Stimulated Rayleigh scattering and slow light generation in a periodically driven Dirac semimetal Cd$_3$As$_2$

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Abstract

Periodic driving of solids by intense light field has recently attracted a great deal of interest for Floquet engineering. Despite significant advances in theory, a comprehensive understanding is still lacking due to limited experimental realization. Electromagnetic response functions in periodically driven materials particularly deserve elucidation, both for fundamental science and potential applications. Here we report on ultrafast multiterahertz pump-probe spectroscopy of a three-dimensional Dirac semimetal, cadmium arsenide. We show that stimulated Rayleigh scattering dominates the optical response under an intense driving field with the assistance of a longitudinal collective mode. The characteristic dispersive lineshape in the transient absorption spectrum is accounted for by optical transitions between light-induced Floquet subbands. We present a convenient model to describe the macroscopic behavior of this phenomenon. Appearance of stimulated Rayleigh scattering with an unprecedentedly large refractive index change may pave the way for slow light generation in conductive solids at room temperature.
Main text
Light has opened various ways to reach interesting nonequilibrium phases of matter, such as light-induced superconductivity\textsuperscript{1,2}, charge density wave\textsuperscript{3}, and excitonic insulator\textsuperscript{4}. The emerging field of Floquet engineering is accelerating new discoveries through the versatility of periodic driving to modify material properties\textsuperscript{5,6}. Examples include control of band topology\textsuperscript{7-10} and excitonic correlations\textsuperscript{11,12}. Floquet engineering is also interesting from the viewpoint of nonlinear optics. The concept of photon-dressed states, which underlies the Floquet picture, has provided an indispensable basis to understand the nonlinear optical response of discrete level systems\textsuperscript{13}. Modern interest in the Floquet formalism has also shed new light on nonlinear optics in solids, e.g., in terms of topology\textsuperscript{14}. It is thus natural to expect novel optical phenomena to emerge from light-induced Floquet states.

Despite remarkable progress in theory, experimental exploration of Floquet states is still limited. Time- and angle-resolved photoemission spectroscopy succeeded in directly observing electron population in photon-dressed Floquet-Bloch bands on a surface of a light-driven topological insulator\textsuperscript{15,16}. Ultrafast transport measurement has recently demonstrated that irradiation by circularly polarized light transforms graphene into a Floquet topological insulator\textsuperscript{7,17}, which partly contributes to anomalous Hall effect\textsuperscript{18}. Manifestations of the light-induced Floquet states in the optical response itself, however, remain unclear. Even the most fundamental properties of light absorption and refraction are to be elucidated. Cadmium arsenide (Cd\textsubscript{3}As\textsubscript{2}), a three-dimensional Dirac semimetal, is an ideal material for these investigations, because it combines high-mobility carriers, a small scattering rate, and low-energy interband transitions\textsuperscript{19}, which allow for coherent dynamics with suppressed dissipation and laser heating. These advantages facilitate an in-depth understanding of Floquet states toward dynamical control of matter. Moreover, Cd\textsubscript{3}As\textsubscript{2} exhibits large optical nonlinearity in a broad frequency region ranging from terahertz to visible\textsuperscript{20-25}, which makes it a promising platform to search for novel functionality in nonlinear optics and optoelectronics from the perspective of Floquet engineering.

To explore the spectroscopic signature and optical functionality of Floquet states in solids, we conduct degenerate pump-probe experiments on Cd\textsubscript{3}As\textsubscript{2} in the multiterahertz frequency region. We observe an unexpected dispersive absorption change at the pump frequency when the pump and probe pulses temporally overlap, even reaching a net optical gain. Stimulated Rayleigh scattering (SRLS) accounts for this phenomenon on a
macroscopic level, which arises from a large reduction of the refractive index due to a light-induced blueshift of a longitudinal collective mode, the plasma oscillation. From a microscopic point of view, the dispersive absorption change can be further traced back to optical transitions between the Floquet subbands allowed by a diamagnetic coupling in the Hamiltonian. These findings provide a breakthrough in both experimental and theoretical study of light-driven materials by unveiling characteristic optical response of Floquet states in solids. Appearance of SRLS also suggests possible applications to lossless slow light generation in semimetals at room temperature.

When light irradiates a semimetal or a semiconductor, it is expected that valence and conduction bands are mixed to form Floquet states, as schematically shown in Fig. 1a. To examine optical properties of such photon-dressed states, we measure transient optical conductivity of an epitaxially grown, (112)-oriented, 140 nm-thick Cd$_3$As$_2$ thin film on a CdTe substrate (see Methods section), exposed to an intense multiterahertz electromagnetic pulse. Figure 1b depicts the experimental setup. As shown in Fig. 1c, Cd$_3$As$_2$ possesses two Dirac nodes on the $k_z$ axis, which are protected by four-fold rotational symmetry$^{19,26}$. Our sample is unintentionally electron-doped so that the Fermi level is shifted to 58 meV above the Dirac nodes$^{25}$. Despite the anisotropy in the low-energy band structure, the linear response in the infrared region is almost isotropic because of the quasi-cubic nature of the structural units that make up the unit cell$^{27,28}$. Figure 1d shows the optical conductivity of the sample in equilibrium, which consists of low-frequency (<15 THz) intraband and high-frequency (>15 THz) interband contributions$^{25,29}$. The narrowband pump pulse drives the latter transitions with a tunable frequency from 16 to 40 THz (66-165 meV in energy, 8-19 μm in wavelength) and with a variable bandwidth, while the probe pulse covers a broad frequency range from 12 to 45 THz (50-186 meV, 7-25 μm) with a duration of 30 fs (see Methods section). Typical waveforms and power spectra are shown in Figs. 1b and 1e, respectively, where the pump pulse is tuned to 29.4 THz (122 meV, 10.2 μm) with a pulse width of 180 fs. The probe pulse after transmitting the sample is spatially separated from the pump pulse and is detected by electro-optic sampling to obtain response functions depending on the pump-probe delay time $\Delta t$ (see Methods section).

