Supporting Information

Cross-Examination of Ultrafast Structural, Interfacial, and Carrier Dynamics of Supported Monolayer MoS_2

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1. Photoluminescence of MoS_2 Monolayer and Bulk

The change in the band structure of MoS_2 from an indirect-gap semiconductor for the bulk to a direct-gap one for a monolayer results in a prominent enhancement in photoluminescence (PL). This is evident in Figure S1 following an excitation at 532 nm. The PL spectra were acquired from different regions of the specimen used for the time-resolved studies and no significant change in the intensity was observed. In addition, optical micrographs of the sample used showed no contrast difference that may originate from stacked multilayer MoS_2. Moreover, no PL peak near 775 nm was observed, which would correspond to indirect-gap PL of bilayer MoS_2.[1] Hence, we confirms by PL, optical microscopy, and reflection electron diffraction that the specimen used in the experiments was a uniform monolayer.

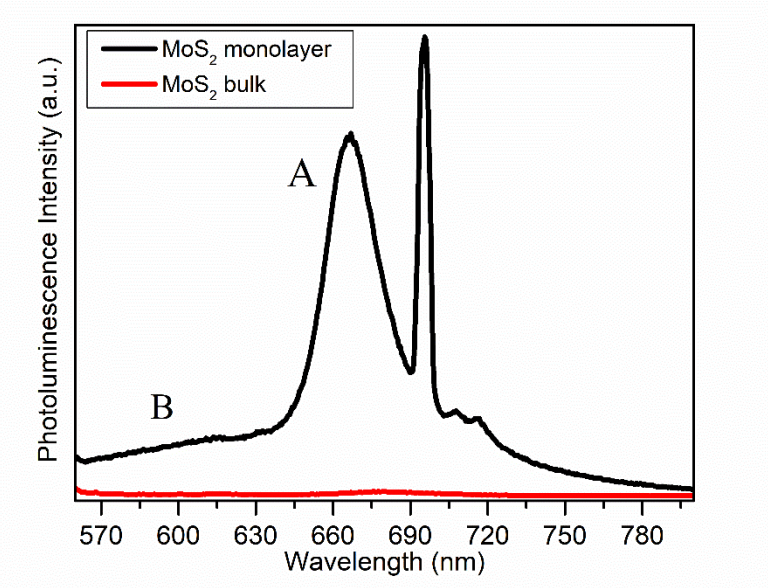

Figure S1. Photoluminescence spectra of MoS_2 monolayer (black) and bulk (red). The two peaks labeled as A and B are characteristics of the exciton emissions of monolayer MoS_2. The peaks near 700 nm originate from the *c*-plane sapphire substrate.

2. Exclusion of Surface Electric Fields as a Possible Cause for Diffraction Dynamics

For reflection ultrafast electron diffraction (UED), it is critical to show that no significant transient electric fields exist to affect the paths of the probe electrons, especially the outgoing part. A direct test is to examine the shadow edge region of diffraction images because its intensity is an integrated result of electron scattering and propagating close to the specimen

