Modern light generation technology offers extraordinary capabilities for sculpting light pulses, with full control over individual electric field oscillations within each laser cycle\textsuperscript{1–5}. These capabilities are at the core of lightwave electronics\textsuperscript{6–11} – the dream of ultrafast lightwave control over electron dynamics in solids, on a few-cycle to sub-cycle timescale, aiming at information processing at tera-Hertz to peta-Hertz rates. Here we show a robust and general approach to valley-selective electron excitations in two-dimensional solids\textsuperscript{12}, by controlling the sub-cycle structure of non-resonant driving fields at a few-femtosecond timescale. Bringing the frequency-domain concept of topological Floquet systems\textsuperscript{13–15} to the few-fsec time-domain, we develop a transparent control mechanism in real space and an all-optical, non-element-specific method to coherently write, manipulate and read selective valley excitations using fields carried in a wide range of frequencies, on timescales that can be much shorter than the valley lifetime, crucial for implementation of valleytronic devices\textsuperscript{16}. 

\textit{Ultrafast topology for strong-field valleytronics}

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Two-dimensional graphene-like systems with broken inversion symmetry, such as monolayer hexagonal boron nitride (hBN) or transition metal dichalcogenides (TMDs), are candidates for next generation quantum materials due to their high carrier mobility and, especially, to their valley degree of freedom, with potential applications in quantum information processing. Valleys are local minima in the crystal band structure corresponding to different crystal momenta: in 2D hexagonal lattices they are located at the \( \mathbf{K} \) and \( \mathbf{K}' = -\mathbf{K} \) points of the Brillouin zone (Fig. 1a). Selective excitation of \( \mathbf{K} \) or \( \mathbf{K}' \) can be achieved using weak circularly polarized field resonant with the direct band gap of the material, it couples to either \( \mathbf{K} \) or \( \mathbf{K}' \) depending on light’s helicity (the optical valley selection rule).

However, such weak fields are ill-suited for manipulating the generated excitations at the ultrafast time-scales desirable due to short valley lifetimes (\( \sim 10^3 - 10^6 \) fsec for excitons and electrons, respectively). Switching excitations between the valleys is also very challenging. A major step towards meeting this challenge has been made recently: Langer et al demonstrated switching of the population between the \( \mathbf{K} \) and \( \mathbf{K}' \) valleys using the combination of a resonant pump pulse, which populated the desired valley, and a strong terahertz pulse, which moved the excited population within the Brillouin zone by controlling the THz field strength.

The feasibility of applying strong non-resonant fields to bulk dielectrics and ultrathin transition metal dichalcogenide (TMD) films without material damage opens major new opportunities in valleytronics. Our approach capitalizes on them. In contrast to previous work, we do not require resonant light, nor do we need precise tuning of the strength of the control field. In-
stead, we use far off-resonant light to modify the topological properties of the system by inducing complex-valued second neighbour hoppings in a topologically-trivial hexagonal lattice, similar to those introduced by Haldane. These hoppings control both the effective, cycle-averaged, band-structure and the Berry curvature in each valley. Exponential sensitivity of multi-photon excitation to the effective bandgap naturally leads to selective excitation in the valley where the bandgap is reduced. We find that rotating the Lissajous figure drawn by the electric field vector of the pulse (Fig. 1b), relative to the lattice (Fig. 1c), controls the magnitude and the phase of the light-induced Haldane-type hoppings, thus controlling the cycle-averaged band structure (Fig. 1d). Thus, valley selection is achieved by tailoring the symmetry of the Lissajous figure to the symmetry of the lattice.

Using light to induce topological properties in solids has led to the concept of topological Floquet lattices generated in the Hilbert space of a laser-dressed quantum system. In this context, our key results are as follows. First, we find that strong circularly polarized fields in the low-frequency regime show opposite valley polarization than those in the high-frequency, resonant regime, extending results obtained for atoms to 2D solids. Second, we initialize and manipulate valley polarization on a few femtosecond time scale in a way that remains consistent for a broad range of frequencies and field intensities, and independent on the specifics of the material. We give two examples, hexagonal boron nitride (hBN) and MoS$_2$. Finally, using an additional linearly polarized probe pulse, we map the valley pseudospin onto the polarization of its harmonics, providing an all-optical measure of the valley asymmetry.
Figure 1: **Light-induced modification of the band structure with strong tailored field.**

