Letter

Floquet engineering of Mott insulators with strong spin-orbit coupling

Naoya Arakawa[1,*] and Kenji Yonemitsu[1,2]
[1]*The Institute of Science and Engineering, Chuo University, Bunkyo, Tokyo 112-8551, Japan*
[2]*Department of Physics, Chuo University, Bunkyo, Tokyo 112-8551, Japan*

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We propose a method for controlling the exchange interactions of Mott insulators with strong spin-orbit coupling. We consider a multiorbital system with strong spin-orbit coupling and a circularly polarized light field and derive its effective Hamiltonian in the strong-interaction limit. Applying this theory to a minimal model of α-$RuCl_3$, we show that the magnitudes and signs of three exchange interactions, J, K, and Γ, can be changed simultaneously. Then, considering another case in which one of the hopping integrals has a different value and the other parameters are the same as those for α-$RuCl_3$, we show that the Heisenberg interaction J can be made much smaller than the anisotropic exchange interactions K and Γ.

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Periodic driving enables us to control the magnetic properties of solids. The solution to the Schrödinger equation for a periodically driven system satisfies the Floquet theorem because of time periodicity of the driving field [1,2]. In particular, the time evolution in steps of the driving period T can be described by a time-independent Hamiltonian [3]. Since its parameters usually depend on the amplitude and frequency of the driving field, the properties could be controlled by tuning the driving field; such control is called Floquet engineering [4–6]. For example, by applying $E(t) = E_0 \cos \omega t$ to a single-orbital Mott insulator and tuning E_0 and ω, we can change the magnitude and sign of the antiferromagnetic Heisenberg interaction between magnetic moments [7,8]. Moreover, for a multiorbital Mott insulator without spin-orbit coupling (SOC), we can control the antiferromagnetic and the ferromagnetic contributions to the Heisenberg interaction via a time-periodic electric field [9,10]. Such control could be used to engineer the magnetic properties of solids because the exchange interactions are key quantities describing magnetization dynamics [11] and spintronics phenomena [12].

Although the magnetic properties of solids are affected by the SOC, Floquet engineering of Mott insulators with strong SOC is lacking. The magnetic properties of Mott insulators with strong SOC are described by spin and orbital coupled degrees of freedom [13,14]. As a result, not only the Heisenberg interaction but also the anisotropic exchange interactions contribute to the effective Hamiltonian [15–19]. For example, the effective Hamiltonians for α-$RuCl_3$ and the honeycomb iridates possess the Heisenberg interaction, the Kitaev interaction, and the off-diagonal symmetric exchange interaction [18,20–22]. Then the combinations of the Heisenberg interaction and the anisotropic exchange interactions cause various types of magnetic order [18,22–27]; if the Kitaev interaction is dominant, the spin-liquid states are stabilized [28]. Controlling the exchange interactions via a time-periodic field may provide a new opportunity to engineer their properties.

*arakawa@phys.chuo-u.ac.jp

Nevertheless, it is unclear how a time-periodic field changes the exchange interactions of Mott insulators with strong SOC.

In this work, we study the exchange interactions of periodically driven Mott insulators with strong SOC. We use a t_{2g}-orbital Hubbard model in the presence of strong SOC and a circularly polarized light field on the honeycomb lattice and derive its effective Hamiltonian in the strong-interaction limit. Applying this theory to the case of α-$RuCl_3$, we show that the magnitudes and signs of three exchange interactions can be changed. Then, studying another case of our model, we show that the Heisenberg interaction can be made much smaller than the anisotropic exchange interactions.

