Comparing the influence of Floquet dynamics in various Kitaev-Heisenberg materials

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In this paper we examine the possibility of Floquet engineering in the three candidate Kitaev materials Na\textsubscript{2}IrO\textsubscript{3}, α-Li\textsubscript{2}IrO\textsubscript{3} and α-RuCl\textsubscript{3}. We derive an effective Floquet Hamiltonian and give an approximation for heating processes arising from doublon holon propagation. This suggests that compounds with stronger Hund’s-rule coupling are less prone to heating. We then investigate the impact of light frequency and amplitude on magnetic interaction terms up to third-nearest-neighbor and find that third-nearest-neighbor Heisenberg coupling \(J_3\) is very susceptible to tuning by circularly polarized light. Finally, we discuss uses of linear polarized light in selectively tuning single bond directions.

I. INTRODUCTION

Light-matter interactions with periodic driving have gathered significant attention recently\textsuperscript{[13]}. In these systems the Floquet theorem is applicable, and they can be described with an effective time-independent Floquet Hamiltonian\textsuperscript{[4–7]}. Frequency, amplitude, and polarization are then capable of changing intrinsic interactions of the system. This paves the way to tune magnetic properties of materials via the driving field. While for large time scales the system thermalizes, the Floquet Hamiltonian has been argued to be valid at short, but experimentally accessible, timescales\textsuperscript{[8]}.

Floquet engineering is especially promising for candidate Kitaev-Heisenberg materials\textsuperscript{[9]}. Those materials are supposed to host a Kitaev spin liquid (KSL) for an idealized structure. However in reality most of them show a zig-zag ordering\textsuperscript{[10,13]}. Tuning these materials from zig-zag to the KSL has been a focal point of theoretical and experimental research. While tuning via pressure\textsuperscript{[14,16]}, magnetic field\textsuperscript{[17–21]}, graphen substrates\textsuperscript{[22]} and chemical doping\textsuperscript{[23,25]} has been investigated extensively, the approach of driving the Kitaev phase via a periodic light field is yet relatively unexplored.

Driving with light periodic in time might allow to alter the relative strength of the interactions, and therefore give rise to a KSL. In particular, materials that are already proximate to the Kitaev phase should be a good starting point. Such materials are, e.g., iridates like Na\textsubscript{2}IrO\textsubscript{3}\textsuperscript{[26]} and α-Li\textsubscript{2}IrO\textsubscript{3}\textsuperscript{[27]} and the ruthenate α-RuCl\textsubscript{3}\textsuperscript{[28]}. Since these materials have already been studied in absence of a light field\textsuperscript{[12,13,27,29,30]}, we base our studies on already well-established models\textsuperscript{[26]}.

Previous studies on Floquet engineering\textsuperscript{[31]} have focused on tuning nearest-neighbor interactions in α-RuCl\textsubscript{3} with circularly polarized light and highlighted the possibility of tuning Heisenberg interactions. However this phenomenon was just found after changing signs of a specific parameter. For iridates studies on Floquet engineering are still lacking.

In this paper we study the capabilities of Floquet engineering, for α-RuCl\textsubscript{3} as well as for α-Li\textsubscript{2}IrO\textsubscript{3} and Na\textsubscript{2}IrO\textsubscript{3}. First we consider an approximate approach to capture heating around the resonances\textsuperscript{[32]} in order to identify frequencies where it is small. This gives an insight on which of these materials are well suited for Floquet engineering. Introducing linear polarized light instead of circular polarized light, interactions become bond dependent. This gives the possibility to tune the interactions via the light angle. We finally discuss third-nearest-neighbor Heisenberg coupling. It was found to be significant in ruthenates (and iridates) and is argued to be responsible for the zig-zag magnetic ordering\textsuperscript{[30]}.

