Photo-induced insulator-metal transition of a spin-electron coupled system

W. KOShibae, N. FURUKAWA, N. NAGAOsa

1 Cross-Correlated Materials Research Group (CMRG), RIKEN-ASI - Wako 351-0198, Japan
2 Aoyama-Gakuin University - 5-10-1, Fuchinobe, Sagamihara, Kanagawa 229-8558, Japan
3 ERATO-Multiferroics, JST, c/o Department of Applied Physics, The University of Tokyo
   Tokyo 113-8566, Japan
4 Department of Applied Physics, The University of Tokyo - Tokyo 113-8566, Japan
5 Correlated Electron Research Group (CERG), RIKEN-ASI - Wako 351-0198, Japan

Received 4 February 2011; accepted 15 March 2011

PACS 71.10.-w - Theories and models of many-electron systems
PACS 71.10.Fd - Lattice fermion models (Hubbard model, etc.)
PACS 78.20.Bh - Theory, models, and numerical simulation

Abstract - The photo-induced metal-insulator transition is studied by the numerical simulation of real-time quantum dynamics of a double-exchange model. The spatial and temporal evolutions of the system during the transition have been revealed including i) the threshold behavior with respect to the intensity and energy of light, ii) multiplication of particle-hole (p-h) pairs by a p-h pair of high energy, and iii) the space-time pattern formation such as (a) the stripe controlled by the polarization of light, (b) coexistence of metallic and insulating domains, and (c) dynamical spontaneous symmetry breaking associated with the spin spiral formation imposed by the conservation of total spin for small energy-dissipation rates.

In correlated electrons such as the transition metal oxides, the collective nature of the system enhances the sensitivity to the external stimuli including photons, and hence even the weak photo-irradiation can trigger the changes of whole system as the phase transition [1–6]. This is because in strongly correlated electronic systems, the rich phases of spin/charge/orbital orderings with nearly degenerate energies compete with each other [7]. For example, in the case of manganites, the metal-insulator transition can be triggered by a very fast optical pulse (~10 fs) observing the coherent orbital wave oscillations [2], which motives theoretical studies on the fast electronic dynamics which we undertake in this paper.

In contrast to these ample examples of photo-induced phase transitions, the theoretical studies are still in the premature stage. Although a classical model for the photo-induced phase transition has been proposed [8], the quantum theory of photo-induced dynamics of correlated electrons are limited to small-size systems or to one-dimensional systems [9–11]. Recently, we have proposed a model for the real-time quantum dynamics of correlated electrons, i.e., a double-exchange model [12] where the electrons are interacting with the classical spins, to simulate the relaxation after the photo-excitation [13]. In that work, we have developed a theoretical method for the fully quantum-mechanical time evolution of electronic wave function combined with the Landau-Lifshitz-Gilbert (LLG) equation for classical spins, which enables the analysis of the larger-scale/higher-dimensional systems. By that theoretical study, the self-organized space-time structure induced by the quantum transitions during the relaxation has been revealed [13].

In this paper, we present the real-time quantum dynamics of the photo-induced insulator-to-metal (IM) transition for a generalized double-exchange model showing the competing antiferromagnetic insulating (AFI) and ferromagnetic metallic (FM) states separated by the first-order phase boundary. Many unexplored issues can be addressed in this model, i.e., the threshold behavior with respect to the intensity/energy of light, the effect of the polarization of light, space-time pattern formation during the transition, the role of the conservation of total spins, and the asymmetry between the two directions of the transition.
We start with the Hamiltonian,
\[ \hat{H} = -\frac{t}{2} \sum_{\mathbf{i}+\mathbf{p},s} \hat{c}^\dagger_{\mathbf{i}s} \hat{c}_{\mathbf{i}+\mathbf{p}s} + \text{h.c.} - J_H \sum_{\mathbf{i},s,s'} \hat{c}^\dagger_{\mathbf{i}s} \hat{c}_{\mathbf{i}s'} \sigma_{ss'} \cdot \hat{S}_{\mathbf{i}} + \frac{J}{2} \sum_{\mathbf{i},i+\rho} \hat{S}_i^z \cdot \hat{S}_{i+\rho} + \langle J_N/2 \rangle \sum_{\mathbf{i},i+\rho} (\hat{S}_i^z \cdot \hat{S}_{i+\rho})^2 \]
\[ = \sum_{\mathbf{i}} \hat{h}_i = \hat{H}_{\text{el}} + H_S, \tag{1} \]
where \( i + \rho \) runs over the nearest-neighbor sites of \( i, s \) and \( s' \) are indices for electron spin, respectively, and \( \sigma = (\sigma^x, \sigma^y, \sigma^z) \) are Pauli matrices. The Hamiltonian \( \hat{H}_{\text{el}} \) involves the creation and annihilation operators for electron but \( H_S \) does not. The local spins, \( \hat{S}_i^z \)'s, are taken to be classical vectors with magnitude \( S \), and other notations are standard. We consider the half-filled case, i.e., the number of electrons \( N_e \) is equal to the number of sites \( N \). In eq. (1), we have introduced the bilocal exchange interaction with coupling constant \( J_N \), which gives an energy barrier between ferromagnetic (F) and antiferromagnetic (AF) states when \( J_N \) is negative.

