Ultrafast Spin Dynamics and Photoinduced Insulator-to-Metal Transition in α-RuCl₃

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ABSTRACT: Laser-induced ultrafast demagnetization is a phenomenon of utmost interest and attracts significant attention because it enables potential applications in ultrafast optoelectronics and spintronics. As a spin−orbit coupling assisted magnetic insulator, α-RuCl₃ provides an attractive platform to explore the physics of electronic correlations and unconventional magnetism. Using time-dependent density functional theory, we explore the ultrafast laser-induced dynamics of the electronic and magnetic structures in α-RuCl₃. Our study unveils that laser pulses can introduce ultrafast demagnetizations, accompanied by an out-of-equilibrium insulator-to-metal transition in a few tens of femtoseconds. The spin response significantly depends on the laser wavelength and polarization on account of the electron correlations, band renormalizations, and charge redistributions. These findings provide physical insights into the coupling between the electronic and magnetic degrees of freedom in α-RuCl₃ and shed light on suppressing the long-range magnetic orders and reaching a proximate spin liquid phase for two-dimensional magnets on an ultrafast time scale.

KEYWORDS: ultrafast spin dynamics, insulator-to-metal transition, α-RuCl₃, two-dimensional magnets, TDDFT

In correlated materials, microscopic degrees of freedom (e.g., electrons, phonons, excitons, and magnons) are intertwined, and their interplay is prominent to understanding the macroscopic properties of quantum materials. Transition in magnetic effects in correlated insulators. Electronic correlations, unconventional magnetism, and opto-electronics provides an attractive platform to explore the physics of macroscopic properties of quantum materials. As a spin−orbit coupling assisted magnetic insulator, α-RuCl₃ enables potential applications in ultrafast optoelectronics and spintronics. As a spin−orbit coupling assisted magnetic insulator, α-RuCl₃ provides an attractive platform to explore the physics of electronic correlations and unconventional magnetism. Using time-dependent density functional theory, we explore the ultrafast laser-induced dynamics of the electronic and magnetic structures in α-RuCl₃. Our study unveils that laser pulses can introduce ultrafast demagnetizations, accompanied by an out-of-equilibrium insulator-to-metal transition in a few tens of femtoseconds. The spin response significantly depends on the laser wavelength and polarization on account of the electron correlations, band renormalizations, and charge redistributions. These findings provide physical insights into the coupling between the electronic and magnetic degrees of freedom in α-RuCl₃ and shed light on suppressing the long-range magnetic orders and reaching a proximate spin liquid phase for two-dimensional magnets on an ultrafast time scale.

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introduce spin dynamics, which is a promising route to understand correlated physics and calls for studies of the underlying mechanism of photoexcitation under extreme conditions.

In this article, we employ ab initio calculations within the framework of real-time time-dependent density functional theory (TDDFT) to investigate laser-driven spin dynamics of the two-dimensional magnet α-RuCl₃. To understand its optical response, we undertake a comprehensive evaluation of the electronic and magnetic properties of α-RuCl₃, and further simulate its dynamical response to laser pulses with different photon energies and intensities. Based on the recently developed ACBN0 functional, the spin dynamics of the correlated insulator as well as ultrafast melting of the bandgap is explored. We find that the photoinduced demagnetization significantly depends on the laser wavelength on account of the photoexcited band renormalization (i.e., insulator-to-metal transitions) and carrier excitation. The delicate interplay of the photodoping effect and the insulator-to-metal transition suggests a way to drive the electronic and magnetic structures out of equilibrium on a time scale of tens of femtoseconds.

**ELECTRONIC AND MAGNETIC PROPERTIES OF α-RuCl₃ IN THE GROUND STATE**

Figure 1 exhibits the atomic, electronic, and magnetic structures of α-RuCl₃. α-RuCl₃ is a two-dimensional system with an ideal Ru honeycomb lattice, with the Ru–Cl–Ru angle being close to 90°. It hosts comparatively modest spin–orbit coupling in the 4d Ru ions of ~0.1 eV. The ground state of α-RuCl₃ displays an in-plane zigzag antiferromagnetic (AFM) order, in which the magnetic moments of Ru ions are parallel to other moments in the same zigzag chain and antiparallel to those in neighboring zigzag chains (Figure 1a).