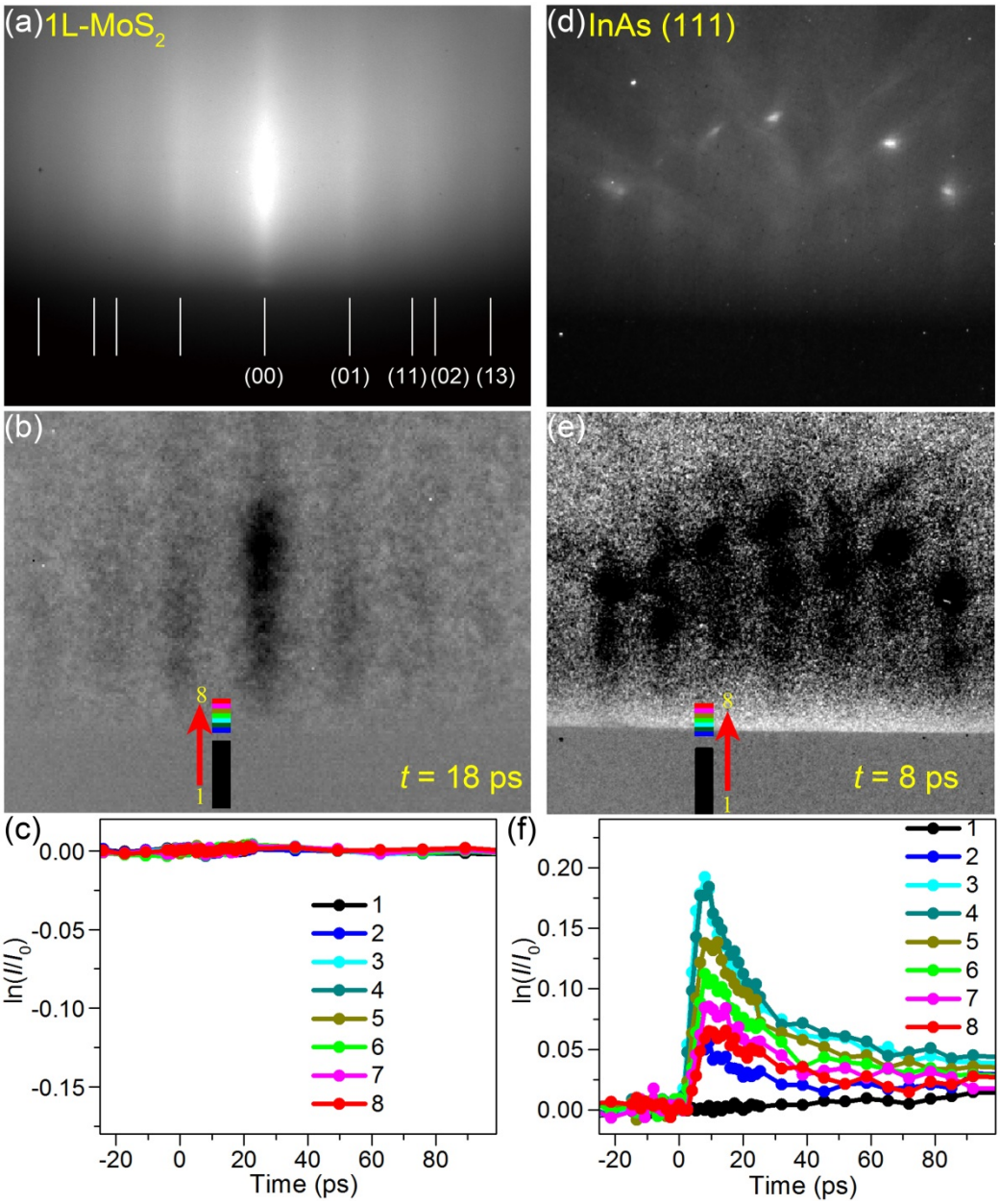

Figure S2. Diffractions and time-dependent intensity changes near the shadow edge for (a–c) 1L-MoS_2 and (d–f) InAs(111). The color codes from black to red are assigned to the horizontal regions labeled as 1 to 8 between diffraction streaks for the integration of scattering intensity.

surface and is thus sensitive to the presence of any surface electric field.[2] Shown in Figure S2 are the diffractions and time-dependent changes near the shadow edge for (i) monolayer MoS_2 (1L-MoS_2) on sapphire and (ii) bulk InAs(111), with the electron footprint well aligned with and fully enclosed by the laser excitation region. It is clear that the level of photoexcitation of 1L-

MoS_2 used in the present study does not produce noticeable surface fields to cause any time-dependent diffraction changes near the shadow edge. In contrast, the significant photoexcitation of InAs(111) leads to clear intensity increase near the shadow edge as a result of the presence of surface transient electric fields pushing the outgoing electrons upward to produce more recorded intensity. In all of our UED experiments and analyses, examination of the shadow edge intensity has always been a crucial step in order to exclude any possible electric effect as a possible cause for the observed diffraction dynamics.

