Floquet engineering is a powerful tool to modify materials by coupling them to periodic light. Traditionally, amplitude and frequency are varied, but the polarization can be tuned to explore a larger phase space. We consider both polarized and several kinds of unpolarized light on insulating magnetic materials, showing that varied polarization protocols enhance different exchange couplings. As an illustration, we couple the triangular lattice Hubbard model at half-filling to periodic light with several polarizations and discuss how to alternately induce Dirac and chiral spin liquids.

Floquet engineering is a powerful tool that allows access to phases and phenomena not realizable in equilibrium, typically using periodic light to drive the system [1–8]. The laser light used generically breaks a symmetry, as its polarization is fixed. Sometimes this symmetry breaking is useful, as new couplings like chiral fields are generated [9–13], or anisotropies and dimensionalities tuned [14–18]. However, sometimes we want to preserve the original symmetries. The degree and nature of the symmetry breaking may be controlled by using unpolarized or partially polarized light. We discuss this role of polarization in tuning magnetic exchange interactions in Mott insulators, and show how different, realistic polarization protocols can give significantly different exchange couplings, even with the same symmetries. As an example, we treat the triangular Hubbard model at half-filling and show how different protocols tune the relative strengths of the Heisenberg exchange couplings, as well as induce chiral fields. In particular, it is possible to boost the ratio of the Heisenberg exchange couplings, as well induce chiral fields. Figure 1. Monochromatic light has a fixed polarization described by a point on the Poincaré sphere. This point is captured by two angles, χ and ψ, as shown. The axes correspond to the Stokes parameters, which describe the degree of horizontal/vertical (S₁), ±45° polarization (S₂) and circular (S₃) polarized. The polarization vector can trace out various paths on the Poincaré sphere, and different kinds of unpolarized light are generated by sampling certain regions of the Poincaré sphere such that ⟨S⟩ = 0, including type II Glauber, which samples the equator and type II ±χ₀, which samples circles at ±χ₀ equally. Type I light samples the entire sphere.

A periodic path on the Poincaré sphere with characteristic time, T₀ = 2π/Ω₀ ≫ T = 2π/Ω, such that the time average of the Stokes parameters is zero, ⟨S⟩ = 0 [20, 29–36].

Practically, these protocols may be implemented using two or more laser frequencies with varying degrees of correlation. Alternately, a uniformly polarized beam may be passed through an optical element called a depolarizer that causes the polarization to vary rapidly over the spot size of the beam, such that the spatial average ⟨S⟩ = 0 [37–39]. Different protocols create unpolarized light differentiated by higher-order correlators of the Stokes parameters, ⟨SᵢSⱼ⟩, (SᵢSⱼSₖ), etc[40], which must also preserve lattice and time-reversal symmetries. Note that we neglect the time dependence given by the trajectory on the Poincaré sphere; we shall show that this can
be made arbitrarily small.

Exchange couplings are sensitive, ultimately, to all higher-order correlators, and so will vary with the type of unpolarized light. The magnetic exchange couplings \( J_{ij} \) for a given protocol are found by averaging the polarized results over the polarization distribution, \( f(\chi, \psi) \),

\[
\langle J_{ij} \rangle = \frac{\int_{-\pi/4}^{\pi/4} d\chi \int_0^{\pi} d\psi \cos 2\chi f(\chi, \psi) J_{ij}(\chi, \psi)}{\int_{-\pi/4}^{\pi/4} d\chi \int_0^{\pi} d\psi \cos 2\chi f(\chi, \psi)} \tag{3}
\]

We fix the intensity, but it may vary, as for natural light \[29\].

To preserve lattice and time-reversal symmetries, we demand that polarization distributions be invariant under rotations and have zero net chirality. Such distributions generate “type II” light \[41\]. Of particular interest is type II Glauber light, which samples all LPs equally, encompassing the equator of the Poincaré sphere. Generic type II light might be constructed from superpositions of distributions with circles at \( \chi = \pm \chi_0 \), \( f(\chi, \psi) = \frac{1}{2} [\delta(\chi - \chi_0) + \delta(\chi + \chi_0)] \). Type I light is even more restrictive, sampling the Poincaré sphere uniformly, \( f(\chi, \psi) = 1 \) \[41\]. Fixed intensity type I light is known as amplitude-stabilized unpolarized light, while natural light has a varying intensity, \( f(I, \chi, \psi) = \frac{2}{I_0} \exp(-2I/I_0) \) \[42\]; for magnetic exchange couplings, these give identical results. It is possible to generate nearly monochromatic type II Glauber \[31\] and type I light \[30, 32\] either using spatial depolarizers or by superimposing slightly frequency detuned incoherent laser beams with orthogonal polarizations.

To illustrate the effect of polarization, we now examine magnetic exchange couplings in a single band Floquet-Hubbard model; more realistic superexchange models will feature similar physics. We consider electrons hopping on a lattice in the presence of a time-dependent electric field, \( E = -\frac{\partial A}{\partial \theta} \), with period \( T = 2\pi/\Omega \). There is a strong penalty for double occupancy, \( U \):

\[
\mathcal{H}_0 = -t_1 \sum_{i, \delta_i} e^{-iA(t)} \delta_i c_i^\dagger c_{i+\delta_i} + U \sum_i n_{i\uparrow} n_{i\downarrow}. \tag{4}
\]

We consider only nearest-neighbor links labeled by \( \delta_i = (\cos \phi_i, \sin \phi_i) \). In the time-independent limit, we can expand in \( U \) either using Brillouin-Wigner perturbation theory \[43, 44\] or a Schrieffer-Wolff transformation \[45\]. On the triangular lattice, to second order, there is only the nearest-neighbor term \[46\], \( J_1 = 4t_1^2/U \), but fourth order terms give corrections to \( J_1 = 4t_1^2/U - 28t_1^4/U^3 \) \[47\], as well as second and third neighbor couplings, \( J_2 = J_3 = 4t_1^4/U^3 \) and a ring exchange term, \( J_{\square} = 80t_1^4/U \) \[47–49\].