We employ the recently proposed ACBN0 functional to exploit the electronic properties of the strongly correlated material. The functional is regarded as a pseudohybrid reformulation of the density-functional theory plus Hubbard U (DFT + U) method, enabling us to compute the Hubbard U and Hund’s J ab initio and self-consistently by solving generalized Kohn–Sham equations (Note 1 in Supporting Information). The method has been recently extended to the real-time case, within the framework of time-dependent density-functional theory. In practice, the functional is an efficient and computationally affordable method to study the optical response of correlated systems driven out of equilibrium.

As to α-RuCl₃, the converged effective Hubbard U terms \((U_{\text{eff}} = U - J)\) with the ACBN0 functional are 1.96 eV for Ru 4d orbitals and 5.31 eV for Cl 3p orbitals, respectively. The parameters yield an indirect bandgap of \(E_{\text{gap}} = 1.05\) eV (Figure 1b), in excellent agreement with the experimental observation (~1.0 eV). In contrast, the indirect bandgap of α-RuCl₃ with on-site Hubbard U on only Ru 4d orbitals reduces to 0.60 eV (Figure 1c), in accordance with previous calculations using empirical Hubbard U terms. Furthermore, without on-site Hubbard corrections, the band structure of α-RuCl₃ displays a metallic state. These results validate the necessity of on-site terms on both Cl 3p and Ru 4d orbitals.

In previous studies, theoretical calculations based on density functional theory plus a Hubbard correction yield a relatively small bandgap (0.6–0.8 eV) in α-RuCl₃, while recent experiments reported a larger bandgap of ~1.0 eV, indicating an evident inconsistency. With respect to magnetic moments, Banerjee et al. reported the ordered moments in Ru ions are around 0.4 \(\mu_B\) for the low-temperature magnetic phase using neutron diffractions. Based on the ACBN0 functional, the magnetic moments of the Ru atoms are 0.33 \(\mu_B\) for the in-plane zigzag AFM order (with Hubbard correction on both orbitals), which is considered to be the ground-state magnetic configuration. These findings reflect that those calculations with both spin–orbit coupling and Hubbard U corrections on the Ru 4d and Cl 3p orbitals are able to capture the microscopic interactions and reproduce the experimental electronic structures.

From the projected band structures of α-RuCl₃ (Figure S1), the orbitals of both the conduction and valence bands are found to exhibit non-negligible contributions from Cl orbitals, validating that on-site Coulomb potentials on both the Ru 4d and Cl 3p orbitals are critical to obtain accurate electronic and magnetic properties. This is interpreted as the on-site Coulomb potential on the chlorine ions increasing the localization of the lone pairs and, hence, the bandgap. Besides, we calculated the amount of charge transfer in the compounds. From Bader analysis (Note 1), the Ru atoms in α-RuCl₃ have the charge of 6.94 e, in contrast to 8 e in the pristine valence orbitals of the pseudopotential. On the other hand, the charge of each Cl atom is 7.35 e (out of 7 e), denoting a significant charge transfer. Therefore, the orbitals of Ru atoms cannot be considered fully localized, and the use of large Hubbard U as a fitting parameter lacks a reasonable physical basis.

Besides the in-plane AFM state, another possible modulated zigzag antiferromagnet order is observed where the magnetic
moments are oriented $\pm 35^\circ$ from the ab plane in experiments.\textsuperscript{21} Our further simulations of $\alpha$-RuCl$_3$ with modulated magnetic orders are presented in Figure S2, where the magnetic orders are fixed as the starting parameters. For the two magnetic states, the energy difference is very small (10 meV/atom energetically lower for the modulated zigzag state). We observe that $\alpha$-RuCl$_3$ with the modulated zigzag state exhibits a slightly smaller bandgap of 1.0 eV while the in-plane ferromagnetic state shows a much smaller indirect bandgap of 0.80 eV. The comparison indicates magnetic states are crucial to determine the electronic structures of the system. It should be mentioned that our calculations do not consider the interlayer magnetic interactions because it is hard to resolve the interlayer structure of $\alpha$-RuCl$_3$ in recent experiments.\textsuperscript{19} The weak van der Waals bonding between the $\alpha$-RuCl$_3$ layers enables several stacking configurations, including a rhombohedral phase with space group R3, as well as a C2/m phase. In this regard, the monolayer $\alpha$-RuCl$_3$ is used as a prototype to investigate the magnetic structure and photoinduced response, as done in other studies.\textsuperscript{28,30}

