Photoinduced itinerant ferromagnetism in copper octacyanomolybdates

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Abstract. We make a microscopic theory of the photoswitchable magnetism in copper octacyanomolybdates. By numerically solving a time-dependent Schrödinger equation based on the relevant extended Hubbard model, we reproduce magnetization by green-light irradiation and subsequent demagnetization by orange-light irradiation. At the onset of the ferromagnetism, the charge-transfer gap disappears. In an attempt to stimulate experimental investigations, we simulate time evolution of the angle-resolved photoemission spectroscopy and optical-conductivity spectra.

1. Introduction
Photomagnetism of cyano-bridged transition metal complexes has been the central issue in both physical and chemical fields. In Fe-Co Prussian blue analogues consisting of hexacyanoferrates and cobalt ions, photoirradiation induces ferrimagnetism [1, 2, 3]. The diamagnetic entity Fe$^{II}$(S = 0) -CN-Co$^{III}$(S = 0) is photoconverted to the magnetic one Fe$^{III}$(S = 1/2) -CN-Co$^{II}$(S = 3/2). The electron transfer mediated by a cyano group triggers the magnetic interaction. The reverse process is also induced by the photoirradiation with longer wavelength [4]. Such photo-manipulation of magnetism by selective charge-transfer excitations is very interesting from the viewpoints of fundamental physics and its application.

Several experimental groups recently discovered photomagnetism in copper-molybdenum bimetallic assemblies Cu$_2$[Mo(CN)$_8$]$n$H$_2$O [5, 6, 7]. These compounds consist of octacyanomolybdates [Mo$^{IV}$(CN)$_8$]$^{4-}$ of the square antiprism configuration [8, 9] and Cu ions. As is shown in Fig. 1(a), the crystal structure is characterized by the $I4/m$ tetragonal space group with $C_{4h}$ point group. The coordination network based on octacyanomolybdates brings about various electron-transfer and magnetic-exchange pathways, in comparison with those in the Prussian blue analogues with simple cubic structures [10]. Fully-filled 4$d_{yz}$ orbitals on Mo$^{IV}$ sites and half-filled 3$d_{yz/xz}$ orbitals on Cu$^{II}$ sites are physically active due to a crystalline-field splitting. Irradiation of the nonmagnetic ground state with a laser light of 473 nm induces a macroscopic magnetization, while subsequent irradiation with a laser light of 658 nm reduces this ferromagnetism. Further photoirradiation with longer wavelength reduces the induced magnetization step by step. This phototunable magnetism is stable from 3 K to 100 K. In the magnetization process, photoirradiation induces electron transfers from the fully-filled Mo orbitals to the half-filled Cu orbitals. As a result, both Mo and Cu ions have magnetic moments and then the ferromagnetic exchange pathway appears. These electron configurations are supposed by the measurements of the magnetization curves before and after photoirradiation [5].
However it remains to be fully elucidated whether the demagnetization can be described as a simple reverse process of the magnetization or not.

In spite of the intriguing observations, neither photoinduced magnetism nor ground-state magnetism has been interpreted yet beyond the phenomenological understanding. In order to reveal a microscopic picture of the phototunable magnetism of Cu$_2$[Mo(CN)$_8$], we draw a ground-state phase diagram and simulate successive photoirradiation of the nonmagnetic ground state.

2. Modelling

In order to describe the dicopper octacyanomolybdates, we employ the 2/3-filling extended Hubbard model represented by

