

Ultrafast Carrier Dynamics in 2D NbTe₂ Thin Films

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Abstract

As one of the representatives of emerging metallic transition metal dichalcogenides, niobium ditelluride (NbTe₂) has attracted intensive interest recently due to its distorted lattice structure and unique physical properties. Here, we report on the ultrafast carrier dynamics in NbTe₂ measured using time-resolved pump-probe transient reflection spectroscopy. A thickness-dependent carrier relaxation time is observed, exhibiting a clear increase in the fast and slow carrier decay rates for thin NbTe₂ flakes. In addition, pump power dependent measurements indicate that the carrier relaxation rates are power-independent, with the peak amplitude of the transient reflectivity increasing linearly with pump power. Isotropic relaxation dynamics in NbTe₂ is also verified by performing polarization-resolved pump-probe measurements. These results provide an insight into the light-matter interactions and charge carrier dynamics in NbTe₂ and will pave the way for its applications to photonic and optoelectronic devices.

KEYWORDS: Layered transition metal dichalcogenides, NbTe₂ flake, ultrafast carrier dynamics, pump-probe spectroscopy.

1. INTRODUCTION

Since the ground-breaking discovery of graphene,¹ two-dimensional (2D) layered materials have undergone a tremendous surge in interest in the past decade, both in fundamental science as well as industrial applications.²⁻⁸ Layered transition metal dichalcogenides (TMDCs), with a formula of MX₂ (M represents transition metal and X is chalcogen element), are a widely studied family of 2D materials that have demonstrated huge potential for electronic and optical devices owing to their novel electrical and optical properties. Thanks to their atomic film thickness and high carrier mobilities, monolayer MoS₂ and WS₂ films have been used for sub-5nm field-effect transistors (FETs).⁹⁻¹¹ A layer-tunable optical band gap that covers a spectral range from the visible to the NIR regions makes TMDCs promising for broadband photodetectors and highly efficient solar cells.¹²⁻¹⁴ In addition, strong light-matter interactions in atomically thin MoSe₂, WS₂, and PdSe₂ above their bandgap gives rise to many fascinating phenomena, such as exotic excitonic properties,¹⁵⁻¹⁷ a strong optical nonlinearity,¹⁸⁻¹⁹ and quantum interference,²⁰⁻²¹ enabling many new photonic and quantum devices.

Recently, metallic 1-*T* phase TMDCs with exotic physical properties, such as charge density waves (CDW) and low-temperature superconductivity, have attracted significant interest.²²⁻²⁸ NbTe₂ is one example that is a semimetal with a topologically protected band crossing.²⁹ Owing to its semimetal nature and ultrahigh electrical conductivity, NbTe₂ has been used as conductive electrode to reduce the contact resistance and improve carrier mobility of other 2D semiconductors.²⁴⁻²⁵ More importantly, NbTe₂ exhibits a coexistence of CDW and superconductivity below 0.74 K, providing a good platform for unconventional superconductivity and strongly correlated electron systems.²⁶⁻²⁸ Linear magnetoresistance and anisotropic magneto-transport properties were also experimentally observed, demonstrating its strong potential for magnetic devices.^{27, 30} Although progress has been promising for electric and magnetic applications, the optical properties of NbTe₂ have yet to be investigated. This includes the ultrafast carrier dynamics and layer-dependent light-matter interaction.

In this work, we characterize the photon-excited carrier dynamics in mechanically exfoliated NbTe₂ flakes having thicknesses from $\sim 15 - 50$ nm via time-resolved transient reflection spectroscopy. Photoinduced bleaching (PB) of 1040 nm probe light is experimentally achieved when the samples are irradiated by a pump at 520 nm. Thickness-dependent carrier relaxation times are observed, where both the fast and slow relaxation rates decrease with sample thickness. We also observe a linear increase in the transient reflection peak amplitude with pump power, whereas the photon-excited carrier decay times are power independent. In addition, polarization-resolved Raman and pump-probe measurements show that the relaxation dynamics are isotropic. Our results present a comprehensive analysis of photon-excited carrier dynamics in NbTe₂ and provide guidance for its applications to photonic and optoelectronic devices.

2. MATERIALS AND CHARACTERIZATION

Sample preparation. NbTe₂ single crystals were synthesized by the chemical vapor transport (CVT) technique.^{27, 31} High purity Nb foil (99.99%), Te power (99.999%), and iodine (99%) were sealed in an evacuated quartz tube, which was subsequently heated to 550 °C and held for one day in a two-zone furnace. After that, the heating temperatures of the two-zone furnace were increased to 850 °C (source side) and 750 °C (sink side) and kept for one week. After cooled naturally, NbTe₂ single crystals were obtained. NbTe₂ flakes with different thicknesses were exfoliated from the bulk crystals using adhesive tape and transferred onto quartz substrates.

Material characterization. Morphology images and thicknesses of the samples were characterized using atomic force microscopy (Alpha 300ras, WITec) in tapping mode. The resolutions in vertical and transverse directions were ~ 0.1 nm and ~ 8 nm, respectively. Raman spectra were characterized with the same instrument with a 532 nm laser excitation. The linear absorbance of the materials was measured by an ultraviolet-visible (UV-vis) spectrometer.