3. Simulation of Heat Diffusion via the Interface

Heat diffusion across the MoS_2–sapphire interface can be described by the following equation:[3]

$$C\rho d\,\frac{\partial T_f(t)}{\partial t} = -\sigma\left[T_f(t) - T_s(t)\right] \tag{S1}$$

$$K\,\frac{\partial T_s(z,t)}{\partial z}\Big|_{z=0} = \sigma\left[T_f(t) - T_s(t)\right] \tag{S2}$$

where C and ρ are the volumetric heat capacity and the density of MoS_2, respectively, d the thickness of the overlaying film, σ the thermal boundary conductance, T_f and T_s the temperatures of the film and the substrate, respectively, K the thermal conductivity of sapphire, and z is along the surface normal direction. In the sapphire substrate, one-dimensional heat diffusion is considered,

$$\frac{\partial^2 T_s(z,t)}{\partial z^2} - \frac{c}{K}\frac{\partial T_s(z,t)}{\partial t} = 0 \tag{S3}$$

with the boundary and initial conditions as follows:

$$T_s(0,t) = T_s(t), \text{and} \tag{S4}$$

$$T_s(z,0) = 297\ \text{K}. \tag{S5}$$

Numerical simulations were carried out based on these equations and conditions.

4. Diffraction Intensity Dynamics of Bare Sapphire

The same experimental condition with a laser fluence of 2 mJ/cm^2 was used to measure the diffraction intensity changes from bare sapphire(0001) (Figure S3). The absence of time-dependent changes is expected because of no absorption of 515-nm photons by sapphire and

therefore shows that all photoexcitation was made in 1L-MoS_2.

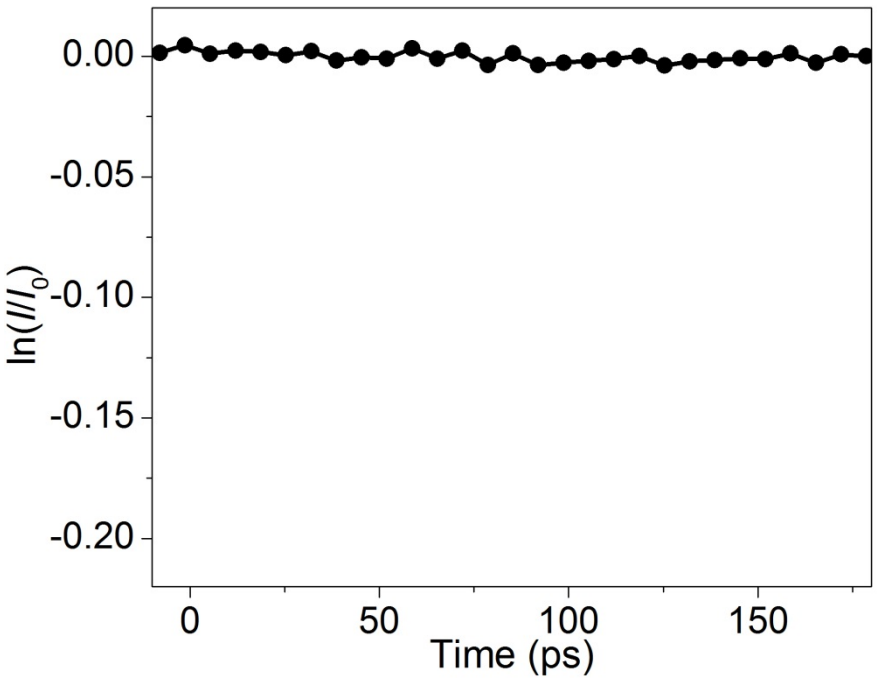

Figure S3. Diffraction intensity change of a bare sapphire(0001) substrate subject to illumination by 515-nm photon at 2 mJ/cm^2.

Additional References:

1. Mak, K. F.; Lee, C. G.; Hone, J.; Shan, J.; Heinz, T. F. Atomically thin MoS_2: A new direct-gap semiconductor, *Phys. Rev. Lett.* **2010**, 105, 136805.
2. Li, J. J.; Wang, X.; Chen, Z. Y.; Zhou, J.; Mao, S. S.; Cao, J. M. Real-time probing of ultrafast residual charge dynamics, *Appl. Phys. Lett.* **2011**, 98, 011501.
3. Stoner, R. J.; Maris, H. J. Kapitza conductance and heat flow between solids at temperatures from 50 to 300 K, *Phys. Rev. B* **1993,** 48, 16373-16387.