

Light-induced valleytronics in pristine graphene

M. S. MRUDUL,¹ ÁLVARO JIMÉNEZ-GALÁN,² MISHA IVANOV,² AND GOPAL DIXIT^{1,*}

¹Department of Physics, Indian Institute of Technology Bombay, Powai, Mumbai 400076, India

²Max-Born Institut, Max-Born Straße 2A, 12489 Berlin, Germany

*Corresponding author: gdixit@phy.iitb.ac.in

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Electrons in two-dimensional hexagonal materials have an extra degree of freedom, the valley pseudospin, that can be used to encode and process quantum information. Valley-selective excitations, governed by the circularly polarized light resonant with the material's bandgap, are the foundation of valleytronics. It is often assumed that achieving valley selective excitation in pristine graphene with all-optical means is not possible due to the inversion symmetry of the system. Here, we demonstrate that both valley-selective excitation and valley-selective high-harmonic generation can be achieved in pristine graphene by using a combination of two counter-rotating circularly polarized fields, the fundamental and its second harmonic. Controlling the relative phase between the two colors allows us to select the valleys where the electron-hole pairs and higher-order harmonics are generated. We also describe an all-optical method for measuring valley polarization in graphene with a weak probe pulse. This work offers a robust recipe to write and read valley-selective electron excitations in materials with zero bandgap and zero Berry curvature. © 2021 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

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1. INTRODUCTION

The realization of atomically thin monolayer graphene has led to breakthroughs in fundamental and applied sciences [1,2]. Charge carriers in graphene are described by the massless Dirac equation and exhibit exceptional transport properties [3], making graphene very attractive for novel electronics applications. One of the most interesting features of graphene and gapped graphene materials is the electron's extra degree of freedom, the valley pseudospin, associated with populating the local minima \mathbf{K} and \mathbf{K}' in the lowest conduction band of the Brillouin zone. This extra degree of freedom has the potential to encode, process, and store quantum information, opening the field of valleytronics [4].

The monolayer graphene, as opposed to gapped graphene materials, presents a fundamental challenge for valleytronics: it has zero bandgap and zero Berry curvature. These aspects are generally considered to be a major impediment for valleytronics. In gapped graphene materials, valley selectivity is achieved by matching the helicity of a circularly polarized pump pulse, resonant to the bandgap, to the sign of the Berry curvature [5–9]. Recently demonstrated [10] sub-cycle manipulation of the electron population in \mathbf{K} and \mathbf{K}' valleys of tungsten diselenide, achieved with the combination of a resonant pump pulse locked to the oscillations of the terahertz control pulse, represents a major milestone. Precise sub-cycle control over the driving light fields opens new opportunities for valleytronics, such as those offered by the new concept of topological resonance, discovered and analyzed in Refs. [11–13]. Single-cycle pulses with the controlled phase of carrier oscillations under the envelope offer a route to valleytronics in gapped graphene-type materials even when such pulses are linearly, not

circularly, polarized [14]. It is also possible to avoid reliance on resonances in gapped graphene-type materials, breaking the symmetry between the \mathbf{K} and \mathbf{K}' valleys via a light-induced topological phase transition, closing the gap in the desired valley [15].

Thus, with its zero bandgap, zero Berry curvature, and identical dispersion near the bottom of the valleys, pristine graphene appears unsuitable for valleytronics—a disappointing conclusion in view of its exceptional transport properties. We show that this generally accepted conclusion is not correct, and that the preferential population of a desired valley can be achieved by tailoring the polarization state of the driving light pulse to the symmetry of the lattice. Our proposal offers an all-optical route to valleytronics in pristine graphene, complementing approaches based on creating a gap by using a heterostructure of graphene with hexagonal boron nitride [16–19], or by adding strain and/or defect engineering [20–24].

While the light configuration we use is similar to that used in Ref. [15] for finite bandgap materials, the physical mechanism underlying valley-selective excitation in a zero-bandgap, centrosymmetric material such as graphene is quite different. In gapped materials, valley polarization is achieved by selectively reducing the effective bandgap in one of the valleys [15]. Here, valley polarization is achieved only when the light-driven electrons explore the anisotropic region in the Brillouin zone.

