Supplementary information

Quantitative Determination of In-plane Optical Anisotropy by Surface Plasmon Resonance Holographic Microscopy

Jiwei Zhang[1], Wenrui Li[1], Jiahao Li[1], Xiaoqing Chen[2], Xiangyuan Luo[1], Siqing Dai[1]*, Xuetao Gan[1]*, and Jianlin Zhao[1]*

[1]*Key Laboratory of Light Field Manipulation and Information Acquisition, Ministry of Industry and Information Technology, and Shaanxi Key Laboratory of Optical Information Technology, School of Physical Science and Technology, Northwestern Polytechnical University, Xi'an 710129, China*

[2]*School of Artificial Intelligence, Optics and Electronics (iOPEN), Northwestern Polytechnical University, Xi'an 710129, China*

E-mail: siqing.dai@nwpu.edu.cn

E-mail: xuetaogan@nwpu.edu.cn

E-mail: jlzhao@nwpu.edu.cn

S.1 Fresnel formulae

As for the four-layer SPR model used in this work, the reflection coefficient $r_{1,N}$ (N=4) of Kretschmann configuration is described by the Fresnel formulae

$$r_{i,N}(\theta)=\frac{r_{i,i+1}(\theta)+r_{i+1,N}(\theta)\exp\left[2\mathrm{j}d_{i+1}k_{zi+1}(\theta)\right]}{1+r_{i,i+1}(\theta)r_{i+1,N}(\theta)\exp\left[2\mathrm{j}d_{i+1}k_{zi+1}(\theta)\right]},\quad \left(\mathrm{j}=\sqrt{-1}, i=1,2,\ldots N-2\right) \tag{S1a}$$

$$r_{i,i+1}(\theta)=\frac{\xi_{i+1}(\theta)-\xi_i(\theta)}{\xi_{i+1}(\theta)+\xi_i(\theta)},\left(i=1,2,\ldots N-1\right), \tag{S1b}$$

$$\xi_i(\theta)=\varepsilon_i/k_{zi}(\theta),\left(i=1,2,\ldots N\right), \tag{S1c}$$

$$k_{zi}(\theta)=2\pi\frac{\sqrt{\varepsilon_i-\varepsilon_1\sin^2(\theta)}}{\lambda},\left(i=1,2,\ldots N\right), \tag{S1d}$$

where i represents the ith dielectric, $r_{i,i+1}$ the reflection coefficient between two adjacent dielectrics, and k_{zi} the wavenumber of transmission light along the z direction in the ith dielectric. The reflectivity and reflection phase shift are

$$R(\theta)=\left|r_{1,N}(\theta)\right|^2, \tag{S2}$$

$$\varphi(\theta)=\arctan\left[\frac{\operatorname{Im}r_{1,N}(\theta)}{\operatorname{Re}r_{1,N}(\theta)}\right]. \tag{S3}$$

S.2 Experimental setup of ASE-SPRHM

The azimuthal scanning excitation SPR holographic microscopy (ASE-SPRHM) was previously proposed to improve the spatial resolution of SPR images[58]. In this work, the experimental setup of ASE-SPRHM has been updated which is shown in Fig. S1.

