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Dirac semimetal saturable absorber with actively tunable modulation depth

Yue Sun,[1,†] Yafei Meng,[1,†] Hongzhu Jiang,[1] Shuchao Qin,[1] Yunkun Yang,[3] Faxian Xiu,[3] Yi Shi,[1] Shining Zhu,[4] and Fengqiu Wang[1,2,*]

[1]School of Electronic Science and Engineering and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, China
[2]Key Laboratory of Intelligent Optical Sensing and Manipulation, Ministry of Education, Nanjing University, Nanjing 210093, China
[3]State Key Laboratory of Surface Physics and Department of Physics, Collaborative Innovation Center of Advanced Microstructures, Fudan University, Shanghai 200433, China
[4]National Laboratory of Solid State Microstructures and School of Physics and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, China
*Corresponding author: fwang@nju.edu.cn

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In this Letter, we demonstrate an electrically contacted saturable absorber (SA) device based on topological Dirac semimetal Cd_3As_2. With a current-induced temperature change in the range of 297–336 K, the modulation depth of the device is found to be significantly altered from 33% to 76% (under the irradiation of a 1560 nm femtosecond laser). The broad tuning of the modulation depth is attributed to the strong temperature dependence of the carrier concentration close to room temperature. The simple tuning mechanism uncovered here, together with the compatibility with III-V compounds substrate, such as GaAs, points to the potential of fabricating broadband, electrically tunable, SESAM-like devices based on emerging bulk Dirac materials.

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Semiconductor saturable absorber mirrors (SESAMs) are widely used as the optical switching devices for picosecond and femtosecond pulse generation [1–3]. In particular, the ease of customizing their various optical properties (including recovery time, modulation depth, and saturation fluence) has made SESAMs one of the most viable solutions for commercial ultrafast fiber and solid-state lasers [3,4]. In addition to property control via engineering material compositions, defects, and resonant structures, SESAMs offer the promising prospect of integrated electrical control of its various optical characteristics, making it an ideal device for in-depth investigation of mode-locking dynamics and optimizing mode-locked regimes with desirable figures of merit [5–8]. However, due to the use of conventional III-V compounds, the current SESAM technology is still limited in terms of access to the long wavelength range [9]. In addition, to achieve active control of the device, rather sophisticated quantum structures have to be fabricated, which limit its prevalent use in practical systems [6,7,10]. Therefore, the quest for new materials that can enable extended spectral coverage, as well as simpler parameter control schemes, is still on.

Three-dimensional Dirac semimetal material cadmium arsenide (Cd_3As_2) has emerged as a novel material for a range of electronic and optical devices [11–14]. In particular, molecular beam epitaxy (MBE) grown Cd_3As_2 has recently been identified as a capable broadband optical switching material [15–17]. Indeed, pulsed operation across a 1–3 μm range has been demonstrated, with a strong nonlinear optical response measured up to 6 μm. What is remarkable about this emerging Dirac quantum material is that the photocarrier lifetime of Cd_3As_2 thin film, an important parameter that is closely related to mode-locking dynamics can be flexibly and accurately controlled by an element-doping approach [15]. It would be highly desirable if the absorption features, particularly the modulation depth, could also be tailored [18,19]. Combining fine control of both the temporal and the absorptive features of a saturable absorber (SA) would allow unmatched flexibility in studying the rather complex mode-locking dynamics, especially those within a semiconductor laser, where transient effects from both the gain medium and the absorber have to be considered [10]. Furthermore, active real-time adjustment of nonlinear parameters of real SAs will open more space for optimizing mode-locked regimes in a designated laser and may impact the design paradigm of ultrafast lasers [20,21].

In this Letter, we have for the first time, to the best of our knowledge, fabricated an electrically contacted SA device with a Cd_3As_2 layer directly deposited on an undoped GaAs substrate, and investigated its nonlinear absorption features, i.e., absorbance as a function of incident light fluence, in the near-infrared range. Surprisingly, it is found that with a simple current-induced temperature change, the modulation depth of the Cd_3As_2 SA device can be significantly tuned at the investigated wavelength of 1560 nm. We also probed the transient dynamics of Cd_3As_2 under different temperatures and found that the

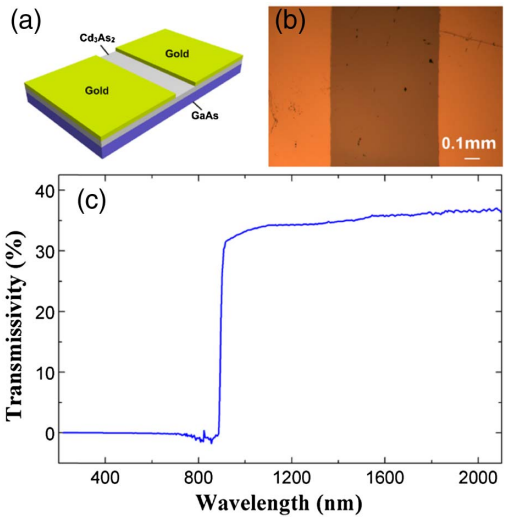

Fig. 1. (a) Schematic of a Cd_3As_2 SA device. (b) Photograph of a Cd_3As_2 SA device. (c) Small-signal linear transmission spectrum of the Cd_3As_2 SA device at room temperature.