Figure 2a shows the transient optical conductivity measured by probe pulses polarized in the same direction as the pump, whose fluence is 0.4 mJ/cm$^2$. During the pump irradiation, an induced absorption (blue) appears just below the pump frequency, while an opposite change (red) occurs on the higher-frequency side. The resulting dispersive lineshape is
clearly seen in Fig. 2b, which plots the optical conductivity at several delay times. This characteristic behavior is distinct both from spectral hole burning and from photon-assisted absorption bands, the two scenarios that have been theoretically considered so far. Note that a net optical gain ($\sigma_1 < 0$) develops from the suppressed absorption at around the maximum pump-probe overlap ($\Delta t \approx 0$ ps). The dispersive structure lasts until the pump pulse leaves the sample and vanishes thereafter, as visualized in Fig. 2c, where the peak and dip values of the conductivity closely follow the pump pulse intensity. We found that no dispersive signal appears when the probe is polarized perpendicularly to the pump (see Supplementary Section S1), implying a coherent nature of the involved processes. To get more insight into this characteristic response, we examined its dependence on the pump intensity. Throughout the studied range, positions of the peak and the dip are almost insensitive to the pump fluence, as shown in Fig. 2d. Following this observation, we plot the fluence dependence of the peak and dip values in Fig. 2e (dots) along with the equilibrium values at 28.2 and 31.3 THz (open circles). In the weak excitation limit ($<0.1$ mJ/cm$^2$), both the peak and the dip develop linearly with the pump fluence, indicating a perturbative origin of the signal. As the pump gets stronger ($>0.1$ mJ/cm$^2$), both features start to saturate. The dip value eventually turns upward against fluence, due to a large background of induced absorption by photoexcited carriers. Figure 2f also verifies that the position of the dispersive structure follows the centre frequency of the pump, excluding the possibility that the signal could arise from some special points in the band structure or specific phonon modes.

In semiconductors, it is known that a dispersive absorption change appears in the early stage of photoexcitation as a result of excitonic effect. This phenomenon can potentially be regarded as a precursor to a light-induced excitonic insulator, where the dispersive structure evolves into strong induced absorption and optical gain. In these cases, however, the absorption peak lies on the higher energy side of the pump photon energy, which is opposite to the behavior observed in Fig. 2. Thus, excitonic effects are of minor importance in Cd$_3$As$_2$, consistent with recent predictions.

From a phenomenological point of view, the dispersive absorption change in Cd$_3$As$_2$ can be understood in terms of SRLS. Suppose that application of the optical field primarily changes the real part of the refractive index. If the pump and probe beams spatially overlap, their interference creates a transient grating and diffracts the pump beam into two directions, one being a new direction often studied in four-wave mixing (FWM) experiments and the other being the propagation direction of the probe, as shown in Fig.
3c. The latter effect suppresses or enhances transmission of the probe beam depending on the phase of the diffracted wave. In case of a negative refractive index change, this process results in an induced absorption (emission) for a probe frequency slightly lower (higher) than the pump, as seen in Fig. 3a. This mechanism, known as two-beam coupling\textsuperscript{13}, accounts for our experimental results in Fig. 2, because interband excitation actually reduces the refractive index through a blueshift of the screened plasma frequency, initially located at 10 THz\textsuperscript{25}. Dynamical narrowing of the separation between the peak and the dip, observed in Figs. 2a and 2b for increasing $\Delta t$, can also be reproduced within this framework, as shown by Figs. 3a and 3b. This behavior can be explained by the detection scheme in our experiment, discussed in Supplementary Section S2. We classify this phenomenon into SRLS as it originates from a kind of density fluctuations, namely, those of the charge carriers. While conventional SRLS is usually weak because it arises from relatively small density fluctuations of the lattice, the much larger change in the carrier density enables unprecedentedly strong SRLS in semimetals and probably also in narrow-gap semiconductors, as discussed later.

Even though the above macroscopic model gives a satisfactory phenomenological description of SRLS, light absorption should be microscopically treated as transitions between quantum levels. We next consider the quantum mechanical aspect of this phenomenon and discuss its connection to the Floquet states. In Fig. 3d, we plot the transient optical conductivity calculated by an effective two-band model for the low-energy band structure (see Methods section). One can clearly recognize the dispersive lineshape. Knowledge of two-level systems helps us to interpret this result using a level diagram. In two-level systems, the well-known ac Stark effect is accompanied by a dispersive structure at the pump frequency, also called SRLS\textsuperscript{13} (Fig. S4a in Supplementary Section S4). It originates from transitions between dressed states in resonance with the driving field (Fig. S4b). Extending this understanding to continuous bands, SRLS in Cd$_3$As$_2$ is attributed to transitions between the Floquet subbands resonant to the pump frequency, as schematically shown in Fig. 3f. Thus, SRLS exemplifies Floquet engineering of a nonlinear optical property in solids. A closer look at its origin, however, reveals that the light-matter interaction responsible for it is different from that in two-level systems. In the latter, a usual coupling between an electric dipole moment and the electric field, also called paramagnetic coupling, induces relatively weak SRLS (see Fig. S4a), with a sign depending on detuning. As a result, SRLS in two-level systems tends to be cancelled out when integrated over continuous bands, leaving a spectral hole stemming from the ac Stark effect. This consequence can be seen in the blue curve in Fig.
which plots the contribution from the paramagnetic coupling only. The dispersive structure in the total optical conductivity arises from a second-order or diamagnetic coupling with the electric field, which gives the red curve in Fig. 3e. This coupling induces a light-induced shift of the screened plasma frequency, so that the microscopic theory actually supports the phenomenological picture presented above. One can also derive the macroscopic model by focusing on the diamagnetic current (see Supplementary Section S5). The derivation tells us that an intermediate frequency between intraband and interband transitions is preferable, because SRLS in this case requires combination of injection and acceleration of photocarriers. Since the above discussion does not rely on details of the band structure, SRLS is expected to occur in general semimetals and narrow-gap semiconductors with low-energy interband transitions, independently of the excitonic effects considered before.

