Ultrafast Control of Spin-Spin Interactions in Two-Dimensional Magnetic Systems: Kane-Mele-Hubbard Model

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We study ultrafast optical control of exchange and Dzyaloshinskii-Moriya interactions in two-dimensional honeycomb lattices described by the Kane-Mele-Hubbard model at half-filling and strongly correlated limit, i.e., Mott insulator regime of a canted antiferromagnet. Based on Floquet theory calculations, we show that by changing the amplitude and frequency of polarized laser pulses, it is possible to tune the amplitude, the sign and even the ratio between these two spin interactions. We find that in this model, the renormaization of spin interactions are helicity independent. Our results pave the way for ultrafast optical manipulation of spins in recently discovered two-dimensional magnetic materials.

Introduction.— Discovery of athermal all-optical control of order parameters in antiferromagnetic (AFM) and ferromagnetic (FM) materials by ultrashort, intense and high-frequency laser pulses has elevated spintronics to a new era of ultrafast magnetism [1 and 2]. Although the underlying microscopic mechanism behind this phenomena is unclear yet, there have been many attempts to uncover its origins. Recently, it has been theoretically proposed that laser pulses can modify and even reverse the sign of exchange interaction [3–10], the strongest spin interaction in magnetic systems. This finding has been supported by recent experiments [11 and 12]. Another important spin interaction is Dzyaloshinskii-Moriya intercation (DMI), an antisymmetric exchange interaction appeared in inversion symmetry broken systems, that breaks the chiral symmetry of the system [13–16]. While this interaction is much weaker than the exchange interaction, its presence has important consequences in magnetic materials such as weak ferromagnetism in AFM materials [13 and 14], stabilizing topological objects like chiral skyrmions [17–20] and chiral domain walls [21–23], and exotic phase of topological magnon insulators [24–28]. The ratio between these two interactions control the tilting angle of canted spins. Proposing a mechanism to tune this ratio has important consequences in ultrafast spin dynamics and switching [5, 7, 8, and 11].

Another important breakthrough in spintronics happened in recent few years is the discovery of two-dimensional (2D) van der Waals magnetic materials with metallic, semiconducting and insulating band structures in both AFM and FM phases [29 and 30]. We can now use the advantage of both low dimensionality and magnetic order in the same material for the first time which promises novel spintronics devices with exceptional performances.

In this Letter, we show that intense and high-frequency laser pulses can change spin-spin interactions and the ratio between exchange interaction and DMI drastically in a class of 2D magnetic materials described by the Kane-Mele-Hubbard model.

Model Hamiltonian.— Effective Hamiltonian of a 2D planar honeycomb lattice in the presence of electron-electron interactions can be described by Kane-Mele-Hubbard model [31–37]. The Hamitlonian in the absence of external perturbations is the sum of the kinetic term $\hat{\mathcal{H}}_t$, the intrinsic SOI $\hat{\mathcal{H}}_{\rm SOI}$ and Coulomb interaction between electrons modelled by extended Hubbard model $\hat{\mathcal{H}}_{\rm int}$,

$$\hat{H}_0 = \hat{\mathcal{H}}_{\rm K} + \hat{\mathcal{H}}_{\rm SOI} + \hat{\mathcal{H}}_{\rm int}, \tag{1}$$

where

$$\hat{\mathcal{H}}_{\rm K} = -t_1 \sum_{\langle i,j\rangle,\tau} \hat{c}^\dagger_{i\tau}\hat{c}_{j\tau} - t_2 \sum_{\langle\langle i,j\rangle\rangle,\tau} \hat{c}^\dagger_{i\tau}\hat{c}_{j\tau}, \tag{2}$$

$$\hat{\mathcal{H}}_{\rm SOI} = i\Delta \sum_{\langle\langle i,j\rangle\rangle,\tau,\tau'} \nu_{ij}\sigma^z_{\tau,\tau'}\hat{c}^\dagger_{i\tau}\hat{c}_{j\tau'}, \tag{3}$$

$$\hat{\mathcal{H}}_{\rm int} = U_{00}\sum_{i=1}\hat{n}_{i\uparrow}\hat{n}_{i\downarrow} + \frac{1}{2}\sum_{\langle i,j\rangle,\tau\tau'} V_{ij}\hat{n}_{i\tau}\hat{n}_{j\tau'}. \tag{4}$$

