Threshold phenomena under photoexcitation of spin-crossover materials with cooperativity due to elastic interactions

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Photo-induced switching from the low-spin state to the high-spin state is studied in a model spin-crossover material with long-range interactions induced by elastic distortions due to different molecular sizes of the two spin states. At a threshold value of the light intensity, nonequilibrium critical behavior is observed in the form of a jump in the high-spin fraction, corresponding to a mean-field spinodal point. Finite-size scaling of the divergence of the transition time is revealed through kinetic Monte Carlo simulations.

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I. INTRODUCTION

In many materials, local elastic distortions induce effective long-range interactions via the macroscopic strain field.[1–3] In addition to elastic crystals,[1] physical realizations include systems as diverse as earthquake faults[4] and long-chain polymers.[5] Phase transitions in such systems belong to the *mean-field* universality class,[4] which has unusual properties, including strong suppression of clusters, compared to transitions caused by purely short-range interactions. Recently it was proposed that molecular solids known as spin-crossover (SC) materials may exemplify this class of systems.[6,7]

The molecules of a SC compound can exist in a low-spin ground state (LS) and a high-spin excited state (HS) of higher degeneracy.[8] If the intermolecular interactions are sufficiently strong, the change between LS and HS can take place cooperatively, and various types of ordering processes may occur.[9–13] The different magnetic and optical properties of the two phases make cooperative SC materials promising candidates for switches, displays, and recording media,[11] and thus their cooperative properties have been studied extensively.[8]

The thermodynamic properties of our SC model can be applied to various systems in which a low-energy state with low degeneracy and a high-energy state with high degeneracy compete. In particular, charge-transfer materials or Prussian-blue analog-type systems possess common thermodynamic properties with the SC systems. Therefore, understanding of the mechanism of the SC system would be widely applicable to those systems.

SC systems also show interesting dynamical properties. At low temperatures and in the absence of light irradiation, the LS phase is the stable phase, while the exited HS phase is unstable or metastable. The HS phase can be induced at low temperatures by several methods, including light exposure, known as light-induced excited spin-state trapping (LIESST).[8,14–16] Magnetization switching at low temperatures by light irradiation promises a method to control functional material properties.[17]

In this paper, we study the photo-induced LS-to-HS transition in a model with local elastic distortions, which are inherent to SC materials and induce effective long-range interactions. In a numerical study of this model without irradiation,[18] we found that the appearance of large compact clusters is suppressed and the system remains uniform, even near the equilibrium critical point and during hysteresis. The observed critical exponents and scaling relations confirm that the model belongs to the mean-field universality class. Under sufficiently strong irradiation at low temperatures an exchange of stabilities may take place, whereby the excited HS phase becomes stable and the LS phase becomes metastable or unstable. In systems with effective long-range interactions we expect the change from metastability to instability of the LS phase to take place at a sharply defined mean-field spinodal point.[4] This leads to a threshold behavior with respect to the irradiation intensity,[4,19,20] which is qualitatively different from the gradual crossover seen in short-range models.[21] Here, we therefore investigate the LS-to-HS transition time and its dependence on the system size as functions of the irradiation intensity. Our results are of general interest as they not only predict nonequilibrium physical properties of a technologically important class of materials, but also because SC materials might serve as laboratory-scale analogs of less readily investigated elastic solids, such as the earth's crust.

In a typical model of a SC material, the energy difference between the HS and LS states is taken as $D>0$, while the degeneracy g_{HS} of HS is larger than g_{LS}, such that HS is preferable at high temperatures. We can express the HS (LS) spin state of the ith molecule by $\sigma_i=+1(-1)$, giving the intramolecule Hamiltonian,

$$\mathcal{H}_{\mathrm{eff}}=\frac{1}{2}\sum_i (D-k_{\mathrm{B}}T\ln g)\sigma_i, \tag{1}$$

where $g=g_{\mathrm{HS}}/g_{\mathrm{LS}}$. The ratio of the degeneracies is expressed by the temperature dependent field, $k_{\mathrm{B}}T\ln g$. In order to pro-

vide the cooperative property in the SC transition, a short-range Ising-type interaction has commonly been adopted.[13,22] Here we instead consider the alternative mechanism with long-range effective interactions induced by local elastic distortions due to the volume difference between the LS and HS molecules.[6,7,18]

We study the dynamics under photoexcitation from the perfect LS state at low temperature. For a single molecule, we find a smooth increase in the HS fraction, and its saturated value is a smooth function of the irradiation intensity. On the other hand, with elastic interactions a threshold phenomenon appears. Under weak irradiation, the system stays in the LS state, but if the intensity exceeds a threshold, it jumps to the HS state. Such threshold phenomena have been pointed out in ordering processes.[23] This sudden change is analogous to the sudden change in magnetization at the coercive field in the hysteresis loop of a ferromagnet following reversal of the applied field, known as a "spinodal point."