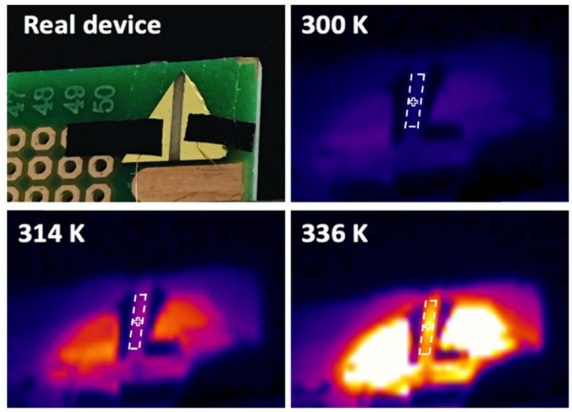

Fig. 2. (Top left panel): Photograph of the real device bound to a PCB board. A hole in the board allows both reflectivity and transmissivity measurements of the Cd_3As_2 film. (Other panels): Illustration of the change of the channel temperature, as the current is increased from 10 (300 K) to 30 mA (314 K) and further to 50 mA (336 K).

temporal response, however, does not exhibit sensitive dependence on the temperature. Our results suggest it is possible to combine different technical approaches to independently control the two most important optical characteristics of Dirac semimetal-based SAs and point out that there is rich photophysics associated with this emerging class of quantum materials.

The Cd_3As_2 thin film, with a thickness of about 30 nm, was grown on an undoped GaAs substrate using a standard MBE [15]. The thickness of the GaAs substrate is about 350 μm. Two gold electrodes separated by approximately 1 mm were deposited on the Cd_3As_2 thin film using electron-beam evaporation. The schematic diagram and microscope photograph of the SA device are shown in Figs. 1(a) and 1(b). The lateral dimension of the device is about 15 mm × 10 mm. A small-signal transmission spectrum of the SA device is shown in Fig. 1(c). In the wavelength range from 900 to 2100 nm, the transmissivity of the device is about 30% and remains almost independent with wavelength. According to the previous literature [22,23], the transmissivity of Cd_3As_2 can remain almost constant from 900 nm to 10 μm, which indicated that the device can work in the whole near- and mid-infrared spectral range. The broadband, constant transmissivity results from the gapless band structure and linear dispersion of Cd_3As_2, while the transmissivity approaches zero with a wavelength shorter than 900 nm because of the strong absorption of the GaAs substrate.

Then we characterized the nonlinear absorption parameters of the electrically contacted Cd_3As_2 SA device using the typical two-arm transmission measurement [24,25]. The incident light is provided by a mode-locked femtosecond fiber laser (Rainbow 1550, NPI Lasers Inc.) operating at a center wavelength of 1560 nm, with a repetition rate of 50 MHz. The pulse duration right before the sample is measured to be approximately 150 fs. The laser output is focused by a lens on the surface of the Cd_3As_2 film between the two gold electrodes. The pump fluence of the laser was tuned between 0.16 MW/cm^2 and 84 MW/cm^2 using a digital variable attenuator (OZ Optics Inc.) operating at 1560 nm. The reflected and transmitted light was measured with a pair of calibrated power meters. In the reflectivity measurements, the gold electrodes were used as the reference for absolute calibration of the reflectivity [10]. In order to alter the device temperature, different electric currents in the range of 0–50 mA are applied to the device via the two gold electrodes. The temperature of the Cd_3As_2 channel (area in between the electrodes) was measured using a high-precision infrared thermometer (875-1i, Testo Inc.), and the relationship between the temperature of the Cd_3As_2 device and the electric current is shown in Fig. 2. It would take approximately 2 min for the device to fully reach thermal equilibrium after ramping up the current and, once equilibrium is established, the temperature would be stabilized and locked to a specific current value, which indicates that the status of the device can be reliably addressed by the electric current. The device is found to be thermally damaged with a current of ~60 mA; therefore, we limit the current to 50 mA, and this corresponds to a temperature change from 297 to 336 K, a range of about 40 deg.

