

Moving Heterointerface with Robustly High Thermal Conductance

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(Received 29 January 2026; accepted 24 August 2026; published 5 October 2026)

Heat generation and conduction at moving interfaces are ubiquitous and fundamentally important. However, despite decades of progress on thermal transport across static interfaces, insights into moving interfaces remain rare due to the complex and ever-changing nature of most sliding contacts. Here, we take advantage of structural superlubricity and achieve simultaneous mechanical manipulation and thermal measurement of the heterointerface between gold micromesas and single-crystal graphite. We uncover a robustly high thermal conductance which is orders of magnitude higher than that in conventional moving contacts and remarkably insensitive to arbitrary interfacial rotation and thousands of cycles of sliding. In conjunction with atomistic simulations, we reveal the critical role of full atomic contact associated with the interfacial superlubricity and the stacking-insensitive out-of-plane phonon modes that dominate heat transfer. Altogether, our findings highlight diverse superlubric heterointerfaces as a pristine platform for exploring phonon dynamics in moving systems and managing heat in emerging devices including twistrionics and slidetrionics.

DOI: [10.1103/yzn5-723b](https://doi.org/10.1103/yzn5-723b)

Subject Areas: Condensed Matter Physics,
 Interdisciplinary Physics, Mechanics

I. INTRODUCTION

The rapid development of embodied artificial intelligence has triggered an explosive demand for high-performance sensors and actuators, where dynamic mechanical contacts are indispensable for motion and tactile perception [1–3]. At these movable interfaces, thermal couplings are ubiquitous and crucial for the overall system performance [4,5]. Historically, heat generation by interfacial friction revolutionized our understanding of the nature of heat [6]. Centuries later, heat transport across moving interfaces has gained ever-growing importance in diverse fields, from bearings and slip rings [7–9] to thermal

scanning probe techniques and micro-electro-mechanical systems [10–12]. However, despite decades of investigations and thousands of papers on interfacial thermal conductance [13], insights into moving interfaces remain rare. Although recent efforts have identified interlayer rotation [14–21] and sliding [22–25] as new degrees of freedom to modulate heat flow, experimental studies have largely been confined to comparing fixed samples of distinct geometries, while exploration of *in situ* movable contacts is still lacking.

With conventional moving interfaces, heat transfer is inevitably throttled by asperity contacts where the actual contact area is typically smaller than 1% of the nominal area, leading to a thermal conductance well below $0.1 \text{ MWm}^{-2} \text{ K}^{-1}$, which may suffer further degradation by wear [26–30]. By enabling full atomic contact at the interface, structural superlubricity—a state of vanishing friction and zero wear—could potentially overcome this limitation and offer unique opportunities for moving contacts [31–33]. Meanwhile, the rapid development of a new generation of movable microdevices with superlubric interfaces also demands accurate measurement and deep

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understanding of thermal transport across moving contacts. Although the experimental investigation of a superlubric homointerface [34] was recently made possible, probing various moving heterointerfaces that are fundamental to device architecture [35–38] remains both a great interest and a significant challenge.

Here, we explore thermal transport across the superlubric heterointerface between gold (Au) and graphite with controlled interfacial rotation and sliding. We choose the Au/graphite interface for three reasons. First, this interface is one of the most important superlubric interfaces beyond purely layered materials, enabling the exploration of many intriguing phenomena [39–43] and emerging devices [44,45]. Further, Au electrodes are widely used in van der Waals devices and often serve as a primary path for heat dissipation. Finally, the Au/graphite interface is ideal for laser pump-probe thermal measurement, offering high sensitivity as no additional transducer layer is needed [46–48].

