Intense-pulse dynamics of massless Dirac electrons

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We identify and describe how intense short light pulses couple to massless Dirac fermions in two-dimensional systems. The ensuing excitation dynamics exhibits unusual scaling with the wavelength of the plane waves and the fact that light coupling is efficient only close to the Dirac points. We exploit these features to achieve valley polarization of more than 70% with simple pulse shapes. Quantitative results are given for pristine graphene.

Electrons in two-dimensional (2D) materials carry pseudospins (of opposite sign) at the minima $K$ and $K'$ (valleys) of the valence band in the 1st Brillouin zone [1,2]. The long lifetime of these degrees of freedom has given their manipulation and transport, in short valleytronics, a boost in the quest to search for usable quantum information encoding [3,5].

Most work has been devoted to finite-mass Fermions in gapped materials [6,10] or gapless material under static fields [11,12] since they render addressing K valley domain (“pedestal”) and thereby making use of the subtleties of the valence band in the 1st Brillouin zone [1,2].

The ensuing excitation dynamics exhibits unusual scaling with the wavelength of light due to the linear dispersion of the band structure and the fact that light coupling is efficient only close to the Dirac points. We exploit these features to achieve valley polarization of more than 70% with simple pulse shapes. Quantitative results are given for pristine graphene.

In the following, we will uncover the general mechanism of coupling Dirac dynamics to intense laser pulses which is ruled by (i) the vector potential $A_0$ of the light pulse in units of the distance $\Delta$ between the Dirac points $\Delta = K - K'$ in momentum space and (ii) by time and momentum scaled with the square root of the light frequency (or wavelength), $\tau = \sqrt{\alpha t}$ and $Q = q/\sqrt{\omega}$. The scaling emerges from the fact that coupling to the light only occurs in the vicinity of the Dirac points with linearized dynamics and dipole matrix elements about these points exhibiting this scaling. Furthermore, pulses with $A_0 \approx \Delta$ are most efficient for inducing large valley polarization since the latter relies on transporting excitation from valley domain $K$ to $K'$ or vice versa where the domain separation in momentum space is characterized quantitatively by $\Delta$.

As an application of this general mechanism we will demonstrate that a conventional half-cycle pulse with linear polarization along $\Delta$ can achieve substantial valley polarization (VP) while including a weaker pre-pulse (“pedestal”) and thereby making use of the subtleties of the mechanism can lead to almost 100% VP.

To be specific we will work with pristine graphene in the usual tight-binding two-band description [2] (conduction and valence band), coupled in dipole approximation to a classical electromagnetic field with the time-dependent vector potential

$$A(t) = A_0 e^{-2 \ln 2 \tau^2 / \pi^2} \cos(\omega t).$$

Mostly, we will apply sub-cycle pulses with a duration of $T = T_\omega / 5$, where $T_\omega = 2\pi / \omega$ is the laser period.

We solve the time-dependent Schrödinger equation for the time-dependent two-band Hamilton operator

$$\begin{equation}
H(t) = H_0 + \frac{\hbar}{2} \left( \frac{\partial}{\partial t} + A(t) \right),
\end{equation}$$

whereby the C-C distance is $a = 1.42 \text{Å}$ [2]. Inclusion of relaxation is easily possible by solving the Liouville–von-Neumann equation for the single-particle density matrix. However, this is not necessary in the present context due to the short light pulses.

We begin with the excitation induced by a half-cycle pulse, with $T = T_\omega / 5$ in Eq. (1), linearly polarized parallel to $\Delta = \Delta e_y$ as this dynamics reveals the ruling principles of intense pulse coupling to massless Dirac electrons quite clearly. Figure 1 resolves this excitation probability $d^2P/dq_x dq_y \equiv \rho(q)$ with respect to all initial conditions in the Brillouin zone, discriminated (in blue and red) according to the $K$ and $K'$ domain (lower and upper triangle).

One sees interference patterns along the polarization direction (but not on the $\Delta$-line directly!) with denser fringes in the lower triangle indicating a larger phase accumulation of the driven electron wave than in the upper triangle. The very fact of the clean pattern suggests interference of two amplitudes with a well-defined phase difference. The underlying momentum space trajectory $\tilde{q}(t) = q + A(t)$ for a specific initial condition $q = \Delta(0,0,1)^T$ in Fig. 1 reveals that indeed, there are two instances of closest proximity to the $K$-point (and hence two excitation “bursts”) with $\tilde{q}_y(t) = 0.1 \Delta + A_y(t) = K_y$, one in the rising and one in the falling part of the pulse (see lower horizontal green-dotted line). At both time instances the excitation suddenly rises, see Fig. 1. Clearly, it is crucial to come close to the Dirac point for excitation. This also explains why there are no excitations for initial conditions with $q_y < K_y$, since the corresponding laser-driven trajectories...
FIG. 1. a) Excitation probability ρ(q) as a function of the initial crystal momentum q for the half-cycle pulse of (1) with λ=2.4 µm and field strength A0=Δ and a pulse length of T=Τc/5. The Dirac points in the centers of the triangular unit cells are marked by crosses. b) Time-dependent crystal momentum ̄q(t) = q unexpected height + A(t) in vertical direction. c) Corresponding excitation probability |aq(t)|² for the initial condition q = Δ(1,0,0) T.