Here $\langle\cdot\rangle$ and $\langle\langle\cdot\rangle\rangle$ denote, respectively, nearest neighbors (NN) and next-nearest neighbours (NNN), $\hat{c}^\dagger_{i\sigma}$ ($\hat{c}_{i\sigma}$) are fermionic creation (annihilation) operators for an electron on site i and spin state $\tau = \{\uparrow,\downarrow\}$, t_1 and t_2 are NN and NNN hopping amplitudes, Δ is the intrinsic SOI constant, $\nu_{ij} = \pm 1$ depending on whether the electron traversing from i to j makes a right $(+1)$ or a left turn (-1), σ^z is the z-component of Pauli matrices $\boldsymbol{\sigma}$, U_{00} and V_{ij} are the on-site and NN Coulomb interactions. The presence of the intrinsic NNN SOI, Eq. (3), reduces the SU(2) symmetry of original Hubbard model to the U(1) spin group. In buckled noncoplanar honeycomb lattices and systems with structural inversion asymmetry, presence of, respectively, NNN and NN Rashba SOIs, furthur reduces the symmetry to Z_2.

Using the variational principle, it has been shown that the NN Coulomb interaction can be effectively approximated by a renormalized local interaction $U = U_{00} - \bar{V}$, where $\bar{V}$ is a weighted average of the NN Coulomb interaction [7 and 38]. Thus, we only take into account the

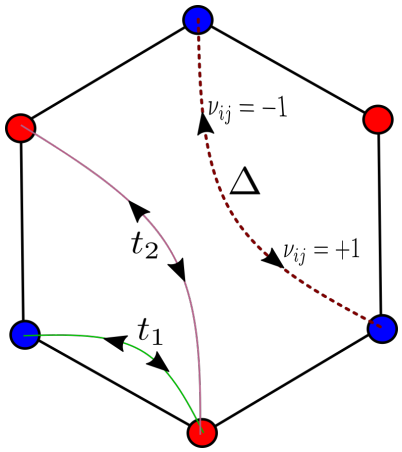

FIG. 1. A honycomb cell with NN hopping t_1, NNN hopping t_2 and intrinsic SOI Δ. $\nu_{ij}=\pm1$ depending on whether the electron traversing from i to j makes a right $(+1)$ or a left turn (-1).

local Coulomb interaction in our total Hamiltonian and rewrite the interaction part of the Hamiltonian (4),

$$\hat{\mathcal{H}}_{\rm int} \approx U\hat{d}, \tag{5}$$

where we define the doublon number operator $\hat{d} = \sum_{i=1}\hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$ with eigenvalues d and projection operators $\hat{P}_d$. We are interested to Kane-Mele-Hubbard model at half-filling in strongly correlated regime $U \gg t_{1(2)}$. In such strong-coupling limit any state with a nonzero number of double occupancies $(d \neq 0)$ will have much larger energy than those with $d = 0$. We obtain an effective Hamiltonian acting on the $d = 0$ subspace by standard second-order perturbation theory in the hopping terms. Using the following relations,

$$\hat{c}^\dagger_{i\tau}\hat{c}_{i\tau'} = \frac{1}{2}(n_{i\uparrow}+n_{i\downarrow})\delta_{\tau\tau'} + \boldsymbol{S}_i\cdot\boldsymbol{\tau}_{\tau',\tau}, \tag{6}$$

$$\hat{c}_{i\tau}\hat{c}^\dagger_{i\tau'} = \frac{1}{2}(2-n_{i\uparrow}-n_{i\downarrow})\delta_{\tau\tau'} - \boldsymbol{S}_i\cdot\boldsymbol{\tau}_{\tau,\tau'}, \tag{7}$$

we find the spin Hamiltonian for 2D AFM Mott insulators,

$$\begin{aligned} H_{\rm S} =& J_1\sum_{\langle i,j\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j + J_2\sum_{\langle\langle i,j\rangle\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j, \\ &+\sum_{\langle\langle i,j\rangle\rangle}\boldsymbol{S}_i\boldsymbol{\Gamma}_{ij}\boldsymbol{S}_j + \sum_{\langle\langle i,j\rangle\rangle}\boldsymbol{D}_{ij}\cdot\boldsymbol{S}_i\times\boldsymbol{S}_j, \end{aligned} \tag{8}$$