Time-resolved pump-probe technique. The transient reflection measurements were performed using a Yb fiber-based laser (Menlo Systems) with a central wavelength at 1040 nm. The repetition rate and pulse width of the laser were 100 MHz and 150 fs, respectively. The laser beam size is ~ 4 μ m. Five percent of the output laser was employed as a probe beam while the balance of 95% provided the pump pulse at 520 nm via frequency doubling. A half-wave plate combined with a linear polarizer was used as a continuously adjustable power attenuator. After passing through a free space time-delay line, the pump and probe pulses were focused with an objective lens (Tu Plan Fluor 50 x NA = 0.8, Nikon) onto the sample surface with a Gaussian spot. The reflected probe beam was separated from the pump light by using a color filter before reaching the silicon photodetector, which significantly improved the signal-to-noise ratio. A lock-in amplifier (SR865A, Stanford Research Systems) referenced to 1.5 kHz mechanically chopped pump (SR542, Stanford Research Systems) was employed to collect the reflection change (ΔR) of the probe beam due to the pump excitation.

3. RESULTS AND DISCUSSION

NbTe₂ is a typical layered CDW material with two different structural phases. At high temperature (above 550 K), it exhibits a high symmetry 1-*T* phase where each Nb atom is coordinated octahedrally by Te atoms.³² Below 550 K, NbTe₂ undergoes a CDW phase transition which results in a displacement of Nb atoms from the octahedral centers to a monoclinically distorted 1-*T'* phase (1-*T'* phase).^{28, 30} This 1-*T'* phase is very stable at room temperature since the phase transition temperature is much higher. The crystal structure of 1-*T'* NbTe₂ is shown in Figure 1(a).

Each monolayer is composed of an Nb layer sandwiched by two Te layers, where the Nb atoms are displaced within the plane to form “trimers,” whereas the Te atoms present an out-of-plane buckling.^{28, 32} The Te-Nb-Te sandwiches stack with weak van der Waals interactions to form a layered structure.

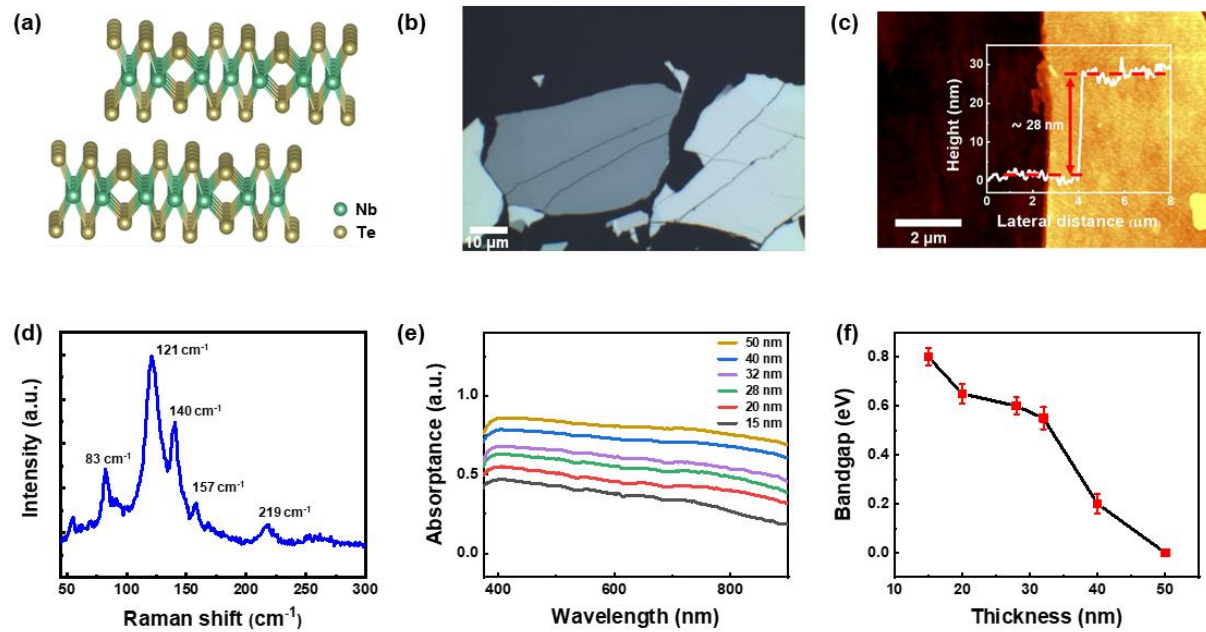


Figure 1. (a) Schematic crystal structure of monoclinic 1-T' NbTe₂. The green and yellow dots represent the Nb and Te atoms, respectively. (b) Optical microscopy image of an exfoliated NbTe₂ flake. (c) AFM height profile of the NbTe₂ flake. The measured thickness is ~28 nm. (d) Raman spectrum excited via a 532 nm laser. (e) UV-vis absorption spectrum. (f) Determined optical bandgaps of samples with different thicknesses.