II. RESULTS AND DISCUSSION

A. Sample preparation and experimental setup

Our experimental scheme is illustrated in Fig. 1(a). The very first obstacle is the preparation of superlubric Au/graphite heterointerfaces that are suitable for simultaneous mechanical manipulations and thermal measurements. To this end, we grew single-crystal Au (111) film directly on epitaxial single-crystal graphite (EG) [49] via thermal evaporation and annealing process, as detailed in Supplemental Material [50]. The Au film was subsequently patterned into square micromesas via wet etching (Fig. S1 [50]). A representative optical image of the fabricated Au mesas of various sizes is shown in Fig. 1(b). The inverse pole figures (IPF) in the electron-backscattered-diffraction (EBSD) mappings for the *x*, *y*, and *z* directions [Fig. 1(c)] show uniform colors, indicating that the Au mesa is single crystal. In Fig. 1(d), we further present a cross-sectional scanning-transmission-electron-microscopy (STEM) image which shows an atomically smooth interface between Au and graphite. The underlying graphite surface is ultrasmooth [Fig. 1(e)] with a root-mean-square surface roughness (R_q) of 83 pm as measured by atomic force microscopy (AFM) after the Au mesa was pushed aside with an AFM tip. We then characterized the interfacial friction following standard procedures in nanotribology [80]. As shown in Fig. 1(f), the measured friction coefficients for four different mesas are all on the order of 10^{-3} or below, demonstrating the superlubric state of the interface.

With the single-crystal Au/EG interface ready, we performed frequency-domain thermoreflectance (FDTR) measurement of the interfacial thermal transport [50]. Briefly, we used a sinusoidally modulated pump laser to heat the Au mesa which served readily as an optothermal transducer. The induced temperature oscillation was then

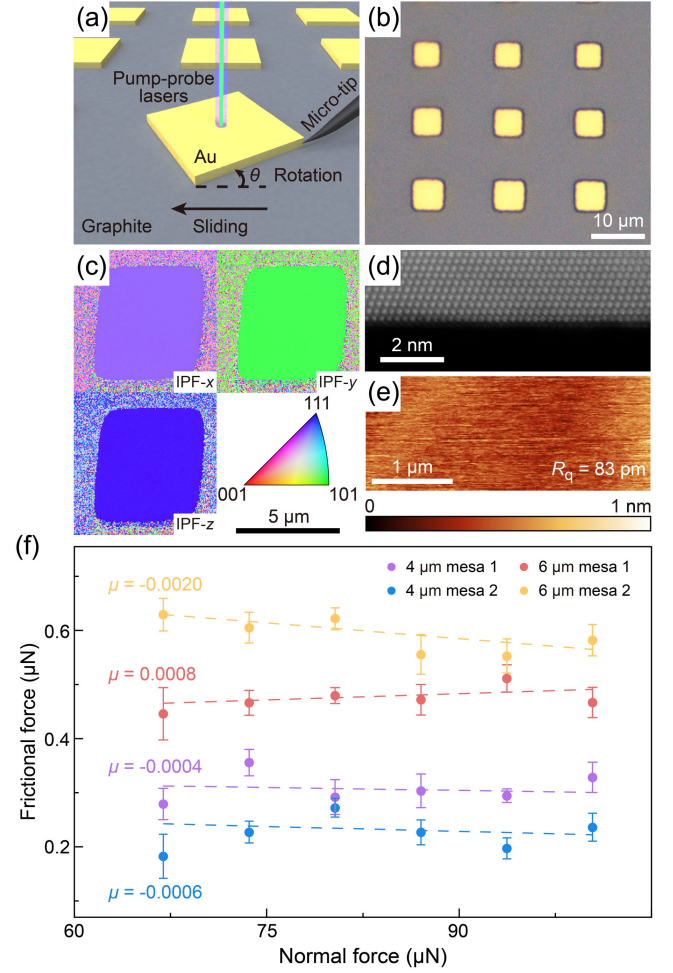


FIG. 1. Basic characterizations of Au mesas on graphite substrate. (a) Schematic of the mechanical manipulations and thermal measurement. The microtip represents either a tungsten tip or an AFM tip. (b) Optical images of Au mesa arrays. (c) EBSD maps of Au mesa on EG. (d) Cross-sectional STEM image of the Au/graphite heterointerface. (e) AFM topography of the EG surface beneath the Au mesa. The R_q was measured over a $3.5 \times 3.5 \mu\text{m}^2$ area. (f) Measurement of the friction coefficient μ at four Au/graphite interfaces.

monitored by a probe laser based on periodic variations in the reflectance of the Au surface. The phase lag between the heat input and the temperature response reflects the overall thermal impedance of the Au/EG system and is particularly sensitive to the interfacial thermal conductance (G) in our case, which therefore could be reliably fitted via the finite-element method (FEM). To begin with, we investigate the effect of *in situ* interfacial rotation by employing a home-built platform with a high-precision sample positioner and a micromanipulator with piezoresistive force sensors (Fig. S8 [50]). By gently pushing at the corner with a tungsten probe (Video S1 [50]), the Au mesa could easily rotate to an arbitrary angle (θ) due to the absence of a perfectly matched lattice structure and the ultralow interfacial friction.