We also show valley selectivity of harmonic generation in graphene, and demonstrate it with the same field we use for valley-selective electronic excitation. High-harmonic generation (HHG) is a powerful method for probing attosecond electron dynamics in systems as diverse as atoms, molecules [25–30], and solids [31–44]. In solids, high-harmonic spectroscopy was used to probe

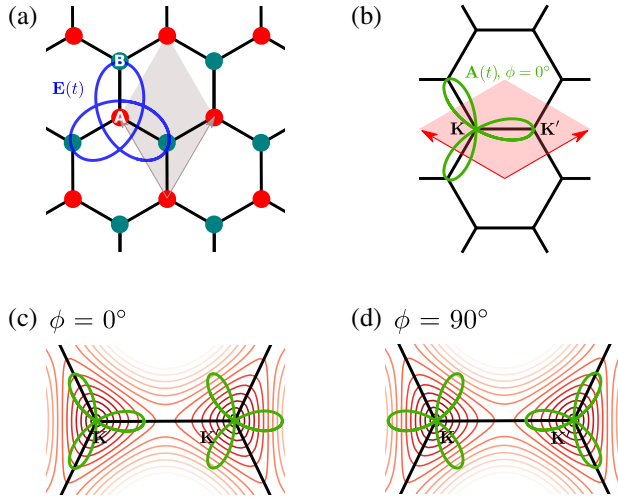


Fig. 1. Physical mechanism of valley polarization in graphene. (a) Graphene lattice in the coordinate space, with the Lissajous figure of the bicircular electric field breaking the symmetry between otherwise identical carbon atoms A and B; two-color phase $\phi = 0^\circ$. (b) Optically induced symmetry breaking viewed in the momentum space; the vector potential of the bicircular field is shown for $\phi = 0^\circ$. (c), (d) Close-up images of the valleys show their asymmetry, leading to different laser-driven dynamics and different excitation rates once the electron leaves the bottom of the valley. Here, the red contours show the conduction band energy in the reciprocal space.

valence electrons [35], atomic arrangement in solids [35–37], defects [45,46], band dispersion [38–40], and quantum phase transitions [41,42,47], and to realize petahertz electronics in solids [35,43,44]. Last but not least, we describe an all-optical method for measuring valley polarization in graphene with a weak probe pulse.

The key idea of our approach is illustrated in Fig. 1, which shows graphene in real (a) and reciprocal (b) space, together with the structure of the incident electric field (a) and the corresponding vector potential (b). The field is made by superimposing two counter-rotating circularly polarized colors at frequencies ω and 2ω . The Lissajous figure for the total electric field is a trefoil, and its orientation is controlled by the relative two-color phase ϕ , i.e., the sub-cycle two-color delay measured in terms of ω . In the absence of the field, the two carbon atoms A and B, in real space, are related by the inversion symmetry. When the field is turned on, this inversion symmetry is broken: the electric field in (a) always points from atom A to the one of the three neighboring atoms B during the full laser cycle, but not the other way around. Indeed, if the center of the Lissajous figure is placed on atom B, the field points in the middle between its neighbors. One can control this symmetry breaking by rotating the trefoil, interchanging the roles of atoms A and B. Thus, the bicircular field offers a simple, all-optical, ultrafast tool to break the inversion symmetry of the graphene lattice in a controlled way. Such controlled symmetry breaking allows one to control the relative excitation probabilities induced by the same laser field in the adjacent \mathbf{K} and \mathbf{K}' valleys of the Brillouin zone.

Figure 1(b) provides the complementary reciprocal-space perspective. In the laser field, the electron crystal momentum follows $\mathbf{k}(t) = \mathbf{k}_i + \mathbf{A}(t)$, where \mathbf{k}_i is the initial crystal momentum, and $\mathbf{A}(t)$ is the laser vector potential, shown in Fig. 1(b) for the electric field shown in Fig. 1(a). The asymmetry between the two valleys with respect to the vector potential is immediately visible.