We prepared single crystal NbTe₂ flakes with different thicknesses via mechanical exfoliation. An optical microscopy image of a representative sample is shown in Figure 1(b). Different contrasts represent areas with different thicknesses. It can be seen that the exfoliated flake presents a flat surface with uniform thickness in the different areas. Figure 1(c) shows the AFM height profile of the NbTe₂ flake, which indicates that the thickness of the flake is ~28 nm. Due to the strong interlayer coupling of NbTe₂, it is very difficult to obtain very thin samples using mechanical exfoliation.²⁵ The thinnest flake obtained in our experiments is ~15 nm. Further AFM images of NbTe₂ flakes with different thickness are shown in Figure S2 (Supporting Information). The Raman spectrum of a NbTe₂ flake is shown in Figure 1(d) with an excitation laser at 532 nm. Characteristic peaks at ~55 cm⁻¹, ~83 cm⁻¹, ~121 cm⁻¹, ~140 cm⁻¹, ~157 cm⁻¹, ~168 cm⁻¹, ~219 cm⁻¹, and ~262 cm⁻¹ can be observed, which correspond to the phonon modes of A_g¹, A_g², A_g⁴, A_g⁵, A_g⁶, B_g⁴, A_g⁷, and A_g⁸ in NbTe₂, respectively.³³⁻³⁴ These results indicate the high crystal quality of the samples. Optical absorption spectra (from 400 nm to 900 nm) of NbTe₂ flakes with different thicknesses were measured by using a UV-vis spectrometer, as shown in Figure 1(e). A broadband absorption response with a smooth absorption band in the wavelength range can be observed for all the thicknesses. The thickness-dependent optical bandgap is estimated from a Tauc plot of $(\alpha h\nu)^{1/2}$ versus $h\nu$ based on the Tauc formula (Figure S3), where α and $h\nu$ represent the optical absorption coefficient and photon energy, respectively. Figure 1(f) shows the measured optical bandgaps as a function of thicknesses, where the bandgap of the NbTe₂ decreases from ~0.8 eV to 0 eV with increasing the sample thickness from 15 nm to 50 nm.

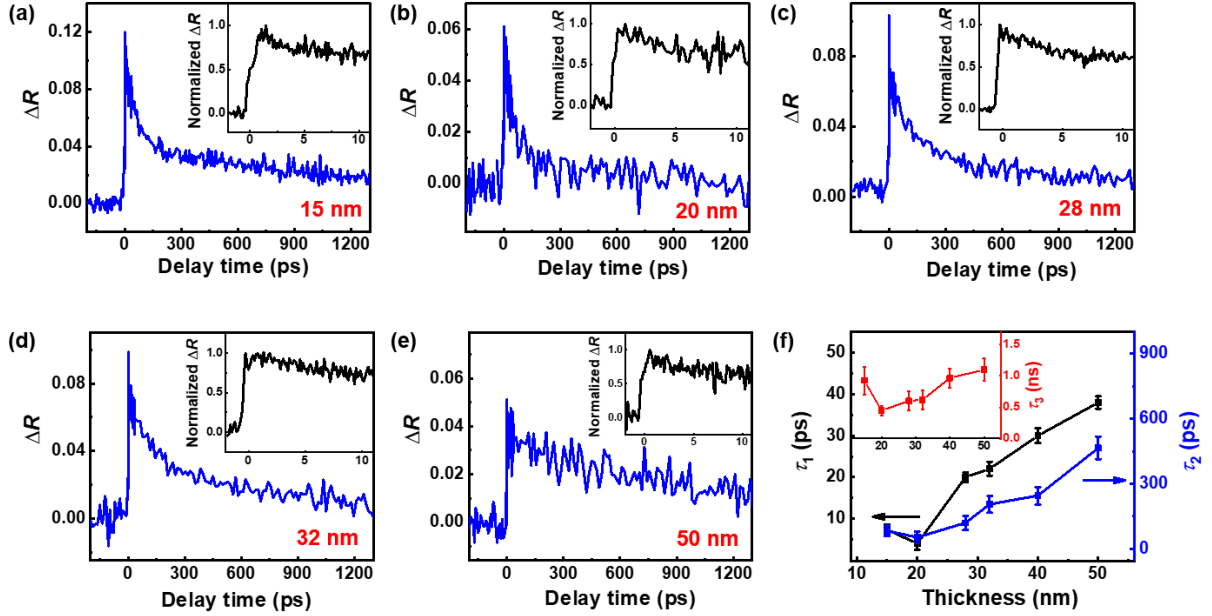


Figure 2. (a)–(e) Time-resolved transient reflection (ΔR) curves of NbTe₂ flakes with different thicknesses. The laser powers of pump (520 nm) and probe (1040 nm) beams are $\sim 40 \mu\text{W}$ and $\sim 35 \mu\text{W}$, respectively. The insets show the normalized ΔR curves around 0 delay time. (f) The measured relaxation time constants with different thicknesses.