B. Robust thermal conductance against rotation

In Fig. 2(a), we show the optical images of a representative Au mesa with a nominal side length of $L_{\text{mesa}} = 6 \mu\text{m}$ at four different rotation angles from 0° to 45° .

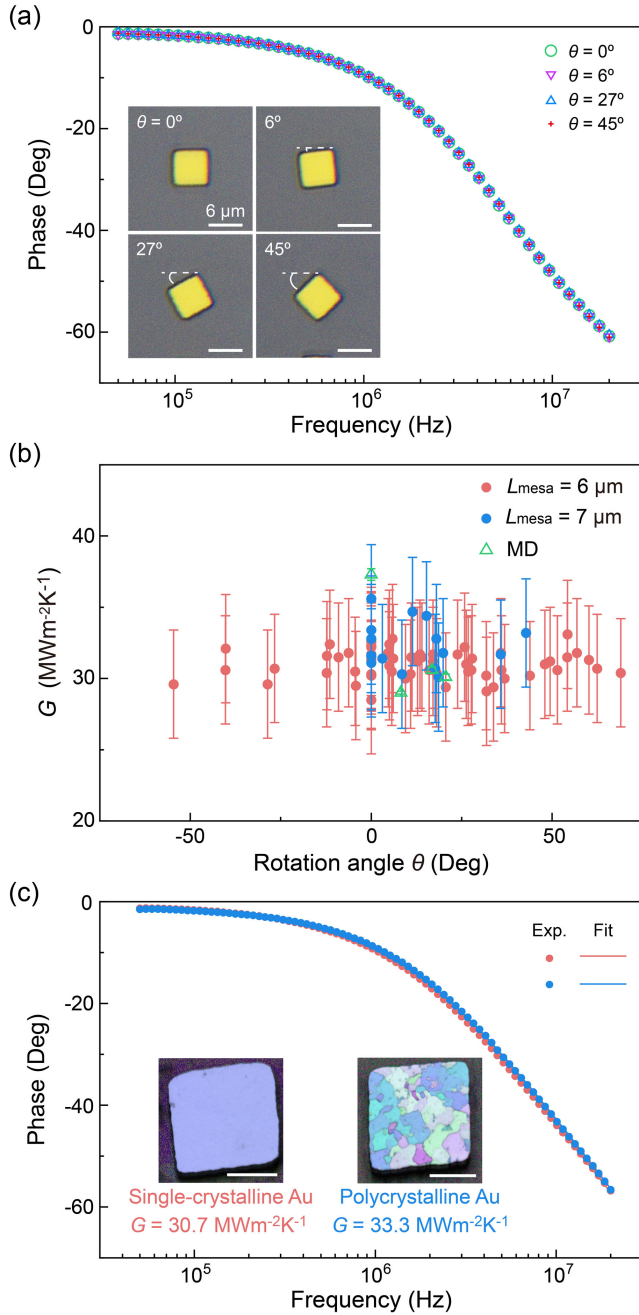


FIG. 2. Thermal measurement of *in situ* rotated Au/graphite interfaces. (a) FDTR phases for a single-crystal Au mesa rotated to different rotation angles. Insets show the optical images. (b) Variations of interfacial thermal conductance (G) with the rotation angle. Results from MD simulations are also shown. (c) FDTR phases together with the fit curves for single-crystal and polycrystal Au mesas on EG. Insets show the EBSD IPF- x maps overlaid on the image quality maps. The scale bars represent $4 \mu\text{m}$.