do not visit any Dirac point. Moreover, the phase difference between the two bursts for this initial condition is close to 5π which effects constructive interference. Since the transition energy along the trajectory is larger below the K’ point (compared to the one between K and K’ point) the phase accumulated between the two excitations is larger there. Consequently, trajectories starting above K’ yield denser fringes than those starting above K, as can be seen in Fig.1b.

Having established that intense pulse optical excitation of massless Dirac electrons occurs only close to the Dirac points, we next turn to the scaling properties of the excitation dynamics. To this end we consider excitation with the same pulse shape as before but at different wavelengths and for three different A0 of the light for all initial conditions in the first Brillouin zone, shown in Fig.2 with a compactified representation. Note that K’ points are now located at the left and right boundaries of the rectangle in the upper half (marked with little crosses in the figure). One sees that the structures have the same shape but are “zoomed out” from left to right for longer wavelengths, most obvious for the two lower rows while in the first row additional interference due to the large excursion of the trajectories masks the underlying similarity of the pattern.

Excitation happening only close to the Dirac points suggests to linearize the dynamics in momentum space and time close to a Dirac point by Taylor expanding about it, qK ≈ q − K with K = [2π/3a](1,−1/√3) T² or qK’ with K’ = [2π/3a](1,−1/√3) T. Hence Hq from (2) may be approximated close to K by the familiar relativistic Hamiltonian for massless spin-1/2 particles 2Hq = vF σ · qK in terms of the Pauli matrices σ = (σx, σy) T and the speed of light replaced by the Fermi velocity vF = 3ℏa/2. The eigenvalues of 2Hq are 2Ec(xqK) = ± vF |qK| with eigenvectors, often referred to as Houston basis 19, Vc(xqK) = (± qKx + iqKy)/|qK|, 1 T/2 for valence (v) and conduction (c) band, respectively.

Within this approximation the time-dependent dipole-coupled Hamilton operator in the Houston basis can be expressed as

\[
\mathcal{H}(q,t) = v_F |\tilde{q}_K(t)| \sigma_z + \frac{F(t) \cdot [e_z \times \tilde{q}_K(t)]}{2 |\tilde{q}_K(t)|^2} \sigma_x, \tag{3}
\]

FIG. 2. a) Excitation probability ρ as a function of the initial crystal momentum q for the pulse (1) with a pulse length of T=Τc/5 and for various wavelengths λ and field strength A0. The Dirac points in the centers of the triangular unit cells are marked by crosses. (For a compact representation we have, in contrast to Fig.1b, nested the unit cells.) The width δqα of the excitation pattern is marked with a black line at the bottom of each contour plot. It is defined as second moment \( δq_α = \sqrt{m_α/m_0} \) with \( m_α = \int_{qK} dq^K q^K^α ρ(q) \). b) Pattern width δqα as a function of the wavelength λ for three values of A0. The dashed lines show δqα = α/√λ, with α fixed at λ=10 µm. The color scale at the bottom right is the same as in Figs.1b and 4b.
where the second term in $\overline{H}$ is a real, reduced Berry-
connection matrix [20], from which the diagonal elements
have been omitted as they are the same and shift only
the total energy of $\overline{H}$, while the off-diagonal terms couple
electron and conduction band through the electric field
$F(t) = \frac{\partial}{\partial t} A(t)$ of the light.

