Light-induced switching of magnetic orders in the anisotropic triangular-lattice Hubbard model

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Using the time-dependent exact-diagonalization method, we study the light-induced phase transition of magnetic orders in the anisotropic triangular-lattice Hubbard model. Calculating the spin correlation function, we confirm that the phase transition from the 120° order to Néel order can take place due to the high-frequency periodic fields. We show that the effective Heisenberg-model Hamiltonian derived from the high-frequency expansion by the Floquet theory describes the present system very well and that the ratio of the exchange interactions expressed in terms of the frequency and amplitude of the external field determines the type of the magnetic orders. Our results demonstrate the controllability of the magnetic orders by tuning the external field.

One of the most significant themes in the condensed matter physics concerns the presence of a variety of long-range orders in correlated electron systems. In particular, by strong laser-pulse irradiation, which leads the systems to nonequilibrium states, the creation and control of long-range orders have recently been made feasible in experiments. Typical examples of such attempts include possible photo-induced superconductivity in cuprates [1, 2], alkali-doped fullerides [3], FeSe [4], and organic salts [5], light-induced charge-density wave in LaTe$_3$ [6], and band gap control in excitonic insulators Ta$_2$NiSe$_5$ via photoexcitation [7, 8].

In the manipulation of nonequilibrium states, the concept of “Floquet engineering” attracts particular attention [9], where we create a nonequilibrium steady state by applying a time-periodic external field and change the state to a desired one by tuning amplitude and frequency of the field. The long-range orders in correlated electron systems can be controlled in this manner. In the Hubbard-model Hamiltonian, e.g., the electric field by light irradiation is introduced via the Peierls phase substitution into the hopping integrals, and thus the control of the states is carried out through modifications of the exchange interactions derived in the strong coupling limit of the Hamiltonian [10, 11]. Theoretical studies have so far included the phase transition from antiferromagnetic to ferromagnetic order [10, 12], switching of superconductivity and charge-density wave in the attractive Hubbard model [11, 13, 14], etc. Related experiments have been carried out in ultracold atom systems [15]. The success of controlling the exchange interactions has also been reported in iron oxides [16].

In this paper, motivated by such developments in the field, we focus on the Hubbard model at half filling defined on the anisotropic triangular lattice (ATL), which is one of the representative systems with geometrical frustration. Since the frustrated systems have many competing orders in their ground states, we can expect the realization of control and switching of the orders by the external field [17–20]. The ground-state phase diagram of our model has so far been investigated well by the variational-cluster approach, where we know that the Néel order, the 120° order, and the collinear order compete to each other [21–23]. Materials described by this model include an inorganic compound Cs$_2$ClO$_4$ [24] as well as organic compounds such as $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl [25] and $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [26].

In what follows, we will first prepare the ATL Hubbard Hamiltonian whose ground state is the 120°-type antiferromagnetic order, and simulate the change in this quantum state under the time-periodic external field, where we use the time-dependent Lanczos method. Then, we will show that by tuning the amplitude and frequency of the field, the initial state with the 120° order can actually be switched to the Néel order. This result will be interpreted using the Floquet effective Hamiltonian obtained from the high-frequency expansion of our model. We will also perform the calculations of the quench dynamics of the Heisenberg model obtained in the strong coupling limit of the Hubbard model, and confirm the validity of our Floquet analysis. We will thus demonstrate the controllability of the magnetic orders in the frustrated spin system by tuning the external field.