Interestingly, the corresponding FDTR phase signals completely overlap, suggesting no difference in G . We further manipulated and measured nine other Au mesas with the same L_{mesa} . As shown in Fig. S7(a) [50], the phase signals at $\theta = 0^\circ$ are highly consistent, except for some small differences at high frequency due to unavoidable shape variations after wet etching. Using all these mesas, we were able to cover the angles from -50° to 70° , with the measured G summarized in Fig. 2(b). Intriguingly, the thermal conductance of the Au/EG interface remains practically the same regardless of the rotation, with no observable difference beyond the experimental uncertainty. At approximately $30 \text{ MWm}^{-2} \text{K}^{-1}$, these G values are much smaller than that of the graphite homointerfaces but surpass many statically measured van der Waals heterointerfaces that can potentially be superlubric [81–85] (Table S4 [50]). Most importantly, the G values of the superlubric Au/graphite interface are over 2 orders of magnitude higher than the thermal conductance of conventional movable contacts [26–30].

The robustness of this interfacial thermal conductance was further confirmed by measurements on mesas with $L_{\text{mesa}} = 7 \mu\text{m}$ [Fig. 2(b)]. We note that the variations caused by *in situ* rotation are even smaller than the differences among mesas of different sizes and batches (Fig. S7 [50]). Apart from single-crystal interfaces, we also experimented with polycrystal Au mesas found on pre-annealed samples for comparison. In Fig. 2(c), we plot representative phase signals for a single-crystal and a polycrystal Au mesa with similar size, together with their EBSD maps showing many grains in the latter. The observed phase difference is rather small, and the fitted G values are close, with $30.7 \pm 3.8 \text{ MWm}^{-2} \text{K}^{-1}$ for single-crystal and $33.3 \pm 3.8 \text{ MWm}^{-2} \text{K}^{-1}$ for polycrystal Au/EG interfaces. These results further suggest that interfacial thermal transport is insensitive to the crystallographic orientations and grain boundaries, in contrast to friction which is highly sensitive [86]. Note that despite the similar G values, the establishment of single-crystal Au/graphite interfaces offers a unique opportunity to unequivocally probe the effect of interfacial rotation.

C. Robust thermal conductance against sliding

In addition to rotation, we further explore the potential of the Au/graphite superlubric interface in sliding motion, which provides insight into arbitrary interfacial movement and also enables long-distance experiments. To this end, we used an AFM tip and applied a small normal force ($15 \mu\text{N}$) on top of the mesa to achieve cyclic sliding while avoiding notable damage of the Au surface, as shown in the inset of Fig. 3(a). The sliding distance for one cycle was set to $2 \mu\text{m}$ with a velocity of $1 \mu\text{m/s}$ (Video S2 [50]). Because of the interfacial superlubricity, the frictional stress remained fairly small and stable [Fig. 3(a)] during 3000 sliding cycles, especially after the initial “running-in”