WAVELENGTH DEPENDENCE OF SPIN DYNAMICS

In the following, we focus on the photoinduced out-of-equilibrium dynamics in $\alpha$-RuCl$_3$ by altering the laser photon energies. The typical shape of the electric field introduced by the applied laser pulse is shown in Figure 2a, with a wavelength of 1180 nm, whose photon energy corresponds to the bandgap (1.05 eV) of $\alpha$-RuCl$_3$. Figure 2b summarizes the real-time evolution of the magnetic moments ($\langle m \rangle$, averaged over the four Ru atoms in the supercell) in $\alpha$-RuCl$_3$ under different photon energies above and below the bandgap. The pump laser intensity ($2.5 \times 10^{12}$ W/cm$^2$) in Figure 2a corresponds to $6.35 \times 10^{-2}$ mJ/cm$^2$, which is on the same order with recent experiments.\textsuperscript{33}

For a photon energy at the bandgap, a closer inspection of the magnetic moments reveals a clear drop in 25 fs, after which they become relatively stable with only a small fluctuation. For lower photon energies, ultrafast melting of the zigzag AFM magnetic order is observed on a time scale of 20 fs. For a photon energy higher than the bandgap ($\hbar \omega = 1.25 \times E_{\text{gap}}$), the averaged magnetic moments reduce to 0.23 $\mu_B$ and oscillate slightly afterward. We find faster demagnetization processes when considering longer wavelengths corresponding to lower photon energies (Figure 2b). Notably, the residual magnetic moment for $\hbar \omega = 0.75 \times E_{\text{gap}}$ is roughly 0.04 $\mu_B$ at the end of laser irradiation and similar to the value for $\hbar \omega = 0.5 \times E_{\text{gap}}$ reflecting saturation of the ultrafast demagnetization (see Figure S3 for snapshots of magnetic moments of $\alpha$-RuCl$_3$ under laser excitation). The wavelength dependence of spin dynamics provides a novel knob for the high modulation of magnetic states under laser excitation and deserves elaborate investigations.

Given the equilibrium results in Figure 1, it is clear that the light-induced reduction of $U$ is crucial for the observed changes in the magnetic structure. In addition, we observe similar demagnetization for a longer laser pulse of 50 fs, as shown in Figure S4. To understand the above findings, we monitored the effective Hubbard $U$ of the Ru and Cl orbitals. It is noteworthy that the laser decreases the effective $U$ for the Ru 4d orbital to 1.50 eV for $\hbar \omega = 0.5 \times E_{\text{gap}}$ (Figure S5). The modification is obviously faster with regard to $\hbar \omega = 1.25 \times E_{\text{gap}}$ in which the residual effective $U$ is 1.39 eV. Since the optical excitation is an ingredient of paramount importance to tune the magnetic properties of correlated materials (e.g., charge-transfer insulators\textsuperscript{38} and Weyl semimetals\textsuperscript{39}), dynamical modification of electron-electron correlations may pave the way to investigate the phase transitions in $\alpha$-RuCl$_3$ from a new degree of freedom.

The variations of the magnetic moments of individual Ru atoms are also provided in Figure 2c,d. For the perpendicular polarization, the photoexcited dynamics demonstrate that the magnitude of spins on different sublattices oscillates with significant out-of-phase components (panel (d) and Figure S3) and the change follows the frequency of the applied laser pulse. It could be attributed to laser-induced symmetry breaking in charge distributions. We should note that laser pulses with different polarizations are capable of breaking the different symmetries of $\alpha$-RuCl$_3$, bringing about different spin sublattices in the dynamics. For the perpendicular polarization, laser excitation breaks the mirror plane vertical to the magnetic moments, and we observe two distinct sublattices for demagnetization of Ru orbitals. This is attributed to the symmetry-breaking and the charge redistribution induced by perpendicular excitations (Figure S6).