$$
\mathcal{H} = \sum_{\langle n,m,i,j \rangle \sigma, \tau} \left\{ t_{MoCu} \left[ (-1)^{i+j+1} c_{n:Cu(i)\sigma}^\dagger c_{m:Mo(j)\sigma} + \text{H.c.} \right] + V_{MoCu} n_{n:Cu(i)\sigma} n_{m:Mo(j)\tau} \\
+ J_{MoCu} c_{n:Cu(i)\sigma}^\dagger c_{m:Mo(j)\sigma} c_{n:Cu(i)\tau} c_{m:Mo(j)\tau} + \text{H.c.} \right\} \\
+ \sum_{\langle n \neq m \rangle \iota=1} \sum_{\sigma, \tau} \left[ \frac{t_{CuCu}}{2} \left( c_{n:Cu(i)\sigma}^\dagger c_{m:Cu(i+4)\sigma} + \text{H.c.} \right) + V_{CuCu} n_{n:Cu(i)\sigma} n_{m:Cu(i+4)\tau} \\
+ J_{CuCu} c_{n:Cu(i)\sigma}^\dagger c_{m:Cu(i+4)\tau} c_{n:Cu(i)\tau} c_{m:Cu(i+4)\tau} + \text{H.c.} \right] \\
+ \sum_{n, \sigma} \left[ \sum_{i=1}^8 (\varepsilon_{Cu} + \frac{U_{Cu}}{2} n_{n:Cu(i)\sigma} - \sigma) n_{n:Cu(i)\sigma} + \sum_{j=1}^4 \left( \varepsilon_{Mo} + \frac{U_{Mo}}{2} n_{n:Mo(j)\sigma} - \sigma \right) n_{n:Mo(j)\sigma} \right]
$$

with $n_{n:A(i)\sigma} = \sum_{\sigma} n_{n:A(i)\sigma}$, where $c_{n:A(i)\sigma}^\dagger$ creates an electron of spin $\sigma = \uparrow, \downarrow \pm \pm \pm \pm$ on the Cu 3$d$ ($A = Cu; i = 1$-8) or Mo 4$d$ ($A = Mo; i = 1$-4) orbitals labelled $i$ at unit cell $n$, as is shown in Fig. 1(a). $\sum_{\langle n,m,i,j \rangle \sigma, \tau}$ and $\sum_{\langle n \neq m \rangle \iota=1} \sum_{\sigma, \tau}$ run all over the nearest neighboring Cu-Mo bonds and Cu-Cu bonds, respectively. $t_{MoCu/CuCu}$, $V_{MoCu/CuCu}$, $J_{MoCu/CuCu}$, and $J_{MoCu/CuCu}$ describe transfer integrals, intermetallic Coulomb repulsions, exchange interactions and pair-hopping interactions, respectively. On-site Coulomb repulsions $U_{Mo/Cu}$ and orbital energies of isolated metals $\varepsilon_{Mo/Cu}$ are considered. All the two-body terms are treated within a Hartree-Fock (HF) scheme.

3. Ground-state properties

Based on the symmetry argument on the crystal structure [11, 12], we obtain possible electronic states as is shown in Fig. 1(b). Both nonmagnetic and magnetic states appear as a ground state. The charge-disproportionate (CDP) and ferromagnetic (FM) states preserve the original translational symmetry property, while the charge-density-wave (CDW) and antiferromagnetic (AFM) states show cell-doubling structures. There are two kinds of charge-disproportionate patterns. Mo-CDP can take the Mo$^{IV}$-Cu$^{II}$ valence arrangement suggested by the observations [5, 7], while Cu-CDP exhibits electron localization in Cu sites. Only FM has a global magnetization.

Figure 1(c) shows a ground-state phase diagram at the sufficiently low temperature $k_B T = 0.001 t_{MoCu}$, where we set $t_{CuCu} = 0.3 t_{MoCu}$, $V_{CuCu} = 2.4 t_{MoCu}$, $V_{MoCu} = 0.8 t_{MoCu}$, $\varepsilon_{Cu} - \varepsilon_{Mo} = 1.0 t_{MoCu}$, and $J_{MoCu} = J'_{MoCu} = -J_{CuCu} = -J'_{CuCu} = 0.6 t_{MoCu}$ unless otherwise noted.
Figure 1. (Color online) (a) A crystal structure of Cu$_2$[Mo(CN)$_8$]. A unit cell contains four Mo ions and eight Cu ions. (b) Possible electronic states, where varying circles indicate electron densities and white arrows describe electron spins. (c) A ground-state phase diagram as a function of on-site Coulomb repulsions. Black and red cross symbols ($\times$) point to ($U_{\text{Cu}}/t_{\text{MoCu}}$, $U_{\text{Mo}}/t_{\text{MoCu}}$)=$($3.2$, $3.2$) and ($3.2$, $5.0$), respectively. (d) Optical conductivity spectra of the Mo-CDP and FM states, where a vertical arrow indicates a pumping photon energy.