(a) Representation of a 2D hexagonal lattice with broken inversion symmetry in real space (left), where red and green represent two different atomic species in the two triangular sub-lattices, and in reciprocal space (right), showing the two valleys, $K$ and $K'$. (b) Strong tailored light field with the symmetry of the sub-lattice, made by the combination of a circularly polarized fundamental field and its counter-rotating second harmonic. The pulse can be rotated by means of the two-color phase delay between the two pulses, $\varphi$. (c) Depending on the orientation of the tailored field (grey trefoil) with respect to the lattice, the two atomic sites are addressed differently by the field (see, e.g., the two atoms inside the trefoil of the pulse). For $\varphi = -\pi/2$, the field interacts equally with the red and green atoms, and the bands show the same structure as in the field-free case, i.e., valley-degeneracy (d, black solid line). For $\varphi = 0$, the field interacts differently with the two atoms (note how the two atoms inside the trefoil are now not interchangeable, irrespective of where the field is placed in the lattice). This leads to a lifting of the valley degeneracy (d, red dashed line). For $\varphi = \pi$, the latter situation is reversed (d, blue dashed dotted line).
We first consider driving excitation with strong circularly polarized fields with frequencies well below the band gap energy. In Fig. 2a,b,c we show the electron populations in the $p_z$ conduction band of hBN after applying a strong ($I = 5 \text{ TW/cm}^2$) circularly polarized field with three different frequencies and the same helicity. The same observable can be obtained, e.g., by angularly-resolved photoemission spectroscopy (ARPES). The most excited valley switches as we transition from the highest frequency (corresponding to multi-photon excitation, Fig. 2a) to the lowest frequency (corresponding to tunneling excitation, Fig. 2c). All panels switch $K$ for $K'$ when the helicity of the laser is reversed (not shown).

The switch in valley polarization in the tunneling regime is reminiscent of the tunnel ionization propensity rule encountered in atomic and molecular systems. There, circularly polarized fields favours depletion of electrons in orbitals that counter-rotate with the field\textsuperscript{32,33}, as opposed to one- or multi-photon ionization, that favours excitation of electrons from co-rotating orbitals. Fig. 2a,c shows that this propensity rule also applies in the context of valleytronics. We thus identify the orbital propensity rule as one mechanism for valley asymmetry in strong, low-frequency helical fields.

This result suggests that one can control the valley polarization in low-frequency fields by controlling the ellipticity of the laser field and, while it may not be trivial to do so in the mid-IR/terahertz range, it is still within experimental reach. However, this approach depends crucially on the relation between the band gap and the frequency of the field, as evidenced in Fig. 2a,b,c, and it is thus material-specific. Using smaller laser frequencies will not solve the problem since,
on the one hand, it will drive some materials into the quasi-static regime (where no propensity rule of valley excitation is present), and on the other hand, it will lead to field oscillation periods comparable to or higher than the valley lifetime.

To solve this problem, we propose to modify and control the cycle-averaged band structures by using tailored fields with the symmetry of the triangular sub-lattices. The most widely used field with such symmetry is the bicircular field\textsuperscript{4,34,35}, composed of the superposition of a circularly polarized field of frequency $\omega$ with its counter-rotating second harmonic $2\omega$, 

\[ F_L = \hat{x} \left[ -F_1 \cos(\omega t) + F_2 \cos(2\omega t + \varphi) \right] + \hat{y} \left[ F_1 \sin(\omega t) + F_2 \sin(2\omega t + \varphi) \right], \]  

(1)

where $F_1$ and $F_2$ are the field strengths of the fundamental and second harmonic drivers, respectively, and $\varphi$ is the sub-cycle phase delay between the two drivers. During one cycle, the field draws the trefoil shown in Fig 1b. Its orientation relative to the lattice is controlled by $\varphi$. The ability to change this geometry during the pulse implies the ability to switch the valley pseudospin on the fly.

Fig. 2d,e,f shows the valley polarization in hBN at fixed $\varphi$, for three different frequencies. In contrast to excitation by circular pulses, now the most populated valley remains robust as we transition from the multi-photon to the tunneling regime (Fig. 2d,e,f). Moreover, reversing the helicity of the two drivers ($\omega$ and $2\omega$) does not switch the valley polarization, suggesting that a different mechanism for valley polarization is playing a role in the bicircular case.