INTENSITY AND POLARIZATION DEPENDENCE OF SPIN DYNAMICS

We also investigated the impact of laser intensity on ultrafast demagnetization. From Figure 3a, it is clear that the laser pulse...
with a stronger intensity introduces a more considerable modulation of the atomic magnetic moments. For an intensity of $I_0 = 0.5 \times 10^{12}$ W/cm$^2$, the residual magnetic moment is $0.21 \mu_B$, and the zigzag AFM state is still stable after the laser illumination. Whereas for $I_0 = 2.5 \times 10^{12}$ W/cm$^2$, we obtain a saturation of the demagnetization at $0.06 \mu_B$, indicating a stronger reduction and complete melting of the magnetic structure.

Figure 3b illustrates the laser-induced spin dynamics for the laser pulses with the polarization parallel to the spins. The spin dynamics follow trends similar to those with perpendicular polarization and introduce a distorted state with a residual magnetic moment of $0.04 \mu_B$. Confining ultrafast demagnetization is robust for the parallel polarization. Our further analysis demonstrates that dynamical modification of the electronic and magnetic parameters in strongly correlated magnets is indeed possible by purely optical means without involving the crystal lattice dynamics. It should be noted that the gap is sensitive to both Hubbard terms and the magnetic orders, indicating that $\alpha$-RuCl$_3$ is a Mott–Slater insulator. Laser polarization may also be a cardinal ingredient of importance to control the magnetic structures.

**PHOTOINDUCED INSULATOR-TO-METAL TRANSITION IN $\alpha$-RuCl$_3$**

We carried out comprehensive calculations for the out-of-equilibrium electronic properties and photoinduced carrier excitations in $\alpha$-RuCl$_3$. Figure 4a exhibits the transient band dispersion of photoexcited $\alpha$-RuCl$_3$ for the photon energy of $\hbar \omega = 0.5 \times E_{\text{gap}}$ (see Figure S7 for the full trajectory and corresponding bandgaps). The transient band structures and bandgaps are computed from the time-evolved density under various laser excitations; see Note 1 in Supporting Information. The bandgap drops strikingly to 0.24 eV before the spin subsystem responds significantly (in 10 fs). After that, the bandgap melts completely when the laser pulse reaches the peak at about 15 fs, revealing that the band renormalization can take place without any structural distortions in $\alpha$-RuCl$_3$. We interpret the ultrafast collapse of the bandgap or insulator-to-metal transition in several tens of femtoseconds as indicating that the strong laser pulses greatly change the electron correlations (illustrated by the effective Hubbard terms) and lead to the modification of magnetic structures.

Furthermore, the ultrafast insulator-to-metal transition is robust for various photon energies at the laser with a strong intensity (Figure S8). Regarding all photon energies, the ultrafast band renormalization takes place within 15 fs. This explains the smaller modulation of magnetic moments. Therefore, the bandgap of $\alpha$-RuCl$_3$ can be easily modulated by optical excitation. As a direct consequence, the required excitation energy for Zener tunneling or multiphoton ionization decreases during laser irradiation in $\alpha$-RuCl$_3$.

In order to elucidate the origin of the optical response, the excited carrier densities are analyzed by characterizing the charge excitation from the valence to conduction bands of Kohn–Sham orbitals, which are calculated by projecting the time-evolved wave functions on the ground-state wave functions of $\alpha$-RuCl$_3$ (see SI for more details), as displayed in Figure 4b. For $\hbar \omega = 1.25 \times E_{\text{gap}}$, the photoinduced carrier population increases to 0.40 e/atom within 15 fs and then oscillates around 0.25 e/atom. Following the shapes of the laser pulses, the peak of the carrier density reaches 1.36 e/atom for $\hbar \omega = 0.5 \times E_{\text{gap}}$. The excited carrier concentration is also sensitive to the photon energies of the laser; i.e., longer wavelengths result in more significant carrier densities. This is attributed to the laser-induced collapse of the bandgaps and the laser pulses with smaller photon energies becoming resonant with the transient bandgaps.

To validate the physical picture, we performed additional simulations from the modulated zigzag antiferromagnet order to track the magnetic dynamics and transient band structures (Figure S9). It is obvious that the laser-induced magnetic and electronic dynamics are similar for the two possible magnetic states (in-plane zigzag and modulated zigzag orders). Therefore, we obtain a robust picture of an ultrafast photoinduced insulator-to-metal transition in $\alpha$-RuCl$_3$. The collapse of the bandgap occurs when the electrons are excited by strong laser pulses. The saturation at the half of the ground-state bandgap is interpreted as the excited carrier density being high enough to modulate the electronic structures and transient bandgaps and introduce the rapid demagnetization. In addition, nonlinear excitation processes can also play a role in the wavelength dependence of the demagnetization, especially at the beginning of the strong laser pulses. Our findings support
that the photoinduced carriers are important to introduce ultrafast melting of magnetic structures. Notably, the recovery process after demagnetization is not traced in this work because the real-time TDDFT method incorporates no effective energy dissipation channel.