The paper is structured as follows: In Sec.\textsuperscript{II} we derive the effective Floquet Hamiltonian for the Kitaev-Heisenberg model, applying the results of\textsuperscript{[7,32–35]} to the already well established Kitaev-Heisenberg model\textsuperscript{[4,53]}. We introduce the off-resonance limit of the model as well as a possibility to describe the system near resonances. In Sec.\textsuperscript{II} we discuss the results for the materials α-RuCl\textsubscript{3}, α-Li\textsubscript{2}IrO\textsubscript{3} and Na\textsubscript{2}IrO\textsubscript{3}. In Sec.\textsuperscript{IIIB} we study heating and determine frequencies where Floquet driving without any considerable heating is possible. For these frequencies we then investigate the effects of Floquet driving on the Kitaev, Heisenberg and Γ terms in Sec.\textsuperscript{IIIC}. Furthermore we also consider third-nearest-neighbor Heisenberg terms (Sec.\textsuperscript{IID}) and the impact of the polarization angle on the bond dependent Kitaev, Heisenberg and Γ interactions (Sec.\textsuperscript{IIIE}). Section\textsuperscript{IV} concludes the paper and gives a summary of the results obtained as well as an outlook to promising future studies.

II. THEORY

A. Effective model

The materials considered in this paper have strong Coulomb repulsion, stabilizing a \(d^5\) configuration with one hole residing on each site. Taking into account virtual excitations with two holes on one site via second order perturbation theory, gives rise to a Kugel-Khomskii–
type model [36]. Due to strong spin-orbit coupling (SOC) present in the considered materials, we can simplify the Hamiltonian further with a projection into the \( j_{\text{eff}} = 1/2 \) pseudospin basis. This leads to the well-known Kitaev-Heisenberg model [57].

Coupling to a periodic light field enters via Peierls substitution into the Hubbard model

\[
H_{\text{kin}}(t) = \sum_{\langle i,j \rangle, \gamma} e^{i(r_{i,j} - r_{0})A(t)} c^\dagger_{i,\gamma} \mathbf{T}^\gamma c_{i,\gamma},
\]

where \( c_i = (c_{i,x,\sigma}, c_{i,y,\sigma}, c_{i,z,\sigma}) \) (\( c_i^\dagger \)) annihilates (creates) a hole at site \( i \) with spin \( \sigma \). For \( d \)-orbitals, we used the notation \( yz(x), zx(y), \) and \( xy(z) \) introduced in [35]. The bond-dependent hopping matrices \( \mathbf{T}^\gamma \) are given by

\[
\begin{align*}
\mathbf{T}^x &= \begin{pmatrix}
\frac{t_1}{2} & \frac{t_2}{2} & \frac{t_3}{2} & \frac{t_4}{2}
\end{pmatrix},
\mathbf{T}^y &= \begin{pmatrix}
\frac{t_1}{2} & \frac{t_4}{2}
\frac{t_2}{2} & \frac{t_3}{2}
\end{pmatrix},
\mathbf{T}^y &= \begin{pmatrix}
\frac{t_1}{2} & \frac{t_3}{2}
\frac{t_2}{2} & \frac{t_4}{2}
\end{pmatrix}.
\end{align*}
\]

(1)

Light interaction \( \Omega(t) \) depends on the vector potential \( \mathbf{A}(t) \) and the vector \( \mathbf{r}_{ij} \), between nearest-neighbors (NN) \( i \) and \( j \). In this paper we will primarily focus on linear polarized (LP) light, i.e. the vector potential \( \mathbf{A}(t) = (E_x, E_y) \), \( \sin(\omega t) \). With this at hand \( \Omega(t) \) becomes \( \sum_{l=\infty} J_{\text{eff}}(u_{ij}) e^{-i\omega t}, \) where \( J_{\text{eff}}(u_{ij}) \) are Bessel functions of first kind and \( u_{ij} = \epsilon/\omega \mathbf{r}_{ij} E_0 (\cos(\phi), \sin(\phi)) \). The on site interactions can be written with a Kitanom Hamiltonian [35]

\[
H_{\text{int}} = U \sum_{i,\alpha} n_{i,\alpha} n_{i,\alpha} + U' \sum_{i,\alpha < \beta} n_{i,\alpha} n_{i,\beta} - J_H \sum_{i,\alpha < \beta} (c^\dagger_{i,\alpha} c_{i,\beta} - c^\dagger_{i,\beta} c_{i,\alpha}),
\]

(2)

with intraorbital interaction \( U \), interorbital interaction \( U' = U - 2J_H \) and Hund’s coupling \( J_H \).