The ATL Hubbard model is defined by the Hamiltonian [see Fig. 1(a)]

$$\hat{\mathcal{H}} = -t_1 \sum_{\langle i,j \rangle, s} \hat{c}^\dagger_{i,s} \hat{c}_{j,s} - t_2 \sum_{\langle\langle i,j \rangle\rangle, s} \hat{c}^\dagger_{i,s} \hat{c}_{j,s} + U \sum_i \hat{n}_{i,\uparrow} \hat{n}_{i,\downarrow},$$

(1)

where $\hat{c}^{(1)}_{i,s}$ is the annihilation (creation) operator of an electron at site $i$ with spin $s$, and $\hat{n}_{i,s} = \hat{c}^\dagger_{i,s} \hat{c}_{i,s}$ is the electron density operator. $t_1$ and $t_2$ are the nearest-neighbor (NN) and next-nearest-neighbor (NNN) hopping integrals, respectively, and $U$ is the on-site Coulomb interaction. The notations $\langle i,j \rangle$ and $\langle\langle i,j \rangle\rangle$ represent the pairs of the NN and NNN sites, respectively. In the strong-coupling limit $U/t_1 \to \infty$ at half filling, the Hubbard model in Eq. (1) is mapped onto the antiferromagnetic Heisenberg model defined by

$$\hat{\mathcal{H}}_{\text{eff}} = J_1 \sum_{\langle i,j \rangle} \hat{S}_i \cdot \hat{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \hat{S}_i \cdot \hat{S}_j,$$

(2)
NNN direction, i.e., in the present study, we use the vector potential parallel to the boundary condition, and adopt a twelve-site cluster illustrated in Fig. 1(a) with periodic boundary condition, and adopt the lattice constant to be unity. Following, we assume $\sigma_p = 2.0/t_1$ and $t_0 = 10/t_1$. We set the Planck constant $\hbar$, the speed of light $c$, the elementary charge $e$, and the lattice constant to be unity.

Since the Hamiltonian depends on time in the presence of the external field, we need to solve the Schrödinger equation to obtain the time evolution of the wave function. We employ the time-dependent Lanczos method [29, 30] for this purpose. The time evolution with a time step $\delta t$ is calculated in the corresponding Krylov subspace generated by $M_L$ Lanczos iterations. We use the twelve-site cluster illustrated in Fig. 1(a) with periodic boundary condition, and adopt $\delta t = 0.01/t_1$ and $M_L = 20$. The time-dependent external field is introduced via the Hamiltonian $H = H_0 + \delta H(t)$, where we assume $\delta H(t) = \delta H(0) \delta(t)$.

The ordering vector $q = (\pi, \pi)$ is illustrated in Fig. 1(b) at $J_2/J_1 < 0.83$, which switches to the 120° order [Fig. 1(c)] at $J_2/J_1 > 0.83$ [27, 28].

The time-dependent external field is introduced via the Peierls phase. Then, the hopping integrals are modified as

$$t_n e^{i A(t)} \mathcal{C}_{j, s} \rightarrow t_n e^{-i A(t)} \mathcal{C}_{j, s},$$

where $A(t)$ is the vector potential at time $t$. In the present study, we use the vector potential parallel to the NNN direction, i.e., $A(t) = t \left( \frac{A_0}{2} (A(t), A(t)) \right)$, where

$$A(t) = \begin{cases} A_0 e^{-((t-t_0)^2/2\sigma_p^2)} \cos [\omega_p (t - t_0)] & (t \leq t_0) \\ A_0 \cos [\omega_p (t - t_0)] & (t > t_0) \end{cases}$$

with the amplitude $A_0$ and the frequency $\omega_p$. In the following, we assume $\sigma_p = 2.0/t_1$ and $t_0 = 10/t_1$. We set the Planck constant $\hbar$, the speed of light $c$, the elementary charge $e$, and the lattice constant to be unity.

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2(b). We find that the 120° order is still preferred in the irradiation of the external field with \( A_0 = 0.1 \) and \( \omega_p = 30/t_1 \) [see Fig. 2(a)], whereas the Néel order is strongly enhanced after the irradiation of the external field with \( A_0 = 4.0 \) and \( \omega_p = 30/t_1 \) [see Fig. 2(b)]. The present results suggest that the light-induced phase transition may occur in the ATL Hubbard model if the intensity of light is sufficiently strong.