From Eq. (3) one sees that only close to the $K$-point,
where $|q_{K}(t)|$ is small, transitions to the other band
happen through the dipole coupling. The correspond-
ing time-dependent dynamics is governed by the approx-
imate Hamilton operator $\overline{H}$ valid close to Dirac points
with intriguing scaling properties regarding the depen-
dence on the light frequency $\omega$: Consider a trajectory in
the vicinity of $K$, e.g., $q_{K}(t) = (b_{x}, c_{y} \omega t)^{T}$, which passes
$K$ at $t=0$ at a distance $b_{x}$ and a velocity $c_{y} \omega$. This tra-
jectory corresponds to a linearly polarized pulse along $e_{y}$
with frequency $\omega$ as in Fig. 2 but the following argument
holds for any close encounter of a Dirac point. In terms
of a scaled time $\tau$ and a scaled momentum $B_{x}$
$$\tau = \sqrt{\omega t} \quad \text{and} \quad B_{x} = b_{x}/\sqrt{\omega},$$
we get from [3]
$$\overline{H}(\overline{q}_{K}, \tau) = \overline{H}(\sqrt{\omega} \overline{Q}_{K}, \tau/\omega) = \sqrt{\omega} \overline{H}(\overline{Q}_{K}, \tau)$$
(5)
with $\overline{Q}_{K}(\tau) = (B_{x}, c_{y} \tau)^{T}$. Obviously, [5] gives rise to a
time-dependent Schrödinger equation $[\overline{H}(\overline{Q}_{K}, \tau) - i\frac{\partial}{\partial \tau}] \psi(\tau) = 0$ which is invariant against changes of the fre-
quency $\omega$. It directly explains the scaling of the widths
$\delta q_{x}$ of the excitation pattern for different wavelengths,
respectively frequencies, encountered in Fig. 2. This is a
universal result for intense light, dipole-coupled to mass-
less electrons.

As a first application of this result we can formulate
simple laser-pulse shapes to control valley polarization
with high efficiency. The latter can be quantified as $\eta$
by the relative excitation probability difference of initial
conditions in the $K$ (red in Figs. 1 and 2) and $K'$ domain
(blue in Figs. 1 and 2),
$$\eta \equiv (P_{K} - P_{K'})/(P_{K} + P_{K'}),$$
(6a)
with
$$P_{K} = \int_{\Delta K} dq^{2} \rho(q), \quad P_{K'} = \int_{\Delta K'} dq^{2} \rho(q)$$
(6b)
integrated over the triangular $q$-domains $\Delta K$ and $\Delta K'$
around $K$ and $K'$, respectively. Note that our definition of
$\eta$ allows values between $-1$ and $+1$ for polarization of the $K$ or $K'$ pseudospin, respectively, while in other definitions [17] the values of $\eta$ range from $-2$ to $+2$.

With the half-cycle pulse discussed so far, one can see
already from Fig. 2 that due to the $\sqrt{\omega}$ scaling smaller fre-
quency (larger wavelength) confines the excitation tighter
to the electron excursion $\overline{q}(t)$ driven by the light pulse
and therefore promises a higher degree of control. Com-
bined with a laser-driven excursion compatible with the
geometry of the Brillouin zone and particular its scale
given by the separation $\Delta$ of the Dirac points, one can
achieve a large asymmetry in the excitation of the $K$
versus the $K'$ domain, manifest in the dominance of red pat-
tern in comparison to blue ones, most prominent for the
right panel in the middle row of Fig. 2 which corresponds
to the largest wavelength displayed and the expected op-
timum near $A_{0} = \Delta$. One sees that for $A_{0} < \Delta$ the
initial conditions in the $K$ as well as in the $K'$ domain
are not exhausted for excitation (lower right panel) while
for $A_{0} > \Delta$ also initial conditions from the $K'$ domain
get excited through the trajectory passing by $K$ (blue in-
tensity in the middle on the top of the right upper panel)
lowering the contrast of valley polarization. The latter is
in general true for shorter wavelength (left row in Fig. 2),
where initial conditions from the $K$ and $K'$ domain are
almost equally excited in the first Brillouin zone.

Next to the “resonance” condition $A_{0} \approx \Delta$ and long
wavelength, the half-cycle nature of the pulse, containing
amplitude dominantly only in one direction, is of course
useful since it breaks the equivalence of $K$ and $K'$ points
for the laser-driven dynamics. Longer pulses with ampli-
tudes in both directions restore this equivalence and di-
minish valley polarization. Not necessary, however, is a
helicity character of the pulse (circular polarization, e.g.)
which was thought to enhance or decrease the interaction
with the pseudospin degrees of freedom depending on the
direction of rotation.

FIG. 3. Valley polarization for half-cycle pulses with 3 wave-
lengths as a function of the peak vector potential in unit of the
$K-K'$ point distance $\Delta$. For comparison we show re-
results for a two-color pulse [21] and a half-cycle pulse with a
pedestal pulse according to Eq. (7) with $\xi = 8$, $T = T_{w}/5$ and
$\lambda = 2.4 \mu m$.

