Ultrafast Mid-IR Carrier Dynamics in Three-Dimensional Dirac Semimetal Cd_3As_2

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Abstract

We investigated ultrafast carrier dynamics in Cd_3As_2 at 2.6 μm. Single-exponential decay and saturable absorption features are observed. The ultrafast optical nonlinearity suggests that Cd_3As_2 is useful for mode-locking lasers in the mid-IR range.

I. INTRODUCTION

Recently, the existence of three-dimensional (3D) Dirac fermions has been theoretically predicted in which the Dirac nodes are developed via the point contact of conduction-valence bands, reminiscence of 3D graphene [1,2]. Cadmium arsenide (Cd_3As_2), as an exemplary three-dimensional Dirac semimetal, has been paid special attentions due to its chemical stability in air and extremely high mobility at both low and room temperatures. Studies based on Cd_3As_2 bulk materials have shown ultrahigh mobility of 9×10^6 $cm^2V^{-1}s^{-1}$ at 5 K and up to 1.5×10^4 $cm^2V^{-1}s^{-1}$ at room temperature [3,4]. Giant and linear magnetoresistance and nontrivial π Berry's phase of Dirac fermions were demonstrated in transport experiments [3,5]. A superconductivity phase was also identified in a Cd_3As_2 crystal making it an interesting candidate of the topological superconductors [6]. It is predicted that Cd_3As_2 hosts topologically nontrivial Fermi-arc states on its surface which serves as a starting point to realize a Weyl semimetal, quantum spin Hall insulator, or axion insulator [7]. Despite the extensive studies on Cd_3As_2, few efforts have been devoted to the ultrafast spectroscopy investigation of photocarriers in this novel material. Here, we report ultrafast carrier dynamics of Cd_3As_2 using a two-color pump-probe technique. The experimental results reveal that Cd_3As_2 exhibit mono-exponential decay and saturable absorption features at 2.6 μm, providing insights into the ultrafast optical properties of this novel Dirac semimetal.

II. EXPERIMENTAL DETAILS

High quality Cd_3As_2 films with 123 nm thickness were grown on mica using molecular beam epitaxy (MBE). The two-color ultrafast pump-probe setup is based on an 800 nm, 1 kHz, Ti: sapphire amplifier. A part of the laser output is used as pump to excite photocarriers in the sample, and the remaining is fed to an optical parametric amplifier (OPA) to generate mid-IR probe beam. In this work, the probe wavelength is fixed at 2.6 μm. Both pump and probe pulses have durations of ~100 fs. The probe beam through the sample is detected by a PbSe photo-detector and a lock-in amplifier referenced to a 500 Hz chopped pump.

III. RESULTS AND DISCUSSIONS

The differential transmission spectra $\Delta T/T_0$ as a function of the delay between the pump and probe pulses with varying pump fluences are shown in Fig. 1. The positive $\Delta T/T_0$ indicates that photobleaching (PB) is occurring in the Cd_3As_2 sample, and this phenomenon can be explained by a simple Pauli blocking effect [8]. To extract relaxation times, the experimental data are fitted by a single-exponential delay. As shown in Fig. 2, the fitted recovery time constants vary in the range of 4.9 - 7.8 ps. Such time scales indicate that the relaxation process may be dominated by carrier-phonon scattering. The elongation of the relaxation time with increasing pump fluences suggests the presence of a bottleneck effect in Cd_3As_2, similar to graphene [9].