We numerically investigate the time (\( \tilde{T} \)) evolution of the electronic states and local spins \cite{13}. We introduce the LLG equation for local spins:
\[ \dot{\hat{S}}_{\mathbf{i}}(\tilde{T}) = -\hat{h}_{\text{eff}}(\tilde{T}) \times \hat{S}_{\mathbf{i}}(\tilde{T}) - \alpha \hat{S}_{\mathbf{i}}(\tilde{T}) \times \dot{\hat{S}}_{\mathbf{i}}(\tilde{T}), \]
where \( \alpha \) is the Gilbert damping constant and
\[ \hat{h}_{\text{eff}}(\tilde{T}) = \langle \Phi(\tilde{T}) | -\partial \hat{H}(\tilde{T})/\partial \hat{S}_{\mathbf{i}}(\tilde{T}) | \Phi(\tilde{T}) \rangle \]
with \( |\Phi(\tilde{T})\rangle \) being the electronic wave function at time \( \tilde{T} \). Note that \( \hat{H}(\tilde{T}) \) is also time-dependent through \( \{\hat{S}_{\mathbf{i}}(\tilde{T})\}_{\mathbf{i}=1-N} \). The time evolution of a single Slater determinant state,
\[ |\Phi(\tilde{T})\rangle = \prod_{m=1}^{N_e} \Phi_m^\dagger(\tilde{T}) |0\rangle, \]
during the small time increment \( \Delta\tilde{T} \) is given by,
\[ |\Phi(\tilde{T} + \Delta\tilde{T})\rangle = \prod_{m=1}^{N_e} \Phi_m^\dagger(\tilde{T} + \Delta\tilde{T}) |0\rangle \]
with
\[ \Phi_m^\dagger(\tilde{T} + \Delta\tilde{T}) = e^{-i\hat{H}_{\text{el}}(\tilde{T})\Delta\tilde{T}/\hbar} \Phi_m^\dagger(\tilde{T}) e^{i\hat{H}_{\text{el}}(\tilde{T})\Delta\tilde{T}/\hbar}. \]
Thus, the state remains a single Slater determinant state upon time evolution if we take a single Slater determinant state as an initial state. The state \( \Phi_m^\dagger(\tilde{T}) |0\rangle \) is not always an eigenstate of \( \hat{H}_{\text{el}}(\tilde{T}) \). More explicitly, we can diagonalize the instantaneous Hamiltonian in a form \( \hat{H}_{\text{el}}(\tilde{T}) = \sum_{\mathbf{i},m} E_{\mathbf{i}m} \phi_m^{\dagger} \phi_{\mathbf{i}}^{\dagger} \phi_{\mathbf{i}} \phi_m, \) i.e., the fermion operator \( \phi_m^{\dagger} \) is for an eigenstate of \( \ell \)-th level \( E_{\mathbf{i}m} \) at time \( \tilde{T} \). Then, the wave function can be expanded as \( \Phi(\tilde{T}) |0\rangle = \sum_{m=1}^{2N_e} c_{\ell m} \phi_m^{\dagger} |0\rangle \). The density of states (DOS) and the occupancy of \( \ell \)-th level at time \( \tilde{T} \) are defined by \( \sum_{m=1}^{2N_e} \delta(\varepsilon - E_{\mathbf{i}m}) \) and \( \langle\Phi(\tilde{T}) | \phi_m^{\dagger} \phi_{\mathbf{i}}^{\dagger} | \Phi(\tilde{T}) \rangle = \sum_{m=1}^{N_e} |c_{\ell m}|^2 \), respectively. This enables us to study the electronic relaxation process beyond the adiabatic approximation \cite{14} where the electronic wave function is always the ground state, \( \prod_{m=1}^{N_e} \phi_m^{\dagger} |0\rangle \), of the instantaneous Hamiltonian \( \hat{H}_{\text{el}}(\tilde{T}) \).