To characterize the photon-excited carrier dynamics, time-resolved pump-probe transient reflection (ΔR) spectroscopy was used with a pump laser at 520 nm and probe laser at 1040 nm. The pump-induced probe reflection change ($\Delta R = R - R_0$) was measured by chopping the pump and monitoring the output of the photodiode with a lock-in amplifier, where R and R_0 are the probe reflections with and without pump light, respectively. Figures 2(a) – (e) show the time-resolved ΔR curves for flakes with thicknesses from ~ 15 nm to ~ 50 nm. The insets of these figures present the corresponding normalized ΔR curves for 0 delay times. It can be seen that, for all thicknesses, a fast increase of probe reflection from zero to its maximum value (positive ΔR) is observed at zero-delay. The positive ΔR indicates photoinduced bleaching (PB) of the probe light.³⁵ Since the NbTe₂ bandgap is much less than the pump photon energy (~ 2.38 eV), the pump can excite electrons directly from the valance to conduction bands. These excited carriers are commonly known to decrease the absorption of the probe light and enhance its reflection due to the filling of states and the Pauli-blocking effect.^{36–39}

After ΔR reaches its maximum, a decay process can be observed in the ΔR curves, which can be mainly separated into two components: a sharp drop of ΔR followed by a slow relaxation process, as shown in Figure 2(a) – (e). By fitting the experimental data, relaxation time constants during the decay process can be obtained. In our case, a tri-exponential decay function was used to fit the measured ΔR curves, as follows:^{40–41}

$$\frac{\Delta R(t)}{R_0} = A \exp\left(\frac{-t}{\tau_1}\right) + B \exp\left(\frac{-t}{\tau_2}\right) + C \exp\left(\frac{-t}{\tau_3}\right) \quad (1)$$

where A , B , and C denote the corresponding amplitudes. t denotes the delay time between the pump and probe, and τ_1 , τ_2 , and τ_3 are the time constants of relaxation processes. Here, we combine the semi-log fit with the tri-exponential fit for better evaluation of the time constants.

The measured values of τ_1 , τ_2 , and τ_3 for different film thicknesses are presented in Figure 2(f). It can be seen that the sample having the fastest relaxation time was 15-nm thick, and had a $\tau_1 \sim 7.4$ ps. This is in the same order of magnitude of other TMDCs, such as MoS_2 ⁴²⁻⁴³ and PdSe_2 .³⁵ This picosecond relaxation process can be attributed to carrier-carrier and carrier-phonon scattering during the carrier-cooling process.⁴⁴⁻⁴⁷ The pump-excited hot carriers initially thermalize to quasi-equilibrium states through carrier-carrier scattering. They then transfer their energy to the NbTe_2 lattice and are cooled mainly by electron-phonon scattering. A thickness-dependent behavior can be observed in τ_1 , where it increases from ~ 7.4 ps to 38.3 ps as the sample thickness increases from 15 nm to 50 nm. It has been demonstrated that an increase in thickness in TMDCs can lead to an enhancement of dielectric screening of the long-range Coulomb interaction, weakening the electron-phonon coupling,⁴⁸⁻⁴⁹ which in turn increases the relaxation time τ_1 for thicker samples.

The time constant τ_2 exhibits a similar trend to τ_1 with increasing sample thickness, although with an overall slower lifetime, ranging from ~ 83.4 ps for 15-nm to ~ 465 ps for the 50-nm flakes, as shown in Figure 2(f). We attribute this relatively longer relaxation process to the anharmonicity-driven phonon-phonon scattering.⁵⁰ As discussed above, τ_1 denotes carrier relaxation to phonons via fast carrier-phonon scattering processes. The subsequent thermalization of these generated phonons with the rest of the phonon subsystem takes a longer time via the anharmonicity-driven phonon-phonon scattering. This phonon dominating process may also explain the thickness-dependent τ_2 because of the slower phonon cooling process occurring in thicker flakes.⁵¹ The longest lifetime τ_3 , is on a nanosecond time scale (inset of Figure 2(f)), which arises from lattice cooling by dissipating the energy to the substrate.^{37, 52-53}

