Propagation dynamics of femtosecond pulse through subwavelength metallic hole arrays

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We have studied the frequency-dependent transmission time delay of femtosecond (fs) laser pulses through subwavelength hole arrays in aluminum films using the up-conversion technique. The pulse delays measured at different wavelengths mainly follow the far-field transmission profile of the surface-plasmon polaron (SPP) resonances. Temporal delays of 60 and 100 fs were found at the major SPP resonance in the single-layer and double-layer samples. A coupled-SPP transmission model is used to explain the temporal dynamics of the transmission process. Our experiment shows that the weak coupling between the SPP waves on different metal-dielectric interfaces leads to the large temporal delay of the transmitted pulses.

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I. INTRODUCTION

The observation of extraordinary optical transmission1 (EOT) through subwavelength holes in metal films has sparked intense experimental and theoretical investigations in recent years. Not only can the transmission be orders of magnitude larger than expected from classic diffraction theory, but also the peak positions in the transmission spectrum can be tuned by adjusting the period of the hole array, the hole size, and the thickness of the film. Moreover, the concentration of light in the subwavelength apertures leads to an electric-field enhancement, which can be used to manipulate the light-matter interactions and nonlinear processes. These effects show promise for use in a wide range of applications in the areas of science and technology, such as near-field microscopy, nanosize laser sources, femtosecond (fs) electron sources, nanolithography, optical modulators, biosensors, and display devices.2–8

The large transmission enhancement has been attributed to a resonant interaction of the incident light with the SPP modes at the metal surface.9,10 For subwavelength holes or slits in a metal film, two distinct paths contribute to the total transmission. The first path is nonresonant scattering of the incident field toward the scattered states through the holes or slits. This part of the transmission is spectrally flat or slowly varying. The second path corresponds to a resonant channel via a discrete SPP excitation in the structure. In the spectral domain, the interference between the resonant and the nonresonant channels gives rise to an asymmetric line shape, the Fano profiles, in the far-field transmission spectrum.11 The linewidth of the SPP mode (Γ) can be estimated from the Fano-line-shaped peaks and can be related to the SPP lifetime as $T_r = (2\Gamma)^{-1}$.12–17 Correspondingly, in the time domain, the SPP-photon coupling leads to distortion and propagation delay of the light pulses at the SPP resonance through the structures. The transmission dynamics of EOT have been studied theoretically13,16,21 and experimentally.12,16,22 A 7-fs pulse transit time was measured through the subwavelength hole array in a 300-nm-thick silver film at the SPP resonance, indicating a group velocity of $c/7$.12 A severe pulse distortion was found in the temporal profile of an fs laser pulse propagating through a metallic plasmonic crystal at the long-life SPP resonances.16 Both theoretical modeling13,20,21 and fs time-resolved measurements14 have observed a two-component structure in the temporal evolution of the transmitted pulse through the subwavelength hole or slit array: a fast transmission of a nearly unperturbed pulse followed by a long tail. Such structures are the temporal fingerprint of a Fano-type process. Moreover, due to the coupling between multiple interfaces, the transmitted pulse exhibits oscillations at the fs time scale. Both the period and the damping of these oscillations were determined by the coupling strength between the interfaces.19 Also, it has been shown that the radiative lifetime of the SPP mode is limited by the SPP scattering at the periodic array.14,23 Hence, the temporal behaviors of the light pulses through diffraction gratings can provide insight into the dephasing dynamics of surface-plasmon modes and the role of the grating structure in the transmission processes, which are crucial for the proposed application of plasmon-based photonics.

In this paper, we present experimental investigations on fs laser pulses transmitted through single- or double-layer aluminum films perforated with subwavelength hole arrays. Temporal delays of light pulses from 1.5 to 1.7 μm, at the optical fiber communication window, have been measured to study the SPP-photon interaction as well as the influence of the structure on the SPP damping dynamics. The coupling effect between SPP waves at multiple interfaces is discussed with a coupled-SPP transmission model. Among different studies of light transmission through perforated metal films, the geometric dependence of SPP modes and the interaction between SPP waves are usually investigated in the frequency domain. Only a few theoretical papers deal with the light transmission dynamics with SPP coupling via photon tunneling through the nanoapertures.19,21 Our experiment shows that the weak coupling between the SPP waves on different interfaces is responsible for the long delay time of the transmitted pulses.

