Supporting Information

Coherent Control of Nanoscale Ballistic Currents in Transition Metal Dichalcogenide ReS_2

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1. Sample Fabrication and Characterization

The thin-film ReS_2 samples are prepared by mechanical exfoliation. First, we repeatedly exfoliate a ReS_2 crystal by a piece of adhesive tape to obtain thin flakes. Next, the tape is pressed onto a SiO_2/Si substrate and subsequently peel off, leaving many flakes on the substrate. Then, the substrate with ReS_2 flakes is placed in acetone solution for about 20 minutes to remove the glue residue on the substrate.

We select two relatively large flakes in our study, as shown in Figure S1. They are labeled as Sample 1 and Sample 2, respectively. Atomic force microscopy (AFM) measurements (shown in Figures S2 and S3) indicate that their thicknesses are about 15 and 18 nm, respectively. The data in the main text are taken from Sample 1. The corresponding data from Sample 2 is given in this document.

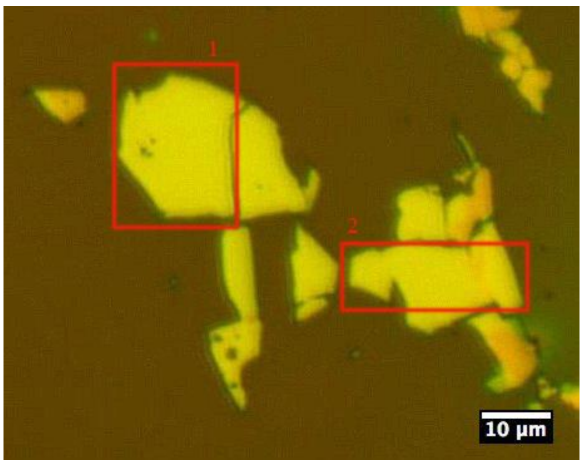

Figure S1: Optical image of the two thin film samples studied.

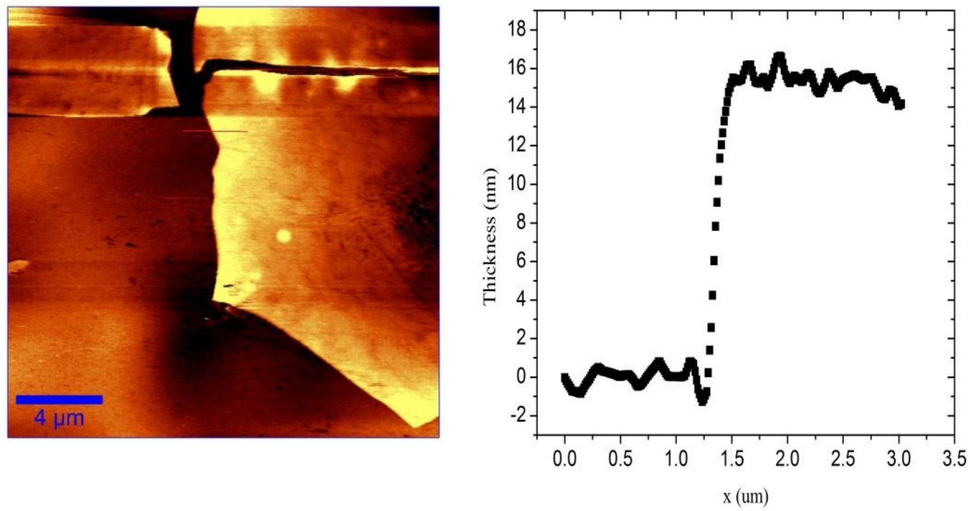

Figure S2: Left: AFM image of Sample 1; Right: A line scan from substrate to sample (indicated as the red line in the left panel), indicating a sample thickness of about 15 nm.