The dependence of the nonlinear absorption α on the pump fluence F_p at different values of temperature is shown in Fig. 3(a). At any fixed temperature, the absorption of the device is observed to drop with the increase of the pump fluence and reach full saturation at a relatively high pump fluence (~10 MW/cm^2), which is expected for SA behavior. With the increasing of the temperature of the electrically contacted Cd_3As_2 device, the absorption increases dramatically for small pump fluence (<1 MW/cm^2), while remaining almost constant for a high pump fluence. In Fig. 3(b), we illustrate the dependency of α on temperature by plotting the value of α for the weakest and the strongest pump fluences. At the weakest pump fluence, α clearly increases linearly with the temperature of the device (from 51.4% to 93.0%). At the strongest pump fluence, α remains nearly unchanged with a value around 17.8% (±0.5%), which means that the temperature does not influence the saturated absorption of the device dramatically. In order to rule out effects from the substrate, we also measured the absorption of the GaAs substrate using the same setup. The GaAs substrate shows no saturable absorption

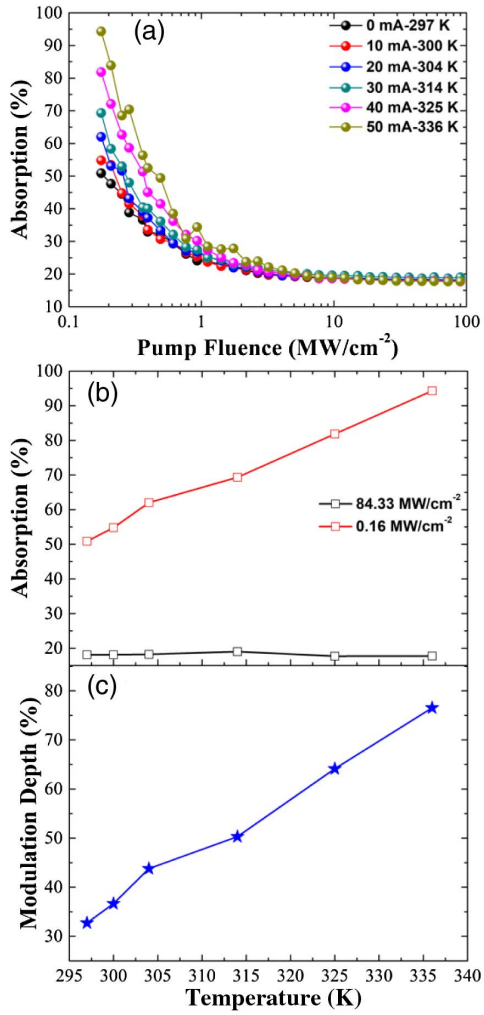

Fig. 3. (a) Absorption of the Cd_3As_2 SA device as a function of the pump fluence, under different values of temperature in the range of 297–336 K. (b) Absorption of the Cd_3As_2 SA device at pump fluences of 0.16 and 84.33 MW/cm2, as a function of temperature. (c) Modulation depth of the Cd_3As_2 SA device as a function of temperature.

behavior and shows a constant absorption around 4% at 1560 nm, indicating that the observed nonlinear response is from the Cd_3As_2 film. The dependency of the modulation depth, or $\Delta\alpha$, on temperature is shown in Fig. 3(c). Apparently, $\Delta\alpha$ is predominantly determined by the values of α at a small pump fluence. The absolute modulation depth is seen to change from 33.4% to 75.9%.

We further measured the nonlinear absorption curves while the device is in the cooling process. Immediately after the current is turned off, the saturable absorption exhibits almost the same nonlinear absorption curve as the current, which means that the presence of the electric current is not the physical origin for the observed change in the small-signal absorption. In several consecutive measurements taken 2–4 min after the current source is turned off, the saturable absorption behavior of the device is seen to gradually return to the initial state at room temperature. These measurements clearly prove that the tunability of the device characteristics is of a thermal nature.

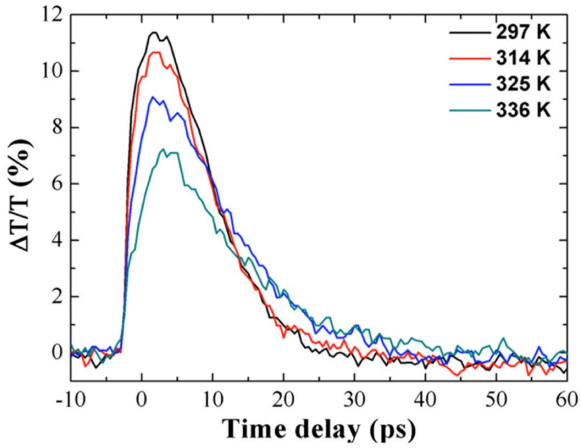

Fig. 4. Time-resolved transient transmission of Cd_3As_2 SA device under different values of temperature in the range of 297–336 K.