Due to the time dependence in \( H_{\text{kin}}(t) \), we apply time dependent perturbation theory. Here we limit ourselves to the subspace of states with zero and one doublon-holon (DH) pair, meaning we only consider virtual \( d^2d^2 \rightarrow d^0d^0 \) excitations [32].

Writing down the time-dependent Schrödinger equations for this subspace yields

\[
\begin{align*}
\frac{i}{\hbar} \frac{\partial}{\partial t} |\Psi_0\rangle &= H_{\text{int}} |\Psi_0\rangle + \sum_m P_0 H_{\text{kin}}(t) |\Psi_{1,m}\rangle, \\
\frac{i}{\hbar} \frac{\partial}{\partial t} |\Psi_{1,m}\rangle &= P_{1,m} H_{\text{kin}}(t) |\Psi_{1,m}\rangle + (T_{nm} + H_{\text{int}}) |\Psi_{1,m}\rangle,
\end{align*}
\]

(3)

with \( P_0 \) the projector on the zero DH pair subspace, and \( P_{1,m} \) the projector on the one-DH pair subspace with energy \( E_m \). \( |\Psi_{1,m}\rangle \) denotes the subspace of one DH pair with energy \( E_m \). Propagation of the doublon and holon through the lattice within the associated subspace is described by \( T_{nm} = P_{1,m} H_{\text{kin}}(t) P_{1,m} \). We dropped \( T_{nm} \) terms, which couple two energetically distinct subspaces with one doublon holon pair, justified by [32].

\[
T_{nm} \text{ can be averaged over time as shown in [7], which yields } T_{nm} = \langle \mathcal{J}_{\text{eff}}(u_{ij}) \rangle_{P_{1,m}} v_{ij}^m v_{ij}^0. \text{ With this at hand we rewrite (5) to}
\]

\[
|\Psi_{1,m}\rangle = \sum_{ij,l} \frac{1}{\Delta E_m - \omega + \Delta E_m} e^{-i\omega t} \mathcal{J}_{\text{eff}}(u_{ij}) v_{ij}^m v_{ij}^0 |\Psi_0\rangle,
\]

(4)

with \( v_{ij}^m \) \( = \) \( v_{ij}^0 \) \( P_{1,m} v_{ij} \). \( \Delta E_m = E_m^1 - E_0 \). Here we performed a partial integration over time and dropped terms \( \mathcal{O}(t/mU^2) \). Plugging (6) into (4), we derive the effective time-dependent Hamiltonian

\[
H_{\text{eff}}(t) = \sum_{ij,\alpha \beta} v_{ij}^m \mathcal{J}_{\text{eff}}(u_{ij}) \mathcal{J}_{\text{eff}}(u_{ij}) e^{i(l-k)t - \omega \Delta E_m} v_{ij}^m \cdot
\]

(5)

In contrast to the time independent case the hopping amplitude is now modulated by frequency \( \omega \), amplitude \( E_0 \) and angle \( \phi \) of the incoming light.

B. Floquet Formalism

In Sec. IIA we derived an effective Hamiltonian periodic in time. It has been shown that Floquet theory can describe these systems with an effective time-independent Hamiltonian \( \tilde{H}_{\text{eff}} \) [35] [39]. For \( \omega >> t \) a Magnus expansion up to first non-vanishing order \( \mathcal{O} \) is sufficient to derive the time independent Floquet Hamiltonian

\[
\tilde{H}_{\text{eff}} = \sum_{ij,\alpha \beta} v_{ij}^0 \mathcal{J}_{\text{eff}}(u_{ij})^2 \Delta E_m - \omega + \Delta E_m v_{ij}^m \cdot
\]

(6)

