Supplementary Information
Exciton-assisted low-energy magnetic excitations in a photoexcited Mott insulator on the square lattice

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SUPPLEMENTARY NOTE

In the main text, we showed time-resolved two-magnon Raman (trTMR) spectrum for a half-filled Hubbard model with the nearest-neighbor hopping t_h and the on-site Coulomb interaction U given by

$$H_0 = -t_\mathrm{h} \sum_{i\delta\sigma} c^\dagger_{i\sigma} c_{i+\delta\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \tag{S.1}$$

where $c^\dagger_{i\sigma}$ is the creation operator of an electron with spin σ at site i, number operator $n_{i\sigma} = c^\dagger_{i\sigma} c_{i\sigma}$, $i + \delta$ represents four nearest-neighbor sites around site i. In insulating cuprates such as $\mathrm{La_2CuO_4}$, the second-neighbor hopping t'_h is necessary for describing its electronic states. Therefore, it is important to investigate the effect of t'_h on trTMR. The corresponding term is given by $-t'_\mathrm{h} \sum_{i\delta'\sigma} c^\dagger_{i\sigma} c_{i+\delta'\sigma}$, where $i + \delta'$ represents four second-neighbor sites around site i. The Peierls substitution of an external electric field given by a pump pulse is done by $c^\dagger_{i,\sigma} c_{i+\delta(\delta')\sigma} \to e^{-i\mathbf{A}(t)\cdot\mathbf{R}_{\delta(\delta')}} c^\dagger_{i,\sigma} c_{i+\delta(\delta')\sigma}$. Here, $\mathbf{R}_{\delta(\delta')}$ is the vector from i to $i + \delta(\delta')$, and $\mathbf{A}(t)$ is the vector potential given by

$$\mathbf{A}(t) = \mathbf{A}_0 e^{-(t-t_0)^2/(2t_d^2)} \cos[\omega_\mathrm{p}(t - t_0)], \tag{S.2}$$

where a Gaussian-like envelope centered at t_0 has a temporal width t_d and a central frequency ω_p.

As is the case of the main text, we set $U = 10$, $t_0 = 0$ and $t_d = 0.5$ by taking $t_\mathrm{h} = 1$ as an energy unit and $1/t_\mathrm{h}$ as a time unit. In addition, we take $t'_\mathrm{h} = -0.25$ [1]. We then calculate $\mathrm{B_1}$ trTMR spectrum $M(\omega; t)$ after turning off a pump pulse for a 4×4 periodic lattice. $M(\omega; t)$ was defined in the main text and numerically calculated by using a Lanczos-type exact diagonalization technique. Supplementary Figure 1 shows the ω_p dependence of $M(\omega; t)$ at $t = 4$. Similar to the case of $t'_\mathrm{h} = 0$ as shown in Fig. 3 of the main text, the pump pulse suppresses the intensity of the two-magnon peak at $\omega = 0.95$, which has been observed in the trTMR experiment for photoirradiated $\mathrm{YBa_2Cu_3O_{6.1}}$ [2]. When $\omega_\mathrm{p} = 6$, we find an enhancement of low-energy spectral weight around $\omega = 0.25$. This is also similar to the case without t'_h discussed in the main text. These similarities clearly indicate that the presence of t'_h does not change the emergence of low-energy magnetic excitations caused by photo pumping. We note that a small shift of both the two-magnon peak and photoinduced low-energy structure toward lower energy as compared with the case without t'_h (see Fig. 3 in the main text) can be attributed to the effect of magnetic frustration induced by second-neighbor superexchange interactions due to finite t'_h.

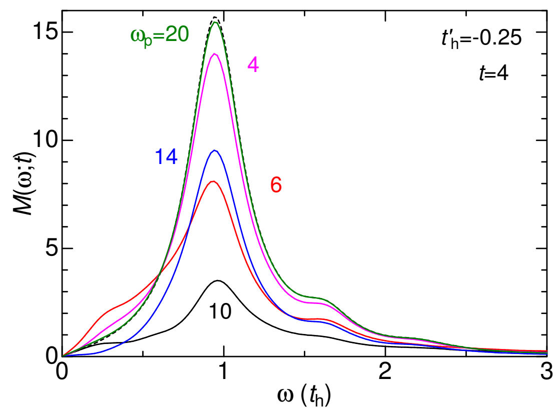

FIG. 1. Pumping frequency ω_p dependence of time-resolved two-magnon Raman spectrum $M(\omega; t)$ at $t = 4$ after pumping for a half-filled 4×4 Hubbard lattice with $U = 10$ and $t'_\mathrm{h} = -0.25$. Dashed back line represents the results before pumping, while purple, read, black, blue, and green solid lines represent the results at ω_p=4, 6, 10, 14, and 20, respectively.

In the main text, the polarization vector $\mathbf{A}_0$ in a pump pulse was set to be along the (1,0) direction, i.e., $\mathbf{A}_0 = (A_0, 0)$. On the contrary, in the trTMR experiment for $\mathrm{YBa_2Cu_3O_{6.1}}$ [2], a polarization vector has been selected to be the (1,1) direction. In order to clarify whether the emergence of photoinduced low-energy magnetic excitations depends on polarization direction or not, we show the ω_p dependence of $\mathrm{B_1}$ trTMR spectrum for $\mathbf{A}_0 = (A_0/\sqrt{2}, A_0/\sqrt{2})$ with $A_0 = 0.5$ in Supplementary Figure 2. The photoinduced low-energy excitations exist at $\omega_\mathrm{p} = 6$ as is the case of the (1,0) direction. Therefore, we conclude that the low-energy excitations are independent of polarization direction and thus

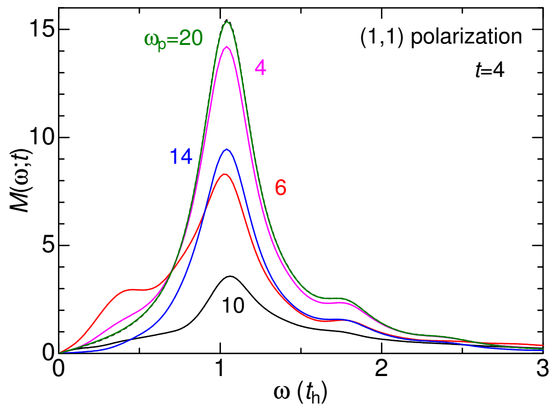

FIG. 2. Pumping frequency ω_p dependence of time-resolved two-magnon Raman spectrum $M(\omega;t)$ at $t=4$ after pumping with (1,1) polarization direction for a half-filled 4×4 Hubbard lattice with $U=10$ and $t'_\mathrm{h}=0$. Dashed back line represents the results before pumping, while purple, read, black, blue, and green solid lines represent the results at ω_p=4, 6, 10, 14, and 20, respectively.

the observed one ranging from 1000cm^{-1} to 1500cm^{-1} in $\mathrm{YBa_2Cu_3O_{6.1}}$ can be explained by our interpretation discussed in the main text.

[1] K. Tsutsui, K. Shinjo, and T. Tohyama, Antiphase Oscillation in Time-Resolved Spin Structure Factor of a Photoexcited Mott Insulator, Phys. Rev. Lett. **126**, 127404 (2021).

[2] J.-A. Yang, N. Pellatz, T. Wolf, R. Nandkishore, and D. Reznik, Ultrafast magnetic dynamics in insulating $\mathrm{YBa_2Cu_3O_{6.1}}$ revealed by time resolved two-magnon Raman scattering, Nat. Commun. **11**, 2548 (2020).