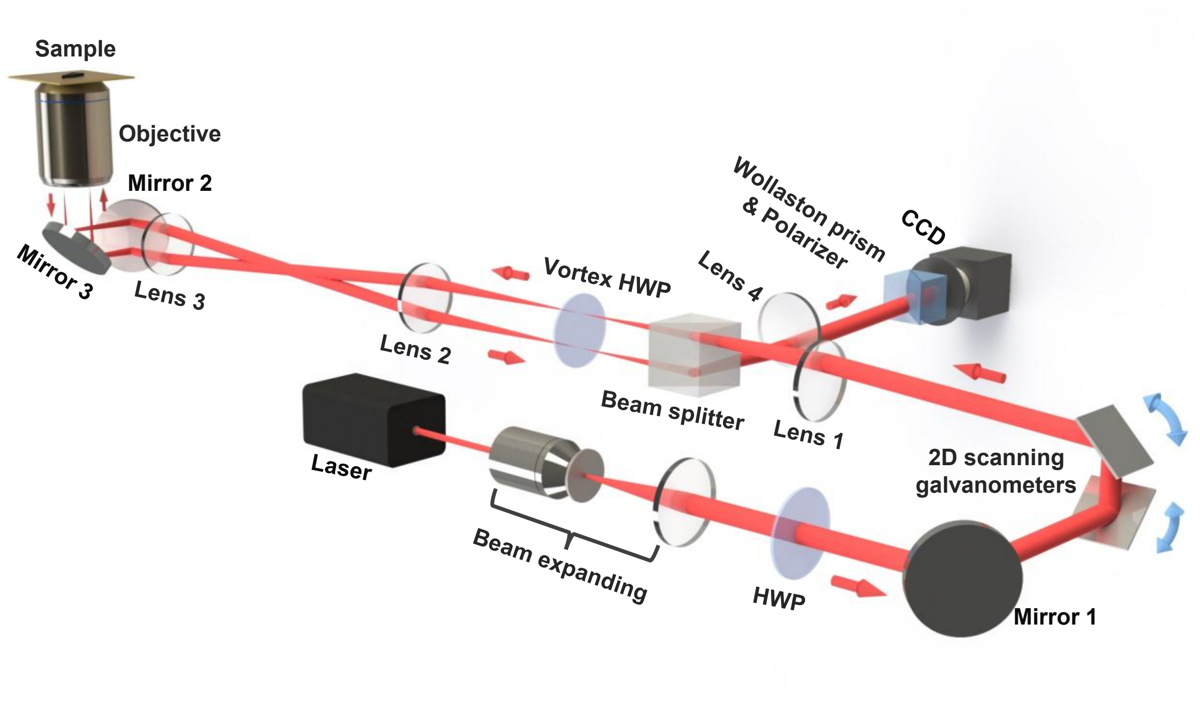

Fig. S1 Experimental setup of ASE-SPRHM

The experimental setup uses wavevector and polarization correlated light fields to excite SPR and common-path hologram recording structure to acquire SPR intensity and phase images. A pair of scanning galvanometers with orthogonal rotating axes are used to adjust the incident angle θ and in-plane angle ϕ of excitation light beam. A vortex half-wave plate (HWP) is used to ensure that the incident beam has both *s*- and *p*-polarization components with equal amplitude for any in-plane angles. The *p*-polarization component excites SPR, and SPW propagating in arbitrary direction on the gold film can be generated. Two identical mirrors 2 and 3 with orthogonal incident planes are used to direct the incident beam into the objective. This ensures that the excitation light beam on the Kretschmann configuration has the right polarization state at all of the in-plane angles.

The beam reflected by Kretschmann configuration carries the sample information and the sample is imaged on the CCD target by lens 4. For any in-plane angle, the excitation beam and reflected one pass through the vortex HWP symmetrically and the corresponding fast axes are perpendicular to each other. Thus, the polarization direction of reflected beam will be rotated to the same as that of incident beam. A Wollaston prism splits the vertical and horizontal, *i.e.*, *s*- and *p*-polarization components of reflected beam with a small angle, of which the *s*-polarization component cannot excite SPR and thus plays as the reference beam. The overlapped area of two beams forms the off-axis hologram after passing through a polarizer.

S.3 Measurement results of in-plane isotropic graphene

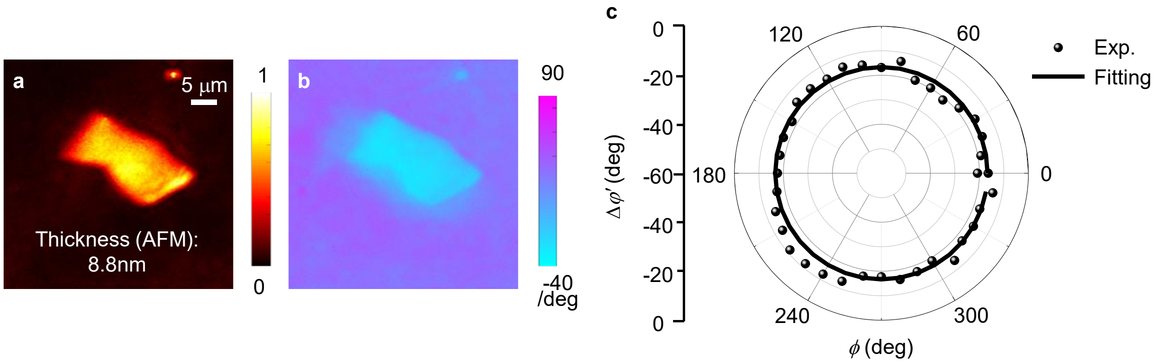

Fig. S2 Measurement results of (a) SPR intensity image, (b) SPR phase image and (c) reflection phase shift difference $\Delta\varphi'$ of the in-plane isotropic graphene sample versus in-plane angle ϕ when the 4th dielectric above sample is water and air.

