Supporting Information

Nano-Imaging of an Edge-Excited Plasmon Mode in Graphene

Guanghui Cheng,^{a,b,§} Dongli Wang,^{a,b,§} Siyuan Dai,^d Xiaodong Fan,^{a,b} Fei Wu,^{a,b} Xiaoguang Li,*^c

and Changgan Zeng*a,b

aInternational Center for Quantum Design of Functional Materials, Hefei National Laboratory for

Physical Sciences at the Microscale, and Synergetic Innovation Center of Quantum Information

and Quantum Physics, University of Science and Technology of China, Hefei, Anhui 230026,

China

^bCAS Key Laboratory of Strongly-Coupled Quantum Matter Physics, and Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, China

^cInstitute for Advanced Study, Shenzhen University, Shenzhen, 518060, China

^dDepartment of Electrical & Computer Engineering, University of Texas at Austin, Texas, 78712,

United States

1. Device characterization



Figure S1. Atomic force microscopy images of the KTaO₃ surface (a) and SrTiO₃ surface (b).

The atomic force microscopic images of the KTaO₃ and SrTiO₃ surfaces after pretreatment are shown in Figure S1, both showing atomically flat terraces.

The monolayer graphene feature and carrier density can be estimated through Raman spectroscopy.¹ From the peak positions of G ($\approx 1594.6 \text{ cm}^{-1}$) and 2D ($\approx 2704.2 \text{ cm}^{-1}$) as well as from the full width at half maximum (FWHM) of G ($\approx 12.43 \text{ cm}^{-1}$) and 2D ($\approx 31.18 \text{ cm}^{-1}$), the graphene flake can be roughly estimated to be hole doping with the carrier concentration $n \approx 6 \times 10^{12} \text{ cm}^{-2}$. Due to the exceptional linear dispersion, Fermi energy of graphene is given by $E_F = \hbar v_F \sqrt{\pi n} \approx 286 \text{ meV}$, where \hbar and ν_F are reduced Planck constant and Fermi velocity of graphene, respectively. Note that such unintentional doping of graphene with similar doping level has also been reported on other substrates,²⁻⁴ which is attributed to the adsorption of H₂O or O₂ molecules in the ambient atmosphere. Such doping level keeps our frequency range of

interest away from the interband damping regime. $(|E_F| > E_P \approx 0.12 \text{ eV})$, where E_F is Fermi energy and E_P is plasmon energy)^{5,6} Thus, it is beneficial to the excitation and detection of plasmon modes. The estimated concentration is also verified by the transport results discussed later.



Figure S2. Raman spectrum of a monolayer graphene flake on the KTaO₃. The red circles are the experimental data, while the red lines are the Lorentzian fitted curves.

2. Transport measurements

To better understand the scattering strength, we also carried out an electronic measurement of graphene device on a KTaO₃ substrate. The graphene flake was etched into standard Hall bar geometry and contacted by Au/Ti electrodes. At 2 K, the Hall mobility can be more than 10,000 cm² V⁻¹ s⁻¹, as shown in Figure S3. The SNOM sample quality should be even better considering the fact that additional disorder or impurities are induced during device fabrication processes for the transport device. Such high mobility is in consistence with the long propagating length of plasmons described in the main text. Here, the back-gate voltage was applied to the device using a 0.5 mm-thick KTaO₃ substrate as the dielectric layer, inferring the tunable carrier densities for

future low-temperature near-field imaging experiment. At zero gate voltage, the carrier density is p-type with $n \approx 4.6 \times 10^{12} cm^{-2}$ close to the value estimated from the Raman results.



Figure S3. (a) Hall bar graphene device on a $KTaO_3$ substrate. (b) The mobility and carrier density extracted from Hall data at T = 2 K.

3. Substrate phonons

To clarify the source of plasmon modes presented in the main text, we have also checked the phonon polariton modes in crystals. We have obtained a broadband nano-infrared spectrum of the KTaO₃ substrate, as shown in Figure S4. Almost zero amplitude is found in the frequency range used in this work, eliminating the possible plasmon-phonon hybridizations.



Figure S4. Broadband nano-infrared spectrum of the bare $KTaO_3$ substrate. Green shade indicates the frequency range used in this work.

4. SNOM measurements of graphene on the silicon oxide

To further clarify the role of substrate in supporting the edge-excited plasmons propagating in graphene, we have carried out control experiments on the conventional SiO₂ substrate. As illuminated in Figure S5 using a silicon tip, the primary fringe can hardly be distinguished, revealing a very weak edge-excited mode, different from that on KTaO₃ and SrTiO₃. Quantitatively, the ratio between the intensity of the principle oscillating peak and the intensity of inner part flake can also reflect the damping rate of graphene plasmons.⁷ It is only 1.15 for graphene/SiO₂ in Figure S5, while the value is as high as 1.92 for graphene/KTaO₃ in Figure 1a and 1.71 for graphene/SrTiO₃ in Figure 4a, showing a stark contrast in plasmon damping rate.



Figure S5. Near-field images of graphene lying on SiO₂ as control experiments, showing a weak and discontinuous fringe along the edge. The white dashed line marks the edge of graphene. IR frequency: $\omega = 946.0$ cm⁻¹. Scale bar, 200 nm.

References

- A. Das, S. Pisana, B. Chakraborty, S. Piscanec, S. K. Saha, U. V. Waghmare, K. S. Novoselov, H. R. Krishnamurthy, A. K. Geim, A. C. Ferrari and A. K. Sood, *Nat. Nanotechnol.*, 2008, **3**, 210-215.
- S. Adhikari, D. J. Perello, C. Biswas, A. Ghosh, N. Van Luan, J. Park, F. Yao, S. V. Rotkin and Y. H. Lee, *Nanoscale*, 2016, 8, 18710-18717.
- X. R. Wang, X. L. Li, L. Zhang, Y. Yoon, P. K. Weber, H. L. Wang, J. Guo and H. J. Dai, Science, 2009, 324, 768-771.
- 4. J. Park, D. W. Kim, J. Y. Woo, J. Lee and C. S. Han, *J. Phys. D: Appl. Phys.*, 2015, **48**, 455102.
- 5. M. Jablan, H. Buljan and M. Soljačić, *Phys. Rev. B*, 2009, **80**, 245435.
- J. Chen, M. Badioli, P. Alonso-González, S. Thongrattanasiri, F. Huth, J. Osmond, M. Spasenović, A. Centeno, A. Pesquera, P. Godignon, A. Z. Elorza, N. Camara, F. J. Garcia de Abajo, R. Hillenbrand and F. H. Koppens, *Nature*, 2012, 487, 77-81.
- A. M. Dubrovkin, J. Tao, X. C. Yu, N. I. Zheludev and Q. J. Wang, Sci. Rep., 2015, 5, 9837.