II. EXPERIMENTAL PROCEDURE

The single-layered (sample A) and double-layered (samples B and C) aluminum films of 0.39 μm in thickness are embedded in silicon oxide ($n \approx 1.47$) and are placed on top of the quartz substrate.23 Each aluminum layer is perforated with
FIG. 1. (Color online) (a) Demonstration diagram of the double-layer aluminum film perforated with subwavelength hole arrays. The lateral shift \(L_x\) is 0 in sample B and 0.5 \(\mu m\) in sample C. (b) Scheme of the up-conversion setup for pulse transmission time-delay measurement.

The temporal profiles of fs laser pulses propagating through the samples were measured using the up-conversion technique. A Ti:sapphire amplifier was used to deliver optical pulses at a center wavelength of 800 nm with a pulse duration of 150 fs. The pulses were then split into two parts. One part went through an optical delay line, and the second harmonics of the other part was used to pump a home-made optical parametric generation/optical parametric amplification (OPG/OPA) system. The idle branch [infrared (IR)] of the OPG/OPA system, with the center wavelength tunable from 1.49 to 1.7 \(\mu m\), was slightly focused onto the sample from the quartz substrate side in the normal direction. The transmitted light through the sample was then collected and was forwarded to a \(\beta\)-barium borate \([\beta-BaB_2O_4\) (BBO)] crystal together with the laser pulses from the delay line. When the laser beam from the delay line and the transmitted pulses through the samples were present simultaneously in the BBO crystal, frequency mixing occurred, resulting in the generation of an up-converted signal. The up-converted signals were measured at different delay times using a photodiode and a lock-in amplifier [Fig. 1(b)].

FIG. 2. (Color online) The up-converted signal with an incident light pulse at 1.548 \(\mu m\) in the cases of a pulse propagated in air (open squares, refers to the x axis on the top), through the bare substrate without aluminum layers (solid curve in black), through sample A (line with squares in purple, SL), through sample B (open circles in red, \(D_0\)), through sample C with the incident field polarized perpendicular to the lateral shift between the upper and the lower aluminum layers (solid circles in blue, \(DY\)), through sample \(C\) with the incident field polarized parallel to the lateral shift (triangles in green, \(DX\)). The inset shows the spectrum of the incident pulse.

The far-field transmission spectra of sample A shows one major SPP resonant peak at 1.53 \(\mu m\). For the double-layered samples, two SPP modes are found, with SPP mode 1 around 1.53 \(\mu m\) and SPP mode 2, the guided mode, above 1.58 \(\mu m\). The resonant frequency of the latter depends strongly on the lateral shift between the hole arrays on the two layers. Figure 3 shows our finite-different time-domain (FDTD) simulations (CONCERTO 6.5, by Vector Fields, Inc.) of the tangent magnetic-
field distribution of the two SPP modes in the double-layered samples. The dielectric function of aluminum was described by the Drude model with parameters extracted from the IR optical response of aluminum. The magnetic field of SPP mode 1 distributed largely on the two outer Al/SiO2 interfaces [Figs. 3(a) and 3(c)], which resembled the field distribution of the symmetric SPP mode in the single-layer structure. The field of SPP mode 2 is concentrated in the gap between the two aluminum layers [Figs. 3(b) and 3(d)]. Similar to the magnetic-field distribution, the corresponding \( E_z \) distribution of SPP mode 2 is present largely within the dielectric gap between the two layers, and the surface charges are confined on the two inner surfaces.

B. Pulse transmission delays

Figure 4 shows the measured pulse delay at different wavelengths together with the far-field transmission spectra and the corresponding Fano-line-shape fittings. Because of the spectral width of the incident pulses and the laser power fluctuations, the overall systematic errors were up to \( \pm 5 \) fs in our experiments at some wavelengths. Negative time delays were occasionally recorded due to the nonuniform thickness of the substrate between different samples or within one sample. As shown in Fig. 4, the magnitude of delay mainly follows the profile of the transmission peaks. At the off-resonance wavelengths, the time delays are nearly zero. When an SPP resonance was excited, a relatively longer delay was observed. The pulse transmission time delay through the single-layer sample is about 60 fs at the SPP resonance [Fig. 4(a), SL]. In the double-layer samples, the relative time delays are about 100 and 40–60 fs at SPP mode 1 and SPP mode 2, respectively [Figs. 4(b)–4(d), D0, DY, DX]. The difference in the relative time-delay curves for DX and DY configurations, especially around SPP mode 2, comes from the polarization dependence of the SPP excitations on the grating. FDTD simulations are also performed for both the transmission spectra and the pulse transmission time-delay calculation (Fig. 5). Consistent with our experimental results, the FDTD simulation shows that the simulated pulse delays vary with the pulse center wavelength, and the magnitudes of the delays mainly follow the profiles of the optical transmission spectra. One obtains the largest pulse delay at the SPP resonance wavelengths. The relative pulse delays are calculated to be about 60–80 fs at SPP mode 1 and about 40–60 fs at SPP mode 2, consistent with the experimental values.