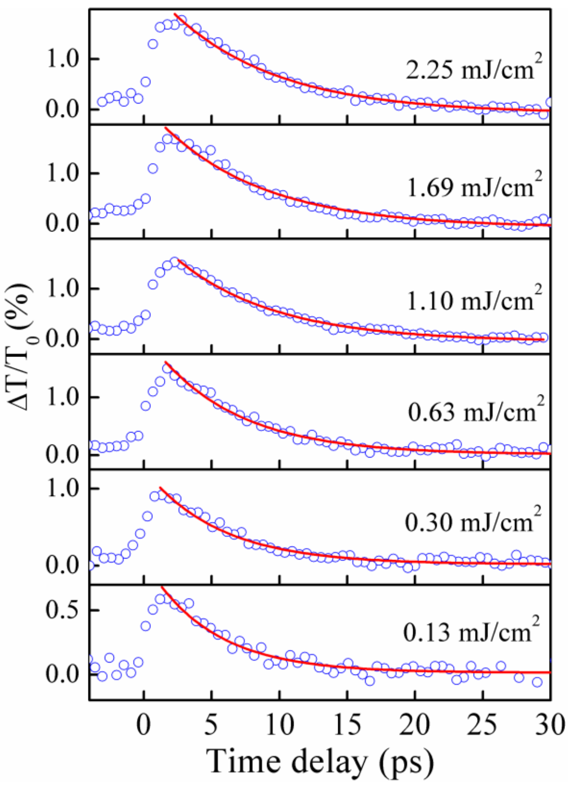

Fig. 1. Time-resolved differential transmission ($\Delta T/T_0$) spectra with varying pump fluences using 800 nm pump and 2600 nm probe. The solid line corresponds to a mono-exponential fit.

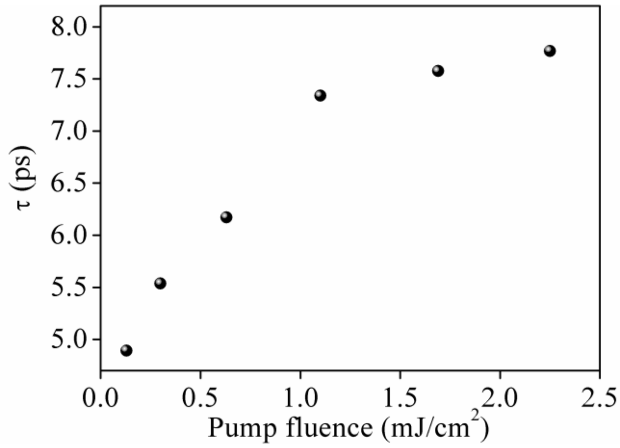

Fig. 2. Pump fluence dependence of the fitted delay time *τ*.

Materials exhibiting saturable absorption are currently intensively researched for ultrashort pulse generation in fiber and solid-state lasers, through an effect known as mode-locking. In particular, saturable absorbers operating beyond 2.5 μm are becoming increasingly desirable for a number of practical mid-IR laser platforms [10]. Fig. 3 shows the pump fluence dependence of the maximum value of $\Delta T/T_0$. The results reveals a clear saturation of absorption at high fluences. Generally the differential transmission is proportional to the pump induced change in the occupation probability, and the saturation behavior can be described with a simple relation [11]

$$\frac{\Delta T_{max}}{T} \propto \frac{I/I_{sat}}{I+I_{sat}} \qquad (1)$$

where I is the pump fluence and I_{sat} is the saturation fluence. The experimental data have been fitted using this equation as shown in Fig. 3 (red line). The inferred I_{sat} is ~0.32 mJ/cm^2. This result experimentally demonstrates that Cd_3As_2 has a saturation behavior at 2.6 μm, and it is promising for pulse generation in the mid-IR wavelength range.

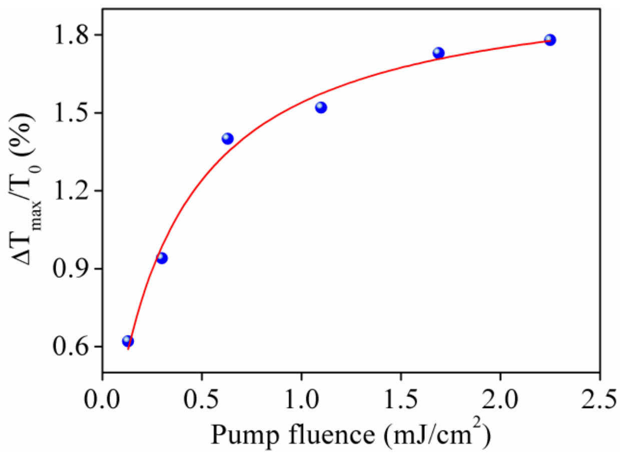

Fig. 3. The maximum transmission change as a function of pump fluences. The solid line is fitted to the experimental data using Eq. (1)

IV. Conclusions

In conclusion, a two-color pump-probe experiment has been carried out to investigate the mid-IR ultrafast carrier dynamics in Cd_3As_2 films at a probe wavelength of 2.6 μm. A mono-exponential relaxation process is observed, and the fitted recovery time constants is found to slow down from 4.9 to 7.8 ps as pump fluence is increased. A pronounced saturable absorption behavior in Cd_3As_2 is identified at 2.6 μm, which is useful for mode-locking lasers operating in the mid-IR range.

Acknowledgment

This work was supported in part by National Natural Science Foundation of China 61378025, 61450110087, 61327812.

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