Setup. We consider a periodically driven multiorbital system described by

$$H = H_{\mathrm{KE}} + H_{\mathrm{SOC}} + H_{\mathrm{int}}, \tag{1}$$

where H_{KE}, H_{SOC}, and H_{int} represent the kinetic energy, the atomic SOC [14], and the local multiorbital Coulomb interactions [29], respectively. The kinetic energy is given by the hopping integrals of the t_{2g}-orbital electrons on the honeycomb lattice (Fig. 1) in the presence of a circularly polarized light field $\boldsymbol{E}(t) = {}^t(E_0 \cos \omega t \quad -E_0 \sin \omega t)$. The effects of $\boldsymbol{E}(t)$ are treated as Peierls phase factors:

$$H_{\mathrm{KE}} = \sum_{i,j} \sum_{a,b} \sum_{\sigma} t_{iajb} e^{-ie(\boldsymbol{R}_i - \boldsymbol{R}_j)\cdot \boldsymbol{A}(t)} c^{\dagger}_{ia\sigma} c_{jb\sigma}, \tag{2}$$

where $\boldsymbol{A}(t) = {}^t(-\frac{E_0}{\omega} \sin \omega t \quad -\frac{E_0}{\omega} \cos \omega t)$; hereafter, we use $\hbar = 1$. Then the atomic SOC of H_{SOC} is given by the LS coupling for the t_{2g}-orbital electrons [14]. The terms of H_{int} consist of the following interactions [29]:

$$H_{\mathrm{int}} = \sum_i \Bigg\{ \sum_{a,b} c^{\dagger}_{ia\uparrow} c^{\dagger}_{ia\downarrow} [U \delta_{a,b} + J'(1-\delta_{a,b})] c_{ib\downarrow} c_{ib\uparrow} + \sum_{\substack{a,b \\ a>b}} \sum_{\sigma,\sigma'} c^{\dagger}_{ia\sigma} c^{\dagger}_{ib\sigma'} (U' c_{ib\sigma'} c_{ia\sigma} - J_{\mathrm{H}} c_{ib\sigma} c_{ia\sigma'}) \Bigg\}. \tag{3}$$

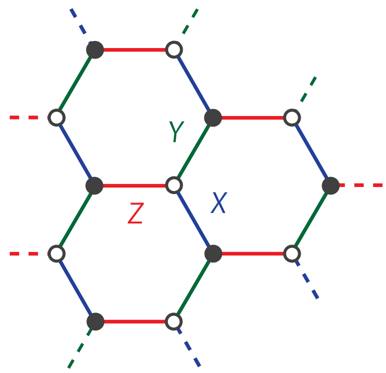

FIG. 1. Structure of the honeycomb lattice. X, Y, and Z denote three different nearest-neighbor bonds. Black and white circles represent the A and B sublattices, respectively.

As a specific example, we consider a minimal model of α-RuCl$_3$ [18]: t_{iajb}'s in H_{KE} can be parametrized by three hopping integrals between nearest-neighbor sites on the honeycomb lattice (Fig. 1). Namely, for the Z bond, the finite t_{iajb}'s are given by $t_{id_{yz}jd_{yz}} = t_{id_{zx}jd_{zx}} = t_1$, $t_{id_{yz}jd_{zx}} = t_{id_{zx}jd_{yz}} = t_2$, and $t_{id_{xy}jd_{xy}} = t_3$; for the X or Y bond, similar relations can be obtained from symmetry arguments [18]. Then the low-energy properties can be described by the $j_{\text{eff}} = 1/2$ states [18,30,31], $|+\rangle_i = \frac{1}{\sqrt{3}}(c^{\dagger}_{id_{yz}\downarrow} + ic^{\dagger}_{id_{zx}\downarrow} + c^{\dagger}_{id_{xy}\uparrow})|0\rangle$ and $|-\rangle_i = \frac{1}{\sqrt{3}}(c^{\dagger}_{id_{yz}\uparrow} - ic^{\dagger}_{id_{zx}\uparrow} - c^{\dagger}_{id_{xy}\downarrow})|0\rangle$, in which the spin and the orbital are entangled by strong SOC. Since such entanglement is the key property of strong SOC [14], this model will be sufficient for analyzing essential physics in the presence of strong SOC.