C. SPP lifetime

According to the Fano model,26 the resonant peaks in the transmission spectra can be described in the form

\[
T_{\text{Fano}} = |t_B|^2 \left( \frac{1 + \sum_r q_r/\varepsilon_r}{1 + \sum_r (\varepsilon_r-1)^2} \right)^2, \quad \text{with} \quad \varepsilon_r = \frac{\omega - (\omega_{\text{SPP},r} + \Delta_r)}{\Gamma_r/2}, \quad q_r = \frac{2\delta_r}{\Gamma_r},
\]

where \(|t_B|^2\) is the nonresonant transmission coefficient, \(\omega_{\text{SPP}}\) is the angular frequency of the SPP excitation, \(\Delta_r\) is the resonant shift originating from the coupling between the SPP state and the far-field continuum, \(\Gamma_r\) is the linewidth of the \(r\)th SPP state, which contains the radiative damping term due to the SPP-continuum coupling and the nonradiative damping terms.
such as the Ohmic losses in the metal, $\delta$, is the ratio between the resonant transition amplitude and the nonresonant transition.

The linewidth $\Gamma$ of SPP mode 1 is found to be about 41 meV in a single-layer structure [sample A, Fig. 4(a)] and about 25 meV in the double-layer structures [samples B and C, Figs. 4(b)–4(d)], corresponding to SPP lifetimes ($T_1$) of 16 and 26 fs, respectively. The linewidths of SPP mode 2 are about 37 and 29 meV in Figs. 4(c) and 4(d), corresponding to lifetimes of 18 and 22 fs, respectively. Compared to the SPP lifetime estimated from the transmission spectra, the measured pulse delay times are nearly four times larger. It should be noted that both SPP mode 1 and SPP mode 2 are the coupled modes of the whole structure instead of SPP excitations at the single metal-dielectric interface. As such, supermodes are built up through the energy exchange between the SPP excitations and the photons trapped in the structure, which requires sufficient interaction time.\(^{29,30}\)

In systems with multiple interfaces, the time required to build up a supermode may be much longer than the lifetime of any modes involved and, thus, leads to a longer pulse transmission time through the structure via the supermode excitation.

### D. SPP coupling

To investigate the relation between the pulse delay and the lifetime of the SPP mode, we study the transmission dynamics under the resonance condition using a simplified coupled-SPP-mode model. Of the two scattering channels, the contribution of the nonresonant channels is proportional to $(D/N)^4$,\(^{31}\) which is very small by subwavelength holes and will be ignored in our model. Figure 6 schematically shows the process of light transmission under the double-resonance condition:\(^{30}\) The scattering of the input state to the transmission continuum takes place with simultaneous excitation of SPPs on two metal-dielectric interfaces. $N_1$ and $N_2$ are the field energy densities of the SPPs on the two coupled interfaces. $I(t)$ is the intensity of the incident pulse, $c$ is the speed of light in vacuum, and $n$ is the refractive index of silicon oxide. $\Gamma_{\text{rad}}$ and $\gamma$ are the radiative and nonradiative dampings of the SPP modes, associated with the linewidth of the SPP mode $\Gamma = \Gamma_{\text{rad}} + \gamma$. $\alpha\Gamma_{\text{rad}} N_i(t)$ is the portion of field energy density that couples to the evanescent field in the hole channel per unit time, ($\alpha < 1$), which induces energy transfer from $N_i$ to $N_j$ ($j \neq i$) at time $t$ while $S_{i-j}(t)$ is the energy flux density per unit time arriving on $N_i$ at time $t$.

Thus, under the resonance condition, the time-dependent equations describing the dynamics of the SPP-photon interaction are as follows:

\[
\frac{dN_1(t)}{dt} = \frac{nI(t)}{c} - N_1(t)[(1 - \alpha)\Gamma_{\text{rad}} + \gamma] - \alpha\Gamma_{\text{rad}} N_1(t) + S_{1-2}(t),
\]

\[
\frac{dN_2(t)}{dt} = -N_2(t)[(1 - \alpha)\Gamma_{\text{rad}} + \gamma] - \alpha\Gamma_{\text{rad}} N_2(t) + S_{1-2}(t),
\]

\[
\frac{N(t)}{c} = N_2(t)(1 - \alpha)\Gamma_{\text{rad}},
\]

where $T(t)$ is the intensity of the transmitted pulse.