Figures 1(c) and 1(d) provide additional support to this qualitative picture. One can observe that the two field-free valleys, \mathbf{K} and \mathbf{K}' , are identical only near their very bottoms. As soon as one moves away from the bottom, the valleys start to develop the trefoil structure, with \mathbf{K} and \mathbf{K}' being mirror images of each other. How the symmetry of the vector potential fits into the symmetry of the valley away from their bottoms will control the dynamics and excitation probability.

In sufficiently strong fields, excitation happens not only at the Dirac point where the gap is equal to zero, but also in its vicinity. For the vector potential in Fig. 1(c), the average gap seen by the electron when following the vector potential from the Dirac point in the \mathbf{K} valley, i.e., moving along the trajectory $\mathbf{K} + \mathbf{A}(t)$, is less than in the \mathbf{K}' valley, i.e., when following the trajectory $\mathbf{K}' + \mathbf{A}(t)$. In sufficiently strong and low-frequency fields, such that the bandgap along trajectories $\mathbf{K} + \mathbf{A}(t)$ and $\mathbf{K}' + \mathbf{A}(t)$ quickly exceeds the photon energy, the excitation probability should be higher in Fig. 1(c). For the same reason, rotating the vector potential as shown in Fig. 1(d) should favor population of the \mathbf{K}' valley. Here, “quickly” means “within a fraction of one-third of the laser cycle,” which is the relevant time-scale for the bicircular field. In this context, lower laser frequencies leading to higher vector potential are better suited to meet this requirement.

2. THEORETICAL FRAMEWORK

In the simulations, we used the nearest-neighbor tight-binding approximation to obtain the ground state of graphene with a hopping-energy of 2.7 eV [48]. The lattice parameter of graphene is chosen to be 2.46 Å. The resultant band structure has a zero bandgap with linear dispersion near the two points in the Brillouin zone known as \mathbf{K} and \mathbf{K}' points.

The density matrix approach was used to follow the electron dynamics in graphene. The time evolution of density matrix element $\rho_{mn}^{\mathbf{k}}$ was performed using semiconductor Bloch equations within the Houston basis $|n, \mathbf{k} + \mathcal{A}(t)\rangle$ as [49,50]

$$\partial_t \rho_{mn}^{\mathbf{k}} = -i\epsilon_{mn}^{\mathbf{k}+\mathcal{A}(t)} \rho_{mn}^{\mathbf{k}} - (1 - \delta_{mn}) \frac{\rho_{mn}^{\mathbf{k}}}{T_2} + i\mathcal{F}(t) \cdot \left[\sum_l \left(\mathbf{d}_{ml}^{\mathbf{k}+\mathcal{A}(t)} \rho_{ln}^{\mathbf{k}} - \mathbf{d}_{ln}^{\mathbf{k}+\mathcal{A}(t)} \rho_{ml}^{\mathbf{k}} \right) \right]. \quad (1)$$

Here, $\epsilon_{mn}^{\mathbf{k}}$ and $\mathbf{d}_{mn}^{\mathbf{k}}$ are, respectively, the bandgap energy and the dipole-matrix elements between m and n energy bands at \mathbf{k} . Dipole matrix elements were calculated as $\mathbf{d}_{mn}^{\mathbf{k}} = -i\langle u_m^{\mathbf{k}} | \nabla_{\mathbf{k}} | u_n^{\mathbf{k}} \rangle$, where $u_n^{\mathbf{k}}$ is the periodic part of the Bloch function. $\mathcal{F}(t)$ and $\mathcal{A}(t)$ are, respectively, the electric field and vector potential of the laser field and are related as $\mathcal{F}(t) = -\partial \mathcal{A}(t) / \partial t$. A phenomenological term accounting for the decoherence is added with a constant dephasing time T_2 . Conduction band population relaxation was neglected [51].