Floquet theory of Mott insulators. We derive an effective Hamiltonian for a periodically driven Mott insulator using the Floquet theory [10]. To derive it, we consider the strong-interaction limit in which t_{iajb}'s are much smaller than the energies of doubly occupied states, $U + 2J'$, $U - J'$, $U' - J_{\text{H}}$, and $U' + J_{\text{H}}$ [19,32]. In this limit, we can approximately express the solution to Schrödinger equation $|\Psi\rangle_t$ as $|\Psi\rangle_t \approx |\Psi_0\rangle_t + |\Psi_1\rangle_t$ [10], where $|\Psi_0\rangle_t$ and $|\Psi_1\rangle_t$ denote the states without and with, respectively, a doubly occupied site. As a result, Schrödinger's equation reduces to a set of simultaneous equations:

$$i\partial_t|\Psi_0\rangle_t = \mathcal{P}_0 H_{\text{KE}}|\Psi_1\rangle_t + H_{\text{SOC}}|\Psi_0\rangle_t, \tag{4}$$

$$i\partial_t|\Psi_1\rangle_t = H_{\text{KE}}|\Psi_0\rangle_t + (\mathcal{P}_1 H_{\text{KE}}\mathcal{P}_1 + \tilde{H}_{\text{int}})|\Psi_1\rangle_t, \tag{5}$$

where $\tilde{H}_{\text{int}} = H_{\text{int}} + H_{\text{SOC}}$; and $\mathcal{P}_0$ and $\mathcal{P}_1$ denote the projections onto the subspaces without and with, respectively, a doubly occupied site. Hereafter, we concentrate on the high-frequency case in which ω is much larger than t_{iajb}'s. In this case, we could replace the time-dependent operator $\mathcal{P}_1 H_{\text{KE}}\mathcal{P}_1$ in Eq. (5) by its time-averaged one $\bar{H}_{\text{KE}} = \sum_{i,j}\sum_{a,b}\sum_{\sigma} t_{iajb}\mathcal{J}_0(u_{ij})\mathcal{P}_1 c^{\dagger}_{ia\sigma}c_{jb\sigma}\mathcal{P}_1$ [10], where $\mathcal{J}_n(u_{ij})$ is the nth Bessel function of the first kind and $u_{ij} = \frac{eE_0}{\omega}\text{sgn}(i - j)$ [33]; the distance between nearest-neighbor sites is set to unity. By using this replacement, we can solve Eq. (5); the result [34] is

$$|\Psi_1\rangle_t = \sum_{i,j,a,b,\sigma}\sum_{n=-\infty}^{\infty}\frac{t_{iajb}\tilde{\mathcal{J}}_{-n}(u_{ij})e^{-in\omega t}}{n\omega - \bar{H}_{\text{KE}} - \tilde{H}_{\text{int}}}c^{\dagger}_{ia\sigma}c_{jb\sigma}|\Psi_0\rangle_t, \tag{6}$$

where $\tilde{\mathcal{J}}_{-n}(u_{ij}) = \mathcal{J}_{-n}(u_{ij})e^{-in\theta_{ij}}$ and $\theta_{ij} = \theta_{ji} = \frac{\pi}{3}$, π, and $\frac{5\pi}{3}$ for the Y, Z, and X bonds, respectively. Furthermore, since H_{int} gives the largest contribution of the terms of $\bar{H}_{\text{KE}}$ and $\tilde{H}_{\text{int}}(= H_{\text{int}} + H_{\text{SOC}})$, we replace $n\omega - \bar{H}_{\text{KE}} - \tilde{H}_{\text{int}}$ in Eq. (6) by $n\omega - H_{\text{int}}$; this replacement may be sufficient if ω is nonresonant, i.e., the denominator of Eq. (6) does not diverge. By using Eq. (6) with this replacement and omitting the constant term (i.e., $H_{\text{SOC}}|\Psi_0\rangle_t$), we can rewrite Eq. (4) as