As shown in ref. \cite{13}, the spin motion is composed of two parts, i.e., \( \hat{S} = \hat{S}^{\text{slow}} + \hat{S}^{\text{rapid}} \), where \( \hat{S}^{\text{rapid}} \) is the rapidly oscillating small-amplitude part around the \( \hat{S}^{\text{slow}} \). Correspondingly, \( \hat{H}_{\text{el}} \) can be decomposed into \( \hat{H}_{\text{el}} = \hat{H}_0 + \hat{H}' \), where \( \hat{H}' \) is Hund’s coupling to \( \hat{S}^{\text{rapid}} \). The characteristic time scale of \( \hat{H}_0 \) is slower than the energy level separation \( \hat{E}_\ell - \hat{E}_{\ell'} \), and hence it makes sense to define the eigenstates and eigenenergy for \( \hat{H}_0 \) at each instant of time \( \tilde{T} \). Also \( \hat{H}' \) can be regarded as the perturbation since \( |\hat{S}^{\text{rapid}}| \ll |\hat{S}^{\text{slow}}| \), which triggers the transition between the states \( \ell \) and \( \ell' \) of \( \hat{H}_0 \) since the transition rate is smaller than the energy separation. Note also that \( \phi_{\mathbf{i}}^{\dagger} \) and \( \hat{E}_{\mathbf{i}m} \) shown below are those obtained for \( \hat{S} \) but the correction by \( \hat{S}^{\text{rapid}} \) is negligible.

Once the perfect ferromagnetic order is established, the energy level separation \( \hat{E}_\ell - \hat{E}_{\ell'} \) is infinitesimally small. However, we are interested in the initial quantum electronic processes where the spin correlation has not been developed to such a long range, and the electronic wave functions are localized due to the disordered spin configurations. The classical dynamics of phase transition at a later stage should be discussed separately. Therefore, our finite size of the system gives effectively the cut-off for the time range where our scheme is valid.

Figure 1(a) shows the phase diagram of the model eq. (1) in the plane of \( S^2 J \) and \( S J_H \) with fixed \( S^4 J_N = -0.2 \) in units of \( t \). There exist three phases, i.e., ferromagnetic metal (FM), antiferromagnetic insulator (AFI) and canted antiferromagnetic insulator (CAFI). (b) The lowest energy per site measured from the ground state as a function of the angle between neighboring local spins in a two-sublattice consideration on an \( 8 \times 8 \) system. A parameter set, \( t = 1, S J_H = 1, S^2 J = -0.043, S^4 J_N = -0.017, S = 1 \) is used.

---

\footnote{For \( S J_H > 4t \), the system is insulating since the energy bands for up and down spin split into two bands separated by a gap \( 2(S J_H - 4t) \) in the FM state.}
Let us focus on the boundary of first-order phase transition. Figure 1(b) shows the energy as a function of the angle $\theta$ between the sublattice spin directions for a $8 \times 8$ size system in periodic boundary condition with a parameter set, $t = 1$, $S J_H = 1$, $S^2 J = -0.043$, $S^4 J_N = -0.017$, $S = 1$. The AF ($\theta = \pi$) and F ($\theta = 0$) states are almost degenerate, and show the local stabilities with the potential barrier between them. This condition is used hereafter and the qualitative behavior of the theoretical result is not sensitive for the parameters on the phase boundary.