S.4 Measurement results of complex RIs of 1L ReS_2 sample

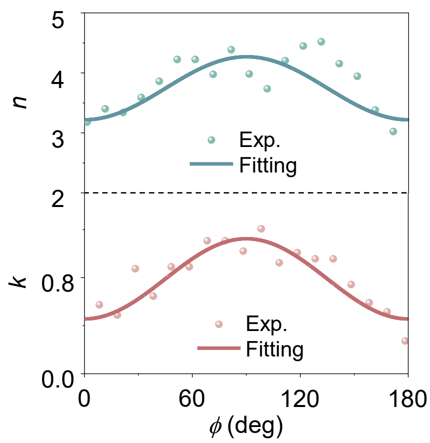

Fig. S3 Measurement results of real part n and imaginary part k of 1L ReS_2 versus in-plane angle ϕ, respectively. Since the prepared 1L sample is located aside the sample with other thicknesses, only 18 in-plane angles of SPR excitation can be realized.

S.5 Thickness characterization results of two multi-layer samples by AFM

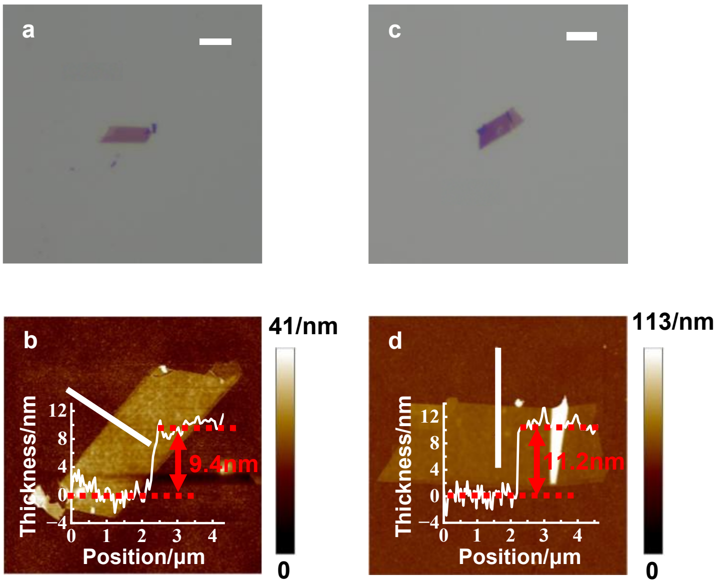

Fig. S4 (a) Optical microscopic image and (b) thickness characterization result of 9.4 nm thick ReS_2 sample by AFM. (c) and (d) The results of 11.2 nm thick ReS_2 sample. Scale bars: 5 μm.

S.6 Original experimental results of complex RIs of two multi-layer samples

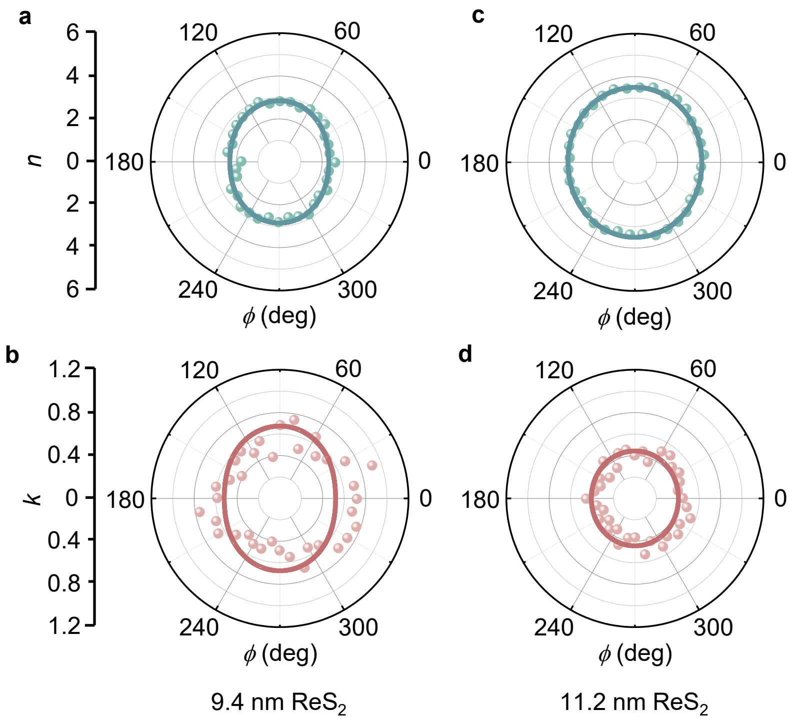

Fig. S5 Measurement results of complex RIs of 9.4 nm and 11.2 nm thick ReS_2 samples at the in-plane angles around a circle. (a, c) Real part n and (b, d) imaginary part k of complex RIs versus in-plane angle ϕ of (a, b) 9.4 nm and (c, d) 11.2 nm thick ReS_2 sample, respectively.