For \( U >> \lambda >> t \) the effective Floquet Hamiltonian can be projected into the \( j_{\text{eff}} = 1/2 \) basis favored by SOC [34] [35]

\[
\tilde{H}_{\text{eff}} = \sum_{i,j,\alpha \beta} \left[ J^\gamma S_i S_j + K^\gamma S_i^\gamma S_j^\gamma + \Gamma^\gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\gamma) + \Gamma^{\gamma\gamma}(S_i^\alpha S_j^\alpha + S_i^\beta S_j^\beta + S_i^\gamma S_j^\gamma + S_i^{\gamma\gamma} S_j^{\gamma\gamma}) \right].
\]

(7)

This takes the form of a Kitaev-Heisenberg model, with the difference being that the factors \( J^\gamma, K^\gamma, \Gamma^\gamma \), and \( \Gamma^{\gamma\gamma} \) depend on frequency \( \omega \), amplitude \( E_0 \) and light angle \( \phi \) in addition to the bond direction \( \gamma \). The interaction
These are described by the term only the Heisenberg term (10) includes circular polarized (CP) light, the bond dependence is only more detail in Sec. III E. We will discuss advantages of both polarization types in A become negligible, meaning the Green's function simply becomes
\( \Phi \) excitations. It has already been shown [30], that \( H \) in (10)-(13) describe a band spin-orbital models for \( \text{YTiO}_3 \) and \( \text{LaTiO}_3 \) [47-50].

Let us first comment on the RPA’s applicability. If there was only the \( \Gamma = \frac{1}{2} \) doublet, the Kitaev-Heisenberg model could be seen as a quasi one-band model. In reality, however, there is also the possibility of \( d_2 \) in addition to \( d_1 \) doublons, i.e., excitation of a hole into the \( \frac{1}{2} \) states. Additionally, the four \( \frac{1}{2} \) - \( \frac{1}{2} \) states correspond to two ‘bands’ already by themselves. A one-band approach can than only work as long as mixing between \( d_1 \) and \( d_2 \) doublons remains small and if the \( \frac{1}{2} \) states can be neatly separated into two weakly mixed bands. Fortunately, resonant inelastic X-ray experiments indicate that this is the case, because trigonal distortion splits the two \( \frac{1}{2} \) sub-bands [42-44], which suppress their mixing. The Green’s functions relating to \( d_1 \) doublons and to the two sub-bands can then be approximated as independent one-band problems. Projecting (15) into the pseudospin basis leaves us then with different Green’s functions, for \( d_1 \) doublons with hopping amplitudes \( t_{B,1} \) and for \( d_2 \) doublons with hopping amplitude \( t_{B,2} \).

In order to perform the RPA we first rewrite the DH Green’s function in terms of a holon \( g_h \) and a doublon \( g_d \). Green’s function, assuming that holon and doublon movements are uncorrelated. This can be done by a convolution integral as shown in [7, 32]. Only doublon and holon paths in the excited subspace that end at their starting point are allowed. In addition, we exclude “loop” paths, leaving only retracable paths [41]. A detailed derivation can be found in [32].

For this approximation the Green’s function becomes complex and no longer diverge at \( \Delta E_m = \omega \). Its imaginary part describes the heating due to DH propagation. The bandwidth of the heating is \( 2\sqrt{z} - 1B \) [2], with the coordination number of the lattice \( z \) and the DH hopping strength \( 1B \). In the remainder of the paper we set \( t_B = t_{B,1} = t_{B,2} \).

III. RESULTS

A. Materials

As promising candidates for a realization of the \( J-K-\Gamma \) model several iridates and ruthenates have been suggested [9, 26, 28, 30, 35, 46]. In this paper we will focus on the three compounds Na$_3$IrO$_3$, \( \alpha \)-Li$_2$IrO$_3$, and \( \alpha \)-RuCl$_3$. In the following discussion we use the hopping parameters of [31] with the only distinction that we chose symmetrized hopping parameters \( t_{ij}^{\sigma} = \frac{1}{2}(t_{ij}^+ + t_{ij}^-) \) and \( t_{ij}^{3\sigma} = \frac{1}{2}(t_{ij}^{+3} + t_{ij}^{-3}) \). The values of Hund’s coupling \( J_H \) and Coulomb repulsion \( U \) for iridates and ruthenates are taken from [45] and [28] respectively. All considered parameters are listed in Tab. 1.