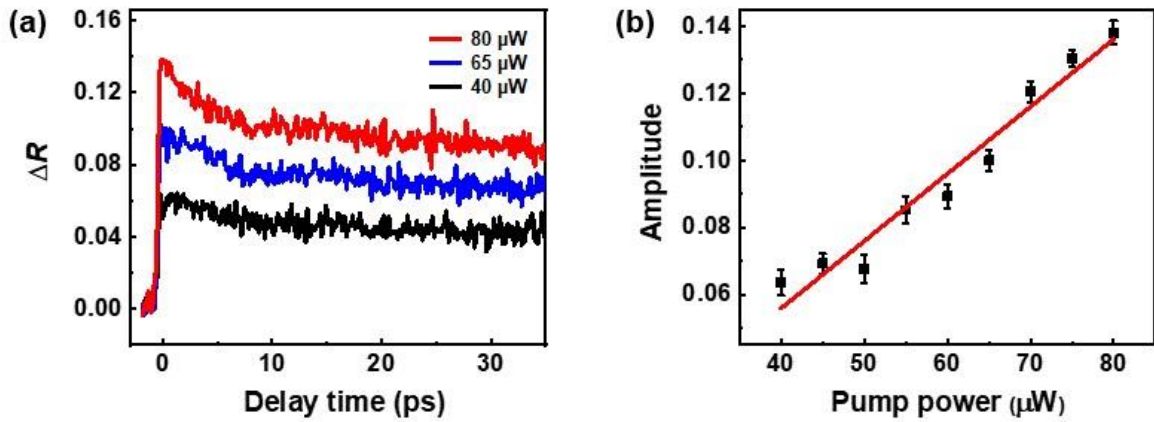


Figure 3. (a) Time-resolved transient reflection (ΔR) curves of a 32 nm-NbTe₂ flake with different pump laser powers. (b) Corresponding peak amplitudes of the ΔR curves as a function of the pump power. The black solid squares represent the experimental data, and the red solid line is the linear fit.

Figure 3(a) shows pump power dependent ΔR measurements for a 32 nm-flake with pump powers from 40 μW to 80 μW , with the probe power fixed at 35 μW . Similar temporal features in the ΔR curves can be observed for different pump powers, indicating that the carrier relaxation dynamics in NbTe₂ are pump power independent, similar to other TMDCs.^{43, 46} In contrast, for the ΔR amplitudes, a clear increase with pump power is observed. Figure 3(b) plots the corresponding peak amplitudes extracted from the ΔR curves in Figure 3(a), demonstrating a linear relationship between the amplitude and pump power. The observed linear contribution of the pump power indicates a one-photon excitation of carriers in NbTe₂ with the pump beam and contribution to Pauling blocking

at the probe wavelength.^{35, 43} The extracted peak amplitudes as a functions of pump power for other thicknesses are presented in Figure S4.