$$i\partial_t|\Psi_0\rangle_t = H_{\text{eff}}(t)|\Psi_0\rangle_t, \tag{7}$$

where

$$H_{\text{eff}}(t) = \sum_{i,j}\sum_{a,b,c,d}\sum_{\sigma,\sigma'}\sum_{n,m=-\infty}^{\infty} t_{jcid}t_{iajb}\mathcal{P}_0 c^{\dagger}_{jc\sigma'}c_{id\sigma'} \times \frac{\tilde{\mathcal{J}}_m(u_{ji})\tilde{\mathcal{J}}_{-n}(u_{ij})e^{i(m-n)\omega t}}{n\omega - H_{\text{int}}}c^{\dagger}_{ia\sigma}c_{jb\sigma}\mathcal{P}_0. \tag{8}$$

The leading term of $H_{\text{eff}}(t)$ is given by the time-independent Floquet Hamiltonian. Since $H_{\text{eff}}(t)$ is time periodic, it can be expressed as the Fourier series $H_{\text{eff}}(t) = \sum_l e^{il\omega t}H_l$. Furthermore, by using a high-frequency expansion of the Floquet theory [4–6], $H_{\text{eff}}(t)$ can be written in the form $H_{\text{eff}}(t) = H_0 + O(\omega^{-1})$. Therefore, the time-averaged $H_{\text{eff}}(t)$, $\bar{H}_{\text{eff}}$, gives the leading term of Eq. (8); $\bar{H}_{\text{eff}}$ is given by

$$\bar{H}_{\text{eff}} = \sum_{i,j}\sum_{a,b,c,d}\sum_{\sigma,\sigma'}\sum_{n=-\infty}^{\infty} t_{jcid}t_{iajb}\mathcal{P}_0 c^{\dagger}_{jc\sigma'}c_{id\sigma'}\frac{\mathcal{J}_n(u_{ij})^2}{n\omega - H_{\text{int}}} \times c^{\dagger}_{ia\sigma}c_{jb\sigma}\mathcal{P}_0. \tag{9}$$

Application to periodically driven α*-RuCl*$_3$. Applying the above theory to the minimal model of α-RuCl$_3$, we derive its Floquet Hamiltonian. This derivation can be performed in a way similar to the derivation in the absence of a driving field. Here we describe the main points of the derivation (for the details, see the Supplemental Material [34]). To derive the expression of $\bar{H}_{\text{eff}}$ for the minimal model of α-RuCl$_3$, we suppose that in the subspace of $|\Psi_0\rangle_t$ a single hole occupies $j_{\text{eff}} = 1/2$ state per site. We also rewrite H_{int} using the irreducible representations of doubly occupied states [19]: $H_{\text{int}} = \sum_i\sum_{\Gamma,g_\Gamma} U_\Gamma|i;\Gamma,g_\Gamma\rangle\langle i;\Gamma,g_\Gamma|$, where $U_{A_1} = U + 2J'$, $U_E = U - J'$, $U_{T_1} = U' - J_{\text{H}}$, and $U_{T_2} = U' + J_{\text{H}}$; $|i;\Gamma,g_\Gamma\rangle$'s are expressed in the Supplemental Material [34]. Then, by calculating the contributions of possible hopping processes to $\bar{H}_{\text{eff}}$, we obtain [34]

$$\bar{H}_{\text{eff}} = \sum_{\langle i,j\rangle}\left[J\boldsymbol{S}_i\cdot\boldsymbol{S}_j + KS_i^{\gamma}S_j^{\gamma} + \Gamma\left(S_i^{\alpha}S_j^{\beta} + S_i^{\beta}S_j^{\alpha}\right)\right], \tag{10}$$

where

$$J = \sum_{n=-\infty}^{\infty}\frac{4\mathcal{J}_n(u_{ij})^2}{27}\left\{\frac{(2t_1+t_3)^2}{U+2J'-n\omega} + \frac{6t_2^2}{U'+J_{\text{H}}-n\omega} + \frac{2\left[(t_1-t_3)^2-3t_2^2\right]}{U-J'-n\omega} + \frac{6t_1(t_1+2t_3)}{U'-J_{\text{H}}-n\omega}\right\}, \tag{11}$$