As shown in the Methods section, energy conserving processes involving both $2\hbar\omega$ and $\hbar\omega$
Figure 2: **Strong-field valley excitation.** (a-c) Electron populations in the first Brillouin zone (red dashed hexagon) of the conduction band of monolayer hBN (band gap of $\Delta = 5.9$ eV) after applying a strong, circular pulse of different frequencies: (a) $\omega = 0.95$ eV, (b) $\omega = 0.68$ eV and (c) $\omega = 0.41$ eV. All other parameters (helicity, peak field strength $F_L = 0.012$ a.u. and duration $\tau = 200$ fs) remain the same. The most populated valley changes as we transition to the low-frequency regime, and all panels switch $K$ for $K'$ when the helicity is reversed (not shown). Panels (d-f) show the same as panels (a-c), but for a bicircular pulse with the same $F_L$, $\tau$ and fundamental frequency $\omega$ as the corresponding panels above. The most populated valley now remains the same for the high frequency (d) and low-frequency case (f), and reversing the overall helicity of the field does not switch $K$ to $K'$ (not shown), indicating a different mechanism for valley polarization.
photons, such as absorption of one $2\hbar\omega$ photon and re-emission of two $\hbar\omega$ photons, lead to complex second-neighbor hopping $t_2$. Its real part is controlled by the field strengths, while the imaginary part is controlled by the two-color phase $\varphi$. A non-zero value of the latter lifts the valley degeneracy. For moderately strong fields (see Methods), the cycle-averaged, laser-induced second neighbour hopping acquires the imaginary component

$$\mathcal{J}\{t_2\} \sim 2J_2\left(\frac{\sqrt{3}a_0 F_1}{\omega}\right)J_1\left(\frac{\sqrt{3}a_0 F_2}{2\omega}\right)\cos\varphi,$$

(2)

where $a_0$ is the lattice constant and $J_n$ is the Bessel function of the first kind of order $n$. Thus, the sub-cycle control over the field geometry, $\varphi$, controls the topological properties of the dressed system. This modification of the band structure is the dominant mechanism contributing to the valley asymmetry for fields with the symmetry of the sub-lattice. In this way, tailored fields allow to switch the valley pseudospin in a few-cycle timescale and in a way that is not material specific.

To illustrate this, we consider monolayer hBN (see Methods for numerical details) and use a bicircular pulse with fundamental frequency $\omega = 0.41$ eV ($\lambda = 3000$ nm), duration of $\tau = 200$ fs, intensity ratio of $I(\omega)/I(2\omega) = 4$, and maximum peak intensity of $I = 5$ TW/cm² (lower than its predicted damage threshold). We change the two-color phase $\varphi$ to control the orientation of the field with respect to the lattice. When $\varphi = -\pi/2$, the field addresses equally both atoms in the lattice, and no valley degeneracy is present in the cycle-averaged band structures (Fig. 3a). Consequently, we find that electrons populate approximately equally the $K$ and $K'$ valleys of the conduction band, leading to a negligible valley polarization. In sharp contrast, when $\varphi = 0$, the cycle-averaged band structure shows a strong valley asymmetry (Fig. 3b). This reflects in the valley populations, that show a 60% contrast. Half a $2\omega$ cycle later, when $\varphi = \pi$, the situation reverses.
and the populations switch to the opposite valley (Fig. 3d). The sense of rotation of the pulse also contributes to the valley asymmetry due to the orbital propensity rule, but its effect is weak. It manifests as a small valley polarization for $\varphi = \pm \pi/2$, and in the fact that $\varphi = 0$ and $\varphi = \pi$ are not exact opposites (compare panels b,d of Fig. 3).

To show that the results above are not system-specific, we performed calculations in MoS$_2$ with the same laser frequencies, which allows us to maintain the same timescale of valley control. Due to the smaller band gap of MoS$_2$, the intensity peak of the total field was kept at $I = 0.15$ TW/cm$^2$, well below its damage threshold. Fig. 3(e)-(h) shows the electron populations after the pulse in the spin-integrated lowest conduction band of MoS$_2$ (see Supplementary Information for spin-resolved), illustrating a similar control as for hBN, but with lower values of the valley polarization. Higher values of valley polarization can be obtained by increasing the field strength or wavelength, while total populations can be controlled with the pulse duration.