Notably all materials possess an anisotropy in z-direction, which intrinsically calls for bond dependent treatment of \( J, K \), and \( \Gamma \).
For Na$_2$IrO$_3$ [Fig. 1(b)] heating is present around a relatively broad area around the resonances, leaving a narrow area around $\omega = 1.1 \text{eV}$, where heating-free driving is possible. In this area, from now on referred to as driving corridor the ORA is expected to be valid. For frequencies $\omega < U - 3JH$ driving without heating becomes difficult due to a multitude of resonances. Evidently the width of the driving corridor is not only dependent on $t_B$ but also the magnitude of $J_H$. Increasing $J_H$ separates the resonances and therefore enhances the driving corridor. A broad driving corridor is a desirable feature for experiments. In Li$_3$IrO$_4$ heating effects are very similar to Na$_2$IrO$_3$. Therefore we omit a more detailed discussion in this section.

In comparison to iridates, $\alpha$-RuCl$_3$ has significant larger Coulomb repulsion and Hund’s interaction. Fig. 1(b) clearly displays that the resonance peaks are further apart than in Fig. 1(a), causing a broader driving corridor. This makes $\alpha$-RuCl$_3$ a more promising candidate for Floquet engineering than iridate compounds. Also the ORA appears to be well justified within the driving corridor.

We conclude that both iridates and ruthenates are susceptible to Floquet engineering within a certain driving corridor. This driving corridor is mainly determined by $J_H$ and $t_B$. Materials with large Hund’s interaction and small $t_B$ should be primed for Floquet engineering. Within this driving corridor the ORA is expected to be valid and differences to the RPA should be minute.

### C. Floquet engineering in the ORA

After considering possible influences of heating, we focus on Floquet driving within the driving corridor between the resonances at $U - 3J_H$ and $U - J_H$. To do so we set the frequency to $\omega = 1.1 \text{eV}$ for iridates and $\omega = 1.6 \text{eV}$ for the ruthenate. As we can see in Fig. 2 varying $E_0$ can change both magnitude and sign of the interaction terms. In addition to ORA results (solid lines), the results for the RPA with $t_B = 0.4$ are displayed (dashed lines). Since the differences are minor and heating is negligible (see Sec. III B), we will from here on only discuss the ORA results.

The interaction terms for Na$_2$IrO$_3$ are shown in Fig. 2(a). The Kitaev interaction is dominant throughout the whole parameter range, while the other terms are considerably smaller. We also note that the absolute magnitude of the interactions decreases with increasing $E_0$. The inset in Fig. 2(a) clearly displays that $J$ can be tuned relative to the $K$, $\Gamma$ and $\Gamma'$ interactions, by changing $E_0$.

On the z-bond of Na$_2$IrO$_3$ this effect is particularly visible since the factor $2t_1 + t_3$ in $B_3(\omega, U, J_H)$ becomes relatively large, due to $t_1 > 0$ and $t_3 > 0$. This effect has already been discussed in [11]. However this has been done with a change of sign in $t_3$ for $\alpha$-RuCl$_3$. It is encouraging that this effect also arises for realistic pa-

### TABLE I. Parameters for Na$_2$IrO$_3$, $\alpha$-Li$_2$IrO$_3$, and $\alpha$-RuCl$_3$. 1$^{st}$- and 3$^{rd}$-nearest-neighbor (NN) hopping parameters in meV are taken from [30]. Coulomb repulsion $U$ and Hund’s coupling in eV taken from [45] for iridates and from [28] for ruthenates

