Reduced damping of surface plasmon polaritons on silicon with intense femtosecond laser pulse

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Received January 22, 2019; revised March 11, 2019; accepted March 27, 2019; published online April 23, 2019

Using intense p-polarized 100-femtosecond laser pulses, the reflectivity of a silicon grating was measured in air as a function of the incident angle. The angular dependent reflectivity was observed to abruptly decrease to create a sharp dip at a specific incident angle, representing the surface plasmon resonance (SPR) induced on the grating. We have found that the sharpness of the dip increases with increasing the laser fluence. The experimental results show that the increase in the laser fluence produces a higher density of free electrons in the surface and decreases the plasmon damping to enhance the SPR. The calculated reflectivity reproduces well the increase in the dip sharpness with increasing the electron density in the silicon grating surface. © 2019 The Japan Society of Applied Physics

Surface plasmon polaritons (SPPs) are collective oscillations of conduction electrons in metal surfaces coupled with electromagnetic waves at the interface between the metal and a dielectric.1,2) The excitation of SPPs is a unique technique to concentrate photon energy on the nanoscale, and has received increased attention for a range of potential applications, including high-sensitivity gas and biosensors,3,4) surface-enhanced Raman spectroscopy,5,6) waveguides,7) extraordinary optical transmissions,8) high-resolution imaging,9) light-emitting diodes,10,11) photovoltaics,12) and photodiodes.13,14) Grating couplers have been used extensively as a method to observe SPPs excited with light,15,16) where the resonantly excited SPPs at the metallic grating surface is confirmed by an abrupt decrease to create a sharp minimum in the reflectivity measured as a function of incident angle or wavelength of p-polarized light. The reflectivity curve with a sharp dip, the so-called surface plasmon resonance (SPR) curve, is known to provide fundamental properties of SPPs, such as plasmon wavelength,15,16) propagation length,17,18) spatial mode and/or plasmonic bandgap,16,19) as well as the generation of enhanced near-fields resulting in the periodic ablation of the grating surface.20) The results show that the incident fs laser pulse ionizes the surface material to form a thin metal-like layer on the Si grating surface and induce the SPR at the interface between the metallized layer and air.

The results obtained for the Si grating20) suggest that the formation of fs-laser-induced periodic surface structures, observed for dielectrics21–27) and semiconductors,28–31) as well as for metals,32–36) can also be ascribed to the excitation of SPPs. In fact, it has been proposed so far that SPPs would be the dominant mechanism responsible for the periodic nanostructures formed with intense fs laser pulses.27,30,36) This mechanism based on SPPs is still under debate, as there have been no observations of SPPs transiently excited on nonmetallic materials with intense fs laser pulses. The measurement of SPR curves provides an effective approach to an extensive study of the excitation of SPPs to form the periodic nanostructures on nonmetallic materials and its application to nanoprocessing with intense fs laser pulses.

This paper presents a characteristic aspect of SPPs excited on a nonmetallic material pumped with intense fs laser pulses, where our attention is focused on the fluorescence dependence of the SPR curves. In the experiment, we observed that the width of the SPR curves decreased with increasing the fluence. Calculations using a simple model target well reproduced the characteristic change in the sharpness of the SPR curve observed with increasing the laser fluence. Experimental and theoretical results clearly show that the damping of SPPs decreases with increasing the laser fluence, and the SPR is induced more efficiently at the higher fluence.

The experimental setup and Si grating were the same as in our previous study.20) Briefly, linearly polarized, 100-fs laser pulses at a wavelength of λ=800 nm were focused on a Si grating surface in air with a 500 mm focal-length lens at an incident angle θ, as shown in the inset of Fig. 1. We used a laminar grating with a groove period of Λ=1300 nm, a width of 650 nm, and a depth of 66 nm, which was fabricated on polished p-type crystalline Si (100) by the photolithography and dry-etching process. The groove direction of the grating was set perpendicular to the incident plane of the laser pulse. The pulse energy on the surface was controlled with partially reflective mirrors and a pair of a half-wave plate and polarizer to adjust the laser fluence F in the range F=500–2000 mJ cm−2. The polarization direction was rotated with a half-wave plate set just before the focusing lens. A microscopic image of the laser beam reflected at the surface and its spatial intensity distribution were acquired with a charge-coupled-device camera to evaluate the reflectivity R at the central beam area. The reflectivity was measured for a single shot of the fs laser pulse as a function of θ in the range θ=10°–40°.