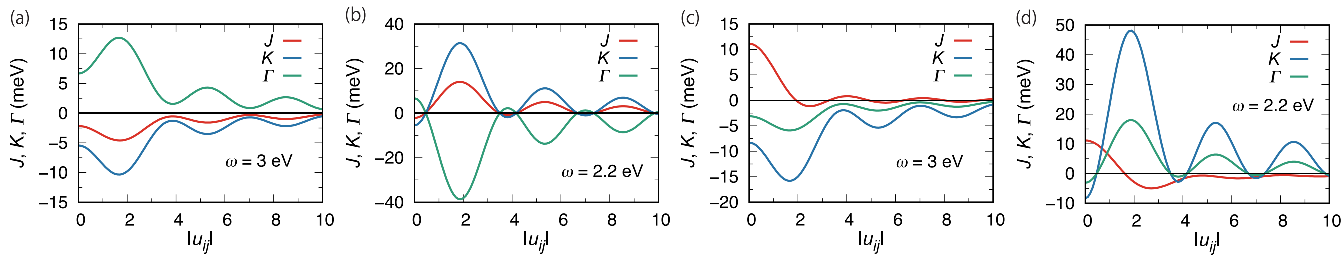

FIG. 2. The $|u_{ij}|(=|\frac{eE_0}{\omega}|)$ dependences of J, K, and Γ in (a) and (b) the case of α-$RuCl_3$ and (c) and (d) another case.

$$K=\sum_{n=-\infty}^{\infty}\frac{4\mathcal{J}_n(u_{ij})^2}{9}\left\{\frac{4t_2^2}{U-J'-n\omega}-\frac{\left[(t_1-t_3)^2+t_2^2\right]}{U'+J_{\rm H}-n\omega}+\frac{\left[(t_1-t_3)^2-3t_2^2\right]}{U'-J_{\rm H}-n\omega}\right\}, \quad (12)$$

$$\Gamma=\sum_{n=-\infty}^{\infty}\frac{16\mathcal{J}_n(u_{ij})^2t_2(t_1-t_3)J_{\rm H}}{9(U'-J_{\rm H}-n\omega)(U'+J_{\rm H}-n\omega)}, \quad (13)$$

and $(\alpha,\beta,\gamma)=(x,y,z),(y,z,x),(z,x,y)$ for the Z, X, and Y bonds, respectively; J, K, and Γ are the Heisenberg interaction, the Kitaev interaction, and the off-diagonal symmetric exchange interaction, respectively. [These expressions hold also for $\boldsymbol{E}(t)={}^t(E_0\cos\omega t\ E_0\sin\omega t)$.]

We now show how J, K, and Γ vary with ω and u_{ij}. To do it, we numerically evaluate Eqs. (11)–(13). We set $t_1=47$ meV, $t_2=160$ meV, $t_3=-129$ meV [35], $J'=J_{\rm H}$, $U'=U-2J_{\rm H}$, $U=3$ eV, and $J_{\rm H}=0.5$ eV; we replace $\sum_{n=-\infty}^{\infty}$'s by $\sum_{n=-n_{\max}}^{n_{\max}}$'s and set $n_{\max}=500$. Figures 2(a) and 2(b) show the $|u_{ij}|$ dependences of J, K, and Γ at $\omega=3$ and 2.2 eV. We see that by changing $|u_{ij}|$, the magnitudes of J, K, and Γ can be changed at $\omega=3$ and 2.2 eV, and that at $\omega=2.2$ eV it is possible to change not only their magnitudes but also their signs.