The time-periodic nature of the Hamiltonian allows us to Fourier-transform to Floquet space, with the discrete set of frequencies \[46, 50–52\], \( m\Omega, m \in \mathbb{Z} \). In this space, the electrons now hop not just between sites, but between Floquet sectors labeled by \( |m| \) \[1, 52\], as shown in Fig. 2 (a),

\[
\mathcal{H} = -\sum_{m, n, i, \delta_i} t_i^{(n-m)} c_i^\dagger c_{i+\delta_i} |m\rangle \langle n| + \sum_m \left( U n_{i\uparrow} n_{i\downarrow} + m\Omega \right) |m\rangle \langle m| \tag{5}
\]

The double occupancy penalty becomes \( U + m\Omega \), with resonances at \( m = -\Omega/U \). The hopping between sectors is given by the Fourier transform (with \( \theta = \Omega t \)), \[52\]

\[
t_i^{(m)} = \frac{t_1}{2\pi} \int_0^{2\pi} d\theta e^{-i\theta} e^{-i\delta_i \cdot A(\theta)}. \tag{6}
\]

The integral gives the Bessel function expression,

\[
t_i^{(m)} = t_1 e^{im(\beta_1 + \pi)} J_{m}(A_1), \tag{7}
\]

where the real space orientation, \( \delta_i \) is incorporated via the amplitude, \( A_1 \) and angle, \( \beta_1 \),

\[
A_1 = A_0 \sqrt{1 + \cos 2\chi \cos [2(\psi - \phi_i)]} \cos \beta_1 = \frac{\sqrt{2} \sin \chi \sin [2(\psi - \phi_i)]}{\sqrt{1 + \cos 2\chi \cos [2(\psi - \phi_i)]}}. \tag{8}
\]

Here, we introduce the dimensionless average fluence \( A_0 = \frac{1}{\pi} \sqrt{t/2} \). Notice that \( A_1 \) is symmetric with respect to \( \chi = 0 \), while \( \beta_1 \) is \( \pi - \beta_1 \) as \( \chi \to -\chi \), which explains the lack of time-reversal symmetry breaking in distributions that sample \( \pm \chi \) equally.

Now we can again calculate the exchange couplings, with modified hoppings and \( U + m\Omega \) denominators. We have done our calculations via Brillouin-Wigner perturbation theory\[49\]. The nearest-neighbor coupling is straightforward\[13, 45, 53, 54\],

\[
J_1^{(\delta_i)} = 4 \sum_m \frac{t_i^{(m)} t_i^{(-m)}}{U + m\Omega} = 4t_1^2 \sum_m \frac{|J_{m}(A_1)|^2}{U + m\Omega}. \tag{9}
\]

Figure 2. (a) Electrons hop between different sites and Floquet sectors, labeled by \( m = 0 \) and \( m = -1 \) shown here. (b) The Hubbard bands are broadened by the hopping of holons and doublons, with bandwidth \( 2\gamma_1 \). To avoid exciting electrons between the bands, the photon frequency \( \Omega \) must be less than the Mott gap, \( U - 2\gamma_1 \), but greater than the overall bandwidth, \( U + 2\gamma_1 \).
The Bessel functions cause $J^{|A_1|}$ to rise to a maximum as a function of fluence, $A_0$, and then oscillate as it decays, as shown in Fig. 3 (b). Note the dependence upon polarization through $A_i$, which can tune the anisotropy of the lattice. Large enhancements may be found by tuning close to the resonances at $U = -m\Omega$, however, heating becomes a concern. Higher-order contributions are more complicated, as superexchange paths proliferate; third-order terms vanish, while fourth-order terms on the triangular lattice are derived in the supplemental material \[64\]. Imaginary hopping terms, if present, generate chiral fields, $J^{|\Delta|}$, where we assume that the polarization and original frequency, $\psi$, precesses with frequency $p\Omega$. We FIX $t_1 = U/(6\gamma)$ and $\Omega = 2/3$ to avoid heating, while maximizing the enhancements. Sufficiently far from resonance, there is minimal heating even for relatively large fluence \[13\], and the Bessel function nature of $t^{(m)}$ means that the enhancement is largest for $A_0$ of order one.

As the Floquet formalism assumes monochromatic light, it is not obvious that time-averaging the polarization gives correct results. A previous work suggests that the Floquet formalism can still be used at least with some types of unpolarized light, as discussed for non-interacting graphene \[64\]. To show that polarization averaging works explicitly, here we consider linearly polarized light with frequency $\Omega$, where the angle of polarization, $\psi$ precesses with frequency $\Omega_p = 2\pi/T_p$. We proceed as before, now with the Floquet frequency $\Omega$, where we assume that the polarization and original frequencies are commensurate. We recover the results for polarization averaged monochromatic light for $T_p \gg T$, as the contributions become effectively broadened by the presence of a second, slower frequency.

Here, we fix the polarization and later average to find the desired unpolarized result.

As a practical concern, we want to enhance the frustrating further neighbor couplings without significantly heating the system by exciting pairs of holons and doublons \[55-60\]. The very resonances that allow large enhancements also lead to problematic heating when one or more photons can excite electrons across the gap \[61, 62\]. To show that polarization, $\psi$, precesses with frequency $p\Omega$, we must keep $\Omega < -U - 2\gamma t_1$, as shown in Fig. 2 (b). However, we also must insist that two photons cannot excite electrons between Hubbard bands, $2\Omega > U + 2\gamma t_1$ \[63\]. This restriction severely limits what materials may effectively be tuned, as only strongly insulating materials with $t_1 < U/(6\gamma)$ may be pumped with $\Omega < -U - 2\gamma t_1$ light without significant heating. For illustration, we fix $t_1 = U/(6\gamma)$ and $\Omega = 2/3$ to avoid heating, while maximizing the enhancements. Sufficiently far from resonance, there is minimal heating even for relatively large fluence\[13\], and the Bessel function nature of $t^{(m)}$ means that the enhancement is largest for $A_0$ of order one.