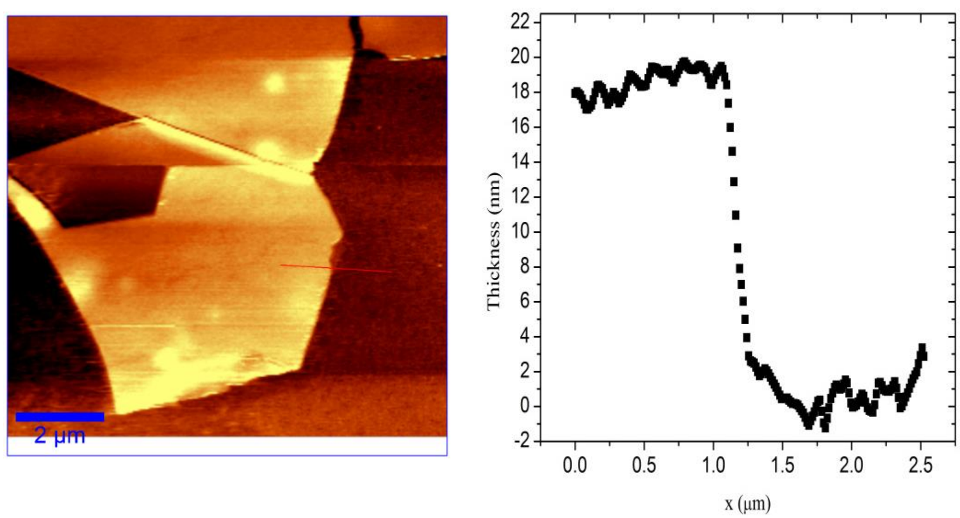

Figure S3: Left: AFM image of Sample 2; Right: A line scan from substrate to sample (indicated as the red line in the left panel), indicating a sample thickness of about 18 nm.

2. Deducing Carrier Density from Pump Fluence

In a laser pulse with an energy fluence of F, the corresponding number density of photons is $F / h\nu$, where h is Planck's constant and ν is the optical frequency. To calculate the excited carrier density, we recall that the transmitted laser intensity through a film of thickness L is $I_t = I_0\, e^{-\alpha L}$, where I_0 is the incident laser intensity. Assuming each absorbed photon excites one electron-hole pair, the density of the excited carriers as a function of the laser penetration depth into the sample, z, is

$$N(z) = \frac{\alpha F}{h\nu} e^{-\alpha z}.$$

Here, $z = 0$ is defined as the front surface of the sample and $z = L$ is at the back surface. The density quoted in the main text are the density at the front surface, $\alpha F / h\nu$.

3. Carrier Lifetime

Since the ballistic transport process only persist for a few picoseconds, discussions in the main text focus on measurements over short time ranges. The carrier lifetime of these samples is determined by measuring differential reflection over a longer time range of several hundred ps. Figure S4 shows results of such a long-time scan. The decay can be fit by a bi-exponential function (red line), with two time constants of 90 and 430 ps, respectively. Both time constants are much longer than the time scale of the measurements on ballistic transport. We attribute the long time constant to the carrier lifetime.

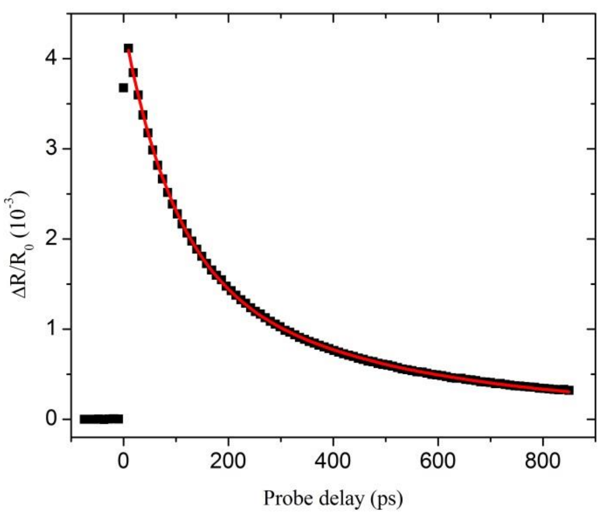

Figure S4: Differential reflection signal over a long time scale under 750-nm pump with a peak energy fluence of 18 μJ/cm^2.

4. Results of Measurements on Sample 2

The main text presents the measurements on the 15-nm sample. Using the same procedure and under same experimental conditions, we studied the 18-nm sample. The results are presented in Figures S5, S6, S7, and S8, in a similar fashion as Figures 2, 3, 4, and 5 of the main text, respectively. From the profiles shown in Figures S6 and S7, we deduced a transport length of about 0.9 nm.

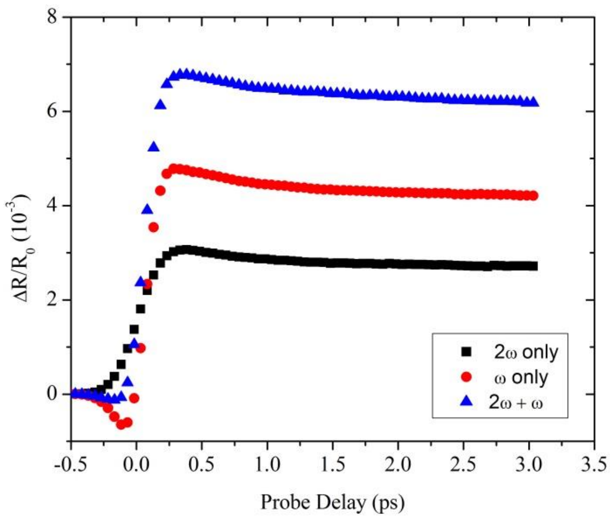

Figure S5: Differential reflection of the 810-nm probe pulse induced by the 2ω pump only (black squares), the ω pump only (red circles), and both pumps (blue triangles).