The total current was calculated as

$$\mathbf{J}(\mathbf{k}, t) = \sum_{m,n} \rho_{mn}^{\mathbf{k}}(t) \mathbf{p}_{nm}^{\mathbf{k}+\mathcal{A}(t)}. \quad (2)$$

Here, $\mathbf{p}_{nm}^{\mathbf{k}}$ are the momentum matrix elements, obtained as $\mathbf{p}_{nm}^{\mathbf{k}} = \langle n, \mathbf{k} | \nabla_{\mathbf{k}} \hat{H}_{\mathbf{k}} | m, \mathbf{k} \rangle$. The off-diagonal elements of momentum and dipole-matrix elements are related as $\mathbf{d}_{mn}^{\mathbf{k}} = i\mathbf{p}_{mn}^{\mathbf{k}} / \epsilon_{mn}^{\mathbf{k}}$.

Finally, the harmonic spectrum was determined from the Fourier transform of the time derivative of the total current as

$$\mathcal{I}(\omega) = \left| \mathcal{FT} \left(\frac{d}{dt} \left[\int_{BZ} \mathbf{J}(\mathbf{k}, t) d\mathbf{k} \right] \right) \right|^2. \quad (3)$$

Here, integration is performed over the entire Brillouin zone.

3. RESULTS AND DISCUSSION

Valley-selective electronic excitation induced by the tailored field is confirmed by our numerical simulations. In the simulations, graphene is exposed to the bicircular field with the vector potential

$$\mathcal{A}(t) = \frac{A_0 f(t)}{\sqrt{2}} \left(\left[\cos(\omega t + \phi) + \frac{\mathcal{R}}{2} \cos(2\omega t) \right] \hat{\mathbf{e}}_x + \left[\sin(\omega t + \phi) - \frac{\mathcal{R}}{2} \sin(2\omega t) \right] \hat{\mathbf{e}}_y \right). \quad (4)$$

Here, $A_0 = F_\omega/\omega$ is the amplitude of the vector potential for the fundamental field, F_ω is its strength, $f(t)$ is the temporal envelope of the driving field, ϕ is the sub-cycle phase difference between the two fields, and \mathcal{R} is the ratio of the electric field strengths for the two fields, leading to $\mathcal{R}/2$ ratio for the amplitudes of the vector potentials. The amplitude of the fundamental field was varied up to $F_\omega = 15$ MV/cm, leading to the maximum fundamental intensity 3×10^{11} W/cm², with the fundamental wavelength $\lambda = 6$ μ m. This laser intensity is below the damage threshold for monolayer graphene [52–54]. We have also varied \mathcal{R} , using $\mathcal{R} = 1$ and $\mathcal{R} = 2$. A pulse with a sin-squared envelope and 145 fs duration (zero to zero) is employed in this work. Our findings are valid for a broad range of wavelengths and field intensities. To obtain the total population of the different valleys, we have integrated the momentum-resolved population over the sections shown in Fig. 2(a).

To quantify the amount of valley polarization, we used the valley asymmetry parameter defined as

$$\eta = \frac{n_c^{\mathbf{K}'} - n_c^{\mathbf{K}}}{(n_c^{\mathbf{K}'} + n_c^{\mathbf{K}})/2}, \quad (5)$$

where $n_c^{\mathbf{K}'}$ and $n_c^{\mathbf{K}}$ are electron populations at the end of the laser pulse in the conduction band around \mathbf{K}' and \mathbf{K} valleys, respectively.

Figure 2(b) shows the asymmetry in the populations of the \mathbf{K} and \mathbf{K}' valleys as a function of the two-color phase ϕ for several values of the fundamental field amplitude and $\mathcal{R} = 2$. Substantial contrast between the two valleys is achieved once the excitations leave the bottom of the Dirac cone, with values as high as $\pm 36\%$ for $\phi = 0^\circ, 90^\circ$, and no asymmetry at $\phi = 45^\circ$. Here, each 180° change in ϕ results in 120° rotation of the trefoil, yielding an equivalent configuration. This is the reason for the periodicity of the valley asymmetry presented in Fig. 2(b). The higher-populated valley is the one where the vector potential “fits” better into the shape of the valley, minimizing the bandgap along the electron trajectory in the momentum space. The same results are obtained when simultaneously changing the helicities of both driving fields [Figs. 2(c) and 2(d)]. The asymmetry in the valley population is negligible up to an intensity of 2×10^{11} W/cm² and gradually increases with intensity [Fig. 2(b)]. This shows that the valley asymmetry is observed only when the laser pulse is able to drive the electrons to the anisotropic part of the conduction band.