with the following spin-spin interactions,

$$J_{1(2)} = \frac{2t^2_{1(2)}}{U}, \tag{9a}$$

$$\boldsymbol{\Gamma}_{ij} = \frac{2\Delta^2}{U}\mathrm{diag}(-1,-1,1), \tag{9b}$$

$$\boldsymbol{D}_{ij} = -\frac{4t_2\Delta}{U}\nu_{ij}\hat{\mathrm{e}}_z. \tag{9c}$$

The first and second terms in the spin Hamiltonian, Eq. (8), are the NN and NNN symmetric Heisenberg AFM exchange interactions, $J_{1(2)} > 0$, respectively; the third term is the NNN anisotropic Heisenberg exchange interaction (XXZ-like term) arising from the intrinsic SOI, and finally, the last term is the intrinsic NNN DMI. The intrinsic SOI in Eq. (3), leads to a DMI vector, $\boldsymbol{D}_{ij}$, perpendicular to the honeycomb plane while it is straightforward to show that the Rashba SOI results an inplane DMI vector perpendicular to the lattice bonds. For the sake of completeness, we explain shortly the effect of disorders by adding an on-site disorder $\sum_{i\tau}\varepsilon_i\hat{c}^\dagger_{i\tau}\hat{c}_{i\tau}$ in the Kane-Mele-Hubbard Hamiltonian 1, where ε_i is an uncorrelated random variable. Following the above procedure, it can be shown that the spin interaction parameters are only renormalized by $U^{-1} \to U/\left(U^2-(\epsilon_j-\epsilon_i)^2\right)$ [39]. In the large U limit, the effect of disorder is negligible and we thus ignore it in the rest of this Letter.

Different terms appeared in the spin Hamiltonian derived in Eq. (8) has already been obtained phenomenologically [24] and even confirmed experimentally [26] but, to the best of our knowledge, this is the first microscopic derivation of the complete spin Hamiltonian $\hat{H}_{\rm S}$ from microscopic Kane-Mele-Hubbard model. It has been shown that this Hamiltonian has interesting features and exotic phases like, existence of magnon spin Nernst effect in collinear AFM layers [40 and 41], topological magnon insulator phase [24–26], spin Hall effects of Weyl magnons [42 and 43] magnonic Floquet topological insulators, spin density wave [44], chiral and topological gapped spin liquid phases [45]. Ultrafast control of DMI and exchange interaction by laser pulses also open a way to engineer all these phenomena.

Laser illumination. The effect of laser irradiation on the honeycomb lattice is introduced in the Kane-Mele-Hubbard Hamiltonian, Eq. (1), via the Peierls substitution method [46]. The electric-field component of a polarized laser pulse is given by $\boldsymbol{E}(t) = \frac{E_0}{2}(e^{-i\omega t}\hat{\epsilon}+\text{c.c.})$, where E_0 is the electric field amplitude, ω is the laser pulse frequency, and $\hat{\epsilon} = (\hat{\mathrm{e}}_x + i\lambda\hat{\mathrm{e}}_y)/\sqrt{1+\lambda^2}$ is the polarization unit vector with $\lambda = 0$ for linear polarization and $\lambda = \pm1$ for right- and left handed polarizations.