The electric field for this scenario is,

$$E(t) = E_0 \left( \frac{\cos \Omega_p t}{\sin \Omega_p t} \right) \Re[e^{-i\Omega t}] = \frac{E_0}{2} \Re \left[ \left( \frac{1}{i} \right) e^{-i\Omega_p t} + \left( \frac{1}{-i} \right) e^{-i\Omega t} \right]. \quad (10)$$

Time-reversal is clearly broken via the precession of the polarization, which leads to a sum of terms with opposite circular polarizations and frequencies, $\Omega_{\pm} = \Omega \pm \Omega_p$. The electric field is periodic with $T_p = NT$ with $N \in \mathbb{Z}$. We now Fourier-transform to Floquet space as before, with frequencies $m\Omega_p$. The calculation proceeds similarly, but now the vector potential, $A(t)$ has two contributions with frequencies, $\Omega_{\pm}$ and amplitudes, $A_{\pm} = \frac{E_0}{\Omega_p}$, $\equiv A_0(1 \pm N^{-1})^{-1}$. The hoppings between sectors are,

$$t_{N=1}^{(m)} = \frac{t_1}{2\pi} \int_0^{2\pi} d\theta e^{-im\theta} e^{iA_+ \sin \theta_+ + iA_- \sin \theta_-}, \quad (11)$$

where $\theta = \Omega t$, and $\theta_\pm = \theta \pm \phi_0$. We perform the integral by twice decomposing $\exp(ix \sin \theta) = \sum_{m'} J_{m'}(x)e^{im\theta}$ into a sum over Bessel functions with different phase factors. $m'$ ranges over all integers. The
\[ t_{i,i+\delta \phi}^{(m)} = t_1 \sum_{m_1,m_2=-\infty}^{+\infty} J_{m_1}(A_+)J_{m_2}(A_-)e^{-i(m_1-m_2)\phi_i} \times \delta_{m-N(m_1+m_2)+m_2-m_1}. \]  

We can now calculate the exchange couplings as before, but with \( \Omega \) replaced by the much smaller \( \Omega_p \). The sum can be calculated numerically for any \( N \), with some \( J_1 \) results shown in Fig. 3 (b). It quickly converges to the average of Eq. (9) over all linear polarizations Eq. (3), for \( N \gg 10 \). This convergence is not intuitive, as we have resonances at \( U = -m\Omega_p \), while the polarization averaged result has far fewer resonances at \( U = -m'\Omega \).

However, the hoppings themselves are dominated by contributions from small \( k/N \). Thus, we can neglect the contributions from deviations from \( m \) very close to integer multiples of \( N \), such that we can define \( m = N\hat{m}+k \), with \( k \) of order one. In the large \( N \) limit, where the sums of Eq. (12) can be truncated, \( A_+ \approx A_- \approx A_0 \), and the hoppings generically take the form,

\[ J_1 = J_2 \text{ is again found by perturbation theory in the excited energies, } U + (N\hat{m}+k)\Omega_p. \]  

As the numerators are dominated by small \( k/N \), we can neglect \( k \) in the denominator,

\[ J_1^{(\delta \phi)} = 4 \sum_{\hat{m},k} t_1^{(N\hat{m}+k)} t_1^{(-N\hat{m}-k)} \frac{\sum_{\hat{m}} f_{\hat{m}}^k}{U + (N\hat{m}+k)\Omega_p} \approx 4t_1^{2} \sum_{\hat{m}} \frac{\sum_{k}|f_{\hat{m}}^k|^2}{U + \hat{m}\Omega_p}. \]  

Thus, for \( T_\phi \gg T \), we can use the original Floquet method and simply average over the polarization distributions.

To demonstrate how varying the polarization protocol can drive materials through different regions of phase space, we explicitly consider the triangular lattice. It provides an apt example, as, in principle, multiple spin liquids are accessible by tuning through different directions in phase space. While the nearest neighbor model has 120° order, there is: a Dirac spin liquid for \( J_2/J_1 \geq 0.1 \); a chiral spin liquid for either \( J_3/J_1 \geq 0.2 \) and \( J_2 = 0 \) or \( J_3/J_1 \geq 0.025 \) for \( J_2/J_1 \sim 0.1 \); and a spinon Fermi surface as \( J_3/J_1 \geq 0.2 \).

We calculated the enhanced couplings to fourth order in Brillouin-Wigner perturbation theory (details in the supplemental material). \( J_2/J_1 \) is maximally enhanced either by type I light; a type II light that consists only of equal parts LCP and RCP light; or CP light, which also generates chiral fields. We show both the absolute changes for CP light with \( \Omega = 2U/3 \), where the enhancement is largest. \( J_2/J_1 \) and \( J_3/J_1 \) can be enhanced by 0.03 and 0.01, respectively. These may seem small, but are nearly 2000% and 500% of the equilibrium values, as shown in (b), and are a significant fraction of the fraction of the \( J_2/J_1 \) required for the Dirac spin liquid. The effective chiral field reaches \( \sim 0.05J_1 \) [49], again a significant fraction of the critical field. Ring exchange, \( J_2/J_1 \) ranges between \(-0.09 \) and \( 0.02 \); positive values eventually induce a spin liquid, but must be ten times larger.