The current generated by the electron injection into the conduction band valleys is accompanied by harmonic radiation and makes substantial contribution to lower-order harmonics, such as H4 and H5 for our two-color driver. These harmonics are stronger whenever the dispersion $\epsilon(\mathbf{k})$ is more nonlinear. In this respect, for the electrons following trajectories $\mathbf{K} + \mathbf{A}(t)$ and $\mathbf{K}' + \mathbf{A}(t)$, the current-driven HHG from the \mathbf{K}' valley is preferred for the vector potential in Fig. 1(c). Conversely, for the vector potential in Fig. 1(d) low-order, current-driven harmonic generation should be preferred from the \mathbf{K} valley. Indeed, following the vector potential, the electron is driven against the steeper walls in the \mathbf{K}' valley in Fig. 1(c) and against the steeper walls in the \mathbf{K} valley in Fig. 1(d).

These qualitative expectations are also confirmed by our numerical simulations, shown in Fig. 3. The same laser parameters used in Fig. 2 are used in this simulation.

In general, the interband and intraband harmonic emission mechanisms in graphene are coupled [55–57], except at low electric fields [57], leading to a complex interplay between the interband and intraband emission mechanisms. The former should be more sensitive to the dephasing than the latter. To this end, we have calculated the harmonic spectrum for different dephasing times T_2 [Fig. 3(a)]. We find that the harmonic emission is essentially T_2 independent, until at least H13, suggesting that the intraband emission mechanism is generally dominant.

Figure 3(b) shows a polarization-resolved high-harmonic spectrum. The $(3n + 1)$ harmonics follow the polarization of the ω pulse (left-handed circular polarization), whereas $(3n + 2)$ harmonics follow the polarization of the 2ω pulse (right-handed circular polarization), while $3n$ harmonics are missing, just as in atomic media [58,59]. In this context, we note that while harmonic

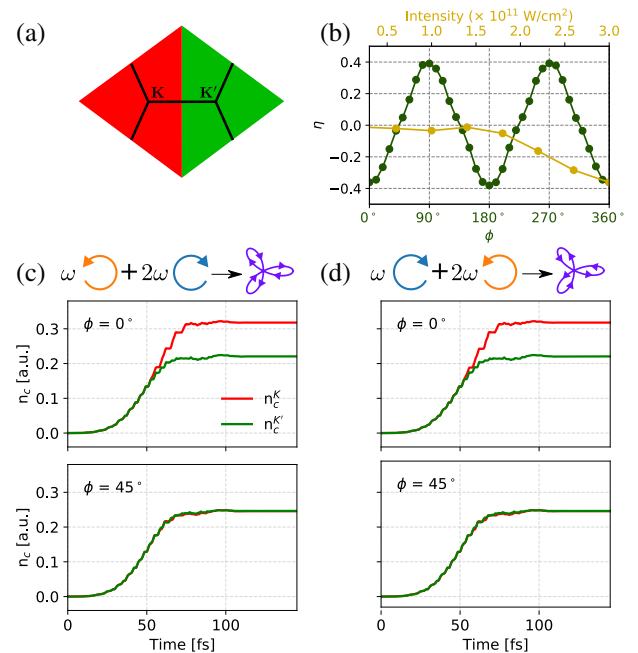


Fig. 2. Valley asymmetry around two valleys \mathbf{K}' and \mathbf{K} . (a) Separation of the Brillouin zone into \mathbf{K}' and \mathbf{K} valleys. (b) Asymmetry in the valley-resolved populations in the conduction band as a function of ϕ (green line with an intensity 3×10^{11} W/cm²) and laser intensity (yellow line with $\phi = 0^\circ$) for a laser with wavelength of 6 μ m, $\mathcal{R} = 2$, and a dephasing time of 10 fs. (c), (d) Excitation dynamics during the laser pulse for $\phi = 0^\circ, 45^\circ$: switching the helicities of both fields simultaneously does not change the outcome.