Finally, from a perspective of Floquet engineering of optical functionality, we discuss the possibility of slow light generation in Cd$_3$As$_2$. As the theory of SRLS predicts, the dispersive structure in transient optical conductivity can be narrowed by reducing the pump bandwidth, as shown in Fig. 4a. Such a narrow structure in absorption is necessarily accompanied with a rapid variation in the refractive index $n$ with frequency $f$, so that the group refractive index $n_g = n + f (dn/df)$ may become large. The resultant slowing down of an optical wave packet is known as slow light generation$^{13,37,38}$. In the present case, a narrow dip develops in the refractive index at, for example, $\Delta t = -0.48$ ps (top panel in Fig. 4b), leading to a group refractive index as large as 40 at 30 THz (bottom panel in Fig. 4b). This corresponds to 40 times deceleration of a wave packet without dissipation, because the extinction coefficient $\kappa$ is not positive there (middle panel in Fig. 4b). Note that $n_g$ less than 1 should not be taken literally since the field decay caused by positive $\kappa$ must be taken into account for those frequencies. An even more interesting situation occurs when a metallic screening ($\epsilon_1 < 0$) by photoexcited carriers coexists with an optical gain ($\epsilon_2 < 0$), where $\epsilon_1$ and $\epsilon_2$ stand for the real and imaginary parts of the dielectric constant. The refractive index $n = \left(\sqrt{\epsilon_1^2 + \epsilon_2^2} + \epsilon_1\right)/2$ then vanishes at the crossover point between absorption and gain ($\epsilon_2 = 0$), which may further enhance the rapid spectral variation in $n$ (top panel in Fig. 4c). The group index correspondingly exceeded 300 at $\Delta t = -0.24$ ps (bottom panel in Fig. 4d), where a metallic screening ($\epsilon_1 < 0$) developed with the help of the SRLS itself. Remarkably,
the extinction coefficient $\kappa = (\text{sgn}\ \epsilon_2) \left[ \left( \sqrt{\epsilon_1^2 + \epsilon_2^2} - \epsilon_1 \right) / 2 \right]^{1/2}$ remains negative in this gain region (middle panel in Fig. 4c), so that a probe wave does not decay in spite of the metallic character in $\epsilon_1$. Thus an electromagnetic pulse might be slowed down more than 300 times without loss under the present experimental condition.

Most previous studies of slow light generation have used electromagnetically induced transparency at low temperature\(^{37}\) and photonic-band engineering\(^{38}\) as the origin of a refractive index change $\Delta n$, which typically amounts to \(\sim 0.01\) and \(\sim 0.1\), respectively. In our case, by contrast, $\Delta n > 1$ is so large that $n$ even vanishes. A relatively large bandwidth $\Delta f \sim 0.5$ THz of the dispersion limits the achievable group refractive index here. This is not necessarily a disadvantage, because a broader dispersion allows a shorter pulse to be slowed down. In fact, photonic-band engineering emerged as a way to generate slow light with a broad bandwidth (~THz)\(^{38}\), compared to a much narrower one (~kHz) achieved by electromagnetically induced transparency. Our results show that lossless and broadband slow light generation is possible by simply shedding infrared light to a single-crystalline semimetal at room temperature. To avoid complication by transient effects, such as the temporal change from Figs. 4b to 4c, continuous-wave or nanosecond CO\(_2\) lasers promise better choice as the pump light source operating at around 30 THz or 10 μm, though optical heating should be suppressed by efficient cooling. The available bandwidth $\Delta f \sim 1/T_1 = 0.13$ THz is then determined by the lifetime of excited carriers ($T_1 = 8$ ps), still keeping a relatively large value. We leave the implementation of slow light generation with this mechanism as a topic of future studies.

In summary, we performed ultrafast pump-probe spectroscopy on a Cd\(_3\)As\(_2\) thin film in the multiterahertz frequency region, to find stimulated Rayleigh scattering to dominate the transient absorption spectrum in the pump-probe overlap. Macroscopically, it originates from a transient grating with a reduced refractive index, formed by photoexcited carriers in the interfering pump and probe fields. The characteristic dispersive lineshape can be further traced back to microscopic optical transitions between the light-dressed electronic bands, the Floquet states, assisted by a diamagnetic coupling with the optical field. The concomitant sharp dispersion in the transient refractive index may be applicable to semimetal-based, lossless, broadband slow light generation at room temperature. From a perspective of Floquet engineering of material states, the application of circularly polarized driving fields promises an interesting future direction, because of its ability to manipulate band topology and magnetic symmetry\(^{8-10,39}\). Berry curvature
induced by the emergent Weyl points may be detectable through the static or optical Hall conductivity \(^7,,17,30,40\). Light-induced excitonic ordering in Dirac semimetals is another fascinating direction \(^41\), though different substances seem to be better for stronger excitonic effect \(^36\). For application, continuous-wave or nanosecond CO\(_2\) lasers may enhance the group refractive index furthermore to facilitate slow light generation, keeping a relatively broad bandwidth (~0.1 THz).

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### Author contributions

R.M. conceived this project. M.G. and S.S. fabricated the sample. N.K. and T.M. evaluated the linear response function. Y.M. and N.K. developed the pump-probe spectroscopy system with the help of J.Y., Y.K., and R.M. N.K. performed the experiment and analyzed the data. Y.M. conducted the phenomenological analysis. Y.M. and T.N.I. performed the microscopic calculations. All the authors discussed the results. Y.M. wrote the manuscript with the substantial help of N.K., T.N.I, and R.M., with the feedbacks from all the coauthors.

### Competing interests

The authors declare no competing interests.
Methods

Sample Preparation. A (112)-oriented Cd$_3$As$_2$ film was grown by molecular beam epitaxy on a (111)B CdTe substrate with a 3° miscut, as detailed elsewhere$^{42}$. The film thickness was 140-150 nm as determined by the growth time. At room temperature, the Hall mobility of the film was $\sim$10,000 cm$^2$/Vs. The linear optical conductivity was measured by terahertz time-domain spectroscopy and Fourier-transform infrared spectroscopy and was fitted by a model function$^{25}$.

Pump-probe spectroscopy. We used a Yb:KGW-based regenerative amplifier (centre frequency 1030 nm, repetition rate 3 kHz, pulse energy 2 mJ, and pulse width 255 fs) as a light source. 80% of the output (1.6 mJ) was used to pump and seed two optical parametric amplifiers (OPAs) emitting signal beams with different colours (1350-1650 nm). Narrowband multiterahertz pump pulses were subsequently generated in a 500 μm-thick GaSe crystal through difference frequency generation$^{43}$. Inserting grating pair stretchers after the OPAs enabled us to vary the pump pulse width$^{44}$ in the range of 180-880 fs, keeping a transform-limited waveform and a large pulse energy of $\sim$1 μJ. The remaining part of the light source (0.4 mJ) was compressed to 14 fs with a throughput of 70 μJ, by a two-stage multi-plate broadening scheme$^{45,46}$. The compressed pulses were further divided into two beams. One irradiated a 30 μm-thick GaSe crystal to generate broadband multiterahertz probe pulses through intra-pulse difference frequency generation. Their waveforms after transmitting the sample were measured by electro-optic sampling in a few-μm-thick GaSe flake on a diamond substrate, using the other beam as gate pulses$^{25}$. The pump and probe beams were focused on the sample with spot sizes of $>125$ μm and 46 μm, respectively, with a relative angle of incidence, 20°. Penetration depth at the pump frequencies exceeded 3 μm, so that the film was uniformly excited in the probed region. In pump-probe measurements, the time difference between the pump and gate pulses was fixed and defined as the pump-probe delay time $\Delta t$, while the arrival time of the probe pulse was scanned to obtain electric field waveforms and transient response functions$^{47}$. We confirmed that another analysis method, i.e., fixing the time difference between the pump and the probe, and scanning the arrival time of the gate, returned qualitatively the same results including appearance of optical gain (see Supplementary Section S3).