(b) shows how different types of unpolarized light drive different paths through phase space, given in terms of the relative enhancement. The initial, equilibrium point \( J_1 = 0 \) is indicated by a dot. Type I light (blue) samples the Poincaré sphere evenly; type II Glauber light (red) samples all linearly polarized light equally; and type II LCP/RCP (green) samples only the poles of the Poincaré sphere, such that there are no chiral fields. Note that the CP light used in (a) gives identical results to type II LCP/RCP for \( J_1, J_2, \) and \( J_3 \).
with different protocols. The absolute changes give an incomplete picture, as the equilibrium values are tiny for the small $t_1/U$ required to avoid heating; the enhancement of $J_2/J_1$ can be as large as 2000%.

Unique paths through the $J_2/J_1 - J_3/J_1$ phase space are traced out by different protocols, as shown in Fig. 4(b), where the ratio $J_3/J_2$ can be tuned by a factor of two. Minimizing $J_3$ is essential to accessing the Dirac spin liquid, as $J_3$ increases the critical $J_2$ [26], and so type I or CP light is more favorable than type II Glauber. Note that while we only show the two extremes of type II light ($\chi = 0, \pm \pi/4$), all type II light lies between these.

In this paper, we have shown that the laser polarization provides a key untapped tuning parameter for Floquet engineering, particularly for strongly interacting materials, which are sensitive to higher order correlations in the polarization. We illustrated this effect on magnetic ex-

We acknowledge useful discussions with Thomas Iadecola, Eduardo Miranda, Peter Orth, Paraj Titum, Thais Trevisan, Chirag Vaswani, and Jigang Wang. V.L.Q and R.F. and were supported by NSF through grant DMR-1555163.

* vquito@iastate.edu

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We must also take care to remain in the regime where perturbation theory makes sense, as the fourth order corrections to $J_1$ can potentially drive it negative for some range of fluences.

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In this section, we discuss the different time scales, frequencies and fluences involved, and discuss experimental feasibility. Here, our degrees of freedom are spins, with interaction scale $J_1 = 4t_1^2/U$, that are experiencing a pulse of light (duration, $T_{\text{pulse}}$) of frequency $\Omega = 2\pi/T$. We assume that the polarization vector oscillates with period $T_p \gtrsim 10T$, such that polarization averaging is expected to be reasonable. The spins will feel the nonequilibrium exchange couplings and relax to their new low energy state within a time scale roughly given by $T_{\text{rel}} \sim 1/J_1$. In order for these spins to “feel” the unpolarized $J$’s, we require $T_{\text{rel}} \gg T_p$. The spins must relax to their new states within the pulse, and be measured. So, most generously, we require $T_{\text{pulse}} \gg T_{\text{rel}} \gg T_p \gg T$. In order to maximally enhance the exchange couplings, $\Omega = 2U/3$, and $t_1 = U/(6\gamma)$, where $\gamma = 2\sqrt{5}$ for the triangular lattice. These time scales can be well separated, with perhaps the most stringent requirement being for the pulse length required to allow the spins to relax,

$$T \sim \frac{1}{U} \ll T_p \sim \frac{10}{U} \ll T_{\text{rel}} \sim \frac{100}{U} \ll T_{\text{pulse}}. \quad (15)$$

These laser frequencies will need to be tuned to the Mott gap, and so are expected to be on the order of electron volts, in the visible range. $T_p$ will therefore be on the order of 10fs, while $T_{\text{rel}} \sim 100$fs, requiring a moderately long pulse. Note that here we consider only how photons affect the electronic degrees of freedom in this single band Hubbard space directly, while in general photons can interact with collective modes, like phonons, or excite electrons into other bands, which may cause additional heating or affect the magnetism more directly [67].

The dimensionless vector potential amplitude can be estimated by restoring the units,

$$A_0 = \frac{a_0 eE}{\Omega \hbar}, \quad (16)$$

where $a_0$ is the lattice spacing, of the order of Angstroms. This amplitude is connected to intensity, with full units, according to

$$I = c\epsilon_0 \left( \frac{\Omega \hbar}{ea_0} \right)^2 |A_0|^2 = 2.6 \times 10^{17} \left( \frac{\Omega \hbar[eV]}{a_0[A]} \right)^2 |A_0|^2 \text{W/m}^2 \quad (17)$$

with $\epsilon_0$ the vacuum permittivity. The electric field strength, $eE$ varies in different experiments, typically ranging from $(0.01 - 1) \text{eV/Å}$ [68, 69], giving to intensities of $I \approx 10^{15} - 10^{17} \text{W/m}^2$. In these experiments, $A_0$ ranges between 0.01 and 1; the slightly larger values of $\sim 2$ that we require are not unreasonable. However, as lasers provide constant power that can be chopped into pulses, either shorter pulses with larger fluences, or longer pulses with lower fluences, at the moment the two requirements of relatively high fluence and relatively long pulses are at odds, given current technology.

In addition to driving the system into a nonequilibrium state, the state itself must be measured via some optical measurements. Ordered phases should be more or less straightforward, as a phase transition should give a clear signal in optical quantities, however, we are proposing to drive materials into spin liquid regions that do not exist in equilibrium materials. Here, the absence of a phase transition would just be the minimal requirement for realizing a spin liquid. Electromagnetic gauge fields do interact with the neutral spinons, albeit often with significantly lower amplitudes than electrons. Gapless spin liquids are predicted to have power-law behavior of the optical conductivity [70], with some evidence in herbertsmithite and others[71, 72], and liquids may have signatures in the magneto-optical Faraday or Kerr effects [73].