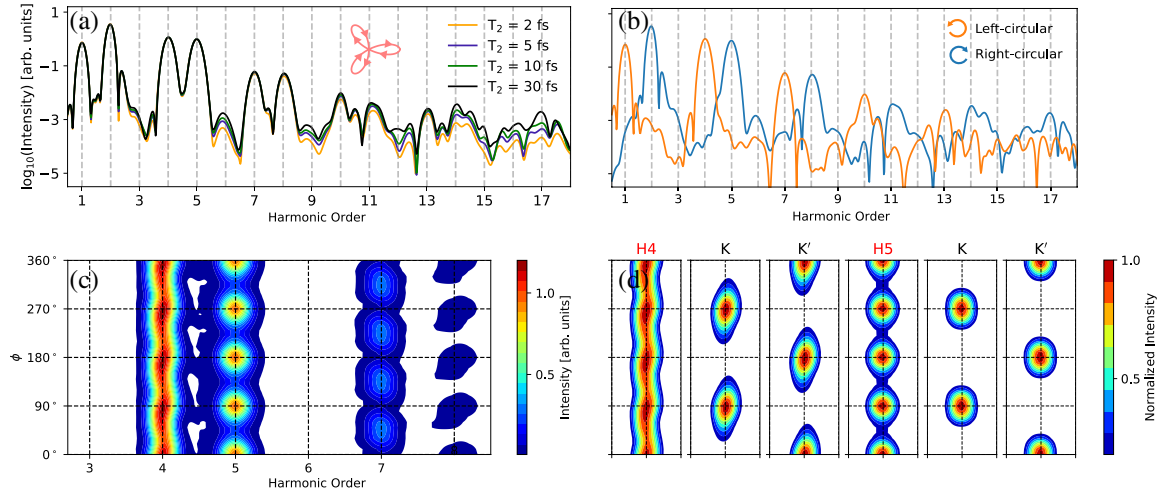


Fig. 3. Valley polarization in high-harmonic emission driven by a bicircular field in graphene. (a) Harmonic spectrum for different dephasing times, for the vector potential shown in the inset ($\phi = 0$). (b) Polarization-resolved high-harmonic emission for left-handed circularly polarized fundamental (orange) and right-handed circularly polarized second harmonic (blue). Alternating harmonics follow alternating helicities: $3n + 1$ follow the fundamental, and $3n + 2$ follow the second harmonic ($3n$ harmonics missing due to symmetry). (c) Harmonic emission as a function of the two-color phase ϕ . (d) Valley polarization of H4 and H5 as a function of the two-color phase. The total field is identical for $\phi = 0^\circ$ and $\phi = 180^\circ$ owing to the threefold symmetry.

generation with single-color circularly polarized drivers is forbidden in atoms, such selection rules do not generally arise in solids [32]. However, the ellipticity-dependence studies on graphene show very weak harmonic yield for drivers with higher ellipticity [52,55,56], as we have also observed. As in atoms, application of bicircular fields allows for efficient generation of circularly polarized harmonics in graphene.

Figure 3(c) shows the dependence of harmonic generation on the orientation of the vector potential relative to the structure of the Brillouin zone. As expected, the total harmonic yield is modulated as the trefoil is rotated, with the lower-order current-driven harmonics H4 and H5 following the expected pattern, maximizing when electrons are driven into the steeper walls in either the \mathbf{K}' or \mathbf{K} valley.

The contribution of different valleys to H4 and H5 as a function of the field orientation is presented in Fig. 3(d). Consistent with the qualitative analysis above, maximum harmonic contribution of the \mathbf{K}' valley corresponds to the vector potential orientations such as shown in Fig. 1(c), while the maximum contribution of the \mathbf{K} valley corresponds to the vector potential orientations such as shown in Fig. 1(d). Therefore, we are able to control the valley polarization of the harmonics by controlling the two-color phase ϕ . We have also checked that these results do not depend on the specific directions of rotation of the two driving fields. That is, we find the same results when simultaneously changing the helicities of both driving fields.

