

# Three-dimensional Dirac semimetal Cd<sub>3</sub>As<sub>2</sub> as high-performance 2-5 μm saturable absorbers

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**Abstract:** 2-5 μm saturable absorbers are highly desirable for constructing convenient mid-infrared ultrafast sources. We reveal that three-dimensional Dirac semimetal Cd<sub>3</sub>As<sub>2</sub> constitutes an ideal mid-infrared saturable absorber, with broadband operation and flexible parameter control.

**OCIS codes:** (190.7110) Ultrafast nonlinear optics; (140.3070) Infrared and far-infrared lasers; (140.3380) Laser materials

## 1. Introduction

The mid-infrared 2-5 μm band has long been devoid of compact and convenient short-pulsed lasers. Although a wide range of applications including spectroscopy, sensing and communications are expected to benefit from such sources, the lack of a robust and capable mid-infrared saturable absorber (SA) seriously impedes the development of ultrashort-pulsed lasers in this spectral range. Semiconductor Saturable Absorber Mirrors (SESAMs) are currently the primary technology employed for passive mode-locked solid-state and fiber lasers [1]. However, their operating range is limited to the near-infrared because of the use of conventional III-V semiconductor materials [2]. A broadband alternative technology relies on low-dimensional materials, for example carbon nanotubes, graphene, transition-metal dichalcogenides and other emerging two-dimensional (2D) materials [3-5], but all of them are faced with practical limitations in terms of scalability and repeatability. Thus, a generalized and highly scalable saturable absorber mechanism covering the entire 2-5 μm range is still yet to be developed.

Here, we propose that this technological challenge for the mid-infrared can be adequately addressed by using Cd<sub>3</sub>As<sub>2</sub>, a representative 3D topological Dirac semimetal (TDS) material, whose electronic band structure is analogous to that of graphene [6,7]. Our experimental results demonstrate that Cd<sub>3</sub>As<sub>2</sub> exhibits saturable absorption covering the entire 2-5 μm range. In addition, its recovery time can be flexibly tailored through Chromium (Cr) doping. The use of Molecular Beam Epitaxy (MBE) growth method further provides the advantage of excellent reproducibility, making our approach well suited for practical exploitation.

## 2. Experimental

Both pure and Cr-doped Cd<sub>3</sub>As<sub>2</sub> films were grown on mica substrates under the ultra-high vacuum in a MBE system. For non-degenerate pump-probe measurements, the pump beam was obtained from an 800 nm, 1 kHz Ti: Sapphire amplifier. The probe beam (2-4.7 μm) was derived from an optical parametric amplifier (OPA). Both the pump and probe pulses have durations of ~100 fs. To detect the differential transmission of probe (induced by the pump), pump beam was chopped to 500 Hz and the fluence of the probe was recorded by a photo-detector and lock-in system. During the measurement, the pump fluence was fixed at 100 μJ/cm<sup>2</sup>. All the measurement performed at room temperature.

## 3. Results and discussion

Fig. 1a shows a typical non-degenerate transient absorption spectroscopy for a pure, un-doped Cd<sub>3</sub>As<sub>2</sub> film with a wideband spectral range from 2 to 4.7 μm. Owing to band filling effect, all of the traces present ultrafast saturable absorption [8]. Note that 4.7 μm is limited by the photodetector used in our setup, not the Cd<sub>3</sub>As<sub>2</sub> sample. Considering that the interband transitions in Cd<sub>3</sub>As<sub>2</sub> start around 200 meV [9], we expect the operating wavelength of Cd<sub>3</sub>As<sub>2</sub> can reach approximately 6 μm. The recovery time constants were fitted by a mono-exponential function, and are found to fall within the range of 5.4-8.2 ps. These ultrafast nonlinear behaviors indicate that Cd<sub>3</sub>As<sub>2</sub> is suitable as SA for pulsed generation in the 2-5 μm.

As recovery time is intrinsic to the saturable absorber materials, achieving flexible tuning plays a key role in realizing high-performance SA devices. Therefore, we next investigate relaxation time tuning ability of Cd<sub>3</sub>As<sub>2</sub> using Cr doping. The normalized pump-probe curves at a probe wavelength of 2 μm for Cd<sub>3</sub>As<sub>2</sub> films having different Cr concentrations are shown in Fig. 2c. For all wavelengths, the decay rate of Cd<sub>3</sub>As<sub>2</sub> films become faster at higher Cr concentrations. This phenomenon is attributable extra trapping centers induced by the dopants [11]. As

listed in Fig. 1d, the relaxation time can be tuned across an order of magnitude, which is unprecedented for a mid-infrared saturable absorber. Such flexible parameter tuning has not been demonstrated in graphene or other emerging low-dimensional materials.