Phenomenological model. For the phenomenological description of SRLS, we used a two-beam coupling framework$^{13}$. Suppose that local optical intensity $I(\mathbf{r}, t)$ proportionally induces the refractive index change $n_{\text{NL}}(\mathbf{r}, t)$, which decays with a time
constant of $\tau$. Spatiotemporal variation of $n_{\text{NL}}$ is then determined by

$$\tau \frac{\partial n_{\text{NL}}}{\partial t} + n_{\text{NL}} = n_2 I,$$

where $n_2$ is a material-dependent constant. As discussed in the main text, $n_2$ is negative in the present case because photoexcited carriers reduce the refractive index around 30 THz with a lifetime of $\tau = 8$ ps due to a blueshift of the plasma frequency from 10 THz$^{25}$. The nonlinear polarization is given by

$$P_{\text{NL}} = 2n_0\varepsilon_0 n_{\text{NL}} E,$$

where $n_0$ is the refractive index at equilibrium, $\varepsilon_0$ the vacuum permittivity, and $E$ the electric field in the film. Now the pump and probe fields are introduced via

$$I(r, t) = n_0\varepsilon_0 c E(r, t)^2 = 2n_0\varepsilon_0 c \left[ |E_0(t)|^2 + |E_1(t)|^2 + [E_1(t) \cdot E_0^*(t)e^{i(k_1-k_0) \cdot r} + \text{c.c.}] \right],$$

where $c$ indicates the speed of light and the overline a time average over several periods. The resulting nonlinear polarization contains a component that propagates in the same direction as the probe field,

$$P_{\text{NL}}^1(t) = 2n_0\varepsilon_0 [n_{\text{NL}}^{00}(t)E_1(t) + n_{\text{NL}}^{10}(t)E_0(t)],$$

where $n_{\text{NL}}^{00}$ and $n_{\text{NL}}^{10}$ obey

$$\tau \frac{dn_{\text{NL}}^{00}}{dt} + n_{\text{NL}}^{00} = 2n_0n_2\varepsilon_0 c|E_0(t)|^2,$$

$$\tau \frac{dn_{\text{NL}}^{10}}{dt} + n_{\text{NL}}^{10} = 2n_0n_2\varepsilon_0 c E_1(t) \cdot E_0^*(t).$$

Among these, $n_{\text{NL}}^{10}$ is induced by spatial interference of the pump and probe fields and diffracts the pump beam into the propagation direction of the probe, which is nothing but SRLS. The resulting change in the effective susceptibility $\Delta\chi$ experienced by the probe pulse is obtained by Fourier transforming $P_{\text{NL}}^1(t)$ against the probe delay time and dividing it by the Fourier component of $E_1(t)$. Change in the optical conductivity is calculated from the relation $\Delta\sigma_1 = \omega\varepsilon_0 \text{Im} \Delta\chi$. Figures 3a and 3b show the simulated results. This effect disappears for orthogonally polarized pump and probe fields due to an implicit assumption of optical isotropy, which is reasonable for the pseudo-hexagonal (112) plane of Cd$_3$As$_2$.

**Microscopic theory.** For microscopic analysis, we adopted an effective two-band model applicable to the low-energy band structure. The Hamiltonian reads
\[ H_{\text{eff}}(\mathbf{k}) = \epsilon_0(\mathbf{k}) + \begin{pmatrix} \epsilon_0(\mathbf{k}) & D(\mathbf{k}) & 0 & 0 \\ D^*(\mathbf{k}) & - \epsilon_0(\mathbf{k}) & 0 & 0 \\ 0 & 0 & \epsilon_0(\mathbf{k}) & -D^*(\mathbf{k}) \\ 0 & 0 & 0 & -D(\mathbf{k}) \end{pmatrix}, \]

where \( \epsilon_0(\mathbf{k}) = C_0 + C_1 k_x^2 + C_2 (k_x^2 + k_y^2) + C_3 k_x^2 + C_4 (k_x^2 + k_y^2)^2 + C_5 (k_x^2 + k_y^2)k_x^2 + C_6 k_x^2 k_y^2, \ M(\mathbf{k}) = M_0 + M_1 k_x^2 + M_2 (k_x^2 + k_y^2) + M_3 k_x^4 + M_4 (k_x^2 + k_y^2)k_x^2 + M_5 (k_x^2 + k_y^2)^2 + M_6 k_x^2 k_y^2, \) and \( D(\mathbf{k}) = (D_0 + D_1 k_x^2 + D_2 k_y^2 + D_3 k_z^2)k_x - i(D_0 + D_1 k_x^2 + D_2 k_x^2 + D_3 k_z^2)k_y. \) Here, we have extended the second-order model in the literature\textsuperscript{26} to the fourth order in \( \mathbf{k}, \) because the second-order model underestimates the interband transition dipole moment at multiterahertz frequencies. We fitted the dispersion relation predicted by a second-order eight-band Kane model\textsuperscript{26} by the above model, to obtain \( C_0 = -0.0107 \text{ eV}, \ C_1 = 11.84 \text{ eV} \cdot \text{Å}^2, \ C_2 = 17.66 \text{ eV} \cdot \text{Å}^2, \ C_3 = 592.7 \text{ eV} \cdot \text{Å}^4, \ C_4 = -413.9 \text{ eV} \cdot \text{Å}^4, \ C_5 = -640.5 \text{ eV} \cdot \text{Å}^4, \ C_6 = -680.1 \text{ eV} \cdot \text{Å}^4, \ M_0 = -0.0215 \text{ eV}, \ M_1 = 19.80 \text{ eV} \cdot \text{Å}^2, \ M_2 = 21.59 \text{ eV} \cdot \text{Å}^2, \ M_3 = 592.7 \text{ eV} \cdot \text{Å}^4, \ M_4 = -672.0 \text{ eV} \cdot \text{Å}^4, \ M_5 = 358.2 \text{ eV} \cdot \text{Å}^4, \ M_6 = -203.9 \text{ eV} \cdot \text{Å}^4, \ D_0 = 1.137 \text{ eV} \cdot \text{Å}, \ D_1 = -133.8 \text{ eV} \cdot \text{Å}^3, \ D_2 = 69.3 \text{ eV} \cdot \text{Å}^3, \) and \( D_3 = -125.7 \text{ eV} \cdot \text{Å}^3. \) The optical field is minimally coupled to the system through Peierls substitution \( \mathbf{k} \rightarrow \mathbf{k} + e\mathbf{A}/\hbar, \) where \( e(>0) \) denotes the elementary charge and \( \mathbf{A} \) the vector potential. To minimize computational efforts, we restricted ourselves to the optical field polarized in the \( z \) direction, i.e., \( \mathbf{A} = (0, 0, A). \) In addition, we assumed the system to be isotropic in the \( xy \) plane, by replacing \( C_6 = 2C_4, M_6 = 2M_4, \) and \( D_3 = D_2. \) We believe that this simplification does not matter significantly, because Cd\textsubscript{3}As\textsubscript{2} is almost optically isotropic\textsuperscript{27,28}. After diagonalization, one obtains an effective \( 2 \times 2 \) Hamiltonian