For a deeper understanding of the above results, we perform some analyses of Eqs. (11)–(13). Since $J'=J_{\rm H}$ and $U'=U-2J_{\rm H}$, J, K, and Γ can be rewritten as follows: $J=J_1+J_2+J_3$, where $J_1=\sum_n\frac{4\mathcal{J}_n(u_{ij})^2(2t_1+t_3)^2}{27(U+2J_{\rm H}-n\omega)}$, $J_2=\sum_n\frac{8\mathcal{J}_n(u_{ij})^2(t_1-t_3)^2}{27(U-J_{\rm H}-n\omega)}$, and $J_3=\sum_n\frac{8\mathcal{J}_n(u_{ij})^2t_1(t_1+2t_3)}{9(U-3J_{\rm H}-n\omega)}$. $K=K_1+K_2$, where $K_1=\sum_n\frac{4\mathcal{J}_n(u_{ij})^2[3t_2^2-(t_1-t_3)^2]}{9(U-J_{\rm H}-n\omega)}$ and $K_2=\sum_n\frac{4\mathcal{J}_n(u_{ij})^2[(t_1-t_3)^2-3t_2^2]}{9(U-3J_{\rm H}-n\omega)}$. $\Gamma=\Gamma_1+\Gamma_2$, where $\Gamma_1=\sum_n\frac{8\mathcal{J}_n(u_{ij})^2t_2(t_1-t_3)}{9(U-3J_{\rm H}-n\omega)}$ and $\Gamma_2=\sum_n\frac{8\mathcal{J}_n(u_{ij})^2t_2(t_3-t_1)}{9(U-J_{\rm H}-n\omega)}$. For the hopping parameters of α-$RuCl_3$, J_1 is much smaller than J_2 and J_3; as a result, $J\approx J_2+J_3$. This is the origin of the in-phase $|u_{ij}|$ dependences of J, K, and Γ [Figs. 2(a) and 2(b)]. Then we can understand the sign changes of J, K, and Γ at $|u_{ij}|\sim 0.4$, 3.5 [Fig. 2(b)] by estimating the dominant contributions. We make the estimate of J because the sign changes of K and Γ can be understood similarly. For $\omega=2.2$ eV, the dominant contributions are given by

$$J\approx\left(J_2^0+J_3^0\right)\mathcal{J}_0(u_{ij})^2+\left(J_2^0c_2-J_3^0c_3\right)\mathcal{J}_1(u_{ij})^2, \quad (14)$$

where $J_2^0=\frac{8(t_1-t_3)^2}{27(U-J_{\rm H})}$, $J_3^0=\frac{8t_1(t_1+2t_3)}{9(U-3J_{\rm H})}$, $c_2=\frac{U-J_{\rm H}}{\delta\omega_2}$, $c_3=\frac{U-3J_{\rm H}}{\delta\omega_3}$, and $\omega=U-3J_{\rm H}+\delta\omega_3=U-J_{\rm H}-\delta\omega_2$ (i.e., $\delta\omega_2=0.3$ eV and $\delta\omega_3=0.7$ eV). At $|u_{ij}|=0$, J is ferromagnetic, i.e., negative, because J_2^0 and J_3^0 satisfy $J_2^0>0$, $J_3^0<0$, and $J_2^0+J_3^0<0$. As $|u_{ij}|$ increases, the term including $\mathcal{J}_1(u_{ij})^2$ in Eq. (14), the positive-sign contribution, becomes considerable and causes a sign change of J. With further increases in $|u_{ij}|$, $\mathcal{J}_1(u_{ij})^2$ approaches zero, and the sign of J changes again.

