Nanoscale mechanical contacts probed with ultrashort acoustic and thermal waves

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Using an ultrafast optical technique we measure coherent phonon-pulse reflection from—a mechanical contact of nanoscale thickness between a thin metal film and a spherical dielectric indenter. Picosecond phonon wave packets at ~50 GHz returning from this interface probe the pressure distribution, the contact area, and the indentation profile to subnanometer resolution, revealing the film deformation in situ. These measurements and simultaneous thermal-wave imaging at ≥1 MHz are consistent with significant enhancement of phonon transport across the near-contact nanogap.

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I. INTRODUCTION

The properties of mechanical contacts between solids depend on how individual asperities mesh on the nanoscale, and a physical understanding is vital in engineering for the study of friction, lubrication, and electrical or heat conduction as well as in biology for the study of cell-membrane adhesion or joints. A generic approach to studying static mechanical contacts that facilitates the separation of elastic and plastic behaviors is the use of microscale or macroscale spherical (Hertz) indenters. This geometry is an apt choice for the lateral imaging of the contact by penetrating wave fields.

The contact area can be imaged by optical interferometry combined with microscopy, also opaque materials, allows one to map in addition the interfacial stiffness or the contact pressure. However, with a spatial resolution on the order of the ~0.1 mm acoustic wavelength at the typical maximum frequencies ~30 MHz used, one cannot effectively probe nanoscale deformations. Neither can one accurately study interfaces with nanoscale roughness. Likewise, probing mechanical contacts using modulated thermoreflectance up to the maximum frequencies ~3 kHz used is limited in spatial resolution by the ~30 µm thermal-diffusion length. One can image nanoindentation processes in a transmission electron microscope with subnanometer resolution, but this technique is not suited to spatially resolved mapping over the contact area.

In this paper we implement much shorter acoustic and thermal wavelengths to map the deformation in or near a Hertz mechanical contact of nanoscale thickness. We introduce an in situ coherent phonon and thermal-profiling technique at frequencies ~100 GHz and ≥1 MHz, respectively, more than 1000 times higher than previously used in contact mechanics.

II. EXPERIMENTAL SETUP

The setup is shown in Fig. 1(a). The sample is a 110-nm polycrystalline Cr film sputtered on a (0001) sapphire substrate of thickness 0.8 mm. The sample lies on a rigid holder pierced with a 1.8-mm radius hole for optical access. A sintered silicon-nitride sphere of diameter D=5.96 mm is indented into the pristine Cr film using a high-precision displacement stage. The applied load, stable at each setting to ±5% over several hours, is measured with a load cell. To investigate contacts with nanoscale separations, we use highly polished surfaces. The rms roughnesses of the film, indenter and substrate, determined by atomic force microscopy (AFM) with 40-nm step size [see Fig. 1(b)], are σf=3 nm, σi=13 nm, and σs=1 nm, respectively. Picosecond longitudinal coherent-phonon pulses are thermoelastically generated at the film-substrate (f-s) interface with linearly polarized optical pump pulses of duration 200 fs, repetition frequency 80 MHz, wavelength 810 nm (optical reflectivity 0.23), and pulse energy E=0.5 nJ from a Ti:sapphire mode-locked laser. This light is chopped at 1.1 MHz for thermal-wave excitation and lock-in detection. The reflectivity change δR(t) is measured with frequency-doubled circularly polarized probe pulses of wavelength λpb=405 nm (reflectivity R0=0.29) and energy 0.06 nJ as a function of the pump-probe time delay t. The coaxial pump
and probe beams are focused at the f-s interface by a laterally scanned ×50 objective lens to spots of radii \( w_1=1.9 \ \mu m \) and \( w_2=1.1 \ \mu m \) (intensity at \( 1/e^2 \)), respectively [effective radius \( w=(w_1^2+w_2^2)^{1/2}=2.2 \ \mu m \)]. Pertinent physical properties are the longitudinal sound velocity \( v_n \), density \( \rho_n \), Poisson’s ratio \( \nu_n \), Young’s modulus \( E_n \), thermal conductivity \( \kappa_n \), and heat capacity \( C_n \), with \( n=i, f, \) and \( s \) for the indenter, ﬁlm and substrate.\(^\text{15} \)