Phase boundaries drawn in black and red denote transitions of the first order and the second order, respectively. With increasing $U_{\text{Cu}}$ and $U_{\text{Mo}}$, FM and Mo-Cu-AFM are stabilized. These magnetic phases are strongly affected by the direct exchange interactions $J_{\text{MoCu}}$ and $J_{\text{CuCu}}$ as well. In the present calculation, we chose positive $J_{\text{MoCu}}$ stabilizing ferromagnetism because the analogue compound Cu$_{1.5}$[Mo(CN)$_8$]·$3\text{H}_2\text{O}$ shows a ferromagnetic correlation between spins on Mo and Cu sites [7]. On the other hand, $J_{\text{CuCu}}$ is taken to be negative. Here we note the identity $\sum_{\sigma} \sum_{i} c_{n,A(i)\sigma}^\dagger c_{n,A'(i')\sigma} c_{n,A(i)\tau} c_{m,A'(i')\sigma} = -2s_{n:A(i)} \cdot s_{m:A'(i')} - n_{n:A(i)} n_{m:A'(i')}/2$, where $s_{n:A(i)}^\lambda = \sum_{\sigma} \sigma c_{n,A(i)\sigma} c_{n,A'(i)\sigma} \tau_{\sigma\sigma'}^\lambda /2$ ($\lambda = x, y, z$) and $\tau^\lambda$ indicates Pauli spin matrices. In a weak correlation regime, Mo-Cu-CDW is most stable due to the relevant intermetallic Coulomb interactions. Mo-CDP predominates in a moderate correlation regime. Assuming that both $U_{\text{Cu}}$ and $U_{\text{Mo}}$ values of Cu$_2$[Mo(CN)$_8$] are in this regime, we chose $U_{\text{Cu}} = U_{\text{Mo}} = 3.2t_{\text{MoCu}}$ as standard parametrization, where the valence arrangement of Mo and Cu ions agrees with the observations [11, 12]. Cu-CDP will never appear as a ground state of Cu$_2$[Mo(CN)$_8$] because it is stabilized only in an unrealistic parameter region as $U_{\text{Mo}} >> U_{\text{Cu}}$. Mo-CDP and FM are sitting adjacently on the phase diagram, and therefore, fluctuation by external fields is likely to induce the phase transition between them.
We further calculate optical conductivities of Mo-CDP and FM. In the FM state, we set \( (U_{\text{Cu}}/t_{\text{MoCu}}, U_{\text{Mo}}/t_{\text{MoCu}}) = (3.2, 5.0) \). Hereafter the intermetallic transfer integral \( t_{\text{MoCu}} \) is set equal to 0.7 eV, which is comparable to that in typical Prussian blue photomagnets [13, 14]. The optical conductivity spectrum is defined as

\[
\sigma(\omega) = -\pi N \left( \langle E_0|T|E_0 \rangle + 2 \sum_{l \neq 0} |\langle E_l|J|E_0 \rangle|^2 \right) \delta(\omega) + \frac{\pi N \omega}{\hbar} \sum_{l \neq 0} |\langle E_l|J|E_0 \rangle|^2 \delta(E_l - E_0 - \hbar \omega),
\]

where \( |E_l \rangle \) is a one-electron-hole-pair excited state from \( |E_0 \rangle \) being either Mo-CDP or FM. \( J \) and \( T \) indicate a current operator and a kinetic energy operator, respectively \([15, 16]\). Considering the electric current in the \( a-b \) plane \([12]\), we obtain the optical conductivity perpendicular to the \( c \) axis as is shown in Fig. 1(d). Mo-CDP shows charge-transfer gap \( \Delta = 0.35 \) eV. A broad absorption band is attributed to the electron excitations from the highest-occupied molecular orbital (HOMO) to the lowest-unoccupied molecular orbital (LUMO) over the whole Brillouin zone. On the other hand, FM shows a significant zero-frequency absorption namely Drude component. The FM state exhibits a metallic conductivity. Thus we find that the insulating Mo-CDP closely competes with the itinerant FM at low temperatures. An electric conductivity serves as a probe of the magnetic transition in Cu[Mo(CN)8].

