surface plasmon polaritons.[22] When a monolayer of dye molecules is deposited on the metal surface directly, the fluorescence intensity is reduced due to nonradiative interactions, but it never disappears completely. A spacer layer in steps of approximately 1 nm thin monolayers of optically inert transparent Langmuir-Blodgett films can be used to reduce direct energy transfer to metal. It has been shown that a monolayer of dye molecules significantly affects the dispersion curve for the surface plasmons[23] even though the plasmons' field is spread far beyond the dye film. In application to fluorescence-based sensors,[24] appropriate radiative decay engineering allows for 20- to 1000-fold improvement of the detection limit when using direct coupling of flurescence to surface plasmons.

In this article we study how the optical gain in the dielectric affects the properties of surface resonant plasmons. In the simulation below we fix the real part of the refractive index at $n_d=1.34$, vary the imaginary part k_d from zero to -0.223 (slightly below the divergence condition), and study how the surface plasmon is modified. Optical properties of metal are described using the optical constants for bulk silver. Although the mean free path of electrons in bulk metallic monocrystals at room temperature is of the order of $30-50$ nm, bulk optical constants are often used for evaluation of light transmission and reflection by thin films. At optical frequencies, displacement of electrons caused by the electromagnetic wave is rather small. For an amplitude of electric field of $E=10^6$ V/cm (corresponding to a light intensity of $P=E^2/(2\cdot 377\Omega)\approx 1.3\times 10^9$ W/cm^2), the classical electron displacement is estimated to be as small as $a=Ee/m\omega^2 \approx 0.6$ pm. This is a reason that estimations based on bulk optical properties usually work well even for very thin films. Quantum confinement phenomena in metallic nanostructures become observable when a characteristic size of the structure is of the order of few nanometers, which results in a noticeable change of electrons' density of states. The estimations done in this article are based on a simple classical theory and aim to reveal the fundamental properties of nanoplasmons.

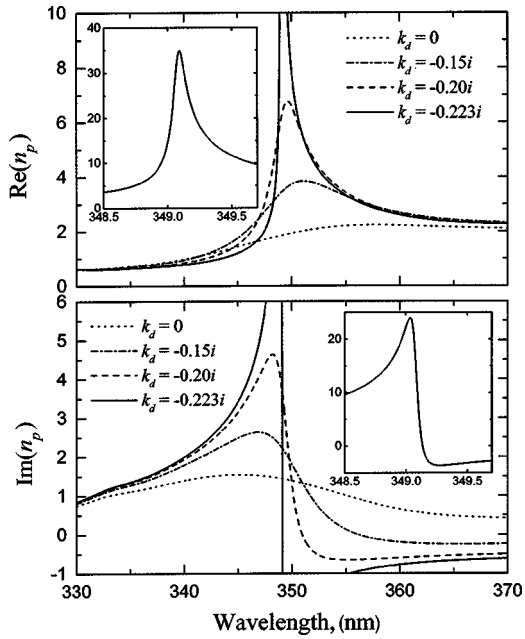

FIG. 1. The real (top) and imaginary (bottom) parts of the effective refractive index of a surface plasmon. The dielectric medium has the real part of the refractive index $n_d=1.34$ and the negative imaginary part k_d ranging from 0 to -0.223. The other medium is silver. The insets show the case of $k_d=-i0.223$, slightly below the divergence condition.

Figure 1 shows the real and imaginary parts of the plasmons' effective refractive index for different values of optical gain in the dielectric. Without the optical gain, the resonance is barely observable. With optical gain (more exactly, the negative imaginary part of the refractive index of the dielectric k_d) approaching the divergence condition ($k_d \to -n_m$; $n_d=k_m$), the plasmons' refractive index increases resonantly, and the spectrum of plasmons' loss reveals resonant behavior changing sharply from large positive to large negative values. Positive plasmon loss means that light amplification in the dielectric does not provide compensation for the optical loss in metal. Negative plasmon loss, in contrast, means that optical gain in dielectric overcompensates for the loss in the metal. Interestingly, for a wavelength slightly below the resonance, the plasmon as a surface excitation still exists, although the optical loss is enormously high. What looks unusual, adding optical amplification to the dielectric results in even stronger damping of the plasmon at