| Material       | Na$_2$IrO$_3$ | $\alpha$-Li$_2$IrO$_3$ | $\alpha$-RuCl$_3$ |
|----------------|---------------|------------------------|-------------------|
| $U$            | 1.7           | 1.7                    | 3.0               |
| $J_H$          | 0.3           | 0.3                    | 0.5               |
| NN             | 1$^{st}$      | 3$^{rd}$               | 1$^{st}$          | 3$^{rd}$         |
| $t_1^\text{rd}$| 33.1          | -9.3                   | 55.0              | -6.4             |
| $t_2^\text{rd}$| 264.3         | -13.8                  | 219.0             | -13.5            |
| $t_3^\text{rd}$| 26.6          | -30.8                  | -175.1            | -33.3            |
| $t_4^\text{rd}$| -11.8         | 16.6                   | -124.5            | 16.6             |
| $t_1^\text{1st}$| 38.8          | -8.4                   | 76.3              | -6.3             |
| $t_2^\text{1st}$| 269.3         | -12.7                  | 252.7             | -13.4            |
| $t_3^\text{1st}$| -19.4         | 35.3                   | -108.8            | -33.0            |
| $t_4^\text{1st}$| -23.4         | 16.0                   | -9.3              | 15.8             |

FIG. 1. Imaginary part of $K_z$ plotted for varying light frequency $\omega$ and amplitude $E_0$ in eV, with the parameter set of Na$_2$IrO$_3$ a) and $\alpha$-RuCl$_3$ b). For the calculations of the Green’s functions the retracable path approximation (RPA) was used.

### B. Heating in dependency of $\omega$ and $E_0$

In this section we investigate possible heating effects arising from DH propagation captured by the RPA. The bandwidth $t_B$ is set to $t_B = 0.04 \text{eV}$. Representative for the other interactions, Kitaev interactions are analyzed in dependency of the frequency $\omega$ and the amplitude $E_0$ of the LP light. We only discuss z-Bond terms to get a qualitative understanding of heating effects. To do so we examine the imaginary part of the Kitaev interaction $K_z$ [11] calculated with RPA.
parameters in Na$_2$IrO$_3$. For a tuning of $J$ it is sufficient to have a comparable energy scale of $A_i(\omega, U, J_H)$ and $B_i(\omega, U, J_H)$. On the z-Bond of Na$_2$IrO$_3$, we therefore have the ability to completely turn off the Heisenberg interactions at $E_0 = 2$ eV and reduce the model to a $K$-$\Gamma$-model. However this is not the case for the x- and y-

Bond in Na$_2$IrO$_3$, due to the fact that $t^{x/y}_i < 0$ for these bonds, which reduces the influence of $B_i(\omega, U, J_H)$.

In Li$_2$IrO$_3$ the Kitaev interaction is far less dominant, and the $J$, $K$, $\Gamma'$ terms all are significant [Fig. 2(b)]. Moreover, the values of $t_1$ and $t_3$ (see Tab. I) imply a low tunability of $J$. In Fig. 2(b) we indeed see that the $J$ term is almost in phase with the other terms. It is however still possible to adjust magnitude and sign of the interaction terms with the help of light frequency and amplitude.

For $\alpha$-RuCl$_3$ results for all interaction parameters are shown in Fig. 2(c). As in the case of $\alpha$-Li$_2$RuO$_3$, the $J$ parameter is mostly in phase with the other terms. Again the reason for that is the sub-optimal combination of the $t_1$ and $t_3$ parameters. Compared to iridates, the frequency of the oscillation is significantly lower. The origin is the larger $\omega$ needed to suppress heating in ruthenates, which enters the Bessel function as $\frac{\omega}{\omega_0}$.

The results of Fig. 2(c) thus confirm [31], in that for realistic parameters and only considering NN interactions a tuning of $J$, $K$, $\Gamma'$, and $\Gamma''$ relative to each other is not possible. However we found that in other materials like Na$_2$IrO$_3$ a tuning of the Heisenberg term is possible due to a more suitable combination of the hopping parameters.

D. Third-nearest-neighbor Heisenberg interactions

Third-nearest-neighbor (3$^{rd}$NN) Heisenberg interactions are argued to have a significant impact on the formation of the zig-zag ground state found in iridates and ruthenates [17]. Especially in Na$_2$IrO$_4$, $J_3$ is a main reason for the absence of the KSL [30]. For this reason, we investigate the possibility of tuning $J_3(E_0, \omega)$ relative to the NN interactions.