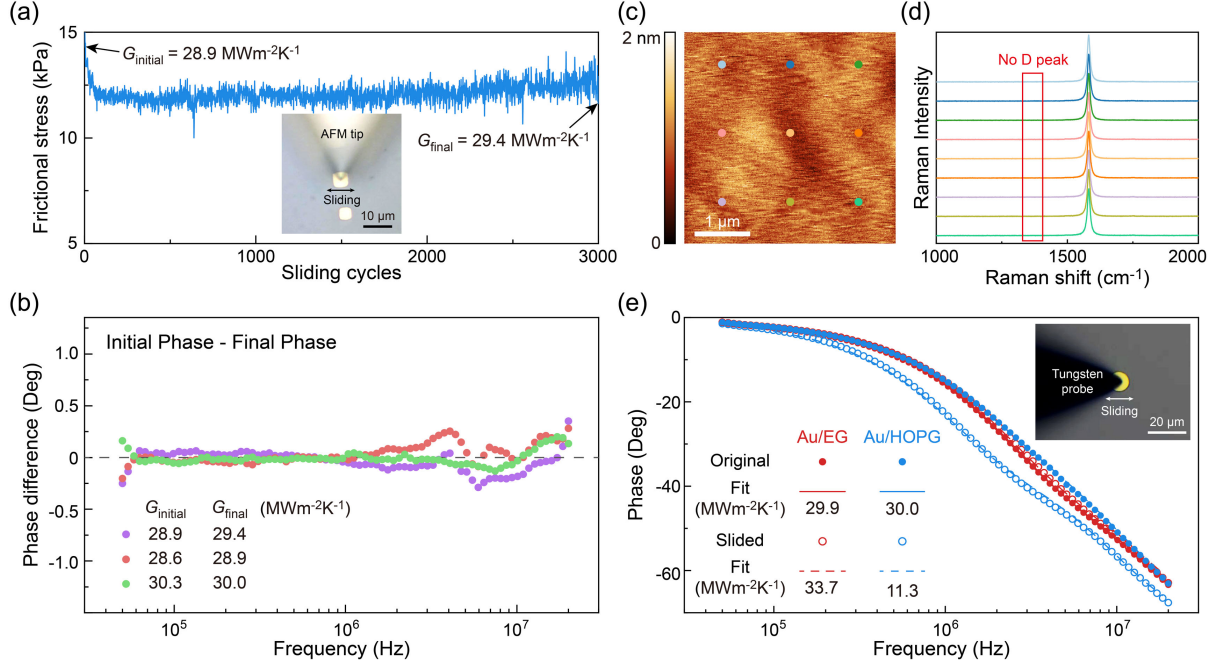


FIG. 3. Measurement of Au/graphite interface before and after sliding. (a) Frictional stress during the long-distance sliding process. The initial and final thermal conductances are also labeled. Inset: optical image of the sliding test with AFM. (b) FDTR phase differences before and after the sliding process for the Au mesa in (a) and two other mesas, together with the fitted G . (c) AFM image of the EG surface after sliding. (d) Raman spectra of EG at nine different points marked in (c). (e) FDTR phases together with the fitted results for Au mesas transferred onto HOPG and EG substrates before and after sliding. Inset: optical image of the sliding process with tungsten probe.

process [87]. We performed FDTR measurements before and after the sliding test, and the fitted G values are labeled in Fig. 3(a). Remarkably, G remained the same after such a long sliding process, with the initial and final values of 28.9 and 29.4 $\text{MWm}^{-2}\text{K}^{-1}$, respectively. A closer inspection is provided in Fig. 3(b) for three different Au mesas by subtracting the signals before and after thousands of sliding cycles, together with the fitted G values. For all three mesas, the sliding-induced phase variations lie within $\pm 0.3^\circ$, confirming that the interfacial thermal conductance is indeed robust against long-distance sliding. After the test, we pushed the mesas aside and characterized the exposed interface using both AFM and Raman spectroscopy. As demonstrated in Figs. 3(c) and 3(d), the EG surface remained atomically smooth, and no D peak was observed in the Raman spectra obtained at nine different spots within the interfacial region, showing the absence of wear during the sliding process.

For further demonstration, we transferred the as-grown Au mesas to other graphite substrates with a tungsten probe (Fig. S8 [50]). Here, freshly cleaved highly oriented pyrolytic graphite (HOPG) was employed, where various defects such as step edges exist on the surface. For comparison, another EG substrate with atomically smooth surface was also used. To facilitate the transfer process, circular mesas with a nominal radius of 5 μm were fabricated instead. As shown in Fig. 3(e), the measured

values G for transferred Au mesas on both HOPG and EG are similar to that of the as-grown interface. Since the nonideal surface condition of HOPG requires a much larger force to establish stable cyclic sliding, we used the tungsten probe instead of the AFM tip. After sliding the Au mesas for 50 cycles with a distance of 4 μm in each cycle, the measured FDTR phase signal for the Au/HOPG interface showed significant decrease, corresponding to a G reduction of over 60%. In contrast, the phase change for the Au/EG interface was small, again suggesting a rather robust G against sliding motion. To understand the substantial difference, we exposed the graphite surface and measured the topography using AFM. Significant wear was observed on HOPG, while the superlubric Au/EG interface remained wear-free, as shown in Fig. S10 [50]. Altogether, the experimental results above highlight the robust thermal conductance across a superlubric heterointerface which surpasses conventional moving contacts by several orders of magnitude and therefore holds unique promise for emerging micro- and nanodevices including twistrionics and slidetronics [88–91].