4. Photomagnetization
Since photomagnetization is hardly realized at a zero magnetic filed, we apply a moderate filed to the system as follow,

\[
\mathcal{H}_{\text{ex}} = -g \mu_B H \sum_{n} \left[ \sum_{i=1}^{8} s_{n,Cu(i)}^z + \frac{4}{3} \sum_{j=1}^{4} s_{n,Mo(j)}^z \right],
\]

where \( H = 2.5 \) T. Then down-spin electrons are selectively photoexcited. Such a photoexcitation is described by multiplying the hopping term \( c_{n,A(i)}^\dagger c_{n',A'(i')} \) by the Peierls phase factor \( \exp[-(ie/\hbar)A(t) \cdot (r_{n,A(i)} - r_{n',A'(i')})] \) \([17]\), where \( e, v \), and \( r_{n,A(i)} \) are the elementary charge, the light velocity, and the position vector, respectively. We consider successive photoirradiation described by the vector potential

\[
A(t) = A_0 \sum_{i=0}^{3} e^{-\gamma^2(t-t_i)^2} \cos \omega_i t,
\]

where centers and photon energies of first~fourth \((i = 0 \sim 3)\) pulses are given by \( t_i = 0.38 + 0.75i \) ps and \( \hbar \omega_i = (1 - 0.25i) \hbar \omega_0 \), respectively. With the experimental observations in mind \([7]\), we set \( \hbar \omega_0 \) equal to 2.6 eV, which is indicated by a vertical arrow in Fig. 1(d). A width and an amplitude of each pulse are \( 2\gamma^{-1} = 0.25 \) ps and \( |A_0| = A_0 = 2.0(h\nu/ea) \), respectively. Furthermore, we consider Dzyaloshinskii-Moriya (DM) interaction breaking the conservation law for the total magnetization:

\[
\mathcal{H}_{\text{DM}} = \sum_{n} \sum_{l=1}^{4} \sum_{p=0}^{1} \sum_{\sigma,\sigma'} (-1)^{l+p+\sigma} D_{l+4\rho}^{(\sigma'\sigma)} \mathcal{S}_{n+\delta(l,\rho,\sigma,\sigma')}:\text{Mo}(1+3\sigma'+(-1)^{\sigma'} \rho) \times s_{n,Cu}(2l-\sigma),
\]

where \( \delta(l, \rho, \sigma, \sigma') = (\sigma'^\text{Re}[f(l)], \sigma'^\text{Im}[f(l)], \sigma') \) with \( f(l) = e^{i\pi/4}[1 + e^{i(1-l)/2}]/\sqrt{2} \). The DM vectors are given as \( D_l^{(\sigma'\sigma)} = g_i \cdot D_l^{(\sigma'\sigma)} \), where \( g_i(e \in C_{4h}) = C_{4z}^{-1}, C_{2z}, C_{4z}, \sigma_h, IC_{4z}, I, IC_{4z}^{-1} \) for \( i = 2 \) to 8, respectively, and \( D_l^{(\sigma'\sigma)} = (D, 0, 0) \) with \( D = 0.7 \) meV. Time evolution of the
electronic wave functions is obtained by integrating the time-dependent Schrödinger equations step by step as
\[
|\psi_{k,\nu}(t + \Delta t)\rangle = \exp\left[-\frac{i}{\hbar} \mathcal{H}_{\text{HF}}(t) \Delta t\right] |\psi_{k,\nu}(t)\rangle,
\]
where the instantaneous Hamiltonian \( \mathcal{H}_{\text{HF}}(t) \) consists of \( \mathcal{H}, \mathcal{H}_{\text{ex}}, \) and \( \mathcal{H}_{\text{DM}} \) within the HF approximation [18]. The time \( t \) is discretized by \( \Delta t = 0.005\hbar/\hbar_{\text{Mo-Cu}} \). The initial state \( |\psi_{k,\nu}(0)\rangle \) is the HF eigenstate of the static Hamiltonian \( \mathcal{H} \), which is specified by a momentum and a band index \((k,\nu)\). The many-body wave function is given by \( |\Psi(t)\rangle = \Pi_{\nu=1}^{16} \otimes |\psi_{k,\nu}(t)\rangle \). We irradiate Mo-CDP with \( a \)-axis-polarized light.