It is more convenient to rewrite the noninteracting part of the Kane-Mele-Hubbard Hamiltonian, Eq. (1), from now on the hopping part, as $\hat{T}_0 = \hat{\mathcal{H}}_{\rm K} + \hat{\mathcal{H}}_{\rm SOI} = -\sum_{i,j,\tau,\tau'}t^{\tau\tau'}_{ij}\hat{c}^\dagger_{i\tau}\hat{c}_{j\tau'}$, where the hopping amplitude is $t^{\tau\tau'}_{ij} = \delta_{\tau,\tau'}t_1$ for i,j being nn and $t^{\tau\tau'}_{ij} = \delta_{\tau,\tau'}t_2 - i\tilde{\Delta}\nu_{ij}\sigma^z_{\tau,\tau'}$ for i,j being NNN. According to the Peierls substitution method, the hopping part of the Hamiltonian in the presence of electromagnetic fields gain an extra phase $t^{\tau\tau'}_{ij} \to t^{\tau\tau'}_{ij}e^{ie\boldsymbol{R}_{ij}\cdot\boldsymbol{A}(t)/\hbar}$, where $\boldsymbol{R}_{ij} = \boldsymbol{R}_i - \boldsymbol{R}_j$, $\boldsymbol{R}_i$ is the position of site i, e is the electronic charge, $\hbar$ is the reduced Planck constant, and $\boldsymbol{A}$ is the vector potential of laser pulse $\boldsymbol{A}(t) = \frac{1}{2}(\boldsymbol{A}e^{-i\omega t}+\text{c.c.})$, with $\boldsymbol{A} = \frac{iE_0}{\omega}\hat{\epsilon}$. The argument of the Peierls phase can be rewritten as

$e\boldsymbol{R}_{ij}\cdot\boldsymbol{A}/\hbar \equiv \alpha_{ij}e^{i\theta_{ij}}$, with $\alpha_{ij} = \pm|e\boldsymbol{R}_{ij}\cdot\boldsymbol{A}|/\hbar$, in a such way that $\alpha_{ij} = -\alpha_{ji}$, $\theta_{ij} = \theta_{ji}$, and $\theta_{ij} \in [0, \pi)$. Now, we use the Jacobi-Anger expansion to rewrite the Peierls phase in the basis of its harmonics,

$$e^{i\frac{e}{\hbar}\boldsymbol{R}_{ij}\cdot\boldsymbol{A}(t)} = \sum_m e^{i(\frac{\pi}{2}-\theta_{ij})m}\mathcal{J}_m(\alpha_{ij})e^{im\omega t}, \tag{10}$$

where $\mathcal{J}_m(x)$ is the m-th Bessel function of the first kind [47].

The total Hamiltonian in the presence of the laser field is time depended through its hopping part, $\hat{H}(t) = \hat{T}(t) + U\hat{d}$. Using Eq. (10), we find $\hat{T}(t) = \sum_m \hat{T}_m e^{im\omega t}$, where $\hat{T}_m$ is the sum of all the m-th Fourier mode of the hopping terms. Finally, we decompose the hopping operator into,

$$\hat{T}(t) = \sum_m (\hat{T}_{-1,m} + \hat{T}_{0,m} + \hat{T}_{1,m})e^{im\omega t}, \tag{11}$$

where $\hat{T}_{dm}(t)$ changes the doublon number by d, for example, if $\hat{P}_d$ is the projection operator into the subspace with doublon number d, then $\hat{T}_{dm}(t) = \sum_i \hat{P}_{i+d}\hat{T}_m(t)\hat{P}_i$.

To find renormalized spin Hamiltonian, we first drive an effective static Hamiltonian using the Floquet formalism [48 and 49]. To do that, we transform the original time-dependent Hamiltonian, $\hat{H}(t)$, using a unitary transformation $\hat{U}(t) = e^{-i\hat{S}(t)}$,

$$\hat{H}'(t) = e^{i\hat{S}(t)}\left(\hat{H}(t) - i\partial_t\right)e^{-i\hat{S}(t)}. \tag{12}$$

We perform the unitary transformation perturbatively in the hopping parameter. We can formally write $\hat{T}(t) = \eta\hat{T}(t)$, where η play the role of a bookkeeping parameter in the perturbation expansion. We expand $\hat{S}(t) = \sum_\nu \eta^\nu \hat{S}^{(\nu)}(t)$ and $\hat{H}'(t) = \sum_\nu \eta^\nu \hat{H}'^{(\nu)}(t)$. Our demand is that the transformed Hamiltonian must have the same periodicity of the original time-dependent Hamiltonian thus the unitary transformation itself must have the same periodicity of the driven laser field so that $\hat{S}^{(\nu)}(t) = \sum_m e^{im\omega t}\hat{S}^{(\nu)}_m$. Additionally, we impose that the transformed Hamiltonian to be block diagonal in the doublon number d while $\hat{S}(t)$ does not contain block-diagonal terms. With these conditions the unitary transformation might be uniquely determined,