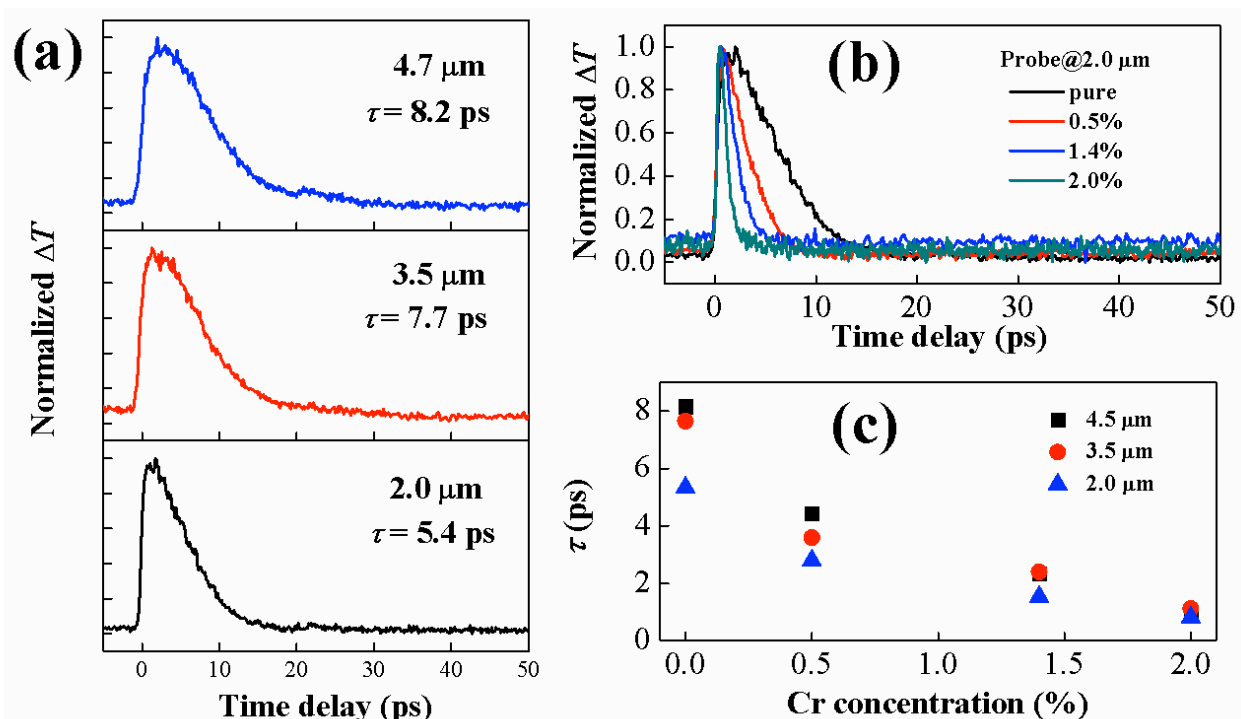


Fig.1 (a) The non-degenerate pump-probe results at 2.0, 3.5 and 4.7  $\mu\text{m}$ . (b) Time-resolved  $\Delta T$  traces at 2  $\mu\text{m}$  for the Cd<sub>3</sub>As<sub>2</sub> samples, (c) The fitted recovery times, as a function of Cr concentrations.

#### 4. Conclusion

In summary, we have for the first time probed the broadband mid-infrared nonlinear optical properties of 3D TDS Cd<sub>3</sub>As<sub>2</sub>. Pronounced saturable absorption covering the entire 2-5  $\mu\text{m}$  band is revealed. Furthermore, relaxation time tuning is also demonstrated in Cd<sub>3</sub>As<sub>2</sub> via element doping during MBE growth. Our findings constitute the first mid-infrared functional optical device harnessing the robust Dirac fermions in 3D TDS systems, opening up the long-sought parameter space for realizing high-performance mid-infrared mode-locked lasers, including those based on fiber, solid-state and quantum-cascade gain platforms.

#### 5. References

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