![Figure 2](image_url)

**Figure 2.** (Color online) (a) Time evolution of a global magnetization \( M \) and an absorbed energy \( \Delta E \). Pumping pulses are also presented. (b) Time evolution of electron densities on Mo and Cu sites. (c) Time-resolved one-particle excitation spectra at \( t = 0.0, 0.5, 1.6 \) ps.

We present time evolution of a global magnetization \( M \) and an absorbed photon energy \( \Delta E \) in Fig. 2(a), where \( M = \sqrt{\sum_{\lambda} (M^\lambda)^2} \) with \( M^\lambda = \sum_{n} \langle \Psi(t) | \sum_{i=1}^{8} s_{n,\text{Cu}(i)}^\lambda + \sum_{i=1}^{4} s_{n,\text{Mo}(i)}^\lambda | \Psi(t) \rangle / N \) and \( \Delta E = \langle \Psi(t) | \mathcal{H}_{\text{HF}}(t) | \Psi(t) \rangle - \langle \Psi(0) | \mathcal{H}_{\text{HF}}(0) | \Psi(0) \rangle \). The electronic energy increases step by step by the successive photoirradiation. Upon the first photoirradiation with \( \hbar \omega_0 \), \( \Delta E \) reads as a double-stepped function of time. At around \( t = 0.38 \) ps corresponding to the pulse center, the photon absorption is suddenly accelerated. The occurrence of a significant magnetization looks simultaneous with the photon absorption at the second step of \( \Delta E \). An incubation time is needed for the rise of ferromagnetism. The second, third, and fourth photoirradiation with longer wavelength reduce the thus-induced magnetization. This step-by-step demagnetization also consists with the observations [7].

Figure 2(b) shows an electron transfer between Mo and Cu sites induced by the photoirradiation, where \( d_{n,\text{Cu}} = \sum_{i=1}^{8} \langle \Psi(t) | n_{n,\text{Cu}(i)} | \Psi(t) \rangle / 8N \) and \( d_{n,\text{Mo}} = \sum_{i=1}^{4} \langle \Psi(t) | n_{n,\text{Mo}(i)} | \Psi(t) \rangle / 4N \). Upon the appearance of the global magnetization, electrons transfer from Mo to Cu sites. These electrons never come back to Mo sites in the subsequent demagnetization processes. This means that the demagnetization can not be interpreted as a simple reverse process of the magnetization. The electron densities on Mo and Cu sites seem to
converge on the average value $16/12=1.33$. The final steady state showing no global magnetization differs from the initial nonmagnetic state.