$J_3$ is calculated in the same manner as $J$ [with (10)]. The hopping parameters considered for $J_3$ are taken from [30] and listed in Tab. I. We note that the requirement for a significant tuning, a large $|2t_1 + t_3|$, is fulfilled for all materials. In addition, the larger distance $|r_{ij}|$ for third nearest neighbors changes the frequency of $J_3(u_{ij})$. Together with the promising hopping parameters, this suggests that $J_3$ can be tuned relative to the other interaction parameters, similar to $J_2$ in Na$_2$IrO$_3$.

Since we want to compare interaction terms of bonds with different directions (3$^{rd}$NN and NN) CP light is more suitable. This is the case since for CP $u_{ij} = e / \omega E_0 |r_{ij}^3|$ becomes bond independent, due to $|r_{ij}^3| = |r_{ij}|$ for NN’s. The only distinctions between NN and 3$^{rd}$NN are therefore the distance $|r_{ij}|$ and the hopping parameters.

In Fig. 3 the normalized $\tilde{K}$, $\tilde{\Gamma}$, $\tilde{\Gamma}'$ and $\tilde{J}_3$ interactions are shown. As normalization factor the maximum of the respective interaction in the considered $E_0$ range is chosen. It becomes obvious that for all materials $J_3$ decreases significantly faster with $E_0$ than the NN interactions. This gives a pathway to suppress $J_3$ via the light frequency of the oscillation is significantly lower. The origin is the larger $\omega$ needed to suppress heating in ruthenates, which enters the Bessel function as $\frac{\omega}{\omega_0}$.

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amplitude $E_0$. Due to the differing frequencies for NN and 3rd NN it is also possible to completely turn off $J^3$ while having finite NN interactions.

In general CP light is well suited for isotropic materials since it does not change the magnitude of bond interactions relative to each other. However if a material has anisotropic interaction terms, LP light has significant advantages over CP light, which we discuss in the following section.

\section*{E. Light Angle}

Up until now we only considered LP light parallel to the $z$-axis. In general LP light can point in an arbitrary direction, making it a parameter that is experimentally adjustable. The angle of the incoming light has an impact on the interaction terms, therefore it should be possible to tune the interactions by just changing the light angle $\varphi$. We expect the light angle $\varphi$ to alter the relative strengths of the $x$, $y$ and $z$ interaction terms, hence we now consider the $x$ and $y$ bond in addition to the $z$ bond.

The focus in this chapter is tuning the Kitaev interactions, which is why for the remainder of the section we only consider $K_x$, $K_y$, and $K_z$. We calculate the Kitaev interactions depending on the light angle $\varphi$ and the driving amplitude $E_0$ with the ORA. Since the interaction terms are $\pi$-periodic we only consider $\varphi \in [0, \pi]$. The results for $\alpha$-RuCl$_3$ at $\omega = 1.6$ eV are displayed in Fig. 4.

The corridors at $\varphi = \pi/6$, $\pi/2$, and $5\pi/6$ for the $y$, $z$- and $x$-bond interaction respectively are the most notable features. For these angles the Kitaev term of the respective bond is independent of $E_0$. We can see that if $\mathbf{r}_{ij} \perp \mathbf{A}(t)$ the interaction decouples from the light, since $u_{ij} = 0$. Therefore it is possible to tune two bond interactions while the coupling along the remaining bond direction remains unchanged. This opens the path to eliminate anisotropies intrinsic to, e.g., $\alpha$-Li$_2$IrO$_3$ and $\alpha$-RuCl$_3$.

The second effect we observe is that depending on the light angle and the driving amplitude, the Kitaev interactions can have different signs for different bonds. Additionally, the magnitude of the interactions also drastically depends on the light angle $\varphi$. As a consequence of these two effects, the Kitaev interactions might no longer support a gapless Kitaev phase, but rather give rise to a gapped Kitaev phase, due to the strong anisotropies.