To explore the parameter regions where the 120° order remains or the Néel order takes over after the light irradiation, we calculate the difference in the spin correlation function \( \Delta S_z(q) \) in the parameter space \((\omega_p/t_1, A_0)\). The results are shown in Fig. 3(a) for the Néel order and in Fig. 3(a) for the 120° order. These results clearly indicate that when the 120° order is suppressed, the Néel order is complementarily enhanced. In addition, both orders are strongly suppressed at \( \omega_p/t_1 < 25 \). In this region, the double occupancy defined as

\[
n_d(t) = \frac{1}{L} \sum_i \langle \psi(t) | \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} | \psi(t) \rangle
\]

increases [see Figs. 2(c) and 2(d)] and therefore the charge excitations occurring across the Mott-Hubbard gap \((\approx U/t_1)\) increase, which leads to the suppression of the spin fluctuations. In Fig. 3(c), we show the calculated result for \( \bar{S}_z(Q_{\text{Néel}}) - \bar{S}_z(Q_{120°}) \). This result indicates which order is realized in the parameter space after the light irradiation; in the red area, the Néel order appears, while in the blue area, the 120° order remains.

In order to discuss the origin of the light-induced phase transition, we analyze the model using the Floquet theory. Applying the high-frequency expansion to our Hubbard-model Hamiltonian with the external field \( A(t) = A_0 \cos \omega_p t \), we obtain the effective Hamiltonian in the strong-coupling limit as [11]

\[
\mathcal{H}_{\text{eff}} = J_1^{\text{eff}} \sum_{\langle i,j \rangle} \hat{S}_i \cdot \hat{S}_j + J_2^{\text{eff}} \sum_{\langle\langle i,j \rangle\rangle} \hat{S}_i \cdot \hat{S}_j,
\]

FIG. 3. Differences in the spin correlation function calculated in the parameter space \((\omega_p/t_1, A_0)\). Shown are (a) \( \Delta S_z(Q_{\text{Néel}}) \), (b) \( \Delta S_z(Q_{120°}) \), and (c) \( \bar{S}_z(Q_{\text{Néel}}) - \bar{S}_z(Q_{120°}) \).

FIG. 4. (a) Difference \( \bar{S}_z(Q_{\text{Néel}}) - \bar{S}_z(Q_{120°}) \) calculated for the effective Heisenberg model Eq. (8) in the parameter space \((\omega_p/t_1, A_0)\). (b) Ratio of the Floquet effective exchange interactions: the region of \( J_2^{\text{eff}}/J_1^{\text{eff}} > 0.83 \) is indicated by red color, while the region of \( J_2^{\text{eff}}/J_1^{\text{eff}} < 0.83 \) is indicated by blue color.
where

\[ J_1^{\text{eff}} = \sum_{m=-\infty}^{\infty} (-1)^m \frac{4 \pi^2 J_m(A_0/\sqrt{2})}{U + m \omega_p} \]

(9)

and

\[ J_2^{\text{eff}} = \sum_{m=-\infty}^{\infty} (-1)^m \frac{4 \pi^2 J_m(\sqrt{2}A_0)}{U + m \omega_p} \]

(10)

are the NN and NNN Floquet effective exchange interactions, respectively. Here, \( J_m(x) \) is the \( m \)-th Bessel function. Using this model, we perform the calculation of the quench dynamics [31], where \( J_1 \) and \( J_2 \) are suddenly changed to \( J_1^{\text{eff}} \) and \( J_2^{\text{eff}} \), respectively, at \( t = 0 \). The result for the difference \( S_2(Q_{\text{Néel}}) - S_z(Q_{120^\circ}) \) thus calculated is shown in Fig. 4(a) in the parameter space \( (\omega_p/t_1, A_0) \), which we find is consistent with the result obtained for the Hubbard model at least in the region \( \omega_p/t_1 \gtrsim 25 \) [see Fig. 3(c)]. The inconsistency found in the region \( \omega_p/t_1 \lesssim 25 \) comes from the enhancement of the double occupancy, which cannot be explained by the strong-coupling expansion. We thus conclude that in the regime of sufficiently high frequency the result obtained from the Floquet effective Hamiltonian Eq. (8) well explains the behaviors of the Hubbard model in the strong coupling region under the time-periodic external field.