\[ H(\mathbf{k}) = \mathcal{E}(\mathbf{k}) - J^z(\mathbf{k})A + \frac{K^{zz}(\mathbf{k})}{2} A^2 + \ldots, \]

with two-fold degeneracy. The field-free part is given by

\[ \mathcal{E}(\mathbf{k}) = \begin{pmatrix} \epsilon_1(\mathbf{k}) & 0 \\ 0 & \epsilon_2(\mathbf{k}) \end{pmatrix}, \]

where \( \epsilon_{1,2}(\mathbf{k}) = \epsilon_0(\mathbf{k}) \pm \sqrt{M(\mathbf{k})^2 + |D(\mathbf{k})|^2}. \) The paramagnetic current operator \( J^z(\mathbf{k}) \) is responsible for optical transitions in the linear response. The second-order coupling constant \( K^{zz}(\mathbf{k}) \) arises from the parabolicity or the inverse effective mass of energy bands; more specifically, \( \mathbf{k} \cdot \mathbf{p} \) perturbation theory tells us that

\[ K^{zz}_{11(22)}(\mathbf{k}) = \frac{e^2}{\hbar^2} \frac{\partial^2 \epsilon_{1(2)}(\mathbf{k})}{\partial k_z^2} - \frac{2|J_{1(2)}(\mathbf{k})|^2}{\epsilon_{1(2)}(\mathbf{k}) - \epsilon_{2(1)}(\mathbf{k})}. \]
where the first term on the right-hand side is proportional to the inverse effective mass. This second-order coupling gives rise to the diamagnetic contribution to the total current operator,

\[ J^z_{\text{tot}}(k) = -\frac{\partial H(k)}{\partial A} = J^z(k) - K^{zz}(k)A + \cdots. \]

Upon this two-band basis, we let the electron population \( n_{1,2}(k) \) and the interband polarization \( P(k) \) follow

\[
\frac{\partial n_1(k)}{\partial t} = -\frac{2}{\hbar} \text{Im}[H_{21}(k)P(k)] - \frac{n_1(k) - f_1(k)}{T_1'} - \frac{n_1(k) - f_1^\text{eq}(k)}{T_1},
\]

\[
\frac{\partial n_2(k)}{\partial t} = +\frac{2}{\hbar} \text{Im}[H_{21}(k)P(k)] - \frac{n_2(k) - f_2(k)}{T_2'} - \frac{n_2(k) - f_2^\text{eq}(k)}{T_1},
\]

\[
\imath \hbar \frac{\partial P(k)}{\partial t} = [H_{11}(k) - H_{22}(k)]P(k) - H_{12}(k)[n_1(k) - n_2(k)] - \imath \hbar \frac{P(k)}{T_2}.
\]

The terms that contain \( T_1, T_1', \) and \( T_2 \) describe scattering effects within a relaxation time approximation. Electron-electron scattering redistributes an initially nonthermal electron population into a time-dependent Fermi distribution function \( f_{1,2}(k) \), within a time interval of \( T_1' \). The targeted chemical potential and electron temperature are determined from the internal energy of electrons,

\[ U = \sum_k [\epsilon_1(k)n_1(k) + \epsilon_2(k)n_2(k)], \]

where the degeneracy factor of 2 is omitted for simplicity. Electron-phonon scattering subsequently equilibrates the electron and lattice subsystems within a time of \( T_1 \), after which electrons settle in a static distribution function \( f_{1,2}^\text{eq}(k) \). Scattering events also destroy the interband polarization with a time constant of \( T_2 \). We note that the above modeling of scattering terms breaks the gauge invariance. For example, adding a constant term in \( A \) may change the final results. Gauge-invariant formulation of the relaxation terms is possible, e.g., on the basis of instantaneous eigenstates, though we do not aim at such elaboration. Current density can be expressed as a sum of paramagnetic, diamagnetic, and higher-order terms,

\[ j^z = j^z_{\text{para}} + j^z_{\text{dia}} + \cdots, \]

where

\[
j^z_{\text{para}} = \sum_k [J^z_{21}(k)P(k) + J^z_{12}(k)P^*(k) + J^z_{11}(k)n_1(k) + J^z_{22}(k)n_2(k)],
\]

\[
j^z_{\text{dia}} = -\sum_k [K^z_{21}(k)P(k) + K^z_{12}(k)P^*(k) + K^z_{11}(k)n_1(k) + K^z_{22}(k)n_2(k)]A.
\]

Pump and probe fields were introduced through \( A = A_0 + A_1 \), where the subscripts 0 and
1 denote pump and probe, respectively. To remove the contribution from four-wave mixing, we averaged the probe-induced current density over the phase of the pump waveform\textsuperscript{48}. The resulting current density was Fourier-transformed with respect to the real time $t$, and divided by the Fourier transform of the probe electric field $E_1 = -\partial A_1 / \partial t$, to obtain transient optical conductivity. This procedure differs from the experimental setup in the main text, where the probe delay time (not the real time) was scanned to perform Fourier transform. However, as shown in Supplementary Section S3, Fourier transform with respect to the gate delay time in the experiment (corresponding to the real time in theory) led to essentially the same results. Therefore, we concentrated on Fourier transform with respect to the real time, which is numerically more feasible. In the simulation, we used $T_1' = 500$ fs and $T_2 = 20$ fs, and neglected the $T_1$ relaxation terms because $T_1 = 8$ ps far exceeds the time scale of interest. Further details of the calculation are presented in Supplementary Section S5. Figures 3d and 3e shows the simulated results. The phenomenological model presented above can be derived from this microscopic framework with some simplifications, as detailed in Supplementary Section S6.