### B. Definition of the magnetic exchange couplings

In this section, we define the exchange couplings of the effective spin Hamiltonian for the triangular lattice. The nearest-neighbor vectors are shown in Fig. 2(a) of the main text, and are given by

$$\delta_1 = \left( \frac{1}{2}, \sqrt{3}/2 \right), \ \delta_2 = (1, 0), \ \delta_3 = \left( \frac{1}{2}, -\sqrt{3}/2 \right). \quad (18)$$

The distinct exchange terms are shown in Fig. 5 yielding the Hamiltonian.
\[
H^{(m\Omega\leq U)}_{\text{spin}} = \sum_{\langle i,j \rangle} J_1^{(i,j)} S_i \cdot S_j + \sum_{\langle \langle i,k \rangle \rangle} J_2^{(i,j,k)} S_i \cdot S_k + \sum_{\langle \langle\langle i,m \rangle \rangle\rangle} J_3^{(i,j,k)} S_i \cdot S_m + \sum_{\langle\langle\langle\langle i,l \rangle \rangle\rangle\rangle} J_4^{(i,j,k,l)} P^{(i,j,k,l)} + J_5^{(i,j,k,l)} P^{(i,j,k,l)} - J_6^{(i,j,k,l)} P^{(i,j,k,l)}
\]

The couplings \( J_1, J_2 \) and \( J_3 \) are the nearest, next-nearest and third-neighbor couplings. \( J_4 \)'s are the ring exchange terms that, in our notation, multiply the 4-body operators

\[
P^{(i,j,k,l)} = (S_i \cdot S_j) (S_k \cdot S_l),
\]

the product of all the spin operators around a given plaquette. For any choice of polarization average that keeps the lattice symmetries,

\[
J^{(i,j,k,l)} = J^{(l,i,j,k)} = J^{(i,k,j,l)}.
\]

The chiral couplings come in two flavors, shown in Fig. 5 (d) and (e). In (e), the electron hops around a closed lattice triangle, while in (d), the three sites form an open path. We call \( J_4^a \) the processes coming from (d) and \( J_4^b \) the ones coming from (e). It becomes natural to find the net chirality of a triangle, by distributing the different fluxes coming from the two terms. Considering four sites forming a parallelogram, like the one shown in (b), the net flux consists of adding two fluxes of (d) and two fluxed of (e). This parallelogram is made of two triangles, implying that the coupling that controls the effective chirality is \( J_4^a + J_4^b \). This is used as the reduced variable in the main text.

Figure 5. Representation of all the sites involved that lead to exchange couplings in fourth order in \( t \) on the triangular lattice, with the bond sites represented in red. (a) \( J_1 \) (b) \( J_2 \) (c) \( J_3 \) (d) \( J_4^a \) (e) \( J_4^b \) (f) \( J_5 \).

C. Magnetic exchange couplings on the triangular lattice

We now present the main features of the perturbative expansion leading to the effective magnetic exchange couplings shown in the main text; an expanded calculation will be shown elsewhere [74]. This calculation can be done two ways, following the Brillouin-Wigner [43] or Schrieffer-Wolff [45, 75]. We take the Brillouin-Wigner approach here.

The Hilbert space of the problem is enlarged when the Floquet modes are introduced. The identity operator in the full Hilbert space formed by joining the Floquet and Fock spaces reads

\[
1 = 1_{\text{Fock}} \otimes 1_{\text{Floquet}} = P + Q,
\]

with \( P \) and \( Q \) the projectors onto the ground state and excited states manifolds of the full Floquet-Fock Hilbert space. The total ground state projector \( P \) is the tensor product of the Fock and Floquet ground state manifolds, \( P = P \otimes P_{F,0} \), while the projector onto excited states is

\[
Q = \sum_{m=-\infty}^{+\infty} P_{F,m} + \sum_{m \neq 0} P_{F,m},
\]

with \( Q \) the excited states of the fermions, only.

When compared to the time-independent case, the novel effects in the structure of the perturbation theory comes from the second term of \( Q \), which projects onto the fermionic ground state manifold as long as \( m \neq 0 \) in Floquet.
space. We now define the resolvent operator $\mathcal{R}$, which encodes the sum over all the excited states and also takes into account the energy denominators as $\mathcal{R} = \mathcal{R}_1 + \mathcal{R}_2$, where

$$
\mathcal{R}_1 = \sum_m P F, m Q \left( \frac{E_0 - H_0}{E_0 - H_0} \right),
$$

$$
\mathcal{R}_2 = \sum_{m \neq 0} P F, m P \left( \frac{E_0 - H_0}{E_0 - H_0} \right),
$$

with $E_0$ is the ground state energy of $H_0$, which is the Hamiltonian of Eq. (5) with the hoppings set to zero.

The information coming from the hopping Hamiltonian is used to construct the wave operator $\mathcal{W}$, which is implicitly defined by

$$
\mathcal{W} = \mathcal{P} + \mathcal{R} (\mathcal{V} \mathcal{W} - \mathcal{W} \mathcal{V} \mathcal{W}).
$$

The effective spin Hamiltonian is obtained from $\mathcal{W}$

$$
H_{\text{spin}}^{(m \Omega \leq U)} = \mathcal{P} H_0 \mathcal{P} + \mathcal{P} \mathcal{V} \mathcal{W} = \mathcal{P} \mathcal{V} \mathcal{W},
$$

where the second equality follows given that the projection of $H_0$ onto the ground state manifold is zero. The equation for the wave operator can be solved recursively to a certain order of the perturbation potential $\mathcal{V}$. The zeroth order term from Eq. (26) to $\mathcal{W}$ is $\mathcal{W}^{(0)} = \mathcal{P}$ [43]. This term gives a vanishing contribution to the effective Hamiltonian Eq. (27) given that $\mathcal{P}$ projects onto the Fock ground state with one electron per site while $\mathcal{V}$ moves electrons creating empty and doubly occupied states. Similar reasoning leads to the conclusion that all terms with an even number of $\mathcal{V}$ insertions in $\mathcal{W}$ will also vanish. The leading contributions to $\mathcal{W}$ are found from first and third order in $\mathcal{V}$ [43],

$$
\mathcal{W}^{(1)} = \mathcal{R} \mathcal{V} \mathcal{P},
$$

$$
\mathcal{W}^{(3)} = \mathcal{R} \mathcal{V} \mathcal{R} \mathcal{V} \mathcal{P} - \mathcal{R}^2 \mathcal{V} \mathcal{P} \mathcal{V} \mathcal{R} \mathcal{P}.
$$

