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Mechanical contacts mapped by picosecond ultrasonic pulses and non-equilibrium electrons

ピコ秒超音波パルスと非平衡電子による機械的接触のマッピング

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1. Introduction

The mechanical contact between solids is sensitive to the surface roughness and to the state of the surface. The contact depends on meshing their individual asperities on the nanoscale, and the real contact area is an important quantity in understanding adhesion, friction and lubrication. Elastic and plastic deformations are often studied with micro- or macroscale indenters, but these methods do not allow one to map the contact area during loading[1,2]. Mechanical contacts can be directly imaged by thermal or ultrasonic methods[3-5]. These methods can be used to map opaque materials over the contact area, and, if very high frequencies are implemented, they potentially allow nanoscale spatial resolution.

We previously presented a GHz-frequency ultrasonic pulse method for mapping contact area based on laser picosecond ultrasonics[4,5]. This method can investigate the elastic and plastic deformations during loading from the ultrasonic echo times and amplitudes. Moreover, it can image the contact area with thermal waves at the pump beam chopping frequency; the image depends on heat conduction through asperities as well as through the gaps between them. In ultrasonic images, however, it is hard to relate ultrasonic reflection or transmission coefficients quantitatively to the real microscopic contact area. And it is also difficult to relate thermal images to the real contact area. Here we propose a different way to map mechanical contacts that is uniquely sensitive to the real contact area between metallic solids with the same laser picosecond ultrasonic setup. This is based on the use of diffusing non-equilibrium electrons probed on femtosecond time scales.

2. Experiment

When femtosecond-duration optical pulses illuminate a metal, the photons are absorbed by the electrons. The excited non-equilibrium electrons undergo electron-electron and electron-phonon scattering, during which they diffuse before being thermalized within ~1 ps[6]. This results in a lattice temperature rise and the generation of ultrasonic pulses[7]. The electron diffusion process governs the maximum optical reflectivity change just after the arrival of the optical pulse. In this study we exploit this effect to probe the mechanical contact between two metal films.

We coated a sapphire ball of radius 1 mm and a sapphire plate of thickness 1 mm with 37 nm gold films, which are selected to be thinner than the non-equilibrium electron diffusion length in gold (~140 nm). The plate is placed with the film facing upwards on a pierced holder to allow optical access from underneath. The principal crystal axes of the ball and plate are arranged to be vertical.

As in a typical laser picosecond ultrasonic experiment, we used the optical pump and probe technique with a 200 fs duration pulses from a mode-locked Ti:Sapphire laser. A pump beam of wavelength 830 nm was chopped at a frequency 1 MHz for synchronous lock-in detection. A probe beam of wavelength 415 nm passes through a delay line. The pump and probe beams are both focused on the plate-film interface from the plate side with a laterally scanning ×50 objective lens. The pump and probe spot diameters were both ~1 μm, allowing the imaging of the contact area. The optical reflectivity change at this interface was monitored while indenting with a gold-coated sapphire ball at loads up to 1 N, or equivalent to pressures up to 1.8 GPa.

3. Results

We first selected the point on the sample corresponding to the center of the mechanical contact, and scanned the delay time t on changing the load F. **Fig. 1(a)** shows the probe relative reflectivity change at F=0 N (before contact) and at F=0.6 N. The maximum reflectivity change owing to the excitation of non-equilibrium electron was observed at t=0.9 ps, and we call this the reflectivity change “spike height”. **Fig. 1(b)** shows the normalized spike height data at the center of the contact area when loading up to 1.0 N and then unloading in steps 0.2 N. One can see that loading even by 0.2 N produces a ~40% drop in the spike

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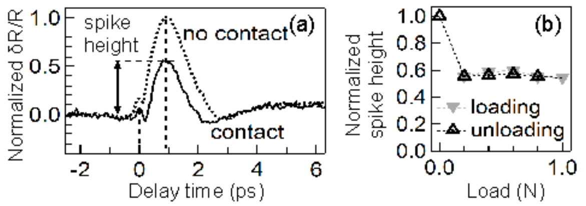

Fig. 1 (a) Normalized relative reflectivity variation at load F=0.6 N (in contact for loading: solid line) and F = 0 N (no contact: dotted line) vs. delay time. A constant background contribution from thermoreflectance has been subtracted. (b) Normalized spike height plotted against load for loading and subsequent unloading.

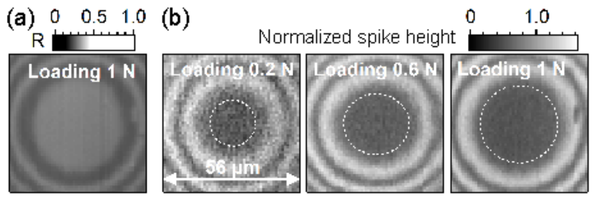

Fig. 2 (a) Mapped probe light reflectivity R images for a 56×56 μm^2 region around the contact area for 1 N loading. (b) Normalized spike-height images for 0.2 N, 0.6 N and 1 N loading, and calculated nominal contact areas (dotted circles).

height.

Then we mapped the ultrafast electron response as well as static probe reflectivity R (**Fig. 2(a)**). The concentric circles in **Fig. 2(a)** are Newton's ring caused by probe beam interference in the gap between the gold films outside the contact region. By fitting to the calculated Newton's ring pattern from Hertz theory[1] and using optical multiple-beam reflection theory, we can derive the nominal contact area and contact pressure distributions. **Fig. 2(b)** shows some images of the ultrafast electronic response as normalized spike-height plots. The dashed line circle shows the calculated nominal contact area.

To understand these results we considered the dynamics of transiently heated electrons in gold. A one-dimensional model of the electron diffusion is appropriate because the electron diffusion length is much smaller than the optical spot size. We assumed that the non-equilibrium electron density obeys the one dimensional diffusion equation which includes an effective electron reflection coefficient r_f at the gold/gold contact interface. The coefficient r_f lies between −1 (perfect reflection) and 0 (perfect transmission), and the corresponding normalized spike height values become 1.0 for r_f=−1 and 0.58 for r_f=0, in good agreement with the experimental results (**Fig. 1** and **Fig. 2(b)**). This method allows us to estimate the real contact areas using the assumed relation between the effective reflection coefficient r_f and the ratio of the real/nominal contact areas.

4. Conclusion

We use a laser picosecond setup to generate non-equilibrium electrons that diffuse across a mechanical contact between two thin gold films deposited on sapphire. We image this process in the contact and near-contact regions to micron resolution *in situ* using transient optical reflectivity changes on femtosecond time scales. By use of a model of the ultrashort-time electron dynamics, we account for an up to ~40% drop in the transient optical reflectivity change on contact. This study not only opens the way to the physics of ultrafast diffusion across contacting surfaces and inside nanoscale asperities, but also to the *in situ* mapping of the real contact area of mechanical contacts.

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