We also calculate the phase diagram of the effective Hamiltonian simply from the ratio of the effective exchange interactions \( J_2^{\text{eff}} / J_1^{\text{eff}} \). The result is shown in Fig. 4(b), where the phase boundary is determined as the line \( J_2/J_1 = 0.83 \), at which the phase transition between the Néel and 120° orders occurs in the ground state of the ATL Heisenberg model. We thus find that the Néel order is preferred in the red region \( (J_2^{\text{eff}} / J_1^{\text{eff}} < 0.83) \), while the 120° order is preferred in the blue region \( (J_2^{\text{eff}} / J_1^{\text{eff}} > 0.83) \). We thus clearly find that the phase diagram obtained by the quench-dynamics calculation is consistent with the phase diagram determined from the ratio of the exchange interactions, implying that the magnetic order realized by the light irradiation can be predicted from the Floquet theory. We note that there are regions where \( J_1^{\text{eff}} < 0 \) and \( J_2^{\text{eff}} < 0 \), i.e. the ferromagnetic order is preferred. Our calculated results, however, do not indicate the presence of such regions. This is because the electric field never flips the spins, or the total spin is conserved by the light irradiation.

In summary, we have investigated the time dependence of the spin correlations of the anisotropic triangular Hubbard model at half filling under the time-periodic external electric field using the time-dependent Lanczos method. We have shown that the 120° order can be switched to the Néel order by tuning the frequency and the amplitude of the external field. To understand the magnetic phase transition under the periodic field, we have introduced the effective Heisenberg-model Hamiltonian by the high-frequency expansion. The phase diagram obtained from the quench dynamics of this effective model is consistent with the results of our Hubbard-model calculations, which implies that the phase diagram obtained by the light irradiation can be interpreted by the Floquet theory. Thus, the switching of the magnetic orders can be realized in the frustrated spin system by tuning the amplitude and frequency of the external field. We hope that our results will shed some light on possible realization of the photo-control of magnetic orders in the frustrated spin systems.