Data availability

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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**Figures and figure captions**

**Fig. 1** | Periodic driving of Cd₃As₂ by light.  
**a**, Schematic picture of Floquet state formation in solids.  
**b**, Setup of the pump-probe experiment.  
**c**, Band structure of Cd₃As₂ around the Γ point.  
**d**, Optical conductivity of the sample. The model fitting (dotted line) takes into account the lower-frequency data outside the panel.  
**e**, Power spectra of the narrowband pump pulse (magenta) and the broadband probe pulse (cyan).
Fig. 2|Pump-probe spectroscopy for transient optical conductivity of Cd$_3$As$_2$. a, Change of the optical conductivity as a function of frequency (horizontal axis) and pump-probe delay time $\Delta t$ (vertical axis). Waveform of the pump pulse is shown on the left. The equilibrium optical conductivity is plotted on the bottom along with the pump power spectrum. Pump and probe pulses are collinearly polarized. b, Transient optical conductivity at several delay times. The equilibrium spectrum is shown as a dotted line. c, Delay time dependence of the peak and dip values extracted from a. Temporal profile of the pump intensity is shown as the shaded curve. d, Positions of the peak and the dip in optical conductivity at $\Delta t = 0.04$ ps, as a function of pump fluence. e, A similar plot for the conductivity values at the peak and the dip. Equilibrium values at 28.2 and 31.3 THz are added as open circles. f, Optical conductivity at $\Delta t = 0.04$ ps for different pump frequencies, i.e., 29.4 THz (the same as in a-e) and 18.2 THz (with a fluence of 0.25 mJ/cm$^2$, a peak electric field of 0.9 MV/cm).
Fig. 3 | Theoretical description of stimulated Rayleigh scattering. a, Change of the optical conductivity $\Delta \sigma_1$ calculated by a phenomenological model. Theoretical details are given in Method. b, Two-dimensional plot of $\Delta \sigma_1$ as a function of frequency (horizontal axis) and pump-probe delay time (vertical axis). c, Geometric picture of stimulated Rayleigh scattering (SRLS) and four-wave mixing (FWM). $k_0$ and $k_1$ denote wavevectors of the pump and the probe, respectively. d, Transient optical conductivity calculated by a microscopic model. Theoretical details are given in Method. e, Contributions from the paramagnetic (blue) and diamagnetic (red) currents in the total optical conductivity (black) at $\Delta t = -0.12$ ps. f, SRLS induced by Floquet states in continuous bands.
Fig. 4| **Slow light generation.** **a,** Transient optical conductivity for broader (thin) and narrower (thick) pump pulses. Pump power spectra are plotted on the bottom with their FWHM indicated in the figure. The broader pump is the same as in Fig. 2a, while the narrower one has a pulse width of 0.88 ps, a fluence of 1.7 mJ/cm$^2$, and a peak electric field of 1.2 MV/cm. **b,** Refractive index $n$ (top), extinction coefficient $\kappa$ (middle), and group refractive index $n_g$ (bottom) measured at $\Delta t = -0.48$ ps for the narrower pump in **a**. Equilibrium spectra are shown as dotted lines. **c,** The same data set for $\Delta t = -0.24$ ps.
Supplementary Information for

Stimulated Rayleigh scattering and slow light generation in a periodically driven Dirac semimetal Cd$_3$As$_2$

Yuta Murotani, Natsuki Kanda, Tatsuhiko N. Ikeda, Takuya Matsuda, Manik Goyal, Jun Yoshinobu, Yohei Kobayashi, Susanne Stemmer, and Ryusuke Matsunaga

S1. Probe polarization dependence

Figure S1 compares the transient optical conductivity measured by probe pulses with a polarization direction parallel (a, $\mathbf{E}_{\text{pump}} \parallel \mathbf{E}_{\text{probe}}$) and perpendicular (b, $\mathbf{E}_{\text{pump}} \perp \mathbf{E}_{\text{probe}}$) to the pump. In stark contrast to S1a featured by a dispersive structure, only broad induced absorption appears in S1b, with a spectral weight gradually shifting to the low-frequency side to develop a Drude-like response. Similar spectra were also observed in the case of near-infrared pump$^{S1}$. This behavior is attributed to real excitation of carriers which cause induced intraband and/or interband absorption. Absence of the dispersive structure is consistent with the interpretation of stimulated Rayleigh scattering (SRLS), since $\mathbf{E}_{\text{pump}}$ and $\mathbf{E}_{\text{probe}}$ perpendicular to each other drive

![Fig. S1 | Probe polarization dependence. a,b. Transient optical conductivity measured by probe pulses with a polarization direction parallel and perpendicular to the pump, respectively. Data in a is the same as that in Fig. 2b in the main text. The dotted lines are the equilibrium spectrum. c, Distribution of carriers excited by the narrowband pump (left) and by the broadband probe (right). An ideal massless Dirac dispersion relation is assumed for simplicity, with the Dirac node located at the centre. Polarization direction of each pulse is indicated by arrows.](image-url)
interband transitions at different regions in the momentum space, as schematically shown in Fig. S1c. Here, distribution of carriers excited by the pump (left) and the probe (right) is calculated for an ideal Dirac dispersion relation, neglecting all scattering processes. A horizontally polarized pulse (the pump in the figure) creates carrier distribution weighted vertically around Dirac nodes, and vice versa (for the probe).

**S2. Temporal change in the peak-to-dip separation**

Figures 2a and 2b in the main text exhibit a narrowing down of the separation between the peak and the dip for increasing pump-probe delay time. This behavior is more clearly seen in Fig. S2a, which plots the temporal variation of the frequencies at the peak and the dip. As already discussed in the main text, the macroscopic model of SRLS reasonably reproduces the experimental result (Fig. S2b).