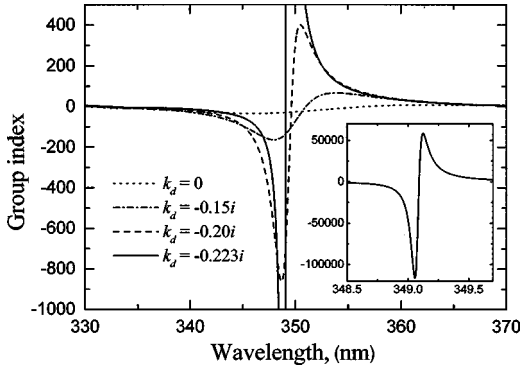

FIG. 2. The real part of the group index of a surface plasmon. Material constants are the same as in Fig. 1. The inset shows the case of $k_d = -i0.223$.

these wavelengths. For $k_d = -0.223$, the exact compensation of loss $[\mathrm{Im}(n_p) = 0]$ happens to be at $\lambda = 349.14$ nm, the plasmons' effective index at this wavelength is equal to $n_p = 28.86$, and its penetration depth into the metal and dielectric is $t_m \approx t_d \approx \lambda / 2\pi n_p = 1.93$ nm. By choosing k_d closer to the divergence condition (a tiny increase of optical gain is required), it is possible to get an even higher value of the plasmons' effective refractive index and, consequently, even stronger surface localization. The spectral interval in which such phenomena can be observed, however, is reducing, which will create another difficulty in experimental studies of resonant plasmons. The spectral resolution of about 0.1 nm must be enough to resolve the spectral features $\mathrm{Re}[n_p(\lambda)]$ and $\mathrm{Im}[n_p(\lambda)]$ shown in Fig. 1 even for the case $k_d = -0.223$, when the plasmons are localized within single nanometers from the interface.

The nanometer-scale localization of resonant surface plasmons near the interface is expected to result in a strong electromagnetic field at the interface and strong optical nonlinearity. Even in isotropic liquid, the polar molecules tend to be oriented near the interface, resulting in a nonzero $\chi^{(2)}$ nonlinearity. The resonant surface plasmon, being localized within one or a few molecule sizes from the interface, will effectively interact with the oriented molecules.

The sharp change of the effective index of the plasmon with wavelength results in another interesting phenomenon: strong reduction of the plasmon group velocity. The group index $n_g = n_p(\lambda) - \lambda dn_p/d\lambda$ becomes huge. The real part of the group index as a function of wavelength is shown in Fig. 2. When $k_d = -0.223$, at the zero loss wavelength (349.14 nm), the group index is as large as $n_g \approx 5.4 \times 10^4$, which corresponds to the plasmon group velocity in single kilometers per second. Further reduction of the resonant plasmon group velocity is possible by approaching closer to the divergence condition. An extremely slow velocity of light has been observed so far in different resonant media, such as the Bose-Einstein condensate of ultracold atomic vapor[25] and even at room temperature in crystals.[26] The surface resonant plasmons thus become another type of optical waves that propagate much more slowly than light in vacuum.

The strong localization of plasmons' field is expected to result in a very efficient interaction with nanometer- and subnanometer-scale perturbations of the surface. Here we

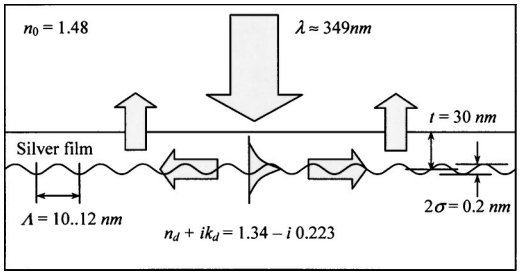

FIG. 3. The scheme of the nanoscale grating structure that shows plasmon-related anomalies in the reflection spectra.

consider the plasmon-related anomalies (often referred to as Wood anomalies[27]), in light reflection from the surface relief gratings. Once the plasmons' effective index is so large $(n_p \approx 29)$, the gratings with period $\lambda / n_p \approx 349\ \mathrm{nm}/29 \approx 12$ nm will provide resonant coupling between the incident light and the plasmon. The simulation is done using the Fourier-Rayleigh approximation: in each layer the electromagnetic field is represented as a linear combination of plane waves corresponding to the diffraction orders, and the strength of electric field in each plane wave is chosen to satisfy the boundary conditions at the interfaces including the sinusoidally corrugated interface between the metal and amplifying dielectric. We used 15 diffraction orders (from −7th to +7th). This approach has been shown to provide accurate quantitative description of light diffraction on metal gratings including extraordinary light transmission through periodical holes in metal films.[28] The structure under analysis was following (Fig. 3): at the top there is a semi-infinite medium with index $n = 1.48$ (fused quartz), then a 50 nm thick silver film (optical indices for each wavelength are determined as discussed above), and a semi-infinite active medium at the bottom with index $1.34 - i0.223$. The interface between the silver film and the active layer is corrugated according to $z = \sigma \cos(\vec{K}\vec{r}_{\parallel})$, where σ is the amplitude of the surface corrugation (half of the peak-to-peak corrugation), $K = 2\pi/\Lambda$ is the grating wave vector, Λ is the grating period, and $\vec{r}_{\parallel}$ is the in-plane radius vector. In all further simulation we set $\sigma = 0.1$ nm, so that peak-to-peak corrugation magnitude is comparable to the inter-atomic distance in solid materials. The period Λ was in the range 10 to 12 nm. Normal incidence was assumed for most of numerical examples. Light polarizations TE and TM refer to polarization of diffracted waves. For the normal incidence, TE (TM) polarization corresponds to the electric field vector of the incident wave normal (parallel) to the grating wave vector.