By calculating one-particle excitation spectra corresponding to angle-resolved photoemission spectroscopy (ARPES) spectra [19], we further investigate a microscopic electronic structure. It is given by $D(t;k,\omega) = H(t;k,\omega) + E(t;k,\omega)$ with $E(t;k,\omega) = \sum_{k',\nu} \delta(\hbar\omega - \varepsilon_{k',\nu})\langle \Psi(t)|\hat{c}_{k',\nu}^\dagger \hat{c}_{k,\nu}|\Psi(t)\rangle$ and $H(t;k,\omega) = \sum_{k,\nu} \delta(\hbar\omega - \varepsilon_{k,\nu})\langle \Psi(t)|\hat{c}_{k,\nu}^\dagger \hat{c}_{k,\nu}|\Psi(t)\rangle$, where $\hat{c}_{k,\nu}^\dagger$ creates an electron in the eigenstate of energy $\varepsilon_{k,\nu}$ for the instantaneous Hamiltonian $H_{HF}(t)$. Figure 2(c) presents $D(t;k,\omega)$ in the photoinduced magnetization and demagnetization processes. The upper panel of Fig. 2(c) shows the band structure of the initial Mo-CDP, where the spin-up and -down electron bands are degenerate. After the first photoirradiation, this degeneracy is so lifted as to induce a net magnetization, as is shown in the middle panel of Fig. 2(c). This is the cooperative effect of the photoexcitation and the spin-mixing due to the DM interaction. After the second photoirradiation, the up- and down-spin bands converge again with a decrease in the magnetization, as is shown in the lower panel of Fig. 2(c). Upon the third and fourth photoirradiation, the band splitting further decreases with decreasing magnetization. In the demagnetization process, we observe photon emission as well as photon absorption. A resonance energy of this stimulated emission of photons also decreases with decreasing magnetization. Therefore, modulation of pumping photon energy is a key ingredient for the step-by-step demagnetization.

We further present optical conductivity spectra at several times in Fig 3. Drude component is clearly observed at $t = 0.3$ ps (before the appearance of the global magnetization) [see Fig. 3(a)]. With an increase in photocarriers, the initial absorption band weakens and the metallic conductivity predominates. Given that the major absorption in Mo-CDP is attributed to the HOMO-to-LUMO electron excitations, this optical feature is reasonable. The magnetic phase transition reads as an insulator-to-metal phase transition, where the metallization slightly precedes the magnetization. In the demagnetization processes, the induced metallic conductivity is almost unchanged [see Fig. 3(b)], while the induced magnetization is pronouncedly reduced [see Fig. 2(a)]. Such a nonmagnetic metallic state never appears in the ground-state phase diagram. The final steady state may thus be quite different from any equilibrium state.

5. Summary

We have demonstrated the tunable photomagnetism of Cu$_2$[Mo(CN)$_8$]. In the formation process of the itinerant ferromagnetism, it is within a few subpicoseconds after metallization that a global magnetization appears. This delay time may be determined by the magnitude of the DM vector giving a spin-mixing effect, where we should be reminded that $|D| \sim$meV. The
demagnetization originates from stimulated emission of photons, where electrons on Mo sites further transfer to Cu sites. The photoswitching of magnetization does not necessarily mean a round trip of electrons. The initial valence arrangement is thus photoconverted to the averaged one. The final steady state is nonmagnetic but metallic, which has never been observed in thermal equilibrium.

The magnetization dynamics is almost unchanged up to \( k_B T = 0.2 t_{\text{MoCu}} \). As is shown in Fig. 4, above this temperature the induced magnetization is monotonically suppressed with increasing temperature. On the other hand, the step-by-step demagnetization process much less depends on temperature. The suppression of the induced magnetization is closely related to the charge-transfer gap of the ground-state Mo-CDP, \( \Delta = 0.5 t_{\text{MoCu}} (=0.35 \text{ eV}) \). Thermal demagnetization effect may become remarkable above the temperature corresponding to the gap \( \Delta \). The induced magnetizations at \( k_B T = 0.3 t_{\text{MoCu}} \) and \( k_B T = 0.5 t_{\text{MoCu}} \) are indeed much different. In the demagnetization process, there are many holes in the valence band and then there is no longer any threshold temperature.

Our findings suggest that the copper octacyanomolybdates must be a functional material in that both magnetism and itinerancy of electrons are phototunable. The spin-charge dynamics will be elucidated by ultrafast pump-probe experiments, such as time-resolved ARPES [20] and conductivity [21] measurements. We hope that the present work will lead to a new insight into photoinduced dynamics in octacyanometalate-based magnets.

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