This narrowing down of the peak-to-dip separation in time is attributed to the measurement scheme of “probe scan” in our experiment. As sketched in Fig. S2c, the time difference between the pump and gate pulses is fixed and defined as the “pump-probe” delay time $\Delta t$, and the arrival time of the probe pulse is scanned to obtain a waveform. In this configuration, causality allows
only a part of the pump pulse before the arrival of the gate (indicated by a one-way arrow in Fig. S2c) to be scattered and detected. As a result, an effective bandwidth of the pump pulse decreases as $\Delta t$ increases, leading to a narrowing down of the dispersive lineshape.

**S3. Probe scan vs. gate scan**

One can also adopt another measurement scheme: time difference between the pump and the probe is fixed and the arrival time of the gate is scanned (Fig. S3a). This “gate scan” procedure is a natural extension of conventional pump-probe spectroscopy with a spectrometer, though time resolution is worse than the probe scan in time-domain spectroscopy. Even in this configuration, we observe a dispersive lineshape in the transient optical conductivity (Fig. S3b), reaching an
optical gain at the largest change. Here, however, the separation between the peak and the dip tends to widen as $\Delta t$ increases (Fig. S3c), unlike the probe scan. This behavior is indeed natural, because only a part of the pump pulse after irradiation by the probe (indicated by a one-way arrow in Fig. S3a) can be scattered by the transient grating. The macroscopic model of SRLS again reproduces this trend, when the gate scan is simulated (Fig. S3d and e).

**S4. Stimulated Rayleigh scattering in two-level systems**

Figure S4a shows a typical absorption spectrum of a two-level system irradiated by a monochromatic light wave. SRLS accompanies the well-known ac Stark effect (AC). Optical gain appearing below the pump frequency ($\omega_{\text{pump}}$) is the three-photon resonance, arising from a higher-order nonlinearity. Energy levels of the original and dressed states, and the related optical transitions, are shown in Fig. S4b.

**S5. Technical details of microscopic theory simulations**

To obtain Figs. 3d and 3e in the main text, we performed numerical simulations of the microscopic theory described in Methods. Here we supplement further technical details for reproducibility.

The vector potentials were taken as

$$A_i(t) = a_i \cos(2\pi f_i t + \phi_i) \exp \left[ -2 \ln 2 \left( \frac{t - t_i}{w_i} \right)^2 \right] \quad (i = 0, 1),$$

where $A_0(t)$ and $A_1(t)$ denote the pump and probe fields, respectively. Considering a typical experimental condition, we chose the parameters as $ea_0/h = 0.24 \text{ nm}^{-1}$, $f_0 = 30 \text{ THz}$, and $w_0 = 180 \text{ fs}$ for the pump and $ea_1/h = 0.05 \text{ nm}^{-1}$, $f_1 = 20 \text{ THz}$, and $w_1 = 20 \text{ fs}$ for the probe. We fixed $t_0 = 0$ and varied the probe delay $t_1 = \Delta t$. To remove the contribution from
the four-wave mixing, we averaged the field-induced electric current over the relative phase \( \phi_0 - \phi_1 \). Specifically, we fixed \( \phi_1 = 0 \) and averaged the results over \( \phi_0 = \left( \frac{2\pi}{N_{\phi}} \right) m \) \((m = 1, 2, ..., N_{\phi})\) for \( N_{\phi} = 8 \). We confirmed that \( N_{\phi} = 16 \) little changed the results.

We numerically solved the time-dependent Schrödinger equation in the presence of the vector potentials. We considered a large \( k \)-space sphere \(|k| \leq k_{\text{max}}\), in which each \( k \)-point is parametrized as \( k = (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)\) with \( \theta \) and \( \varphi \) being the polar and azimuthal angles, respectively. We chose \( k_{\text{max}} = 1 \text{ nm}^{-1} \) large enough that appreciable interband transitions occur only within the sphere. Since the cylindrical symmetry of the model guarantees the \( \varphi \)-independence of the response, we discretized the system into \( 100 \times 30 \) points in the \( k \)-\( \theta \) plane, for \( 0 \leq k \leq k_{\text{max}} \) and \( 0 \leq \theta \leq \pi \) (we confirmed the convergence of the results). We solved the set of equations with the 4th-order Runge-Kutta algorithm and calculated the total electric current by summing contributions from all \( k \)-points. Note that, in the \( T_1^{\prime} \) relaxation terms, \( f_{1,2}(k) \) are determined by the time-dependent temperature \( T(t) \) and chemical potential \( \mu(t) \), which are common for both bands. During time evolution, \( T(t) \) and \( \mu(t) \) were solved self-consistently with the system’s internal energy \( U(t) \) and the total number of electrons (kept constant throughout the simulation).

**S6. Derivation of the macroscopic model from the microscopic one**

Figure S5 shows the outline of the derivation. We start from the microscopic equations of motion presented in the Method section,
\[
\frac{\partial n_1(k)}{\partial t} = -\frac{2}{\hbar} \text{Im}[H_{21}(k)p(k)] - \frac{n_1(k) - f_1(k)}{T'_1} - \frac{n_1(k) - f_1^\text{eq}(k)}{T_1},
\]
\[
\frac{\partial n_2(k)}{\partial t} = +\frac{2}{\hbar} \text{Im}[H_{21}(k)p(k)] - \frac{n_2(k) - f_2(k)}{T'_1} - \frac{n_2(k) - f_2^\text{eq}(k)}{T_1},
\]
\[
i\hbar \frac{\partial P(k)}{\partial t} = [H_{11}(k) - H_{22}(k)]p(k) - H_{12}(k)[n_1(k) - n_2(k)] - i\hbar \frac{\partial P(k)}{T_2}.
\]

Within the rotating-wave approximation, the electron-hole pair density \( \Delta N = \sum_k [n_1(k) - f_1^\text{eq}(k)] = \sum_k [f_2^\text{eq}(k) - n_2(k)] \) follows
\[
\frac{\partial \Delta N}{\partial t} = \frac{2}{\hbar} \text{Im} \left[ A_-(t) \sum_k J_{21}(k)p(k) \right] = \frac{\Delta N}{T_1},
\]
where \( A_+(t) \) denotes the rotating and counter-rotating components of the vector potential \( A(t) \).