III. RESULTS FOR THE ACOUSTIC EXPERIMENTS

A. Shape of the acoustic echoes

Figure 1(c) shows the amplitude \( \Delta R(t)/R_{0} \) (where \( \Delta R \)<0) for the load \( F=4.2 \) N in and out of contact.\(^\text{16} \) \( \Delta R \sim E \) was checked up to pump-pulse energy \( E=0.5 \) nJ. For \( t<0 \), \( \Delta R \) is lower in contact due to thermal-wave effects, as discussed later. At \( t=0 \) we observe a peak followed by a dip and then a rise in \( \Delta R(t)/R_{0} \) at \( t=\tau=20 \) ps. This is due to the cumulative effects of a transient-temperature rise\(^\text{17} \) \( \Delta T \sim 500 \) K (\( \Delta T/dT<0 \)) and strain generation.\(^\text{12,18} \)

Initially a hydrostatic compressive stress field is generated in Cr over a depth \( z_{ac}=35 \) nm determined by the optical skin depth \( \approx 30 \) nm (Ref. \( 19 \)) and the electron diffusion depth \( \approx 15 \) nm.\(^\text{14} \) The acoustic impedances \( Z_{a}\rho_{a}v_{a} \) of Cr and \( Al_{2}O_{3} \) imply a low ﬁlm-substrate reﬂection coefﬁcient \( rf=Z_{a}-Z_{b}/(Z_{a}+Z_{b})=-0.04 \), so two unipolar compressive coherent-phonon pulses (of strain amplitude \( \approx 10^{-5} \)) are launched perpendicular to the f-s interface in both materials.\(^\text{20} \) In \( Al_{2}O_{3} \) these pulses create oscillations in \( \Delta R \) (visible after \( 40 \) ps) at the Brillouin frequency \( 2n_{a}v_{a}/\lambda_{ph}=100 \) GHz, with \( n_{a}=1.76 \) the \( Al_{2}O_{3} \) refractive index.\(^\text{20,21} \) In Cr a broadband picosecond strain pulse of frequency \( f<0 \) \( \approx 100 \) GHz (Ref. \( 23 \)) and wavelength \( \lambda \approx 70 \) nm is reﬂected from the ﬁlm-indenter (f-i) interface with a reﬂection coefﬁcient \( rf \) (determined later). This pulse is detected at the f-s interface as an echo \( \approx 30 \) ps. Because of the low value of \( rf \), only one echo is resolved. Echoes normalized using \( rf \) in and out of contact are shown in Fig. 1(c). When in contact the echo arrives earlier, is reduced in height, and is asymmetrically distorted, although the echo spectrum is not signiﬁcantly changed. The echo shape depends on the longitudinal strain \( \eta \) that in the near-surface region can be written in the approximate form of the stress wave speed \( c_{s}=\sqrt{E/\rho} \) in the Cr film, and compare it to theory in Fig. 2(b). The data lie above the predictions, consistent with theories for elastically indented thin ﬁlms for \( E_{f}/E_{i}<1.2 \). To apply Hertz theory, the combined surface roughness \( \sigma=(\sigma_{1}^{2}+\sigma_{2}^{2})^{1/2}=13 \) nm should also be considered. In our case the roughness parameter \( \alpha=\sigma D/2a^{2} \)<0.07 \( \approx 1 \) for \( F>1.1 \) N, so Hertz theory is valid.\(^\text{1} \) We thus use the Hertz pressure

\[
p(r) = p_0 [1 - (r/a)^2]^{3/2},
\]

with maximum value \( p_{0}=3F/2\pi a^{2} \), to derive \( |r_{f}|p_{0} \) from the measured \( |r_{f}|F \) at \( r=0 \). We see a linear decrease \( |r_{f}|p_{0} \) \( =1 - p_{0}/p_{1} \) in Fig. 2(c) with \( p_{1}=6.1 \) GPa. This pro-
vides a calibration to obtain \( p(r) = p_f [1 - |r_f(r)|] \), showing

good agreement with theory in Fig. 2(d). Before discussing

these results in detail we first present the results for film

indentation.

C. Profiles of the indentation

Uniquely, with picosecond ultrasonics we can directly ex-

tract the film indentation \( \Delta h = v_d \Delta t / 2 \) from the echo arrival-

time variation \( \Delta t \) during loading.27 \( \Delta h(r) \), plotted in Fig.