From Eq. (27), $\mathcal{W}^{(1)}$ and $\mathcal{W}^{(3)}$ lead to the effective spin Hamiltonians in orders two and four,

$$
H^{(2)} = \mathcal{P} \mathcal{V} \mathcal{R} \mathcal{V} \mathcal{P},
$$

$$
H^{(4)} = \mathcal{P} \mathcal{V} \mathcal{R} \mathcal{V} \mathcal{R} \mathcal{V} \mathcal{P} - (\mathcal{P} \mathcal{V} \mathcal{R} \mathcal{P}^2 \mathcal{V} \mathcal{P}) H^{(2)}.\n$$

1. Second-order perturbation theory

The second-order correction $H^{(2)}$ can be calculated by decomposing $\mathcal{R}$ as the sum of $\mathcal{R}_1$ and $\mathcal{R}_2$ and noticing, from Eq. (25), that $\mathcal{R}_2 \mathcal{V} \mathcal{P} = 0$ since, explained earlier, $\mathcal{P} \mathcal{V} \mathcal{P} = 0$. In second-order perturbation theory, therefore, $\mathcal{R}_2$ does not enter the calculation and the structure is identical to the time-independent model, except for the energy denominators and renormalized hoppings. By plugging the resolvent $\mathcal{R}_1$ explicitly, Eq. (30), and defining $\mathcal{V}_{m_1 \rightarrow m_2} = \langle m_1 | \mathcal{V} | m_2 \rangle$ we arrive at

$$
H^{(2)} = - \sum_m (P \mathcal{V}_m Q) \frac{1}{(U + m \Omega)} (Q \mathcal{V}_{-m} P).
$$

By inserting $\mathcal{V}_m$, we arrive at Eq. (9).

2. Third-order perturbation theory

Even though the Floquet fields break time-reversal symmetry dynamically, the contributions in third-order perturbation theory sum out to zero, including the chiral terms. This is true for any choice of polarization and was previously addressed for circularly polarized light [13].
3. Fourth-order perturbation theory

Since the third-order corrections vanish, we now proceed to fourth order. By plugging the resolvent $\mathcal{R}$ into Eq. (31), we find that the first term leads to two possible intermediate steps, with either $\mathcal{R}_1$ or $\mathcal{R}_2$ in the middle. By separating all the contributions, we arrive at

$$
\mathcal{H}^{(4)} = \mathcal{P} \mathcal{V} \mathcal{R}_1 \mathcal{V} \mathcal{R}_1 \mathcal{V} \mathcal{P} + \mathcal{P} \mathcal{V} \mathcal{R}_1 \mathcal{V} \mathcal{R}_2 \mathcal{V} \mathcal{P} - (\mathcal{P} \mathcal{V} \mathcal{R}_1^2 \mathcal{V} \mathcal{P}) \mathcal{H}^{(2)}.
$$

(33)

After using equations (24) and (25) for the resolvent, the Hilbert space of the problem is again the Fock space of the fermions, as only the projectors $P$ and $Q$ are left in the calculation. By plugging them explicitly, we arrive at

$$
\mathcal{H}^{(4)}_a = - \sum_{m_1,m_2,m_3} \frac{P \mathcal{V}_{m_2} \mathcal{Q} \mathcal{V}_{m_3-m_2} \mathcal{Q} \mathcal{V}_{m_2-m_1} \mathcal{Q} \mathcal{V}_{m_1} P}{(U + m_3 \Omega) (U + m_2 \Omega) (U + m_1 \Omega)} - \sum_{m_1,m_2,m_3} \frac{P \mathcal{V}_{m_3} \mathcal{Q} \mathcal{V}_{m_3-m_2} \mathcal{Q} \mathcal{V}_{m_2-m_1} \mathcal{Q} \mathcal{V}_{m_1} P}{(U + m_3 \Omega) (2U + m_2 \Omega) (U + m_1 \Omega)}
$$

(34)

$$
\mathcal{H}^{(4)}_b = - \sum_{m_1,m_2 \neq 0,m_3} \frac{P \mathcal{V}_{m_2} \mathcal{Q} \mathcal{V}_{m_3-m_2} \mathcal{P} \mathcal{V}_{m_2-m_1} \mathcal{Q} \mathcal{V}_{m_1} P}{(U + m_3 \Omega) (m_2 \Omega) (U + m_1 \Omega)},
$$

(35)

$$
\mathcal{H}^{(4)}_c = \sum_{m_1,m_2} \frac{P \mathcal{V}_{m_2} \mathcal{Q} \mathcal{V}_{m_2} \mathcal{P} \mathcal{V}_{m_1} \mathcal{Q} \mathcal{V}_{m_1} P}{(U + m_2 \Omega)^2 (U + m_1 \Omega)}.
$$

(36)

In the proceeding equations, we decomposed $Q$ as

$$
Q = Q_U + Q_{2U} + Q_{3U} + \ldots,
$$

(37)

with $Q_{kU}$ projecting onto the fermionic manifold of energy $kU$.