The simple half-cycle pulse discussed so far is also more
efficient at a wavelength of $\lambda = 7.2 \mu m$ with $\eta = 43 \%$,
than a two-color clover-shaped pulse [17] with $\eta = 27 \%$, as
can be seen in Fig. 3 where VP is shown as a function of
$A_{0}$ for different pulse forms and different wavelengths
for the half-cycle pulses. Apart from the shortest wave-
length (800 nm) where the blurred fringe pattern leads to excitation delocalized in $q$-space and therefore to irregular changes of $\eta$ with increasing $A_0$. All other pulses behave qualitatively similar with the achieved VP as a function of $A_0$: VP sets in for $A_0 > \Delta/2$ and reaches a maximum around $A_0 = \Delta$. The delayed onset becomes understandable by realizing that a Dirac point is located at a distance larger than $\Delta/2$ from any edge of its triangular domain. However, only if initial conditions $q$ from a different Dirac domain reach the $K$ point through a laser-driven trajectory which crosses the boundary of the triangular domain of $K$, one can expect finite VP.

The half-cycle pulse

$$A_\xi(t) = \frac{A_0}{2} \left[ e^{-2\ln2 t^2/\tau^2} + e^{-2\ln2 t^2/\xi^2 T^2} \right].$$

(7)

with a pedestal, another half-cycle pulse that is $\xi$ times longer, however, stands out as it produces a VP of more than 70% (dashed line in Fig. 3 with $\xi = 8$) which may reach close to 100% by optimizing the pulse shape which we have not done since here we are interested in the principles of controlling VP with intense pulses.

In that respect, it is important to fulfill one more criterion for efficient excitation when passing a Dirac point at time $t^*$. The field strength $F(t^*)$, cf. Eq. (7), must be large, otherwise even a large dipole matrix element does not help. Vice versa, large field strengths can compensate to some extent small dipole matrix elements giving rise to “uncontrolled” excitation, further away from the Dirac points. The pulse (7) fulfills this criterion as one can see from Figs. 4b and c. There, the initial conditions of the $K$ and $K'$ domains and the $K$ and $K'$-point encounters of the respective light driven trajectories are shown in red and blue, respectively. Large VP can only be achieved if excitation through the $K'$ point (blue trajectories) is also achieved for initial conditions from the $K$ domain (red area). The times $t^*$ of the Dirac point encounters for all initial conditions $q$ in Fig. 4 show that this is indeed the case for the single half-cycle pulse (dashed) as well as for the one with a pedestal (solid). However, what difference the latter makes becomes apparent if one looks with which field strengths $F(t^*)$ these encounters happen as illustrated in Fig. 4: Now it is clear that with the half-cycle pulse also unfavorable initial (blue) conditions from the $K'$ domain encounter the $K'$ point with large $F$ (dashed blue line in the blue area) and are therefore strongly excited, while for the pedestal pulse these initial conditions encounter $K'$ with field strength $F(t) \approx 0$ leaving the large field strengths encounters with high excitation to initial conditions from the $K$ domain. This implies an increase of the VP contrast and leads to the high degree of $\eta = 72\%$ in Fig. 3 as illustrated with the excitation probabilities in Fig. 4.

To summarize, we have formulated the principles of intense-laser-pulse excitation in gapless 2D materials. The realization that substantial excitation is only possible close to the Dirac points suggests a linearization about these points and in time leading to the familiar relativistic Hamilton operator for massless Dirac fermions with linear dispersion augmented by a linearized Berry connection which represents the dipole coupling to the light. Scaling momenta and time in this effective time-dependent Hamilton operator renders it globally proportional to $\sqrt{\omega}$, which leads to a time-dependent Schrödinger equation in the scaled time $\tau = \sqrt{\omega}t$ completely independent of the frequency of the light, provided the pulse shape function is also formulated as a function of $\tau$.

An obvious application of this insight into the strong-field-driven dynamics of massless electrons is the control of valley polarization. As we have demonstrated, it can be achieved with half-cycle pulses linearly polarized along the vector $\Delta$ connecting the $K$ and $K'$ Dirac points to break their equivalence transiently. A peak amplitude of the pulse $A_0 \approx \Delta$ leads to the highest VP values. In general the VP contrast can be enhanced by increasing the wavelength to get more localized excitation in momentum space according to the scaling. A third control parameter optimizing VP is to design the pulse shape in such a way that the highest field strength during the pulse is achieved when laser-driven electron trajectories pass a Dirac point starting with initial conditions from the domain of the other pseudospin symmetry.

We expect these principles to also hold for systems with band gaps small in relation to the strong driving laser field, a conjecture which is supported by the report of VP with linearly-polarized short pulses in the gapped
hBN and MoS$_2$ systems \cite{JimenezGalan2021} for ponderomotive energies considerably larger than the band gap. Investigations of intense pulse dynamics in 2D-systems with more exotic topological properties are underway to explore the limitations and possible extensions of the universal behavior induced by the relativistic massless dynamics.

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