2(e), varies over regions slightly larger than the contact radii

\( a \), as expected. Striking correlation at each load \( F \) between

the fluctuations in \( \Delta h(r) \) and \( -p(r) \) [see dashed line in Fig.

2(e)] confirms that these are indeed induced by roughness.

Finer scale data at \( r = 0 \) is shown in Fig. 2(f) for a first load-

unload cycle in steps of 0.5 N lasting 10 min per point. The

overall shape of \( F \) vs \( \Delta h \) is evidently governed by elasticity.

The application of linear elasticity theory for a thin Cr film

on a rigid substrate indented by a rigid sphere leads to the

following equation:28

\[ F = \pi D \rho v_f^2 \Delta h^2 / 2h. \] (4)

The predicted load-indentation curve is shown by the dashed

line in Fig. 2(f). Unsurprisingly, the measured \( \Delta h \)

(\(<6 \text{ nm}\)) is smaller because of the indenter and substrate

elasticity. In comparison, the deformation of the substrate-

indenter system is much larger: \( \delta = 2a^2 D / 180 \text{ nm} \) for

\( F > 1.1 \text{ N} \).

A permanent plastic deformation \( \sim 1 \text{ nm} \) remains after

unloading [see arrows in Fig. 2(f)]. However, the Vickers

hardness \( H = 15 \text{ GPa} \) of thin-film Cr,29 the softer material,

indicates that the maximum pressure \( p_f = 1.5 \text{ GPa} \) is not suf-

ficient for bulk-film yielding.1 This justifies the use of linear

elastic theory for the bulk deformation of Cr. In order to

understand the origin of the plasticity, one should consider

the asperity deformation. For our Gaussian distribution of

asperity heights, this is governed by the so-called plasticity

index:230

\[ \psi = (E^*/H) (\sigma r_a) \sqrt{2}. \] (5)

Here \( \psi \geq 3 \), where \( r_a \geq 0.3 \text{ \( \mu \text{m} \) is the AFM-measured aver-

age Si3N4 asperity radius; \( \psi > 1 \) indicates Cr plastic com-

pression by the harder Si3N4. For noninteracting asperities,

overall force balance dictates that \( S_r H = S_0 \rho_f \), where \( S_r \) and \( S_0 \)

are the real and nominal contact areas, respectively.24 At the

maximum load \( F = 4.2 \text{ N} \), \( S_r / S_0 = 0.1 \) at \( r = 0 \). Using the

measured Si3N4 rms roughness \( \sigma_r = 13 \text{ nm} \) and rms surface

slope 0.2 rad, we estimate a corresponding contact clearance

\( u \sim 15 \text{ nm} \), with an average asperity contact radius \( R_s \sim 60 \text{ nm} \) at a density \( N_a \sim 10 \text{ \( \mu \text{m}^{-2} \).} \) This order of magni-

tude for \( u \) demonstrates that picosecond ultrasonics is ex-

quisitely sensitive to the closing of nanoscale gaps between

solids.

D. Interpretation of the results for acoustic reflection

coefficient

Precise modeling of \( |r_f| \) from contacts is complicated by the

broadband strain pulse. At very low frequencies, it is

common to assume the presence of a thin interlayer of thick-

ness \( l \), density \( \rho \), and longitudinal sound velocity \( v \), or a

contact stiffness per unit area \( K = pv^2 / l). \) For plastic de-

formation of asperities, each of Hertz stiffness \( 2E/HR_a \)

\( K = 2E/HR_a \leq 0.2 \text{ GPa} \). This implies a reduced \( |r_f| \) for

\( f \leq K / (2\pi Z_f) = 0.7 \text{ GHz} \) \( (A \leq 10 \text{ nm}) \). However, this results

in a negligible change in \( |r_f| \), inconsistent with the observed

\( \sim 25\% \) reduction at \( F = 4.2 \text{ N} \). Since wavelengths

\( \Lambda \approx 70 \text{ nm} \sim R_a \) are present at the \( f-i \) interface, this model

should be replaced by a theory of acoustic scattering from

nanoscale contacts.