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[1] D. Fausti, R. I. Tobey, N. Dean, S. Kaiser, A. Dienst, M. C. Hoffmann, S. Pyon, T. Takayama, H. Takagi, and A. Cavalleri, Science 331, 189 (2011).
[2] W. Hu, S. Kaiser, D. Nicoletti, C. R. Hunt, I. Gierz, M. C. Hoffmann, M. Le Taon, T. Loew, B. Keimer, and A. Cavalleri, Nat. Mater. 13, 705 (2014).
[3] M. Mitrano, A. Cantaluppi, D. Nicoletti, S. Kaiser, A. Perucchi, S. Lupi, P. Di Pietro, D. Pontiroli, M. Riccò, S. R. Clark, D. Jaksh, and A. Cavalleri, Nature Phys. 530, 461 (2016).
[4] T. Suzuki, T. Someya, T. Hashimoto, S. Michimae, M. Watanabe, M. Fujisawa, T. Kanai, N. Ishii, J. Itatani, S. Kasahara, Y. Matsuda, T. Shibauri, K. Okazaki, and S. Shin, Commun. Phys. 2, 115 (2019).
[5] M. Buzzi, D. Nicoletti, M. Fechner, N. Tancogne-Dejean, M. A. Sentef, A. Georges, T. Biesener, E. Uykur, M. Dressel, A. Henderson, T. Siegrist, J. A. Schnizler, K. Miyagawa, K. Kanoda, M.-S. Nam, A. Ardavan, J. Coulthard, J. Tindall, F. Schlawin, D. Jaksh, and A. Cavalleri, Phys. Rev. X 10, 31028 (2020).
[6] A. Kogar, A. Zong, P. E. Dolgirev, X. Shen, J. Straquardine, Y. Q. Bie, X. Wang, T. Rohwer, I. C. Tung, Y. Yang, R. Li, J. Yang, S. Weathersby, S. Park, M. E. Korina, E. J. Sie, H. Wen, P. Jarillo-Herrero, I. R. Fisher, X. Wang, and N. Gedik, Nat. Phys. 16, 159 (2020).
[7] S. Mor, M. Herzog, D. Golez, P. Werner, M. Eckstein, N. Katayama, M. Nohara, H. Takagi, T. Mizokawa, C. Monney, and J. Stähler, Phys. Rev. Lett. 119, 086401 (2017).
[8] K. Okazaki, Y. Ogawa, T. Suzuki, T. Yamamoto, T. Someya, S. Michimae, M. Watanabe, Y. Lu, M. Nohara, H. Takagi, N. Katayama, H. Sawa, M. Fujisawa, T. Kanai, N. Ishii, J. Itatani, T. Mizokawa, and S. Shin, Nat. Commun. 9, 4322 (2018).
[9] T. Oka and S. Kitamura, Annu. Rev. Condens. Matter Phys. 10, 387 (2019).
[10] J. H. Mentink, K. Balzer, and M. Eckstein, Nat. Commun. 6, 6708 (2015).
[11] S. Kitamura and H. Aoki, Phys. Rev. B 94, 174503 (2016).
[12] N. Dasari and M. Eckstein, Phys. Rev. B 100, 121114(R) (2019).
[13] M. A. Sentef, A. Tokuno, A. Georges, and C. Kollath, Phys. Rev. Lett. 118, 087002 (2017).
[14] R. Fujiuchi, T. Kaneko, K. Sugimoto, S. Yunoki, and Y. Ohta, Phys. Rev. B 101, 235122 (2020).
[15] A. Eckardt, Rev. Mod. Phys. 89, 011004 (2017).
[16] R. V. Mikhaylovskiy, E. Hendry, A. Secchi, J. H. Mentink, M. Eckstein, A. Wu, R. V. Pisarev, V. V. Kruglyak, M. I. Katsnelson, T. Rasing, and A. V. Kimel, Nat. Commun. 6, (2015).
[17] S. Kitamura, T. Oka, and H. Aoki, Phys. Rev. B 96, 014406 (2017).
[18] M. Claassen, H.-C. Jiang, B. Moritz, and T. P. Devreux, Nat. Commun. 8, 1192 (2017).
[19] S. Jana, P. Mohan, A. Saha, and A. Mukherjee, Phys. Rev. B 101, 115428 (2020).
[20] N. Bittner, D. Golež, M. Eckstein, and P. Werner, arXiv:2005.11722.
[21] A. Yamada, Phys. Rev. B 89, 195108 (2014).
[22] M. Laubach, R. Thomale, C. Platt, W. Hanke, and G. Li, Phys. Rev. B 91, 245125 (2015).
[23] K. Misumi, T. Kaneko, and Y. Ohta, J. Phys. Soc. Jpn. 85, 064711 (2016).
[24] R. Coldea, D. A. Tennant, K. Habicht, P. Smeibidl, C. Wolters, and Z. Tylczynski, Phys. Rev. Lett. 88, 137203 (2002).
[25] S. Lefebvre, P. Wzietek, S. Brown, C. Bourbonnais, D. Jérôme, C. Mézière, M. Fourmigué, and P. Batail, Phys. Rev. Lett. 85, 5420 (2000).
[26] Y. Shimizu, K. Miyagawa, K. Kanoda, M. Maesato, and G. Saito, Phys. Rev. Lett. 91, 107001 (2003).
[27] Z. Weihong, R. H. McKenzie, and R. R. P. Singh, Phys. Rev. B 59, 14367 (1999).
[28] S. Yunoki and S. Sorella, Phys. Rev. B 74, 014408 (2006).
[29] T. J. Park and J. Light, J. Chem. Phys. 85, 5870 (1986).
[30] N. Mohankumar and S. M. Auerbach, Comput. Phys. Commun. 175, 473 (2006).
[31] M. Kawamura, K. Yoshimi, T. Misawa, Y. Yamaji, S. Todo, and N. Kawashima, Comput. Phys. Commun. 217, 180 (2017).