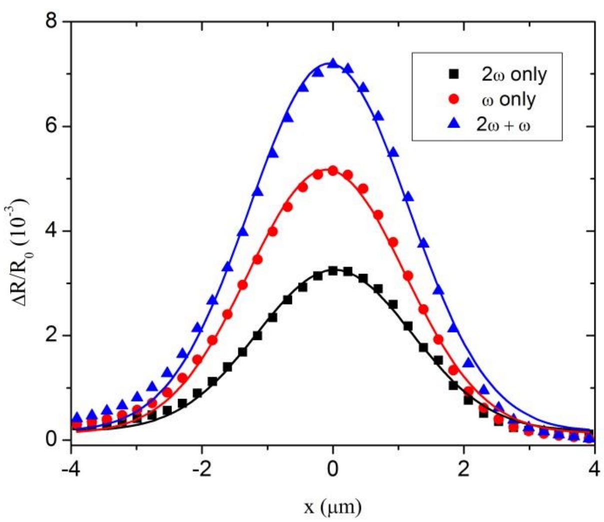

Figure S6: Spatial profiles of differential reflection injected by the 2ω (black squares), ω (red circles); and both (blue triangles) pump pulses, as the probe spot is scanned along x axis with fixed probe delay of 0.8 ps. Here x=0 is defined where the centers of the pump and probe spots overlap.

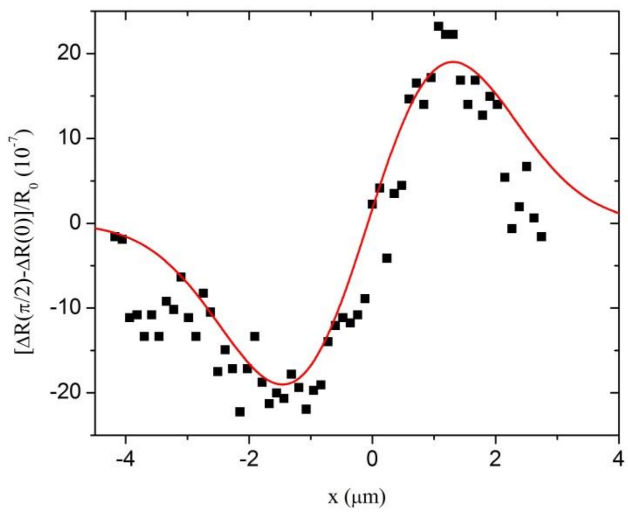

Figure S7: The difference between the differential reflection signals with $\Delta\phi = \pi/2$ and $\Delta\phi = 0$ as a function of the probe position. The probe delay is 0.8 ps.

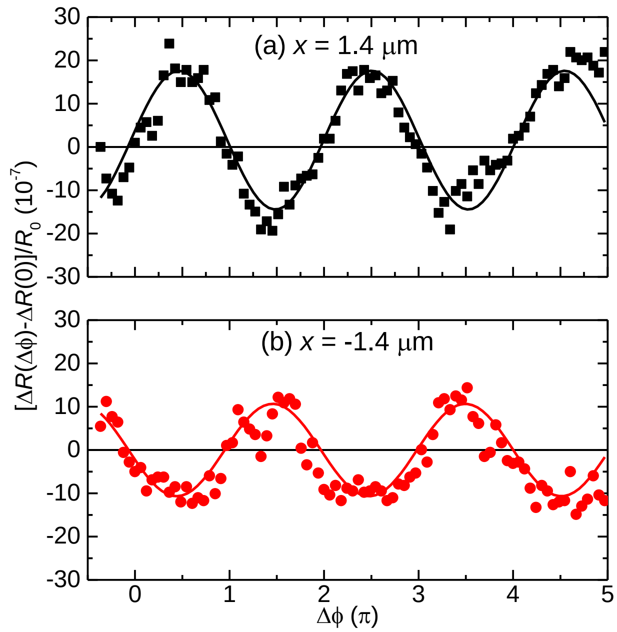

Figure S8: The difference between the differential reflection signals with $\Delta\phi$ and $\Delta\phi = 0$ measured at probe positions of 1.4 μm (a) and -1.4 μm (b), respectively. The probe delay is 0.8 ps.