Because of the coupling between the SPPs on the two interfaces, temporal oscillations occur in the energy flow between $N_1$ and $N_2$ at a beating period of $T_{\text{osc}} = \pi/\Omega$, where $\Omega = \sqrt{\Delta \omega^2/4 + |V|^2} \approx |V|$, $|V|$ is the coupling strength, and $\Delta \omega$ is the detuning between the two SPPs on different interfaces.\(^{21}\) Given the energy exchange yield\(^{21}\) $\rho = |V/\Omega|^2$, the evanescent field density $\alpha\Gamma_{\text{rad}} N_1(t)$ flowing from $N_1$ arrived at $N_2$ after half a period, among which a portion of $\rho$ was transferred to $N_2$, whereas, the rest $(1 - \rho)$ was reflected back to $N_1$ after another half period, and so on. Thus, the energy transferred from $N_1$ to $N_2$ at $t = t_0 - t_0 + \Delta t$ is

\[
S_{1-2}(t_0)\Delta t = \Delta t \cdot \alpha\Gamma_{\text{rad}} \left[ N_1(t_0 - \frac{1}{2} T_{\text{ex}}) \rho + N_1(t_0 - \frac{3}{2} T_{\text{ex}}) \rho(1 - \rho)^2 + \cdots \right]
\]

\[
+ \Delta t \cdot \alpha\Gamma_{\text{rad}} N_2(t_0 - T_{\text{ex}}) \rho(1 - \rho) + N_2(t_0 - 2T_{\text{ex}}) \rho(1 - \rho)^2 + \cdots \right]
\]

\[
\approx \Delta t \cdot \alpha\Gamma_{\text{rad}} \rho \left[ \sum_{j=0}^{\infty} N_1(t_0 - \left(j + \frac{1}{2}\right) T_{\text{ex}}) \times (1 - \rho)^{2j} + \sum_{j=1}^{\infty} N_2(t_0 - jT_{\text{ex}}) \times (1 - \rho)^{2j-1} \right]
\]

where $N_i(t_0 - t_a)$ is the field energy density of the SPP mode on interface $i$ at $t = t_0 - t_a$. If the asymmetry in the hole arrays between the bottom and top surfaces of each aluminum layer is ignored, one obtains $\Delta \omega \approx 0$. It follows that $\rho = |V/\Omega|^2 \approx 1$. Thus, only the first term in the summary in Eq. (3) is nonzero, i.e.,

\[
S_{1-2}(t_0)\Delta t \approx \Delta t \cdot \alpha\Gamma_{\text{rad}} N_1(t_0 - \frac{1}{2} T_{\text{ex}}).
\]

Since the emitted photons from SPPs are radiated in both the forward and the backward directions, we assume one half the radiation is coupled to the far-field scattering states, whereas, the other half is coupled toward the hole channels contributing to the transmission continuum.
FIG. 7. (Color online) Simulations on the transmitted pulse profile through sample A (SL) and sample B (D0) at SPP mode 1. The incident pulse width is 150 fs. Parameters of (profile through sample A (SL, SPP1) and (D0, SPP1) and (profile through sample B (D0, SPP1) and sample C (square symbols) are also plotted on the contour map, whose T pulse squared symbols indicate the value of T. The T pulse parameter set of (T1 = 26 fs, T2 = 85 fs) have been used in the SL configuration and the D0 configuration, respectively. The transmitted pulse profiles are multiplied by a factor of 2 for reasons of clarity.

FIG. 8. (Color online) Contour line plot of the pulse transmission time delay T_delay as a function of SPP lifetime T1 and energy exchange period T_ex between the coupled SPPs, calculated from Eq. (3). The curves labeled from 35 to 95 fs are the calculated pulse delays. (Square symbols) The measured pulse delays at SPP mode 1 and SPP mode 2 for sample A (SL, SPP1), sample B (D0, SPP1), and sample C with incident light polarized along the lateral shift (DX, SPP1, SPP2) and perpendicular to the lateral shift (DY, SPP1, SPP2). The positions of the symbols are according to the measured pulse delay T_delay and the SPP lifetime T1 estimated from the FWHM of the resonance peaks in the transmission spectra.

