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Femtosecond laser generation of microbumps and nanojets on single and bilayer Cu/Ag thin films

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Estimation of the absorbed laser fluence for experimental conditions

The conversion of absorbed fluence used in the TTM-MD modeling to incident fluence usually specified in the presentation of experimental results is performed based on the results of a series of TTM simulations accounting for the electron temperature dependence of the reflectivity and optical absorption coefficient. The laser parameters such as wavelength and pulse duration, as well as the temperature dependences of electron-phonon coupling coefficient, electron heat capacity, and electron thermal conductivity are the same as in the TTM-MD model described in Methods. The lattice heat capacity of Cu and Ag is taken to be constant and equal to 24.44 J/mol K and 35.35 J/mol K, respectively [1].

The reflectivity and the optical absorption depth are determined by the complex permittivity ε that is parameterized based on the Lorentz-Drude model [2],

$$\varepsilon = 1 - \frac{\Omega_P^2}{\omega\left(\omega - i\Gamma_0\right)} + \sum_{j=1}^{k} \frac{f_j \omega_P^2}{\left(\omega_j^2 - \omega^2\right) + i\omega\Gamma_j},$$

where ω_P is the plasma frequency, which is 1.64×10^{16} Hz for Cu and 1.37×10^{16} Hz for Ag, ω is the angular laser frequency, $\Omega_P = \sqrt{f_0}\omega_P$ is the plasma frequency associated with intraband transitions with oscillator strength f_0 and damping constant Γ_0, k is the number of oscillators with frequency ω_j, strength f_j, and lifetime $1/\Gamma_j$. The complex permittivity is described as a function of temperature by replacing Γ_0 by an effective electron collision frequency, $\nu_{ef} = \min\left(\nu_e, \nu_c\right)$. In this expression, $\nu_e = AT_e^2 + BT_l$ is the electronic collision frequency defined as a sum of the electron-electron and electron-phonon collision terms,

where T_e is the electron temperature, T_l is the lattice temperature, and the values of the coefficient A (2.66×10^6 $s^{-1}K^{-2}$ for Cu and 3.57×10^6 $s^{-1}K^{-2}$ for Ag) and B (2.41×10^{11} $s^{-1}K^{-1}$ for Cu and 1.12×10^{11} $s^{-1}K^{-1}$ for Ag) are provided and justified in Ref. [3]. $v_c = \left(4\pi n_0 / 3\right)^{1/3} \sqrt{v_f^2 + k_B T_e / m_e}$ is the maximum critical electron collision frequency [4], where n_0 (8.34×10^{28} m^{-3} for Cu and 5.74×10^{28} m^{-3} for Ag) is the electron density, and v_f (1.57×10^6 m/s for Cu and 1.39×10^6 m/s for Ag) is the Fermi velocity. The reflectivity R and the optical absorption depth l_{opt} can be calculated from the following equations:

$$R = \left|\frac{\sqrt{\varepsilon}-1}{\sqrt{\varepsilon}+1}\right|^2, \qquad l_{opt} = \frac{\lambda}{4\pi\,\mathrm{Im}\left(\sqrt{\varepsilon}\right)},$$

where λ is the laser wavelength. As described in Methods, the effective range of laser energy deposition l_{eff} is calculated as a combination of l_{opt} and a ballistic electron penetration depth l_b. In order to evaluate the sensitivity of calculated absorbed fluence to the assumed value of the ballistic penetration depth, the results obtained with a constant value of l_{b0} (15 nm for Cu and 53 nm for Ag) [5] are compared with the ones where the ballistic range is estimated as the mean free path of the electrons, $l_b\left(T_e, T_l\right) = v_f / v_e$, which is a function of both electron and lattice temperatures. Fig. S1 shows the values of reflectivity R, optical penetration depth l_{opt}, and ballistic penetration depth l_b for a broad range of electron temperatures at a fixed lattice temperature of T_l = 300 K. The dependences of the optical properties, R and l_{opt}, on electron temperature are plotted in Fig. S1a. The two assumptions of ballistic penetration depth, l_{b0} and $l_b(T_e, T_l)$, are illustrated in Fig. S1b.