The simulation results are shown in Fig. 4. No spectral features are observed for TE-polarized light, while for the TM polarization, the reflection spectrum shows a characteristic resonant behavior when the incident wave is in resonance with the surface plasmon. These spectral features are located between 349.1 and 349.2 nm, where the imaginary part of the plasmons' effective refractive index is close to zero. When the grating period is changing from 10 to 12 nm, the resonance is slightly shifted to longer wavelength mainly because the sharp change of the real part of the plasmons' effective index, which controls the location of the resonance. A shift to longer wavelengths results in a

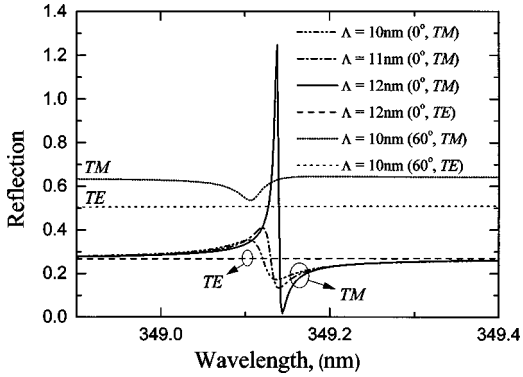

FIG. 4. Reflection spectra of a thin silver film with a nanoscale corrugated boundary at the interface with the dielectric medium with strong optical amplification.

larger amplitude of the resonance due to smaller propagation loss at longer wavelengths. The reflection spectrum at $\Lambda = 12$ nm shows a peak larger than unity, which is associated with optical amplification of the plasmons' field while it propagates along the interface. In contrast to the spectra of conventional plasmon-related anomalies the incident angle only slightly affects the resonance conditions due to a very large value of the effective plasmons' index. At 60° incident angle and $\Lambda = 10$ nm the reflection minimum is observed in practically the same wavelength range. The shape of the resonance is changed, though. At oblique incidence, reflection for TE-polarized light shows no variations, as expected. Qualitatively, all these features in reflection spectra are easy to understand: they are nothing else but Wood anomalies associated with the surface resonant plasmons at the interface between metal and amplifying dielectric. The striking peculiarity is that they are produced by 10–12 nm gratings with 0.1 nm half-depth, while the light wavelength is 349 nm.

The conditions at which the surface resonant plasmons are expected to be observed are rather unusual. Achieving such large optical gain in the dielectric in close proximity to the metal surface would be a challenging task. It will obviously require an appropriate pump scheme. To achieve high gain, pumping by short intense optical pulses with very low duty cycle will be preferable. Further, when pumping a liquid layer on top of a metal film, the interference between the incident and reflected waves will always strongly reduce the intensity of pumping light near the interface. To achieve a strong optical pump at the very surface, the pumping wave itself should be a plasmon with the maximum of its intensity at the interface. Another challenging task would be to fabricate a nanoscale grating at the metal surface. Lateral resolution of the order of 10 nm and better is achievable with modern tools such as focused ion beam (FIB) milling. Once the grating height is comparable to the interatomic distance, the metal surface must be atomically flat. Fortunately, this flatness is required in a relatively small area. In the FIB system, usually it is possible to observe the sample image using secondary electrons produced by the ion beam. Due to the strong channeling effect in metals, individual monocrystalline grains are clearly distinguished. Thus, it must be possible to select a single monocrystalline grain, say, a few micrometers across, and fabricate the nanograting within the grain. A 1 μm grain can accommodate 100 grating grooves, which is enough for observation of an optical resonance with a Q-factor of about 100. Other technical solutions are also possible.

The resonant surface plasmons have many interesting and unique properties, and thus deserve further study. They could be useful for characterization of surface defects at the (sub) nanometer scale and for the development of a new type of extremely sensitive optical sensors capable of detecting small number of molecules adsorbed to the metal surface. Another potential study field would be a new type of optical nonlinear interaction at the surface.

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