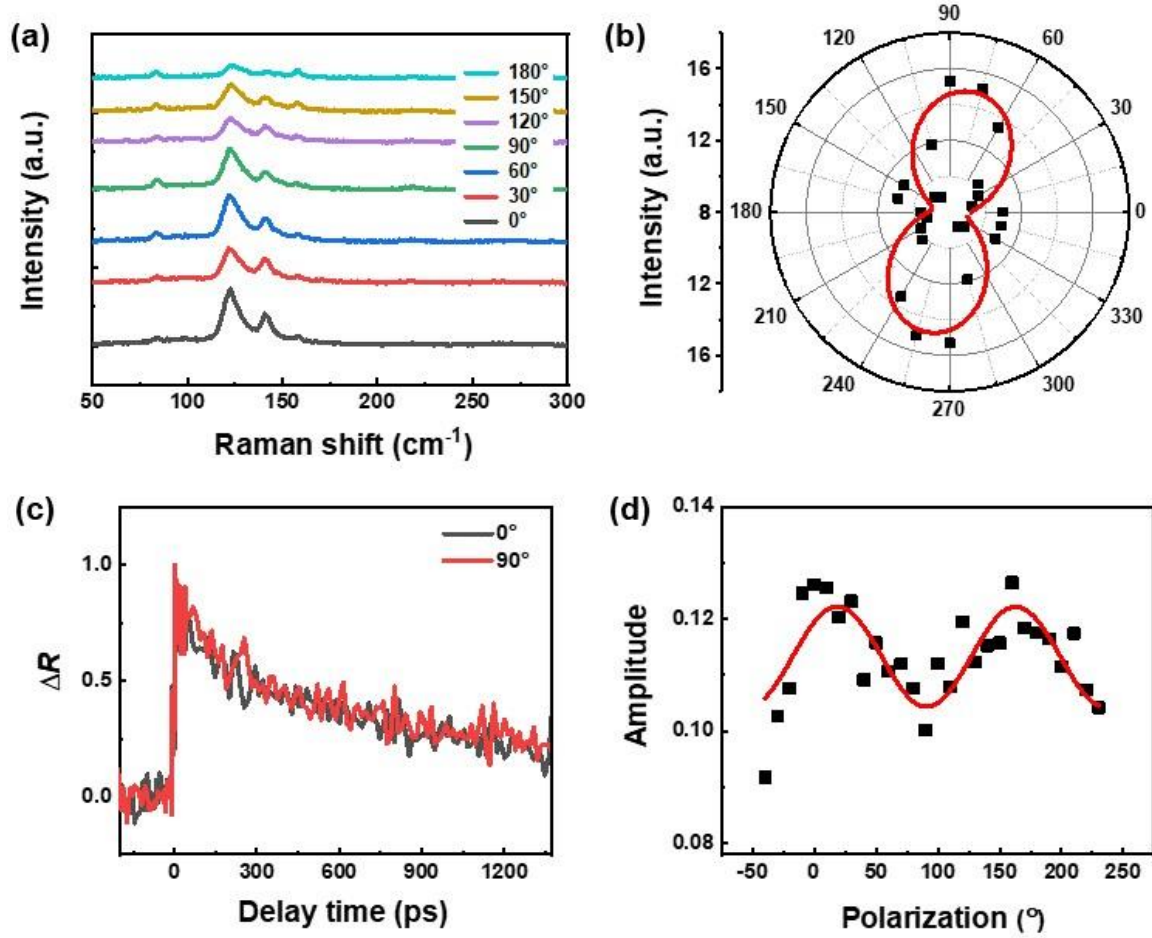


Figure 4. (a) Polarization-dependent Raman spectra of NbTe₂ flake. (b) Polarization diagram of the Raman intensities of A_g² mode ($\sim 83.4 \text{ cm}^{-1}$) was extracted through the fitting of the Raman spectra of each polarization angle under the parallel configurations. (c) Normalized ΔR curves under different pump polarization, where the probe polarization is fixed at 0°. (d) Peak amplitudes as a function of pump polarization angles with respect to the sample orientation.

We investigated the anisotropic ultrafast carrier dynamics via polarization-dependent pump-probe measurements. Angle-resolved polarized Raman spectroscopy was used to analyze the crystal axis of NbTe₂ flakes under a parallel configuration, with an excitation laser wavelength of 532 nm. In the experiment, we fixed the sample and rotated the polarizers in the incident and scattered light paths to vary the angle between the sample crystallographic orientation and the polarizations of beams. Figure 4(a) shows the Raman spectra of a flake for different excitation laser polarization angles. To better illustrate the polarization trend, the polarization diagram of A_g² mode of the sample is plot in Figure 4(b). It can be seen that the peak intensity of the Ag mode oscillates with a periodicity of 180° as the orientation of the polarization is rotated. Therefore, by using this polarization diagram, the crystallographic orientation of the flakes can easily be determined.

After determining the crystal directions, we conducted the polarization-resolved pump-probe measurements. The pump and probe powers were 40 and 35 μW , respectively, with their polarization angles controlled by rotating

a half-wave plate. Figure 4(c) shows the normalized ΔR curves of the 40-nm sample for pump polarization angles of 0° and 90° with respect to the sample orientation. Varying the pump polarization did not change their temporal response, indicating that the photon-excited carrier relaxation process is isotropic in NbTe₂ flake. We also measured the peak amplitudes of the ΔR curves under different pump polarization angles (Figure 4(d)) where a sinusoidal dependence on the polarization angles is observed, originating mainly from the anisotropic pump absorption. This is further verified by the polarization-dependent transmission of pump light in the sample, as shown in Figure S5.

4. CONCLUSIONS

In summary, by using time-resolved transient reflection spectroscopy, we characterize the photon-excited carrier dynamics in mechanically exfoliated single crystal NbTe₂ flakes. A typical photoinduced bleaching (PB) of probe light at 1040 nm and thickness-dependent relaxation dynamics of excited carriers in NbTe₂ flakes are observed when the samples are irradiated with a 520-nm pump beam. The influence of the pump power is also investigated, showing a linear increase in the transient reflection peak amplitude with pump power, with a power-independent carrier decay. Polarization-resolved pump-probe measurements indicate that the carrier relaxation dynamics in NbTe₂ is isotropic. These properties demonstrate the potential of NbTe₂ as a novel and interesting 2D material for photonic and optoelectronic applications. In particular, these results indicate that the ultrafast response of single crystal NbTe₂ flakes could be useful for integrated photonic chips based on CMOS compatible platforms for microcomb devices [55-70] for high bandwidth applications [71-190].