We are aware that in all considered materials the Kitaev phase is not yet realized. In this section we therefore want to discuss the nature of the Kitaev phase if it were
to be realized. Interactions giving rise to a gapped (gapless) Kitaev phase are referred to as gapped (gapless) interactions.

The criterion for gapless interactions $K_\alpha + K_\beta > K_\gamma$, with $\alpha, \beta, \gamma \in \{x, y, z\}$, does not hold for arbitrary $\varphi$ and $E_0$. Applying this condition to the Kitaev terms of Fig. 4 we see where gapped and gapless interactions arise. The results for the different materials are displayed in Fig. 5. We also added isotropic interactions with the condition $|K_\alpha - K_\beta| < \epsilon$, for all combinations of $\alpha, \gamma$ and $\epsilon = 10^{-3}$. This isotropic interaction is included to highlight that Floquet engineering is capable of increasing the isotropy in materials [Fig. 5(c)].

For Na$_2$IrO$_3$ we observe that without driving the NN Kitaev interactions are already almost isotropic. Turning on the LP light then breaks the isotropy and also gives rise to gapped interactions. Around $\varphi = \pi/6$, $\pi/2$, and $5\pi/6$ the interactions are very likely to be gapless.

In case of $\alpha$-Li$_2$RuO$_3$ the system has gapless interactions in the absence of LP light. Again turning on LP light can lead gapped interactions. In contrast to Na$_2$IrO$_3$, there is only a preference of gapless interactions at $\pi/2$. This can be explained due to the large intrinsic anisotropy in $z$-direction. Also the areas of a gapped interactions are significantly larger than in Na$_2$IrO$_3$ leading to the conclusion that deriving a gapless interaction in the presence of a LP light field might be complicated.

Like in $\alpha$-Li$_2$RuO$_3$, the high intrinsic anisotropy in $\alpha$-RuCl$_3$ allows a stable gapless interaction only at $\pi/2$. However unlike the case in the iridate, the ruthenate actually can be driven into an isotropic interaction at $\varphi = \pi/2$ and $E_0 \approx 7$ eV.

IV. DISCUSSION AND CONCLUSIONS

We have investigated Floquet engineering for a variety of candidate Kitaev materials. To do so we derived an effective Floquet Kitaev-Heisenberg model for LP light similar to the CP light model of [35]. The Green’s functions in [15] were approximated with the ORA and RPA.

With help of the RPA we were able to determine a frequency range where the ORA is valid. In addition, we also identified frequencies where heating can be assumed to be negligible. This lead us to the conclusion that Floquet engineering in ruthenates should in principle be more promising than in iridates.

For suitable off-resonance frequencies, we investigated the influence of the light amplitude on the nearest-neighbor interaction terms (Fig. 2), where our results qualitatively agree with the results reported in Ref. [31]. We thus observe that tuning in magnitude and sign of the interactions in dependency of the light amplitude is possible. For instance, Heisenberg $J$ can be tuned relative to the other parameters, especially on the $z$-bond of the Na$_2$IrO$_3$ compound.

Going beyond nearest-neighbors, we find that third-nearest-neighbor Heisenberg interactions $J_3$ are quite susceptible to Floquet tuning. With the right setting of frequency and amplitude, a significant decrease of the third-nearest-neighbor interaction terms should be achievable in all considered materials. Circular polarized light is expected to work better than linear polarization, and could be used to suppressing zig-zag order, which is stabilized by $J_3$ and competes with the Kitaev spin liquid.

Linear polarized light, on the other hand, allows us to tune the interactions in different directions, with the help of the angle of the incoming light. Starting from a pure Kitaev model, this can either lead to a gapped model or more isotropic Kitaev interactions. It is also possible to decouple single bonds completely from the light so that they remain unaffected, so one can specifically “erase” anisotropies in one direction.

After investigating the more traditional Kitaev candidate materials, it would be interesting to consider materials which at first glance seem unattractive due to e.g.
high anisotropies, which could be tuned into promising candidates with the help of Floquet engineering. Finally, materials with strong $U$ and $J_H$ are particularly promising, due to a wide driving corridor. This suggests materials based on 3d elements [48][49], where these interactions tend to be stronger.

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