D. Atomistic simulations

To understand the physical mechanisms behind the robustly high thermal conductance of the Au/graphite heterointerface, we first emphasize the critical role of the

structural superlubric state. The superlubric interface enables full atomic contact, eliminating the geometric constrictions on heat flow caused by the multiple asperities in traditional contacts. To directly demonstrate the importance of the contact area, we prepared another HOPG substrate with a rough surface via oxygen plasma treatment (Fig. S11 [50]), and then transferred a circular Au mesa for FDTR measurement. With $R_q = 2.8$ nm, we obtained an interfacial thermal conductance which is over 40 times lower than that of the Au/EG interface. It is important to note that intimate contact usually leads to strong adhesion or even cold welding, thus rendering moving impossible. Structural superlubricity reconciles this conflict by the unique force cancellation mechanism [31], offering an unprecedented avenue for realizing thermally conductive moving contacts. In addition, the wear-free nature of superlubricity ensures a perfect atomic contact throughout the moving process, which prevents the degradation of G as observed on the Au/HOPG interface.

In the regime of perfect contact, interfacial thermal transport becomes intrinsically governed by the dynamics and coupling of various energy carriers—namely, electrons and phonons, which could provide further insights into the robust G of the Au/graphite interface. We first look into various heat conduction channels including electron-electron (G_{e-e}), electron-phonon (G_{e-ph}), and phonon-phonon (G_{ph-ph}) coupling across the interface [Fig. 4(a)]. Previous work has shown that G_{e-ph} is usually negligible [92]. Despite the metal or semimetal nature of the Au/graphite system, the electron-mediated contribution G_{e-e} [93,94] is also estimated to be only approximately $0.4 \text{ MWm}^{-2} \text{ K}^{-1}$, as limited by the low group velocity of electrons in graphite along the out-of-plane direction [50]. In contrast, a previous theoretical study based on the diffuse mismatch model incorporating the full phonon dispersions of Au and graphite predicted a G value of $59 \text{ MWm}^{-2} \text{ K}^{-1}$ [95], indicating that G_{ph-ph} alone could be sufficient to explain the experimentally observed thermal conductance despite the pronounced mismatch in the phonon density of states.

Subsequently, we performed nonequilibrium molecular dynamics (NEMD) simulations by employing machine-learned potentials [96–98]. To begin with, we examined the influence of interlayer rotation on thermal transport. For simplicity, potential herringbone reconstruction of the Au(111) surface was not considered [50,99–102]. As shown in Fig. 2(b), the simulated G values with quantum corrections [103] at several representative θ align well with the experimental results. Although a slightly higher G at the pseudocommensurate angle of $\theta = 0^\circ$ is predicted, it is too small to be experimentally resolved. Such a weak angle dependence differs markedly from the graphite homointerface, the conductance of which varies by over 30-fold with rotation [34]. Intuitively, this insensitivity may be readily explained by the mismatched lattice parameters of Au and

graphite, which forbids the formation of truly commensurate interfaces. For a deeper understanding of the unusually robust G of the Au/graphite interface, we first computed the spectral thermal conductance [104–106] in Fig. 4(b). For all three rotation angles, the peaks of the spectral G remain within the frequency range of 1 to 4 THz and practically overlap, with only a small shift to higher frequency for $\theta = 0^\circ$. Further, we decompose the spectral G into contributions from in-plane and out-of-plane vibrations (Fig. S16 [50]). Upon integration [Fig. 4(c)], we find that the out-of-plane modes dominate heat conduction across the Au/graphite interface, which is corroborated by the relative magnitude of the binding and sliding energy associated with the interface as obtained via first-principles simulations [Figs. 4(d) and 4(e)]. The binding energy barely changes with interfacial rotation which explains why G is insensitive to θ . Meanwhile, variations in the sliding energy are consistent with the weak but noticeable angle dependence of G [Fig. 2(b)].

To gain further insights, we constructed a Au/graphite superlattice toy model to focus on the effect of interfacial rotation on phonon dynamics (Fig. S17 [50]). Specifically, we calculated the phonon spectral energy density (SED) along the out-of-plane direction ($\Gamma - A$) at $\theta = 0^\circ$, 13.2° , and 21.8° in Fig. 4(f), which captures the collective vibrations of Au and carbon atoms and reveals the phonon dispersions [107]. Our results reveal that for the longitudinal acoustic (with out-of-plane atomic vibrations) phonons, both the group velocities and linewidths show only minor variations as θ varies [42]. In contrast, the transverse acoustic (TA, in-plane vibrations) phonons show localization behavior with increasing θ , with their group velocities reduced to nearly zero. In previous studies of thermal transport in layered systems, it was also recognized that the rotation angle predominantly affects the TA modes, although the variations in G can be notably larger depending on the specific materials involved [17,34].