$$\hat{S}^{(\nu)}(t) = \sum_{d\neq 0}\sum_m \eta^\nu \hat{S}^{(\nu)}_{d,m}e^{im\omega t}, \tag{13}$$

where $\hat{S}^{(\nu)}_{d,m}$ changes the double occupancy number by d. We expand the transformed Hamiltonian 12 in power series of η and determine $\hat{S}^{(\nu)}(t)$ iteratively in ν so that $\hat{H}'^{(\nu)}(t)$ is diagonal in the doublon number. After tedious but straightforward calculations, the transformed Hamiltonian up to the second order in the hopping parameter is obtained $\hat{H}'(t) = \hat{T}'(t) + \mathrm{U}\hat{d}$, where

$$\hat{T}'(t) \approx -\sum_m \hat{T}_{0,m}(t)e^{im\omega t} + \\ + \frac{1}{2}\sum_{mn}\left(\frac{\left[\hat{T}_{1,n},\hat{T}_{-1,m-n}\right]}{\mathrm{U}+n\omega} - \frac{\left[\hat{T}_{-1,n},\hat{T}_{1,m-n}\right]}{\mathrm{U}-n\omega}\right)e^{im\omega t}. \tag{14}$$

Now, we are in the situation that we can calculate the effective static Hamiltonian by time averaging of the transformed Hamiltonian $\hat{H}_{\text{eff}} = \hat{P}_0\hat{H}'(t)\hat{P}_0$, where $\hat{P}_0$ is the projection operator to the $d = 0$ subspace. Doing some algebra, the effective static Hamiltonian in terms of creation and annihilation operators is obtained,

$$\hat{H}_{\text{eff}} = -\sum_{i,j,\tau,\tau'}\left(t^\tau_{ij}t^{\tau'}_{ji}\sum_n \frac{\mathcal{J}^2_n(\alpha_{ij})}{\mathrm{U}+n\omega}\right)\hat{c}^\dagger_{i\tau}\hat{c}_{j\tau}\hat{c}^\dagger_{j\tau'}\hat{c}_{i\tau'} + \mathrm{U}\hat{d}. \tag{15}$$

Finally, the spin Hamiltonian, at half-filling and strong interaction limit of the effective Hamiltonian $\hat{H}_{\text{eff}}$, using the relations Eqs. (6) and (7), is given by,

$$\tilde{H}_{\mathrm{S}}(\omega) = \tilde{J}_1\sum_{\langle i,j\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j + \tilde{J}_2\sum_{\langle\langle i,j\rangle\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j, \\ + \sum_{\langle\langle i,j\rangle\rangle}\boldsymbol{S}_i\tilde{\boldsymbol{\Gamma}}_{ij}\boldsymbol{S}_j + \sum_{\langle\langle i,j\rangle\rangle}\tilde{\boldsymbol{D}}_{ij}\cdot\boldsymbol{S}_i\times\boldsymbol{S}_j, \tag{16}$$

with the following renormalized spin interactions,

$$\tilde{J}_{1(2),ij} = 2t^2_{1(2)}\frac{\mathcal{J}^2_n(\alpha_{ij})}{\mathrm{U}+n\omega}, \tag{17a}$$

$$\tilde{\boldsymbol{\Gamma}}_{ij} = 2\Delta^2\mathrm{diag}(-1,-1,1)\frac{\mathcal{J}^2_n(\alpha_{ij})}{\mathrm{U}+n\omega}, \tag{17b}$$

$$\tilde{\boldsymbol{D}}_{ij} = -4t_2\Delta\frac{\mathcal{J}^2_n(\alpha_{ij})}{\mathrm{U}+n\omega}\nu_{ij}\hat{\mathrm{e}}_z. \tag{17c}$$

All spin interaction parameters are renormalized by the same function but α_{ij} is different for NN and NNN parameters. Thus, the ratios between these renormalized parameters are different than non-perturbed parameters which is a peculiar of honeycomb lattices described by the Kane-Mele-Hubbard model. The renormalized spin interactions presented in Eq. (17) are helicity independent.