The magnitude of the empirical parameter, Gilbert damping constant, is typically in the range $1.0 \geq S_0 \geq 0.01$ [15]. Let us first examine the photo-excitation and relaxation dynamics for $S_0 = 1.0$ (see fig. 2). The dimensionless time $T = (t/h)T$ is defined, i.e., the time is measured in the unit of $h/t$ which is typically $\sim 10^{-15}$ s assuming $t = 0.4$ eV. We show DOS and electronic occupancy at $T$ as defined above in figs. 2(a)–(e). We start with the AFI state, i.e., the electronic state has an insulating energy gap $2SJ_H$ and the lower energy band is occupied by electrons and the upper energy band is empty at $T = 0$. In order to mimic the thermal fluctuation, we introduce a random tilting of each spin from the perfect AF configuration up to 0.1 rad which corresponds to the state with an excitation energy of $\sim 0.0003t$ from the ground state.

The photo-excitation is achieved by introducing the time-dependent vector potential into the hopping matrix element: for the nearest-neighbor pair $(i, i + \rho)$ in the $y$-direction, we use $t \rightarrow t e^{iA(T)}$ where the phase $A(T)$ is given by the vector potential. In the early stage, during the period $T = 0 \sim T_f$, we have applied $A(T) = A_0 \sin(\omega T)$. (Figure 2 shows the results for $A_0 = 0.1$ and $T_f = 80$.) The frequency $\omega$ is tuned for the energy difference between the second highest and the second lowest energy levels of the AF state on the finite-size system. The particles are excited from the lower to the upper energy band (see fig. 2(a)) and the particle-hole (p-h) pairs are created on the second highest and the second lowest energy levels (see fig. 2(b)). In the excited state, the spatial distribution of excitation energy is uniform as seen in fig. 2(f). At $T = T_f$, total amount of the excited electron is about 4, and after that, $A(T) = 0$.

Up to $T \sim 200$, the local-spin structure is almost AF. However, the electron occupation shows a time evolution and the spatial distribution of excitation energy forms a stripe pattern as seen in figs. 2(c) and (g). Correspondingly, the transverse component of the local spins shows a dynamical pattern as in fig. 2(j). During the photo-excitation by the light polarized in the $y$-direction, the electrons are accelerated along the direction. The polarized motion of electrons derives a deformation of the local-spin structure to be favorable for the electron motion.

In the period $T = 200 - 300$, the system shows a drastic change, that is, AFI-to-FM transition. For $T > 300$, on the other hand, $S(\pi, \pi)$ is almost 0, and the gap closes. In fact, the energy level structure is very close to that of the tight-binding Hamiltonian for free-electrons with a Zeeman splitting. (The sparse energy level distribution around zero in energy axis is due to the finite-size effect.) As seen in fig. 2(k), the local-spin almost becomes F corresponding to the metallic electronic-states. The electrons, however, remain still highly excited. As seen in figs. 2(a) and (d), $n_{ex}$ increases and shows a peak at $T \sim 430$. After that, it decreases but as large as the number of initially excited electrons, up to $T \sim 660$. On the other hand, it has been confirmed that the total energy of the system, i.e., the electrons and the local spins, decreases monotonically due to the Gilbert damping.

Figures 2(b)–(e) show that the change of DOS is almost characterized by the AF-to-FM transition. However, the local spins show tiny oscillation which period is much smaller than the time-scale of the AF-to-FM transition.
This rapid-motion of local spin $\vec{S}_i^{\text{rapid}}$ representing the electron-electron interaction [13], brings about the enhancement of $n_{cz}$ for $T_f < T$. (Small intensities around $\vec{q} = (0,0)$ in fig. 2(k) represent the tiny component of $\vec{S}_i^{\text{rapid}}$.) This is a representation of “Auger” process which is absent in the non-interacting system. This multiplication of p-h pairs offers a possibility to enhance the photo-current generation and solar-cell action.

This AFI-to-FM transition, however, does not occur when the excitation is not strong enough. For the results shown in fig. 2, the photo-excitation has been done in the early stage, $0 \leq T \leq T_f$, with $\omega \approx 7S J_{HH}$. By decreasing the period $T_f$, we can reduce the number of the p-h pairs by the photo-excitation $n_{cz}(T_f)$. We find that the IM transition hardly occurs for $n_{cz}(T_f) \lesssim 2$, in the present numerical condition.