For the explicit calculation of all the couplings that appear from Eqs. (34)-(36) for the triangular lattice, it is a matter of summing over all possible paths. For notation, we refer again to Fig. 5. The effective magnetic exchange couplings are expressed in terms of the functions $A_i$, defined in Eq. (8), and we define $\tilde{t} = t_1/U$ and $\Omega = \Omega/U$, for simplicity.

$$
A_{ijkl} (m) = (-1)^{m_2} \tilde{p}_3 \frac{J_{m_3} (A_{i_1}) J_{m_3-m_2} (A_{i_2}) J_{m_2-m_1} (A_{i_3}) J_{m_1} (A_{i_4})}{(1 + m_1 \Omega) (1 + m_2 \Omega) (1 + m_3 \Omega)},
$$

(38)

$$
\mathcal{L}_{ijkl} (m) = (-1)^{m_1+m_3} \tilde{p}^3 \cos^2 \left( \frac{m_2 \pi}{2} \right) \frac{J_{m_2} (A_{i_1}) J_{m_3-m_2} (A_{i_2}) J_{m_2-m_1} (A_{i_3}) J_{m_1} (A_{i_4})}{(1 + m_1 \Omega) (2 + m_2 \Omega) (1 + m_3 \Omega)},
$$

(39)

$$
\mathcal{B}_{ij} (m) = (-1)^{m_1+m_3} \tilde{p}^3 \cos^2 \left( \frac{m_2 \pi}{2} \right) \frac{J_{m_3} (A_{i_1}) J_{m_3-m_2} (A_{i_2}) J_{m_2-m_1} (A_{i_3}) J_{m_1} (A_{i_4})}{(1 + m_1 \Omega) (m_2 \Omega) (1 + m_3 \Omega)}, \quad m_2 \neq 0,
$$

(40)

$$
\mathcal{G}_{ij} (m) = \tilde{p}^3 \delta_{m_2,0} \left[ J_{m_1}^2 (A_{i_1}) J_{m_3}^2 (A_{i_2}) + J_{m_1}^2 (A_{i_1}) J_{m_3} (A_{i_2}) J_{m_3} (A_{i_2}) \right] \frac{1}{(1 + m_1 \Omega)^2 (1 + m_3 \Omega)},
$$

(41)

where we define $m \equiv (m_1, m_2, m_3)$.

The next-nearest neighbor coupling $J_2$ [Fig. 5(b)] reads

$$
J_2^{(i,k)} = \sum_m -8 \left\{ A_{1,2,2,1} (m) \cos^2 \left( \frac{m_1 + m_3 \pi}{2} \right) \cos ((1 - \beta_0) (m_1 - m_3)) + A_{1,2,1,2} (m) \cos^2 \left( \frac{m_1 + m_2 + m_3 \pi}{2} \right) \times \cos [m_1 - m_2 + m_3 (\beta_1 - \beta_0)] \right\} + 8 \mathcal{L}_{2,2,1,1} (m) \cos [m_2 (\beta_1 - \beta_0)] - 16 \mathcal{B}_{2,1} (m) \cos [(\beta_1 - \beta_0) m_2] + 8 \mathcal{G}_{2,1} (m),
$$

(42)

while the plaquette terms [Fig. 5(f)] reads

$$
J_4^{(i,j,k,l)} = \sum_m 32 \left\{ A_{1,2,2,1} (m) \cos^2 \left( \frac{m_1 + m_3 \pi}{2} \right) \cos [(\beta_1 - \beta_0) (m_1 - m_3)] + A_{1,2,1,2} (m) \cos^2 \left( \frac{m_1 + m_2 + m_3 \pi}{2} \right) \times \cos [(m_1 - m_2 + m_3) (\beta_1 - \beta_0)] \right\} + 32 \cos [m_2 (\beta_1 - \beta_0)] \mathcal{L}_{2,2,1,1} (m).
$$

(43)
The $J_3$ coupling [Fig. 5(c)] is

$$J_3^{(i,l,m)} = \sum_m -4A_{2,2,2} (m) + 8B_{2,2} (m) + 4G_{2,2} (m). \quad (44)$$

The chiral term reads [Fig. 5(d)]

$$J_x^{(i,j,k)} = \sum_m 16 \left[ L_{2,2,1,1} (m) - B_{2,1} (m) \right] \left[ \sin [m_2 (\beta_1 - \beta_0)] - \sin [m_2 (\beta_1 - \beta_3)] + \sin [m_2 (\beta_2 - \beta_3)] \right]. \quad (45)$$

It might be surprising, at first sight, that the expression of (45) has only terms proportional to $\mathcal{L}$ and $\mathcal{B}$, with the terms proportional to $\mathcal{A}$ vanishing exactly. An interesting sanity check that this is the case consists of expanding (45) in powers of $1/\Omega$, assuming $\Omega \gg U$. The leading contribution comes from $1/\Omega^3$ and not $1/\Omega$, as would be naively expected. This is in agreement with the fact that, at the limit of high $\Omega$, the $1/\Omega$ corrections to the hoppings on the triangular lattice vanishes. $J_x^b$ [Fig. 5(e)] gives

$$J_x^b = -3J_x^a. \quad (46)$$

These results are generic for light of arbitrary fixed polarization. In the main text, we address the vanishing of the chiral terms for the light profile shown in Fig. 3 and Eq. (10), which presents a slowly varying periodic polarization. This requires generalizing the above expressions for hoppings that do not follow Eq. (7) of the main text, but instead, Eq. (12). For $J_x^b$ it reads

$$J_x^b = \sum_{m_1,m_2,m_3} \frac{1}{(1+m_1\Omega)(2+m_2\Omega)(1+m_3\Omega)} - \sum_{m_1,m_2\neq 0,m_3} \frac{1}{(1+m_1\Omega)(m_2\Omega)(1+m_3\Omega)} g (m) \quad (47)$$

with

$$g = \left[ \begin{array}{c} t_0^{m_1} (t_0^{m_1-m_2}) * + (t_0^{m_1}) * t_0^{m_2-m_1} \left[ (t_3^{m_2-m_3}) * t_3^{m_3-m_2} + t_3^{m_3-m_2} (t_3^{m_3}) * \right] \\
+ \left[ t_3^{m_1} (t_3^{m_1-m_2}) * + (t_3^{m_1}) * t_3^{m_2-m_1} \left[ (t_3^{m_2-m_3}) * t_3^{m_3-m_2} + t_3^{m_3-m_2} (t_3^{m_3}) * \right] \\
+ \left[ (t_3^{m_1}) * + (t_3^{m_1}) * (t_3^{m_2-m_1}) \left[ (t_3^{m_2-m_3}) * t_3^{m_3-m_2} + t_3^{m_3-m_2} (t_3^{m_3}) * \right] \\
+ \left[ (t_3^{m_1}) * + (t_3^{m_1}) * (t_3^{m_2-m_1}) \left[ (t_3^{m_2-m_3}) * t_3^{m_3-m_2} + t_3^{m_3-m_2} (t_3^{m_3}) * \right] \\
\end{array} \right] \right. \quad (48)$$

It is easy to verify that it reduces to Eqs. (46) and (45) in the monochromatic limit.