Finally, we conducted additional NEMD simulations to investigate the interfacial thermal conductance for different sliding configurations of Au and graphite. As illustrated in Fig. 4(g), G remains almost unaffected as the Au layer slides along the graphite surface. This robustness is consistent with the above physical picture of thermal transport dominated by out-of-plane vibrations, while the in-plane modes barely contribute and are largely impeded at the interface [25]. In conjunction with the structural superlubricity, the thermal conductance thus stands rather robust against arbitrary interfacial movement.

III. CONCLUSIONS

In summary, we have presented a unique study of thermal transport across the superlubric Au/graphite heterointerface. Remarkably, an ultrarobust thermal conductance against interfacial rotation and sliding is observed, which surpasses that of conventional moving contacts by at

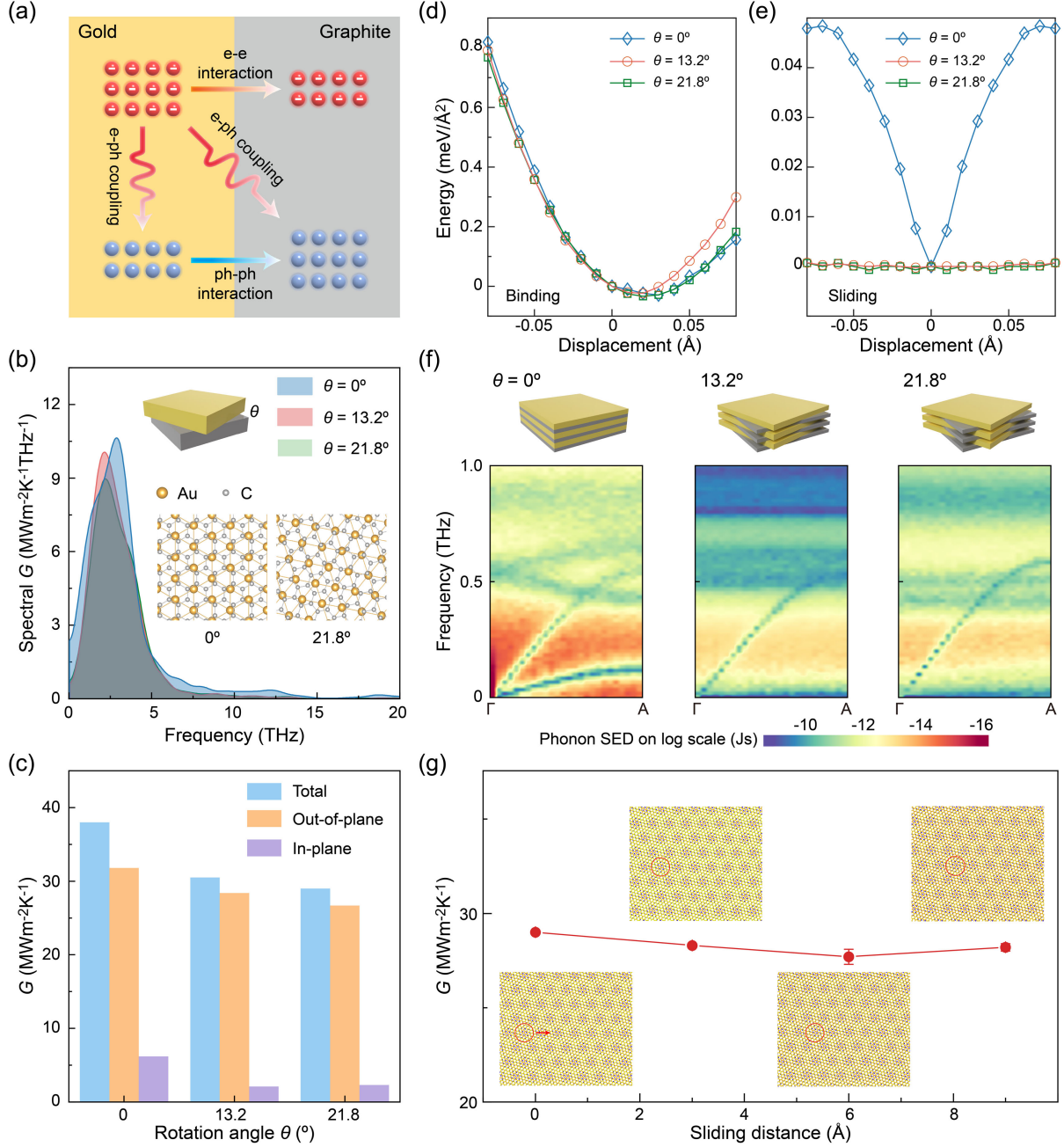


FIG. 4. Atomistic simulations of the Au/graphite interface upon rotation and sliding. (a) Schematic illustration of various potential heat conduction channels across the interface, with e and ph denoting electron and phonon, respectively. The influence of e-ph coupling in Au is negligible [50]. (b) Spectral interfacial thermal conductance (G) for three rotation angles. Inset: atomic models of rotated interfaces at $\theta = 0^\circ$ and $\theta = 21.8^\circ$. (c) Mode-resolved G for different angles. (d) Binding and (e) sliding energy profiles. (f) Effects of rotation on the phonon SED along the out-of-plane direction, together with schematics of the superlattice toy model. (g) Simulated G for Au sliding to different relative positions on the graphite surface. Insets show the corresponding Moiré patterns which reflect the atomic stacking configurations. The red circle and arrow help visualize the left-to-right movement of the Au atoms.

least 2 orders of magnitude. We attribute this robustly high thermal conductance first to the structural superlubricity that helps maintain an interface with full atomic contact throughout the moving process, and second to the particular phonon dynamics where the out-of-plane vibrations dominate regardless of the stacking configurations. In terms of