To this end, consider the diffraction of a strain pulse, of

radial dependence \( \eta_i = \exp(-r^2 / w_i^2) \) governed by the pump

spot of radius \( w_i \), generated at the \( f-s \) interface. The \( z \) and \( t \)

dependences in \( \eta_i \) are implicit here. This pulse reaches the

\( f-i \) interface in the near field without lateral spreading since

the Cr thickness satisfies the relation \( h \ll w_i / \Lambda \). Provided

\( R_a > 2 / 2\pi, \) reasonable only at frequencies \( f \geq 20 \text{ GHz} \), each

contacting asperity can be approximated to have a reflection

coefficient \( |r_f| = 0.17 \), assuming negligible mode conversion.36 We define a corresponding normalized top-hat function \( T(r - d_i) \), where \( r \) and \( d_i \) are, respectively, the in-

plane position vector and that of the \( i \)th asperity. The re-

lected field \( \eta_i(r) \) at the \( f-i \) interface is then

\[ \eta_i(r) = \eta_i(r) - (1 - |r_f|^2) \sum_i T(r - d_i) \eta_i(d_i). \] (6)

Applying Babinet's principle to the diffraction of this discon-

tinuous source—analogous to a holey cheese—over the

propagation length \( h \) and positioning randomly distributed as-

perities over the probe spot of radius \( w_2 \), we get the follow-

ing simple result based on relative areas:

\[ |r_f|^2 = 1 - (1 - |r_f|^2) |R_f^2| S_a, \] (7)

where \( h \ll w_i / \Lambda \). Using \( \pi R_f^2 S_a = S_0 / S_r \), this implies

\[ |r_f(r = 0)| = 1 - (1 - |r_f|^2) p_0 / H. \] (8)

However, even using extrapolation to \( f = 0 \) only leads to an

8% drop in \( r_f \) at \( F = 4.2 \text{ N} \). So this model of acoustic dif-

fraction only partially explains our finding of a \( \sim 25\% \) reduc-

tion in \( r_f \) at this load. An additional mechanism for en-

hanced acoustic transmission or scattering should therefore

be sought.

The above model is limited to high frequencies but also in

that it ignores the asperity vibrational eigenmodes.37,38 Even

for frequencies well below their longitudinal resonance

\( v / (2\pi) \sim 300 \text{ GHz} \), enhanced transmission and echo dis-

tortion can result when the lateral mode structure and the

contribution of evanescent waves to \( r_f \) are considered. The

required numerical analysis of this process is beyond the

scope of this paper. It is clear, however, that our experimen-

tal approach is apt for probing the physics of acoustic dif-

fraction from and resonance in nanoasperities.

IV. RESULTS FOR THE THERMAL EXPERIMENTS

A. Thermal profiles of the contact

To obtain contact profiles based mainly on thermal waves

with diffusion lengths \( \mu = (2 \kappa_0 / C_0 \omega) / 2 \lesssim 3 \text{ \( \mu \text{m} \), we make

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shown for contact value, and phase lead below a flat and parallel radii. was solved for a sinusoidal spatially Gaussian heat source radius \( w \). Both remain constant over the contact area for variations in pump-light absorption on residual transmission as well as low-contrast Newton’s rings. These arise from measured air gap only those with frequencies give a non-negligible contribution here, but is in phase with the chopping at 1.1 MHz and \( t \). The derived air gap agrees with Hertz theory and Hertz theory [see Fig. 3(b)].

**B. Interpretation of the thermal profiles**

The three-dimensional (3D) thermal-diffusion equation was solved for a sinusoidal spatially Gaussian heat source (of radius \( w = 2.2 \, \mu m \)) at \( t = -10 \) ps to avoid acoustic effects. The probe temperature variation is \( A \sin(\omega t + \phi) \), where \( \sin \omega t \) is in phase with the chopping at 1.1 MHz and \( \phi \) is the phase lead recorded by the lock-in amplifier. \( |\partial R/R_0| \propto A \) \([-70 \, \text{K} \, \text{(Ref. 17)}] \), normalized with respect to its out-of-contact value, and phase lead \( \phi \) are plotted vs \( r \) in Fig. 3(c). The contact radius, determined from the initial rise in \( A \) or decrease in \( \phi \) [arrows in Fig. 3(c)], agrees with Hertz theory. At larger \( r \), \( A \) saturates, whereas \( \phi \) shows a sharp minimum. Both remain constant over the contact area for \( F \approx 1.1 \, N \).