In the preliminary experiment, the ablation threshold F_ab for a single shot was measured in air to be F_ab=400(±10) mJ cm−2 for the flat Si substrate. In the present experiment at F>F_ab we moved the target after each shot in the direction along the grating surface so that every shot of the laser pulse hits a fresh surface area of the target. The surface morphology
polarized fs pulses shows a characteristic change with a SPM. of the target was observed with a scanning probe microscope (SPM).

We measured $R$ for the Si grating as a function of $\theta$ with a single shot of $p$- and $s$-polarized fs laser pulses at $F = 500\text{ mJ cm}^{-2}$ larger than $F_{ab}$. The results obtained are shown in Fig. 1. At $F = 500\text{ mJ cm}^{-2}$ as shown in Fig. 1(a), $R$ measured with a $p$-polarized fs pulse decreases to create a small dip at $\theta \sim 24.0^\circ$, where $R$ for both $p$ and $s$ polarizations has increased in the whole range of $\theta$, compared with $R$ measured at $F \ll F_{ab}$. 20 With increasing $F$ to 700 mJ cm$^{-2}$, as seen in Fig. 1(b), $R$ measured with $p$-polarized fs pulses shows a characteristic change with a pronounced dip at $\theta \sim 23.5^\circ$. Increasing $F$ to 2000 mJ cm$^{-2}$, $R$ for $p$-polarization exhibits an abrupt decrease to create a sharp minimum at $\theta \sim 24.0^\circ$, as shown in Fig. 1(c). In addition, we notice that $R$ in the range of $\theta$ in Fig. 1 is observed to increase with increasing $F$ for both $p$ and $s$ polarizations. The increase in $R$ with increasing $F$ suggests that the Si grating surface is more strongly metallized with increasing the laser intensity.

As shown in our previous work, 20 the dip in the $\theta$-dependent $R$ seen in Fig. 1 indicates the resonant excitation of SPPs on the metallized Si grating surface. It is noted in Fig. 1 that the dip sharpness in the SPR curve increases with an increase in $F$. In Fig. 2, the full width $\Delta \theta$ at half maximum of the SPR curves is plotted as a function of $F$. The results show the monotonic decrease in $\Delta \theta$ from $\sim 4^\circ$ to $\sim 2.5^\circ$ with an increase in $F$ from 500 to 2000 mJ cm$^{-2}$.

Using the SPM, we observed a Si grating surface irradiated with the fs laser pulse. With increasing $F$ of the $p$-polarized fs pulse, the inner side of the grating grooves was more strongly ablated at $\theta = 23^\circ$–24$^\circ$, corresponding to the increasing peak of the dip in the SPR curves shown in Fig. 1. 37 In contrast, for $s$ polarization, neither the dip of $R$ nor the deep ablation of the grooves was observed at $F = 500$–2000 mJ cm$^{-2}$. This indicates that the free-electron density $N_e$ increased in the grating surface with increasing $F$ of the $p$-polarized fs-laser pulses, 20 as well as the increase in $R$ at the larger fluence.

It is well known that an SPP induced on a metal surface with light can theoretically be described as a damping oscillator that is driven by the electric fields of light and decays at the rate $\Gamma = \Gamma_{rad} + \Gamma_{i}$ with the radiation damping $\Gamma_{rad}$ and the internal damping $\Gamma_{i}$. 40 In this description, the SPR structure can approximately be expressed by a Lorentzian function with the full width $\Gamma$ at half maximum (FWHM). 18,38–41 It is noted that the decrease in the width $\Gamma$ corresponds to a decrease in the damping or the decay rate of SPPs. Based on the fundamental nature of SPR, we may conclude that the decrease in $\Delta \theta$ observed with increasing $F$ in Fig. 2 is attributed to a decrease in $\Gamma$ of the SPPs excited on the Si grating, i.e., the better momentum matching for the excitation of SPR 20 takes place with an increase in $F$.
To confirm the nature of SPR observed with increasing \( F \), we calculated the reflectivity \( R_{\text{cal}} \) at the Si grating surface irradiated with fs laser pulses. The method and procedure of calculating \( R_{\text{cal}} \) are the same as in our previous study. Briefly, we first calculate the density \( N_e \) of the free electrons produced in the Si surface at \( F \) and the resulting change in the permittivity \( \varepsilon_a \) of the surface material. As is well known for such ablative ultrafast-laser interactions, \( N_e \) at the target surface rapidly increases to reach a maximum just after the peak of the laser pulse intensity, and the high value of \( N_e \) is maintained during the interaction, followed by ablation after the end of the interaction. In the calculation, for simplicity, we consider that the effective value of \( N_e \) is dominated by the maximal value during the interaction, contributing to \( R_{\text{cal}} \) observed in the experiment.