We next list the fourth-order corrections for $J_1$. For circular polarization, $\delta J_1^{(4)}$ is

$$\delta J_1^{(4)} = \sum_m 8A (m) f_\triangle^{(CP)} (m) - 8\mathcal{L} (m) \left[ \cos \left( \frac{\pi m_2}{3} \right) + 2 \cos \left( \frac{2\pi m_2}{3} \right) \right]$$

$$- 16\mathcal{B} (m) \left[ \cos \left( \frac{\pi m_2}{3} \right) + 2 \cos \left( \frac{2\pi m_2}{3} \right) + 2 \right] - 40\mathcal{G} (m), \quad (C P) \quad (49)$$

with

$$f_\triangle^{(CP)} (m) = 2 + \cos^2 \left( m_1 + m_3 \frac{\pi}{2} \right) \left[ \cos \left( \frac{1}{3} \pi (m_1 - m_3) \right) + 2 \cos \left( \frac{2}{3} \pi (m_1 - m_3) \right) \right]$$

$$+ \cos^2 \left( m_1 + m_2 + m_3 \frac{\pi}{2} \right) \left[ \cos \left( \frac{1}{3} \pi (m_1 - m_2 + m_3) \right) + 2 \cos \left( \frac{2}{3} \pi (m_1 - m_2 + m_3) \right) \right]. \quad (50)$$

The correction $\delta J_1^{(4)}$ to a bond along the $\delta_3$ direction coupled to linearly polarized light is
The couplings \( t \) slightly negative (see Fig. 4 of the main text for the parametric plot of exchange term consisting of an ensemble of linearly-polarized light (type II Glauber light). The main difference regards the ring-protocols: by averaging over the entire Poincare sphere (type I light) and by averaging over the equator of the sphere, \( \delta J \) is achieved. This poses a disadvantage as compared to the average over linear polarization when the goal is to destabilize the 120 phase and transition to a SL regime, but may lead to other phase transitions.

One concern that arises from examining these corrections in Brillouin-Wigner theory is that we generically find \( nU/m \geq 1 \) for the fluences considered in this work, \( J_c \) becomes negative for \( A_0 = 1.68 \) before the maximum enhancement of \( J_{2,3} \) is achieved. This poses a disadvantage as compared to the average over linear polarization when the goal is to destabilize the 120 phase and transition to a SL regime, but may lead to other phase transitions.

4. Higher-order corrections

We now comment about the effects corrections from higher orders in perturbation theory. Given that the odd powers of \( t_1/U \) lead to vanishing contributions, the next finite order in perturbation theory is sixth order. By keeping the ratio \( t_1/U < 0.04 \), as we must to avoid heating, higher orders will contribute only small corrections to the fourth-order results. To justify the truncation of the perturbative expansion in the presence of the Floquet field, we may examine the relative contributions to \( J_1 \). Generically, there are two contributions: the second and the fourth-order ones, \( J_1 = J_1^{(2)} + J_1^{(4)} \). By computing the ratios \( |J_1^{(2)}/J_1| \) and \( |J_1^{(4)}/J_1| \) for the fluences considered in this work, 80% or more of the total contribution to \( J_1 \) comes from the second order term, \( |J_1^{(2)}/J_1| \geq 0.8 \). For higher values of fluence, \( J_1^{(2)} \) becomes small and can even pass through zero and go negative. In this region, the sixth-order corrections must be incorporated, but otherwise are negligible.

\[
\delta J_1^{(4)} = \sum_m 8 \left\{ \cos^2 \left[ \frac{(m_1 + m_2 + m_3) \pi}{2} \right] + \cos^2 \left[ \frac{(m_1 + m_3) \pi}{2} \right] \right\} (A_3,2,3,2 + A_2,3,3,2 + A_3,1,3,1 + A_1,3,3,1) \\
- 8 \cos^2 \left[ \frac{(m_1 + m_3) \pi}{2} \right] (A_{1,2,1,2} + A_{2,1,1,2}) 16 A_{2,2,2,2} - 8 (L_{2,2,3,3} + L_{3,3,2,2} - L_{2,2,1,1}) \\
- 16 [2 (B_{3,2} + B_{2,3}) + 2B_{3,3} - B_{1,2}] - 8 (2G_{3,2} + 2G_{2,3} + 2G_{3,3} - G_{1,2}), \quad \text{(LP)}
\]

Notice that \( \delta J_1^{(4)} \to -28t_1^2/U^3 \) as \( A_0 \to 0 \), recovering the time-independent limit. The corrections for linearly polarized light in other directions are found by permutations of the sub-indices.

In Fig. 6, we show the modification of the exchange couplings as function of the fluence \( A_0 \) for two polarization protocols: by averaging over the entire Poincare sphere (type I light) and by averaging over the equator of the sphere, consisting of an ensemble of linearly-polarized light (type II Glauber light). The main difference regards the ring-exchange term \( J_c \). When the average is performed over the entire sphere, \( J_c \) becomes negative for \( A_0 = 1.68 \) before the maximum enhancement of \( J_{2,3} \) is achieved. This poses a disadvantage as compared to the average over linear polarization when the goal is to destabilize the 120 phase and transition to a SL regime, but may lead to other phase transitions.

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