We have neglected pair generation and recombination caused by the carrier-carrier scattering, or the \( T'_1 \) terms. In the lowest order, \( P(k) \) is given by
\[
P(k) = \int_0^\infty \frac{d\omega}{2\pi} A(\omega)e^{-i\omega t} \frac{[f_1^\text{eq}(k) - f_2^\text{eq}(k)] |J_{12}(k)|^2}{\hbar(\omega + i/T_2) - \epsilon_1(k) + \epsilon_2(k)},
\]
where \( A(\omega) \) is the spectral amplitude of \( A(t) \). Using Eq. (6), we obtain
\[
\sum_k J_{21}(k)p(k) = \int_0^\infty \frac{d\omega}{2\pi} A(\omega)e^{-i\omega t} \sum_k \frac{[f_1^\text{eq}(k) - f_2^\text{eq}(k)] |J_{12}(k)|^2}{\hbar(\omega + i/T_2) - \epsilon_1(k) + \epsilon_2(k)}
\]
\approx A_+(t) \sum_k \frac{[f_1^\text{eq}(k) - f_2^\text{eq}(k)] |J_{12}(k)|^2}{\hbar(\omega_0 + i/T_2) - \epsilon_1(k) + \epsilon_2(k)},
\]
where \( \omega_0 \) in the denominator is replaced by \( \omega_0 \), a representative frequency included in \( A(t) \).

Substitution of Eq. (7) into Eq. (5) yields
\[
\frac{\partial \Delta N}{\partial t} = \frac{\omega_0 \sigma_\text{inter}(\omega_0) A(t)^2}{\hbar} \frac{\Delta N}{T_1},
\]
where the interband optical conductivity,
\[
\sigma_\text{inter}(\omega) = \frac{1}{\omega} \text{Im} \sum_k \frac{[f_1^\text{eq}(k) - f_2^\text{eq}(k)] |J_{12}(k)|^2}{\hbar(\omega + i/T_2) - \epsilon_1(k) + \epsilon_2(k)},
\]
has been defined. Using \( A(t)^2 = E(t)^2/\omega_0^2 \) and \( I(t) = n_0 e_0 c E(t)^2 \), we obtain
\[
\frac{\partial \Delta N}{\partial t} = \frac{\sigma_\text{inter}(\omega_0) I}{\hbar}\frac{\Delta N}{T_1}.
\]
This is the well-known rate equation for electron-hole pair excitation.

We next turn to the diamagnetic current density,
\[
j_{\text{dia}} = -\sum_k [K_1(k)n_1(k) + K_2(k)n_2(k)] A.
\]

The interband matrix elements are omitted because numerical analysis shows their minor importance. Neglecting \( k \)-dependence of \( K_{1,2}(k) \), the light-induced change in \( j_{\text{dia}} \) is given by
\[ \Delta j_{\text{dia}} = -(K_1 - K_2) \Delta N \cdot A. \]  

This relation is alternatively expressed in terms of the electric polarization,

\[ \Delta P_{\text{dia}} = \int_{-\infty}^{t} dt' \Delta j_{\text{dia}}(t') \]

\[ \approx -\frac{1}{\omega_0^2} (K_1 - K_2) \Delta N \cdot E, \]

where the second equality derives from the slow variation in \( \Delta N \) compared to the fast oscillation in \( A \) or \( E \). Equating the above expression with \( \Delta P_{\text{dia}} = 2n_0 \varepsilon_0 \varepsilon_{\text{NL}} E \), the refractive index change is given by

\[ n_{\text{NL}} = -\frac{K_1 - K_2}{2\omega_0^2 n_0 \varepsilon_0} \Delta N. \]  

Now we are ready to end up with the macroscopic model, by combining Eqs. (10) and (14) to obtain

\[ \tau \frac{\partial n_{\text{NL}}}{\partial t} + n_{\text{NL}} = n_2 I, \]  

with

\[ \tau = T_1, \]

\[ n_2 = -\frac{T_1 (K_1 - K_2) \sigma_{\text{inter}}^{1}(\omega_0)}{2\hbar \omega_0^3 n_0^2 \varepsilon_0^2 c}. \]

A factor \( \omega_0^2 \) in the denominator of Eq. (17) suggests that lower frequency is preferred for SRLS, as long as the intraband response does not obscure it. This dependence stems partly from (10) and partly from (14). First, Eq. (10) shows that a larger number of carriers are generated by lower-frequency fields, because the optical energy is distributed to a larger number of photons to be absorbed. Second, Eq. (14) indicates that the injected carriers change the refractive index more efficiently at lower frequencies. This behavior is easily expected from the Drude model, which predicts a frequency-dependent change in the dielectric constant,

\[ \Delta \varepsilon = -\left( \frac{1}{m_e} + \frac{1}{m_h} \right) \frac{e^2 \Delta N}{\varepsilon_0 \omega^2}, \]  

at frequencies higher than the damping rate. Here, \( m_e \) and \( m_h \) denote the effective mass of electrons and holes, respectively. \( \mathbf{k} \cdot \mathbf{p} \) perturbation theory tells us that the inverse effective mass in the above equation is contributed by both the diamagnetic current and the paramagnetic current, the latter arising from virtual excitation of interband transitions. At high frequencies, the paramagnetic current fails to contribute to the inverse effective mass because interband transitions are no longer virtual. In such a situation, \( e^2/m_e \) and \( e^2/m_h \) in Eq. (18) are replaced by \( K_1 \) and \( -K_2 \), so that Eqs. (13) and (14) follow. We note that the frequency dependence of Eq. (17) is consistent with the experimental tendency seen in Fig. 1f, where a weaker driving (0.9 MV/cm)
at a lower frequency (18.2 THz) and a stronger driving (1.3 MV/cm) at a higher frequency (29.4 THz) led to absorption changes with a similar magnitude, though saturation effect hinders quantitative comparison.

The paramagnetic current density, neglected so far, has been extensively studied in the conventional nonlinear optics. Therefore, we only discuss its consequences without equations. It is known that the paramagnetic current also leads to SRLS in two-level systems\textsuperscript{52}. In this case, however, relative positions of the peak and the dip are inverted depending on the sign of detuning, so that the dispersive lineshape tends to be cancelled out when integrated over a continuum.

Spectral hole burning, arising from a combination of Pauli blocking and ac Stark effect, survives such a cancellation. Our numerical analysis revealed the tendency of hole burning to be suppressed when the dephasing time $T_2$ is short. It is also possible that the diamagnetic coupling $K_{1,2}$ is so large that it can hide the spectral hole burning. Since either the value of $T_2$ or the correct parameters in the effective Hamiltonian are not known, these two possibilities are not resolved at present.

References

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