E. Nonradiative SPP damping

Equations (2)–(5) discussed above are only applicable in the case of weak coupling where SPPs on a single interface are nearly unchanged. This is justified for SPP mode 1 in our samples. However, for SPP mode 2, the resonant coupling occurs between the two inner Al/SiO2 interfaces, and the field extension is about 3 μm in SiO2, which is much larger than the dielectric gap (300 nm) between the two interfaces. Thus, the small-perturbation approximation may not be appropriate. Here, we consider N_f = (N_1 + N_2)/2 as a combined SPP mode of N_1 and N_2 in the whole structure. Thus, Eqs. (2) are reformulated as

\[
\frac{dN_f(t)}{dt} = \frac{n I(t)}{2c} - N_f(t)(\Gamma_{rad}/2 + \gamma),
\]

\[
\frac{n}{c} T(t) = N_f(t)\Gamma_{rad}/2.
\]

With the coupled-SPP-mode transmission model, T_ex is estimated to be about 40 fs in the single-layered sample and about 80–100 fs in the double-layered sample for SPP mode 1, corresponding to a coupling strength of 52 and 23 meV, respectively (Fig. 8). As we mentioned before, the surface charge oscillation at SPP mode 1 is confined on the two outer Al/SiO2 interfaces, suggesting the transmission at SPP mode 1 is mediated via the coupling of the SPPs on these two outer Al/SiO2 interfaces. The doubled T_ex in the double-layered structures reflects the relative weak coupling due to the doubled film thickness. For SPP mode 2, the estimated T_ex (15 fs in the DY configuration and 40 fs in the DX configuration) is much smaller than that of SPP mode 1 in the double-layered samples. It suggests a stronger interaction between SPP waves at the two inner interfaces upon the excitation of SPP mode 2, associated with the localized electromagnetic field in the dielectric gap between the aluminum layers. Compared to the DY configuration, the localized field intensity is lower in the DX case due to the x-directional shift between the upper and lower hole arrays. Accordingly, the electromagnetic coupling between the two inner interfaces is relatively smaller in the DX configuration, which is consistent with our coupled-mode model calculation (Fig. 8). Since the incident pulse duration in the measurement was twice that of the estimated energy exchange period T_ex at the two SPP resonances, the oscillation feature due to periodic energy exchange was not resolved in the temporal evolution of the transmitted pulse in our experiments (Fig. 2).
which suggests the combined system reacts like a super-SPP mode but with a radiative damping term $\Gamma_{\text{rad}}/2$, half that on an isolated interface. This also suggests the nonradiative term $\gamma$, for example, the Ohmic loss, will have a stronger effect in the transmission process at SPP mode 2. This result is consistent with the FDTD simulation. As shown in Fig. 9, by reducing $\text{Im}(\varepsilon_{\text{Al}})$ to 1/10 the original value ($\varepsilon_{\text{Al}}$ is the dielectric function of aluminum, and $\text{Im}(\chi)$ is the imaginary part of $\chi$), the linewidth of SPP mode 2 decreases from 38 to 16 meV while the linewidth of SPP mode 1 decreases from 25 to 23 meV. Moreover, the resonance peak at SPP mode 2 is largely suppressed as the losses increase to ten times the original value. This also explains the relatively lower transmission amplitude at SPP mode 2 in our experiments.

IV. SUMMARY

To summarize, we have measured the pulse delays through single- and double-layered aluminum films perforated with hole arrays using an up-conversion technique. The frequency-dependent pulse transmission time delay mainly follows the profile of the transmission spectra. The maximum temporal delays at the SPP resonances are about 60 and 100 fs for the single-layer and the double-layer samples, respectively, which is consistent with the FDTD simulations. A coupled-SPP-mode transmission model is used to understand the temporal dynamics of pulse propagation through hole arrays via the interactions between SPPs on different interfaces. According to the measured time delays, we deduce that the coupling strength between the two outer Al/SiO$_2$ interfaces is about 52 and 23 meV in the single- and double-layered samples. The coupling strength between the two inner Al/SiO$_2$ interfaces is much stronger, which leads to a fast energy exchange between the SPPs on these two interfaces and a relatively shorter pulse delay upon the excitation of SPP mode 2. The longer temporal delays of light pulses achieved by weakly coupled metal-dielectric interfaces may help improve the performance of nanoscale plasmonic structures and may help design new photon devices.

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