fundamental research, our findings highlight structural superlubric interfaces as a pristine playground for exploring phonon physics in dynamic systems, free from the constantly evolving interfacial conditions due to roughness and wear in traditional moving contacts. With the present study as an initial step, future advancement in experimental

techniques such as operando probing of phonon transport during movement may lead to unexpected insights into the interplay of force and heat at the atomic limit. Meanwhile, with the rich diversity of superlubric heterojunctions already available [87] and the progress in scaling up superlubricity to the macroscale [86,108,109], the possibilities for engineering applications are vast.

W. Z. appreciates Ting Liang and Weitao Wang for helpful discussions on the SED and MD calculations. This work was financially supported by the Science Fund for Creative Research Groups from the National Natural Science Foundation of China (Grant No. 52521007), the Scientific Research Innovation Capability Support Project for Young Faculty (Grant No. ZYGXQNJSKYCXNLZCXM-E1) from the Ministry of Education of China, and the National Key R&D Program of China (Grant No. 2024YFA1207900). D.P. acknowledges support from the National Natural Science Foundation of China (Grant No. 12574037) and of Guangdong Province (Grant No. 2025A1515012226). W.Z. acknowledges support from the National Natural Science Foundation of China (Grant No. 525B2087) and the China Association for Science and Technology. B.S. acknowledges support from the New Cornerstone Science Foundation through the XPLOER PRIZE. We thank the Molecular Materials and Nanofabrication Laboratory of Peking University and Shenzhen Key Laboratory of Superlubricity Technology (Grant No. ZDSYS20230626091701002) for supporting our experiments. We appreciate the High-Performance Computing Platform of Peking University for supporting our simulations.

B. S., Q. Z., and F. Y. conceived the idea and designed the research. Y. W. under the guidance of D. P. prepared the Au mesa samples. F. Y. conducted the mechanical and thermal measurements and analyses. W. Z. performed the MD and DFT calculations. F. Y., Y. W., and Y. W. performed the optical, AFM, Raman, SEM, and STEM characterizations. B. S., Q. Z., and D. P. supervised the work. F. Y. and B. S. prepared the manuscript with input from all authors.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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