We have also examined the frequency $\omega$ and $A_0$ dependence for the IM transition. The rate of p-h pair creation for $T < T_f$ is smaller for smaller $A_0$. In the case that $\omega = 2S J_{HH}$, the threshold of the IM transition is lying on $4 < n_{cz}(T_f) < 5$ in the numerical simulations. Although the IM transition occurs even in this case, we do not find the “Auger” process. Comparison with the above case of $\omega \approx 7S J_{HH}$, it is concluded that the higher-energy photon is more effective to induce the IM transition through the “Auger” process. When $n_{cz}(T_f)$ is close to the threshold, we find the coexistence of the insulating and metallic domains. The results for $\omega = 2S J_{HH}$ and $n_{cz}(T_f) \approx 4.13$ are summarized in fig. 3. For the photo-excitation, $A_0 = 0.15$ and $T_f = 5$ are used. In fig. 3(a), different time scales are used for $T < T_f$ and $T_f < T$, respectively, to zoom up the initial stage for the photo-excitation. During the photo-excitation, total energy increases, and later on ($T_f < T$) it decreases due to the Gilbert damping. In a long time period, $150 \lesssim T \lesssim 1100$, the local-spin state changes gradually (see the time dependence of $S(\pi, \pi)$ in fig. 3(a)).

A long time later, the AF (blue) and F (yellow) domains are spatially separated as seen in fig. 3(d). (The stick with dot indicates the local spins. Each spin is identified to belong to F and AF domains expressed by yellow (gray in print) and blue (dark gray in print) regions, respectively. In the figure, the spin belongs to F (AF) domain when all the neighboring spins are in F (AF) alignment, and the other spins are put into the white area.) The corresponding excitation energy density is shown in fig. 3(e). The energy levels and occupation numbers before and after the transition are shown in figs. 3(b) and (c), respectively. The residual p-h pairs in fig. 3(c) are confined in the F metallic domain, so that the electronic state form a slightly higher-energy density region on the F domain and the stable AFI domain (see figs. 3(d) and (e)). This meta-stable state continues for a long time within our simulation (at least up to $T \sim 6000$).

We have also studied the case that $S_0 = 0.01$ (fig. 4). Other parameters are the same with the case in fig. 2. Despite the reduced relaxation rate, the real-time dynamics of the electronic and local-spin states is not silent.
and is rather active. During the relaxation dynamics, the direction of the staggered magnetization changes drastically. And the spatial inhomogeneity of the electronic state has also been developed (see fig. 4(f)). The slow energy relaxation is advantageous for the “Auger” process resulting in the large amount of p-h pair creation at $T \sim 8000$ when the gap closing occurs (see fig. 4(a)). At this transition, all the local spins play very active dynamics with developing spatial inhomogeneity of the electronic state (see fig. 4(g)). The change of spin configuration from AF state is clearly seen in the equal-time spin structure factor shown in figs. 4(a) and (j): the intensity at $(\pi, \pi)$ becomes small and that at around the center region is grown. Near the center, there exist four spots, and two of them disappear later on in the time evolution. Finally, a spiral spin-structure is obtained in this case (see fig. 4(k)). Due to the Gilbert damping, the total spin-angular-momentum is not conserved. For large $S_\alpha$ the spin state can be easily changed from AF to F as in fig. 2(g) but not for small $S_\alpha$ because the conservation law becomes effective. Therefore, the F state is avoided and the spiral spin configuration appears characterized by $\mathbf{S}(\mathbf{q})$ in fig. 4(k). The numerical simulation shows a dynamical symmetry breaking: The model eq. (1) has a symmetry, i.e., the $x$ and $y$ directions are equivalent. In the case shown in fig. 4, however, the $x$-direction is chosen for the propagation vector of the spiral spin-state. Tiny details of numerical condition, e.g., $A_0$ and $T_f$ of the photo-excitation, the random tilting of local spins, chooses the direction $x$ or $y$ randomly as the propagation vector.