To reveal potential impact on the transient behavior of the device, the recovery times of the Cd_3As_2 SA device under different temperatures were also investigated with nondegenerate pump-probe spectroscopy. The samples were pumped at 800 nm and probed at 2 μm, and heated by applying the electric current. In Fig. 4(a), the dependency of transient transmission $\Delta T/T$ on time delays is shown for different values of temperature. It is clearly seen that the magnitude of the transient transmission signal decreases appreciably with increasing T. Such a signature means that for the high temperature regime, relatively fewer states are occupied for the upper states corresponding to the probe photon transitions. On the other hand, the recovery time is seen to only change slightly under different temperatures (in a range of 6.1–8.8 ps). This is desirable, as it suggests such a thermal approach can be used to selectively control only the modulation depth feature of the SA devices.

While elucidating the underlying mechanism for modulation depth tuning involves a detailed understanding of the selection rules, as well as the initial ultrafast thermalization dynamics for such an emerging Dirac system, we believe two factors may play a major role here. First, when the pump fluence is low, the relaxation of carriers is dominated by electron-phonon (*e-p*) scattering, as for the case of graphene [16,26]. With the temperature rising, the phonon number density will increase, leading to stronger e-p scattering, which contributes to faster emptying of electrons in the excited states. According to the report in Ref. [27], light absorption can be enhanced if the excited state can already be emptied during the pump pulse [27]. Secondly, transport experiments carried out on samples with similar thicknesses suggest that the carrier concentration of Cd_3As_2 film almost doubles, i.e., from 4.5×10^{17} to 7.5×10^{17} cm^{-3}, when the temperature increases from room temperature to 350 K. As the device is based on n-type Cd_3As_2, the Fermi level will be raised slightly with the increasing carrier concentration. With a higher Fermi level, the excited state will be closer to the Fermi level, therefore facilitating faster relaxation of excited electrons through electron-electron (*e-e*) scattering [16]. This is consistent with literature where faster relaxation of photocarriers is observed in graphene with an increasing Fermi level [27,28].

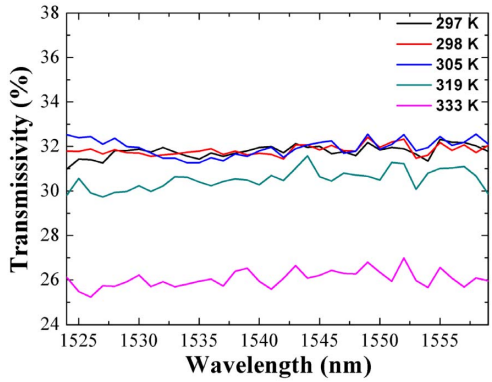

Fig. 5. Small-signal transmissivity spectra of the Cd_3As_2 SA device under different values of temperature in the range of 297–333 K.

The small-signal transmission spectrum was also experimentally verified with a home-built amplified spontaneous emission (ASE) light source, which emits across 1525 to 1560 nm. As shown in Fig. 5, the transmissivity of the device remains constant as a function of wavelength, which agrees with the linear dispersion of Cd_3As_2. Because the ASE source is a continuous-wave light with much wider spectrum, the transmissivity is expected to be qualitatively similar to the nonlinear absorption measurement (using femtosecond pulses) at a weaker pump fluence. Indeed, as the temperature is increased, the reduction of transmissivity was observed for all wavelengths of the spectra (power of ASE ~ 3 mW), and this again confirms that the changes of modulation depth are largely determined by the absorption at a weaker pump fluence. However, the reduction of transmissivity measured by ASE was not as strong as that measured by a femtosecond laser, which is consistent with the physical picture that the small-signal absorption change is fundamentally linked with a transient effect.

In conclusion, we have demonstrated a parameter tunable SA device based on electrically contacted Cd_3As_2 thin film deposited on a GaAs substrate. The modulation depth of the as-fabricated device can be tuned in a wide range from 33.4% to 75.9% with the current-induced temperature changing from 297 to 336 K. Such an unprecedented capability, achieved with a simple planar structure, is highly desirable to the investigation of mode-locking dynamics in a range of laser platforms. For example, it would be useful to employ such a device for actively controlling the laser pulsation regimes (mode-locked/*Q*-switched), as well as studying the dependence of various figures of merit (i.e., self-starting performance) on absorber modulation depth. We attribute the observed effects to the temperature-induced stronger *e-p* scattering and higher carrier concentration, which both contribute to the faster emptying of electrons in the excited states. Combined with the broadband feature of Dirac semimetal Cd_3As_2, we expect the proposed electrically contacted SA device to enable a range of new fundamental explorations of ultrafast lasers, especially those operating in the mid-infrared to terahertz.

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†These authors contributed equally to this Letter.

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