Figure 2, shows the dependence of normalized nn exchange interaction $\tilde{J}_{1,ij}/J_{1,ij}$ and normalized NNN DMI $\tilde{D}_{ij}/D_{ij}$ in the dimensionless electric field parameter $\mathcal{E} = \frac{eaE_0}{\omega}$, where a is the lattice constant. The results are obtained for dimensionless material parameters $t_1 = 1$, $\mathrm{U} = 10$ and $\omega = 4$and14 (equivalently, $T = \frac{2\pi}{\omega} \approx 10^{-16}$s). Figure 2, shows that it is possible not only change the sign and amplitude of the exchange interaction as already reported in Ref. [5] but its is possible to change the signa nd amplitude of intrinsic DMI. More importantly,

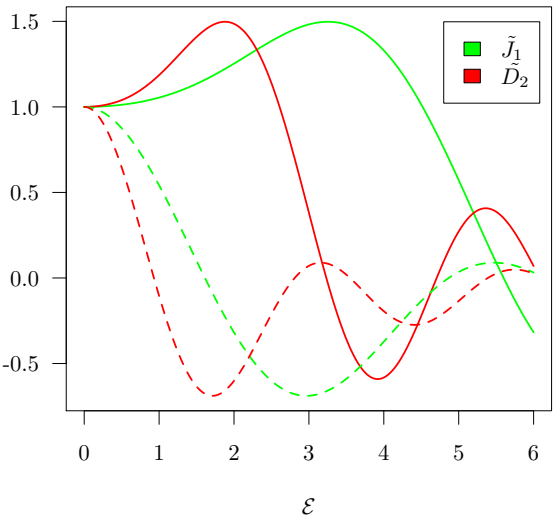

FIG. 2. Dimensionless exchange interaction (green lines) and DMI (red lines) as a function of $\mathcal{E} = \frac{eaE_0}{\omega}$, for two laser frequencies $\omega = 4$ (solid lines) and $\omega = 14$ (dashed lines).

the ratio $\tilde{J}_{1,ij}/\tilde{D}_{ij} \neq J_{1,ij}/D_{ij}$ can be tuned by laser excitations which is responsible for ultarafast photoinduced spin dynamics and magnon scattering phenomena.

Eqs. (16) and (17) explicitly show that the spin Hamiltonian in the presence of a time-dependent external field can be effectively written as $\tilde{H}_{\rm S} = H_{\rm S} + g_{\alpha\beta ij} S_i^{\alpha} S_j^{\beta} E^{\alpha} E^{*\beta}$, where α and β stand for spatial component of vectors, i and j refer to the lattice cite, and g is the opto-magnetic coupling tensor which can be read from Eqs. (17). Thus, the dielectric permittivity tensor which determine optical properties of the medium, is given by $\varepsilon_{\alpha\beta} = \partial^2 \tilde{H}_{\rm S}/\partial E^{\alpha} \partial E^{*\beta}$. This opto-magnetic effect described by the dielectric permittivity ε, can be detected by measuring the intensity of the scattered light $I_{\rm sc} \propto (\varepsilon_{\alpha\beta} E_0)^2$ [50].

In ultrafast spin dynamics experiments, very intense laser pulses are used and one might think about heating and concerning on the validity of our approach. Recent theoretical [51 and 52] and experimental [53] works show that the energy absorption rate is suppressed exponentially for high frequency laser pulses $\omega/W \gg 1$, where $W \propto t_1$ is the fermionic bandwidth, which is the case in ultrafast experiments with optical laser pulses. Thus, rapidly driven systems have a very long prethermalization period and this implies that the evolution of these systems in the presence of short laser pulses can be described by our formalism safely.

Possibility of ultrafast optical modification of exchange interactions in the bulk of iron oxides has been recently reported [11]. We hope that our work stimulate new experiments on measuring both exchange interaction and DMI in novel 2D magnetic systems.

In summary, we have investigated the effect of polarized and intense high-frequency laser pulses on 2D canted antiferromagnetic Mott insulators using Floquet theory. We have found that both sign and amplitude of the ratio between the exchange interaction and DMI is changed in honeycomb lattices irrespective to the helicity of laser pulses. Our calculations open a new way to ultrafast and energy-efficient control of spins and engineering of topological objects and topological properties of 2D van der Waals magnetic materials.

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