Application to another case. We consider another case and study the effects of the driving field on the exchange interactions. In this case, we set $t_3=129$ meV and use the same values of the other parameters as those used in the case of α-$RuCl_3$; in a set of these values, J_1 is comparable to J_2 and J_3. Although it may be difficult to change the value of t_3 in α-$RuCl_3$, we study this case to clarify how the driving field changes J in the presence of non-negligible J_1. Figures 2(c) and 2(d) show the $|u_{ij}|$ dependences of J, K, and Γ in this additional case. We see that the $|u_{ij}|$ dependence of J differs from that of K or Γ. In particular, J can be very small in magnitude, while K and Γ are finite [see, for example, their values at $|u_{ij}|=1.6$ in Fig. 2(d)].

Discussion. We comment on the validity of our theory. First, the hopping integrals of our model are simplified compared with those obtained in the first-principles calculations [21]. However, since the leading terms are t_2 and t_3 [21], our model may be appropriate for a minimal model of α-$RuCl_3$. Then the obtained $|u_{ij}|$ dependences of J, K, and Γ might be affected by the doublon-holon hoppings described by $\bar{H}_{\rm KE}$. Nevertheless, we believe our results remain qualitatively unchanged. This is because the previous studies [9,10] show that in the frequency range where the correction due to $\mathcal{J}_1(u_{ij})^2$ is important and the corrections due to $\mathcal{J}_n(u_{ij})^2$'s for $n\geqslant 2$ are less important (the range of $U-2J_{\rm H}<\omega<U$ in Ref. [9]), the effects of the driving field on the exchange interactions remain qualitatively unchanged even if the doublon-holon hoppings are taken into account.

We also remark on heating effects. The periodically driven system eventually approaches an infinite-temperature state [36,37]. However, at intermediate times $t\lesssim\tau$ [38], it can be approximately described by the Floquet Hamiltonian as long as ω is nonresonant [10] and much larger than the exchange interactions [3,39–42]. Since these conditions hold in our study, the properties similar to our results could be realized experimentally.

We now discuss the implications of our results. First, our results in the case of α-$RuCl_3$ indicate that by tuning ω and changing E_0, one can change the magnitudes and signs of the three exchange interactions of periodically driven α-$RuCl_3$. In particular, by using this method, the Kitaev interaction can be made ferromagnetic (negative) or antiferromagnetic (positive). Since its sign drastically affects the magnetic properties of materials with strong SOC [22,43], our results will provide an opportunity for connecting the ferromagnetic and the antiferromagnetic Kitaev physics. Such control of the exchange interactions could be achieved by pump-probe measurements. Then our results in another case suggest that if the contribution from the doubly occupied state with A_1 symmetry is non-negligible, it is possible to make the Kitaev interaction much larger in magnitude than the Heisenberg interaction. Therefore, the periodically driven Mott insulator with strong SOC and hopping integrals that lead to such a contribution may be suitable for realizing the Kitaev model [28] and the spin liquid.

Conclusions. We have studied the exchange interactions of Mott insulators with a circularly polarized light field and strong SOC in two cases. In the case of α-$RuCl_3$, we have shown that J, K, and Γ have similar $|u_{ij}|$ dependences, and that their magnitudes and signs can be changed by tuning ω and varying E_0. These properties can be utilized for changing the exchange interactions of α-$RuCl_3$ and controlling its magnetic properties. In another case, we have shown that the $|u_{ij}|$ dependence of J differs from those of K and Γ, and that J can be made much smaller than K and Γ by tuning $|u_{ij}|$. The latter property suggests a new possibility of realizing the Kitaev spin liquid. Our results will provide the first step towards controlling the exchange interactions and magnetic properties of periodically driven Mott insulators with strong SOC.

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[34] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.103.L100408 for derivations of Eqs. (6) and (10), which includes Refs. [10,18,19,32].

[35] Our t_1, t_2, and t_3 correspond to the averages of t_1, t'_{1a}, and t'_{1b} of Ref. [21], of t_2 and t'_2, and of t_3 and t'_3, respectively.

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