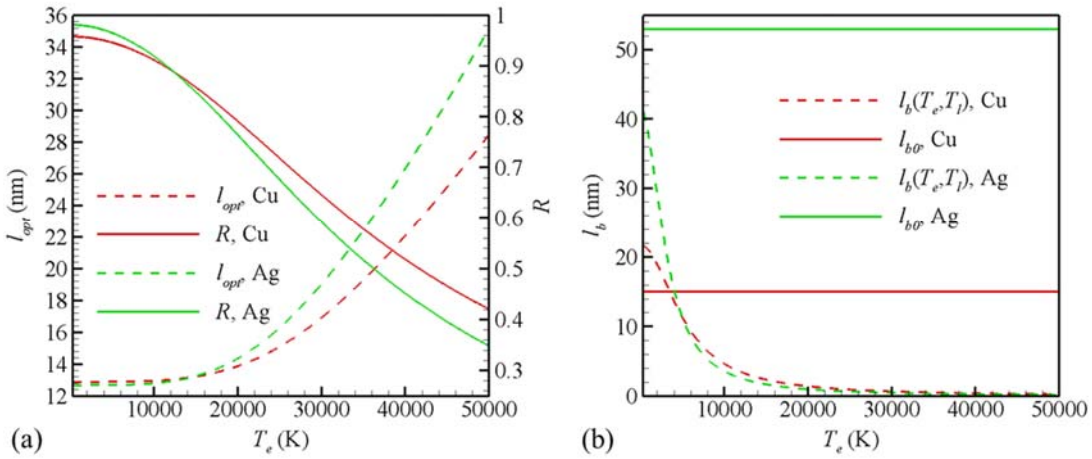

Fig. S1. (a) The electron temperature dependence of reflectivity R and optical penetration depth l_{opt} calculated for Cu and Ag at a laser wavelength of 800 nm and a fixed lattice temperature of 300 K. (b) Comparison of two assumptions for the ballistic penetration depth, a function of electron and lattice temperatures $l_b\left(T_e,T_l\right)=v_f/v_e$ with lattice temperature at 300 K, and a constant value l_{b0}, for Ag and Cu.

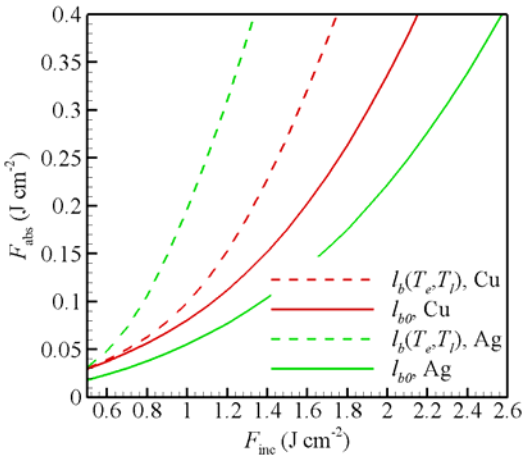

Fig. S2. The relation between the incident fluence F_{inc} and absorbed fluence F_{abs} calculated for λ = 800 nm based on the assumption of temperature dependent ballistic penetration depth $l_b(T_e, T_l)$ and a constant value of l_{b0}, respectively.

The TTM simulations are performed for bulk Cu and Ag systems irradiated by 60 fs laser pulses at a wavelength of 800 nm, to build the relationship between the absorbed fluence and incident fluence. Fig. S2 shows the absorbed fluences calculated from a range of incident ones with two assumptions of the ballistic penetrating depth indicated in Fig. S1b. Due to the ultrashort laser pulse duration and shallow optical penetration depth, the laser energy is expected to be rapidly deposited in the surface region of the target. For a bilayer system composed of 50-nm-thick layers, the optical properties of the bilayer are largely

defined by the properties of the top layer, since most of the laser energy is confined within the top layer during the laser pulse. Therefore, the absorbed fluence for a bilayer system can be expected to be close to that of a bulk system exposed to the same incident fluence, and the relationships between the incident fluence and absorbed fluence predicted for the Cu and Ag bulk systems, Fig. S2, can be used as reasonable approximations for the Cu on Ag and Ag on Cu bilayer systems investigated in the present study.

The temperature dependence $l_b(T_e, T_l)$ is based on the assumption of electron equilibration within the characteristic time of a single scattering event. Since a single scattering event cannot be expected to result in the complete equilibration of the excited electron, this assumption can only be considered as a lower bound limit of the ballistic range, which results in overestimation of the surface heating, underestimation of surface reflectivity, and corresponding overestimation of absorbed fluence. At the threshold absorbed fluence for complete ablation of the bilayer film, about 0.4 J/cm^2 as predicted in the MD-TTM simulations, the incident fluence is calculated as approximately 2.2 J/cm^2 for Cu and 2.6 J/cm^2 for Ag with constant values of l_b, which is in a reasonably good agreement with the experimental values of 2.5 J/cm^2 for Cu on Ag bilayer and 2.8 J/cm^2 for Ag on Cu bilayer, respectively, as shown in Fig. 1. Therefore, the relations between the absorbed and incident fluence predicted with an assumption of constant l_b (solid lines in Fig. S2) are used as rough estimations for comparison of computational and experimental results.

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