The permittivity changing in a fs-time scale has been reported to depend on various kinds of processes such as free-carrier response, band filling, electron-hole plasma screening, Auger recombination, and band-structure renormalization, which are mainly characterized by \( N_e \). However, it is known that at \( F > F_{\text{ab}} \), the permittivity and resulting reflectivity are dominantly modulated by the free-carrier response. Then, using the Drude model, the permittivity of the Si surface including \( N_e \) can be described as

\[
\varepsilon_a = \varepsilon_{\text{Si}} - \frac{\omega_p^2}{\omega^2 + i \omega / \tau},
\]

where \( \varepsilon_{\text{Si}} = 13.5 + i \, 0.0384 \) is the incident light frequency in vacuum, \( \tau = 1.1 \) fs is the Drude damping time of free electrons, and \( \omega_p = \sqrt{2N_e(\varepsilon m m)} \) is the plasma frequency with the permittivity \( \varepsilon_0 \) of vacuum, electron charge \( e \), electron mass \( m \), and optical effective mass of carriers \( m' = 0.18 \). For a Si grating with \( \varepsilon_a \) given by Eq. (1), \( R_{\text{cal}} \) was calculated as a function of \( \theta \) for different values of \( N_e \), using the rigorous coupled-wave analysis method. The results of \( R_{\text{cal}} \) for \( N_e = 0.6 \times 10^{22} \text{ cm}^{-3} \) \( (\varepsilon_a = - 3 + i \, 7) \), \( 0.8 \times 10^{22} \text{ cm}^{-3} \) \( (\varepsilon_a = - 8 + i \, 9) \), and \( 1.5 \times 10^{22} \text{ cm}^{-3} \) \( (\varepsilon_a = - 30 + i \, 20) \) were almost coincident with or slightly smaller than \( R \) observed at \( F = 500 \text{ mJ cm}^{-2} \), respectively. In the present experiment, the actual peak value of \( N_e \) at each of \( F \) would be larger than the calculated density, because the measured value of \( R \) was time-integrated during the ultrafast interaction of the laser pulse. The increase in \( N_e \) to \( N_e \geq 0.6 \times 10^{22} \text{ cm}^{-3} \) would lead to a large change in \( \varepsilon_a \) from \( \text{Re} \left[ \varepsilon_a \right] = 13.5 \) of crystalline Si to \( \text{Re} \left[ \varepsilon_a \right] < -3 \). The calculated results of \( N_e \) and \( \varepsilon_a \) indicate that the Si surface would certainly be metallized by the fs laser pulse at \( F = 500 - 2000 \text{ mJ cm}^{-2} \), and \( N_e \) of the Si grating surface increases with an increase in \( F \).

To see the \( F \)-dependence of \( \Delta \theta \), we estimated the width \( \Delta \theta_{\text{cal}} \) of the calculated SPR curve, where \( \Delta \theta_{\text{cal}} \) is defined by the FWHM of the SPR curve. The results are shown in Fig. 3, where the inset shows an example of the SPR curve at \( N_e = 2 \times 10^{22} \text{ cm}^{-3} \). As seen in Fig. 3, \( \Delta \theta_{\text{cal}} \) decreases monotonically from 9° to 1° with increasing \( N_e \) in the range \( N_e = 0.8 - 3 \times 10^{22} \text{ cm}^{-3} \). The result reproduces well the observed \( F \) dependence of \( \Delta \theta \) as seen in Fig. 2, whereas the decrease in \( \Delta \theta \) with increasing \( F \) is smaller than that in \( \Delta \theta_{\text{cal}} \). The smaller decrease observed in the experiment would arise from the time-integrated measurements of \( R \), as mentioned above.

The results demonstrate that the plasmon damping is rapidly reduced to more effectively excite the SPR at the Si grating surface, when the surface is more strongly metalized with an increase in \( F \) of the fs laser pulse. In summary, we have measured the incident angle dependent reflectivity of the Si grating surface, using intense fs laser pulses at different fluences. The results demonstrate that SPPs are resonantly excited on the metallized Si grating surface, and the SPP damping decreases with increasing the fluence for the better momentum matching to excite the SPR. Calculations using a simple model target reproduced well the characteristic changes in the SPR curve and its sharpness at the different laser fluences, supporting the results and conclusion obtained in the experiment.

**Acknowledgments** The authors gratefully acknowledge K. Miyazaki for helpful comments and useful discussions, and S. Oku of the NTT Advanced Technology Corporation for the fabrication of the Si grating. This work was partially supported by Grant-in-Aid for Young Scientists (A) Grant No.24686011, the Murata Science Foundation 2016, and Nanotechnology Platform Program of the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan (Grant No. S-15-MS-3004).

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