To read-out the induced valley polarization, we employ a probe pulse of frequency 3ω linearly polarized along the x direction (parallel to $\Gamma - \mathbf{K}$ and $\mathbf{K}' - \mathbf{K}$ directions in the Brillouin zone). The amplitude of the probe field is 1.5 MV/cm ($F_\omega/10$). Since the \mathbf{K}' , \mathbf{K} valleys in graphene are related by space inversion, the even-order harmonics generated by individual (asymmetric) valleys are equal in magnitude but opposite in phase [Fig. 4(a), red and green lines]. In the absence of valley polarization, their interference leads to complete cancellation of even harmonics [Fig. 4(a)] (full Brillouin zone signal). In the presence of valley polarization, the cancellation of even harmonics is

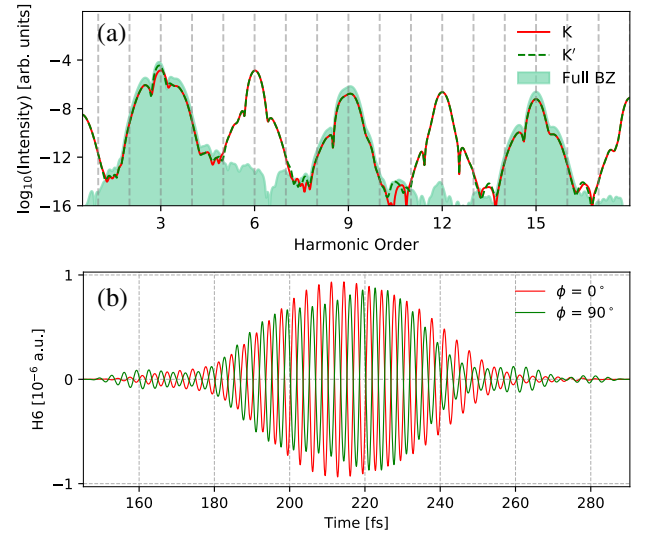


Fig. 4. Read-out of the induced valley polarization in graphene. (a) Harmonic spectrum corresponding to a linearly polarized field with amplitude of 1.5 MV/cm and frequency of 3ω for monolayer graphene. The red and green lines show the valley-resolved HHG spectrum. (b) Sixth harmonic (H6) generated by 3ω pulse after the bicircular field broke the inversion symmetry in graphene.

quenched, and even harmonics signal scales proportional to valley polarization (see also Refs. [14,60]). The phase of the even harmonics follows the dominant valley. Importantly, $3N\omega$ harmonics are absent in the spectra generated by the bicircular $\omega - 2\omega$ field [Fig. 3(b)]. Thus, even harmonics generated by the 3ω probe provide background-free measurement of valley polarization.

Figure 4(b) shows generation of the second harmonic of the 3ω probe pulse (labeled H6) for the two-color phases of the bicircular pump $\phi = 0^\circ$ (red curve) and 90° (green curve), which switches the valley polarization between \mathbf{K} and \mathbf{K}' valleys. While the H6 intensity measures valley polarization, its phase clearly identifies the dominant valley. This phase can be measured by interfering the signal with the reference second harmonic of 3ω generated,

e.g., from a beta barium borate (BBO) crystal. Controlling the delay of the reference second harmonic generated in the BBO crystal, we can map the phase of H6 generated by graphene on the amplitude modulation of their interference.

4. CONCLUSION

In summary, valley polarization in both electronic excitation and harmonic generation can be achieved in pristine graphene by tailoring the Lissajous figure of the driving pulse to the symmetry of the graphene lattice. This allows one to both break the inversion symmetry between the adjacent carbon atoms and also exploit the anisotropic regions in the valleys, taking advantage of the fact that the energy landscapes of the valleys are mirror images of each other. The present work opens an avenue for a new regime of valleytronics in pristine graphene and similar materials with zero bandgap and zero Berry curvature.

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