We now turn to optical reading of the valley pseudospin. Since the Berry curvature is opposite at the $K$ and $K'$ valleys, when an in-plane electric field is applied, the carriers generate a current perpendicular to the electric field (anomalous current), with opposite direction at each valley. For equal valley populations, the anomalous current will thus cancel, leading to a zero anomalous Hall conductivity (AHC),

$$\sigma_{xy} = -\frac{e}{h} \sum_n \int_{BZ} \frac{d^3k}{(2\pi)^3} f_n(k) \Omega_{n,z}(k),$$

where $f_n$ and $\Omega_n$ are the population and Berry curvature of the $n$-th band. If the valley populations are not equal, the perpendicular currents originating from $K$ and $K'$ do not compensate each other,
Figure 3: **Strong-field manipulation of valley polarization.** Electron populations in the lowest conduction band of hBN (top panels) and (spin-integrated) MoS$_2$ (lower panels), after applying a bicircular field with a fundamental wavelength of 3 µm, at four different values of $\varphi$: (a,e) $\varphi = -\pi/2$, (b,f) $\varphi = 0$, (c,g) $\varphi = \pi/2$ and (d,h) $\varphi = \pi$ (sketch of the relative orientations of the field with respect to the hexagonal cell are shown above correspondingly). The insets at the center of panels (a)-(d) show the cycle-averaged band structures for hBN calculated using the laser-induced hoppings $\tilde{t}_1$ and $\tilde{t}_2$ (given in Methods), which vary as a function of $\varphi$. 
leading to a non-zero AHC \cite{25}. This so-called valley Hall effect has been demonstrated for MoS\textsubscript{2} monolayers by measuring the direction of the transverse Hall voltage \cite{27}. Alternatively, the sign of the anomalous current can be retrieved all-optically from the helicity of the harmonics of a linearly polarized probe \cite{28}.

To read the valley polarization all-optically, we use a probe field linearly polarized along the \textbf{G}-\textbf{M} direction of the lattice, carried at a 3N multiple of the bicircular fundamental frequency \( \omega \). This guarantees a background-free measurement, i.e., without interferences of the harmonics generated by the bicircular field, since the latter does not generate \( 3N\omega \) harmonics due to symmetry. We show that probe frequencies of both \( \omega \) and \( 3\omega \) work. The probe field is introduced after the interaction with the bicircular pump pulse that generates the valley polarization. The harmonics emitted parallel to the probe field are not affected by the different valley populations at \( \textbf{K} \) and \( \textbf{K}' \), while those emitted perpendicularly undergo a phase jump of \( \pi \) as the population switches from one valley to another (see Methods), leading to a rotation in the helicity of the harmonics. When the dynamics occur mainly in one conduction band, as the case of monolayer hBN, the harmonic helicity is able to read the AHC and, consequently, the valley polarization (Fig. 4a). In the case of MoS\textsubscript{2} (Fig. 4b), the helicity follows qualitatively the valley polarization, but the agreement with the AHC is not as good due to the influence of higher bands.

In conclusion, we have shown that strong-field valleytronics is a solid alternative to schemes based on resonant excitation. We have found that valley polarization with circularly polarized fields is opposite in the low-frequency regime, which extends the orbital-dependent propensity rule of
Figure 4: **Optical reading of valley polarization.** Valley population asymmetry (dotted solid blue curve), helicity of the third harmonic of the probe (faint blue curve) and anomalous Hall conductivity (faint red curve), as a function of $\varphi$ (for a full rotation of the trefoil), in hBN (left panel) and MoS$_2$ (right panel). The valley population asymmetry is calculated as $A = \pm 2(f_{n,K} - f_{n,K'})/(f_{n,K} + f_{n,K'})$, where $f_{n,K}$ ($f_{n,K'}$) is obtained by integrating the electron population inside the black dashed circles encircling $K$ ($K'$) in Fig. 3 and the $+$ ($-$) sign is used for hBN (MoS$_2$), due to their opposite Berry curvatures (see inset). The helicity is calculated as $h = 2(I_\circ - I_\circ)/(I_\circ + I_\circ)$, where $I_\circ$ ($I_\circ$) is the component of the harmonic intensity rotating clockwise (anticlockwise). The probe field is carried at $3\omega$ frequency for hBN and $\omega$ frequency MoS$_2$, where $\omega$ is the fundamental frequency of the bicircular pulse.
tunnel ionization in atoms and molecules to valleytronic devices. Using strong, low-frequency fields with the triangular symmetry of the sub-lattice, we have been able to initialize a valley polarization and manipulate it within a timescale much shorter than the reported valley lifetimes, and without requiring resonant conditions. We have shown that this a consequence of a light-induced complex second neighbour hopping that modifies the cycle-averaged band structures, opening the way to non-resonant light-induced topological phase transitions controlled at few-cycle rates. Moreover, we have shown that the valley polarization and the anomalous Hall conductivity are mapped onto the polarization properties of the light emitted by a linearly polarized probe pulse, demonstrating that high harmonic generation can be used to read the light-induced anomalous Hall conductivity and valley polarization in an all-optical way. Our work lays the grounds of a new regime for valleytronics and, possibly, light-induced topology.

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