Confirmation of these results in two spatial dimensions is shown for \( |\partial R/R_0| \) in Fig. 3(a) for a 200 \( \times \) 200 \( \mu m^2 \) region scanned in 1-\( \mu m \) steps. We resolve the circular contact area as well as low-contrast Newton’s rings. These arise from variations in pump-light absorption on residual transmission to the \( f-i \) air gap. The derived air gap agrees with Hertz theory [Fig. 3(b)].

\[
\frac{K_d}{K_{nc}} = 1 + \frac{4 \gamma}{\gamma + 1} \left( \frac{\alpha_f - \alpha_i}{\alpha_f} \right) \frac{K_n}{Pr}
\]

with \( \gamma = 7/5 \) for a diatomic gas and \( Pr = 0.72 \) the Prandtl number in air. \( \alpha_f \) and \( \alpha_i \) are the film and indenter accommodation coefficients (<1). Gap heat transport should therefore be overestimated \( (K_d/K_{nc} \approx 3) \) (Ref. 47), so kinetic theory fails to explain the anomaly in \( A \). Another possibility is near-field thermal radiation. But to account for our results would require \( K_d/4\sigma g T_3 \sim 10^7 \) enhancement compared to the blackbody value, where \( \sigma g \) is the Stefan-Boltzmann constant, an implausible factor for dissimilar materials. Although Hertz theory explains the acoustically determined contact radius \( a \), it may be that surface roughness plays a role in explaining the anomalies. The roughness parameter \( \alpha \approx 0.05 \) implies some contact even at radii \( 1.3a \). Compared to the coherent acoustic phonons, the efficient transmission of the much shorter wavelength incoherent thermal phonons through the asperities should therefore broaden the thermal-wave-probed contact area. However, the anomalies in amplitude and phase occur at a gap thickness of \( \sim 50 \, \text{nm} \), a value that is seemingly too large to be accounted for by the smaller rms surface roughnesses of the contacting Cr and \( \text{Si}_3\text{N}_4 \). A theory accounting for the detailed geometry of the asperity penetration into the gap, an intermediate region in which asperities and air coexist, is required. To elucidate the thermal anomalies we observe, measurements with different indenters in evacuated chambers should be carried out to reveal the physics of asperity deformation and thermal transport through the nanowire-likes connections that connect the contacting bodies.

**V. CONCLUSION**

In conclusion, we demonstrate that nanoscale acoustic waves excited with ultrashort optical pulses show enhanced coherent phonon transport between or enhanced scattering from solids in mechanical contact. In addition, we demonstrate that optically excited microscale thermal waves show enhanced incoherent phonon transport across the gap between solids under near-contact conditions. These findings should be investigated in the future by experiments on different contacting materials, on a range of surface roughnesses, and with a variety of gases or a vacuum in the contact region. In addition, further measurements using coated
transparent indenters with the pump and probe light incident on opposite sides of the interface will allow phonon transmission to be monitored. Accessing nanometer indentation with high-frequency phonons also opens the way to mapping deformation or plastic flow in a wide variety of materials—whether metallic, semiconductor, or dielectric—and to direct probing of the effects of wear and friction in nanomechanical systems.

#References#

38. Acoustic transmission at ~50 GHz only occurs for air gaps <0.1 mm (see Ref. 7). Intermittent contact at our acoustic displacements ~0.1 nm has negligible influence as does a ~2 nm water layer under our conditions of 35% humidity. See J. Israelachvili, *Intermolecular and Surface Forces* (Academic, London, 1985).
39. This data was taken at t = 40 ps. The contrast at this time is mainly determined by the temperature changes.
41. For air at 300 K, ρ0 = 1.01 kg m⁻³, κ0 = 0.026 W m⁻¹ K⁻¹, and C = 1.02 × 10³ J m⁻³ K⁻¹ (see Ref. 21).
42. Reduction in Rř leads to similar results, consistent with observed saturation in A and φ for F ≥ 1.1 N.
44. The predicted φ in and out of contact are ~14.4° and ~17.9°.
45. Preliminary measurements of A(δ) for an h = 215 nm and σ = 7 nm film [T. Dehoux, R. Li Voti, and O. B. Wright (unpublished)] showed similar trends.