The direct electronic and spin dynamics obtained from our first-principles calculations enable us to simulate the photoinduced response of \(\alpha\)-RuCl\(_3\), at the atomistic spatial scale on a femtosecond time scale. Strong photoexcitation leads to an ultrafast insulator-to-metal transition and creates a high density of electron–hole pairs. The magnetic interactions are modulated as an effective nonmagnetic state is obtained. It should be noted that the ultrafast process is not the result of collective magnetic excitation (e.g., magnons), which would keep the magnitude of the magnetic moment fixed while decreasing its components along the local order parameter directions. We note that extracting information about the collective excitation should demand more effort from the model Hamiltonian and TDDFT methods.\(^{34,35}\)

For \(\alpha\)-RuCl\(_3\), laser excitations significantly modify the Hubbard terms and generate a substantial number of electron–hole pairs, triggering a rapid insulator-to-metal transition. In the meantime, excess energy in the electron system transfers to the spin subsystem, inducing ultrafast demagnetization. The interplay between the photodoping effect and the insulator-to-metal transition plays a vital role in the magnetic dynamics, while the collapse of magnetic structures causes the reduced bandgap in return. Notably, we cannot simply view the demagnetization as only a consequence because the reduced magnetic moments contribute to the decreased bandgaps.\(^{25}\) Upon significant modification of the Hubbard terms, the local magnetic moments and the Mott gap are found to vanish simultaneously. Our results indicate that both the magnetic ordering and the Hubbard terms are crucial to stabilize the insulating state.

Regarding the laser energies exceeding the ground-state bandgap, photoexcitation creates a dense population of electron–hole pairs. This, in turn, triggers the closure of the bandgap and initiates an ultrafast demagnetization process. In contrast, when laser energies below the ground-state bandgap are employed, a concentration of electron–hole pairs is generated as well, coupled with the presence of in-gap states and orbital excitations. Consequently, the excited carriers play a substantial role in modifying the electronic structures, eventually inducing an intriguing insulator-to-metal transition. As the energy of the pump laser surpasses the transient bandgaps, the trends and time scales governing laser-induced magnetic dynamics exhibit comparable behavior and minimal discrepancies for both above- and below-gap excitations. In experiments, the magnetic transition could be probed by the time-resolved linear magnetic dichroism and X-ray magnetic circular dichroism,\(^{31,32}\) which is able to illustrate the magnetic difference. Photoinduced insulator-to-metal transition in \(\alpha\)-RuCl\(_3\) can be detected by time- and angle-resolved photoemission spectroscopy. It is noteworthy that a femtosecond laser-induced quantum-spin-liquid state is beyond the current study and will be the subject of future studies.

In conclusion, our \textit{ab initio} simulations revealed the nature of photon-driven electron and spin dynamics in \(\alpha\)-RuCl\(_3\). We demonstrated that laser pulses can provoke a magnetic transformation between zigzag AFM magnetic order and disordered magnetic states with much smaller magnetic moments. In addition, the spin response is remarkably sensitive to the laser wavelength and polarization, on account of the photoinduced insulator-to-metal transition and different excited carrier distributions. This subtle interplay suggests a way to modulate the electronic and magnetic structures by using ultrashort laser pulses. Our work provides new insights into photoexcitation-induced magnetic phase transitions and may pave the way for suppressing the long-range magnetic order and realizing a quantum-spin-liquid state at ultrashort time scales.

### ASSOCIATED CONTENT

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.3c02668.

Detailed methods, parameters, and additional figures (PDF)

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**Author Contributions**

AR conceived the research direction and JZ proposed the project. Most of the calculations were performed by JZ, with contributions from all authors. NTD implemented the ACBNO
functional into the OCTOPUS code. All authors contributed to the analysis and discussion of the data and the writing of the manuscript.

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**Notes**

The authors declare no competing financial interest.

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