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SUPPORTING INFORMATION

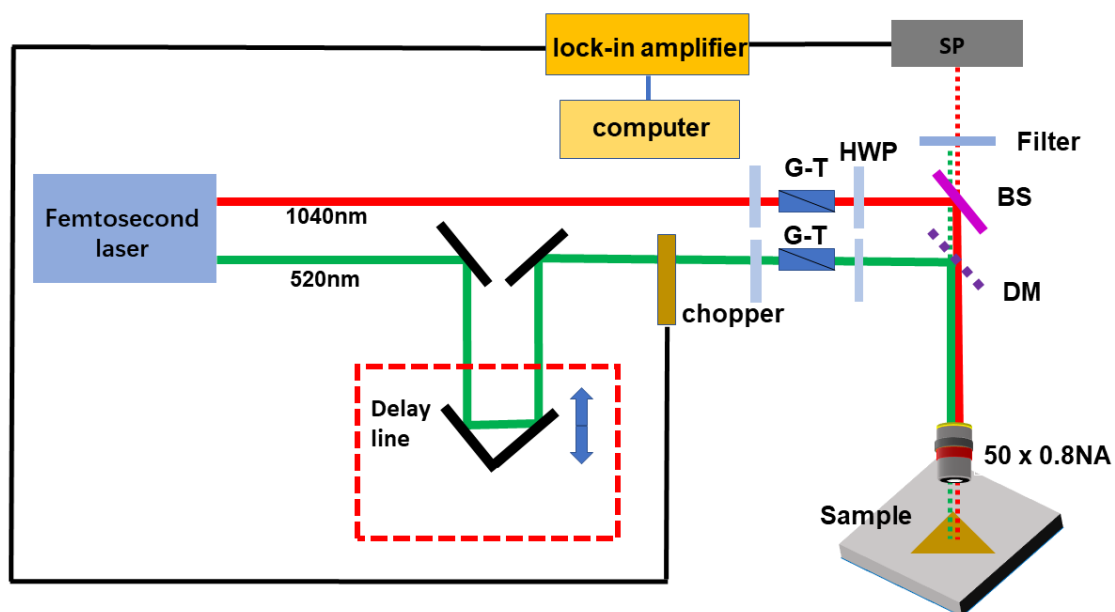


Figure S1. Schematic diagram for the time-resolved transient reflection measurement system. BS: beam-splitter; HWP: half-wave plate; DM: dichroic mirror; SP: Silicon photodiode detector; G-T: Glan-Taylor prism.

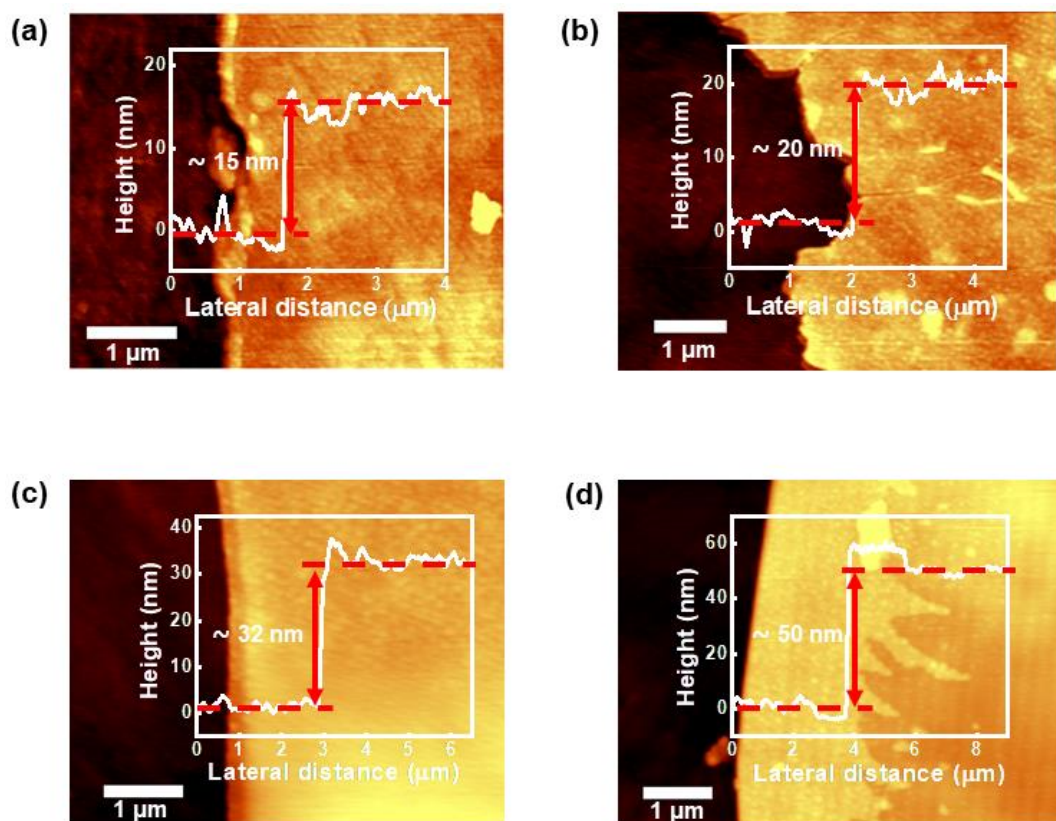


Figure S2. (a)–(d) AFM images and height profiles for NbTe₂ samples with different thicknesses.

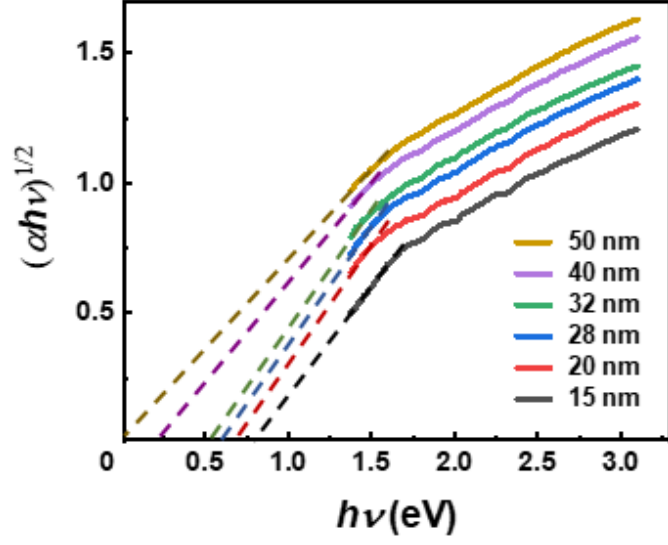


Figure S3. Tauc plots of NbTe₂ flakes with different thicknesses.