So far, we have discussed the AFI-to-FM transition. As shown in fig. 1(b), FM and AFI states are almost degenerate, so that FM-to-AFI transition of this system is worth to be examined. In the F state, however, the excitation with $\mathbf{q} = 0$ is prohibited. Therefore, we have prepared the initial excited state within the F state by putting the initial electron occupation by “hand”. Although we have examined a number of numerical simulations, FM-to-AFI transition has not been obtained. The numerical results suggest that FM-to-AFI transition is much more difficult than AFI-to-FM transition in the present model.

Now we discuss the relevance of the present study to the experiments. In the manganites, the photo-induced electronic dynamics including the coherent oscillations of the orbital occupancy, i.e., the orbital waves, has been observed with the period of $\sim 40$ fs [2]. This is an evidence of the coupled dynamics between the quantum electron and the classical order-parameter-field by orbital degree of freedom. Although our model does not include the orbital degree of freedom and overestimates the timescale due to the absence of the direct interaction-terms between electrons, we find a similar fluctuation of the classical field, i.e., the rapid motion of the spins in our simulation. Also the asymmetry between insulator-to-metal and metal-to-insulator transitions well corresponds to our result. Furthermore, our results suggest that the spatial pattern formations of quantum-mechanical origin occur accompanied by characteristic electronic structures, which might be revealed by time-resolved X-ray diffraction and/or angle-resolved photo-emission spectroscopy. These issues, however, are left for future studies.

***

The authors are grateful to Y. Tokura, M. Kawasaki, H. Matsueda, T. Tohyama, S. Ishihara and K. Tsutsui for useful discussions. This work is supported by KAKENHI (Grant Nos. 19048015, 19048008, 17105002, 21244053, and 21360043), a High-Tech Research Center project for private universities from MEXT, the “K” computer project of Nanoscience Program, JST-CREST, SICP-(Joint Research Type) from JST, NEDO and Funding Program for World-Leading Innovative R & D on Science and Technology (FIRST Program).

REFERENCES

[1] Miyano K., Tanaka T., Tomioka Y. and Tokura Y., Phys. Rev. Lett., 78 (1997) 4257; Fiebig M., Miyano K., Tomioka Y. and Tokura Y., Science, 280 (1998) 1925.
[2] Polli D., Rini M., Wall S., Schoenlein R. W., Tomioka Y., Tokura Y., Cerullo G. and Cavalleri A., Nat. Mater., 6 (2007) 643.
[3] Rini M., Tobey R., Dean N., Itatani J., Tomioka Y., Tokura Y., Schoenlein R. W. and Cavalleri A., Nature, 449 (2007) 72.
[4] Mitsumori Y., Osawa A., Slupinski T., Maruki H., Kashemura Y., Minami F. and Munekata H., Phys. Rev. B, 69 (2004) 033203.
[5] Kivelson S., Su Y., designated author.
[6] Okimoto Y., Peng X., Tamura M., Morita T., Onida K., Ishikawa T., Ishihara S., Todoroki N., Kyomen T. and Itoh M., Phys. Rev. Lett., 103 (2009) 027402.
[7] Imada M., Fujimori A. and Tokura Y., Rev. Mod. Phys., 70 (1998) 1039.
[8] Nagaosa N. and Ogawa T., Phys. Rev. B, 39 (1989) 4472.
[9] Matsueda H., Ando A., Tohyama T. and Maekawa S., Phys. Rev. B, 77 (2008) 193112.
[10] Kanamori Y., Matsueda H. and Ishihara S., Phys. Rev. Lett., 103 (2009) 267403.
[11] Yonemitsu K. and Maeshima N., Phys. Rev. B, 79 (2009) 125118.
[12] de Gennes P.-G., Phys. Rev., 118 (1960) 141.
[13] Koshida H., Furukawa N. and Nagaosa N., Phys. Rev. Lett., 103 (2009) 266402.
[14] Heger A. J., Kivelson S., Schrieffer J. R. and Su W.-P., Rev. Mod. Phys., 60 (1988) 781.
[15] Malozemoff A. P. and Sloczewsiki J. C., Magnetic Domains Walls in Bubble Materials (Academic Press, London) 1979.