Surface Plasmon Assisted Control of Hot-Electron Relaxation Time

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Abstract: Using time-resolved reflection measurements in the Kretschmann configuration under fixed absorbed power, we observe a slowing of the hot-electron relaxation in gold films associated with coupling to surface plasmons. © 2020 The Author(s)

1. Introduction

The optical generation of hot carriers in metallic components has attracted interest for applications such as solar energy conversion [1], nanoscale heat sources [2], photothermal cancer therapy [3], and biosensors [4]. For the excitation of hot carriers in metals, the incident photon energy is typically lower than the energy of the band-to-band transition, thus the efficiency of hot carrier generation is reduced as a result of the poor absorption of light within the metals. To overcome this limitation, surface plasmons have been broadly utilized to enhance absorption which results in increase in the measurement sensitivity. Transient reflectivity measurement using pump-probe spectroscopy is a powerful method to characterize carrier dynamics. Here, we investigate the relationship between the hot carrier relaxation time and the characteristics of surface plasmon coupled on the surface of gold (Au) thin film, measured in the Kretschmann configuration. Under the fixed absorbed power in the Au film, we observe that the hot-electron relaxation time reaches its maximum at the resonance wavelength, which suggests that the modified intensity profile of the internal electric field plays a role in hot carrier relaxation time.

2. Results and Discussion

To study the effect of surface plasmons on hot-electron relaxation time, we combine the prism coupling technique using the Kretschmann configuration (i.e. prism coupling) with degenerate pump-probe optical spectroscopy, as illustrated in Fig. 1a. For the 44 nm gold film considered here, surface plasmon excitation occurs at 745 nm (1.66 eV) wavelength, where the photon energy is lower than the d-transition of Au (2.4 eV). As schematically is shown in Figure 1a, once the surface plasmon is excited in the Au film, the electric field is strongly confined at the interface between the Au film and air. Figure 1b shows the absorption measurement (solid circles) and the FDTD simulation (solid line) as a function of incident wavelength ranging from 730 nm to 775 nm, with resonance wavelength at 745 nm.

![Fig. 1. (a) Schematic of the pump-probe setup with surface plasmon coupling using the Kretschmann configuration. (b) Absorption measurement (solid circles) and simulation (solid line) after surface plasmon coupling.](image-url)
To rule out the effect of absorbed light power in the control of the hot carrier relaxation dynamics, we sweep the wavelength ($\lambda = 730 \sim 775$ nm) with the fixed absorbed power ($P_{\text{abs}} = 120$ mW) and record the transient differential reflectivity measurements. To ensure that the absorbed power remains the same over the entire incident wavelength range, we adjust the incident pump intensity according to the absorption spectra (Fig. 1b).

For the numerical modeling, we first convert the recorded transient reflectivity measurement ($\Delta R/R$) to the electron temperature under the assumption of intra-band optical pumping, which results in a non-equilibrium hot electron distribution that can modify the optical properties of the Au film. We use a free electron model by considering a simplified parabolic electron density of states for Au to convert the reflectivity data to the corresponding electron temperature. We propose a “Modified Two-Temperature Model” which relates the interaction of ultrashort pulse with two subsystems (electrons and phonons) by adding the unique internal electric field profiles into the source term definition within the metal film and employ it to fit the temperature converted data. Finally, the hot-electron relaxation time is extracted based on the results from the best fits of the temperature converted data and our modified two temperature model.

Figure 2a presents the results of the hot electrons relaxation time obtained from the fits for the case of the fixed absorbed power ($P_{\text{abs}} = 120$ mW) with the resonance wavelength at $\lambda = 745$ nm. Our results show that regardless of the amount of absorbed power, the hot carrier relaxation time is strongly dependent on the intensity of the electric field at the metal - air interface (Fig. 2b). (See the trend of hot carrier relaxation time (Fig 2a) and the electric field enhancement (Fig 2b)).

Fig. 2. (a) Experimentally measured hot-electron relaxation time under the fixed absorbed power. (b) Field enhancement is computed from the FDTD simulation for wavelengths ranging from 730 nm to 775 nm under the fixed 120 mW absorbed power normalized by the intensity of the input field.

3. References
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