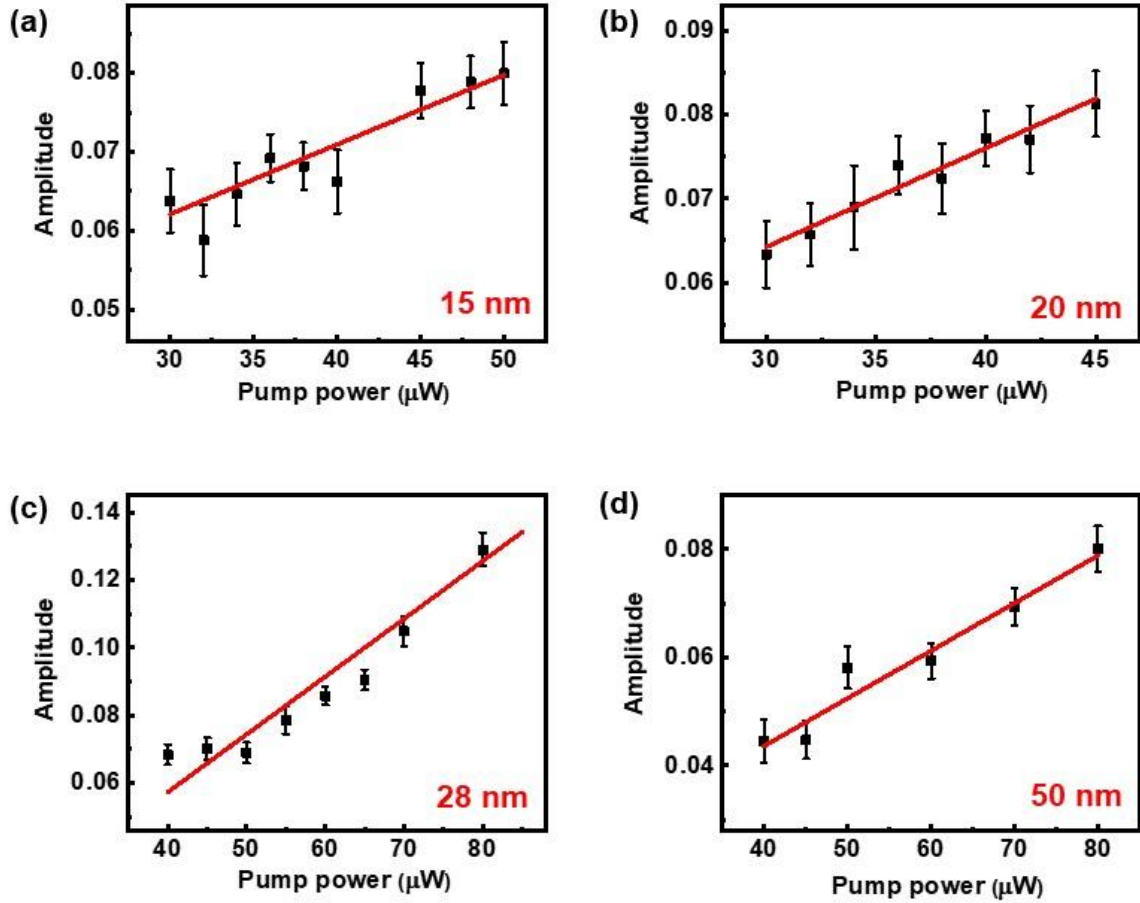


Figure S4. (a)–(d) Peak amplitudes of the ΔR curves as a function of the pump power for NbTe₂ flakes with different thicknesses. The black solid squares represent the experimental data, and the red solid line is the linear fit.

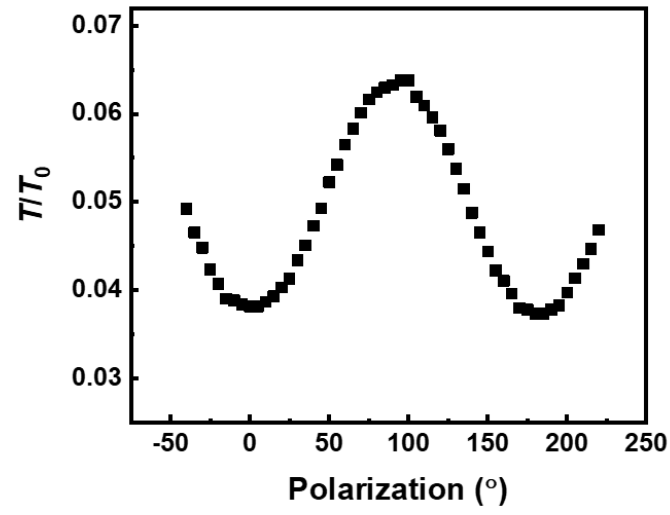


Figure S5. Polarization-resolved transmission of 520 nm pump light in NbTe₂ sample. T and T_0 are measured transmissions for the sample and quartz substrate, respectively.