The dynamics of field-driven first-order phase transitions have previously been studied in detail for models with short-range interactions. In that case, the nucleation and growth of compact clusters play important roles, and different regimes have been identified, depending on the relative time scales of nucleation and growth.[21] If the average time between nucleation events is longer than the time it takes a single cluster to grow to a size comparable with the system, the transformation will happen stochastically in a Poisson process via a single cluster (single-droplet, or SD, regime), and the average lifetime of the metastable phase is inversely proportional to the system volume. If the average time between nucleation events is shorter than the growth time, many clusters nucleate while the first one is growing, resulting in an almost deterministic process (multidroplet, or MD, regime), and the metastable lifetime is independent of the system volume (Avrami's law).[21]

The rest of this paper is organized as follows. In Sec. II we present the details of our model, in Sec. III we present a phenomenological description of the switching process under photo-irradiation, and in Sec. IV we present a finite-size scaling analysis of the dynamics near the nonequilibrium spinodal critical point. Finally, in Sec. V we present a summary and discussion of our results.

II. MODEL

We perform kinetic Monte Carlo (MC) simulations by the constant-pressure method.[7] As the intramolecular interaction, we adopt the form (1) with the parameters $g=20$ and $D=0.5$. In addition, the Hamiltonian contains intermolecular elastic interactions between nearest and next-nearest neighbors[7]

$$V=\frac{k_1}{2}\sum_{\langle i,j\rangle}[r_{ij}-(R_i+R_j)]^2+\frac{k_2}{2}\sum_{\langle\langle i,j\rangle\rangle}[r_{ij}-\sqrt{2}(R_i+R_j)]^2, \tag{2}$$

where r_{ij} is the distance between the ith and jth sites, and R_i and R_j are the molecular radii: R_{HS} and R_{LS} for HS and LS, respectively. Their ratio is set to $R_{\mathrm{HS}}/R_{\mathrm{LS}}=1.1$. The elastic constants are $k_1/k_2=10$ with $k_1=50$. To this model we add the kinetic process of photo-irradiation. The photoexcitation is simulated as follows. A site is randomly chosen and changed to the HS state with probability a if it is previously in the LS state. We identify a with the irradiation intensity. (Strictly speaking, a increases monotonically with the radiation intensity.) Although this MC method does not represent the microscopic transition processes in detail, we do not expect this to change the universal aspects of the nonequilibrium critical properties.[24]

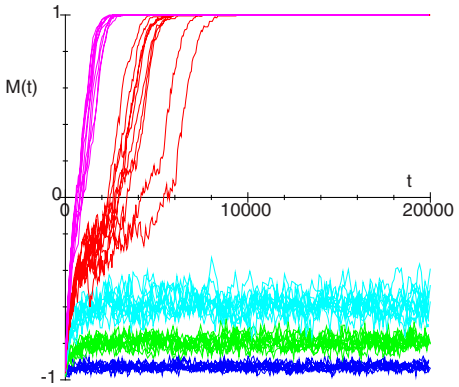

FIG. 1. (Color online) Time series of the order parameter $M(t)$ under irradiation. Irradiation intensities $a=0.001$, 0.002, 0.003, 0.004, and 0.005 from bottom to top. We plot 10 samples for each value of a. The system size is $L=50$. The time t is measured in MCSS.

The order parameter is the fraction of HS molecules, f_{HS}, which is expressed by the "magnetization"

$$M=\frac{1}{N}\sum_{i}^{N}s_i=\frac{1}{2}(f_{\mathrm{HS}}-1). \tag{3}$$

We study a two-dimensional model on a square lattice at the low temperature of $k_{\mathrm{B}}T=0.01$.

III. PHOTOEXCITATION PROCESS

In Fig. 1, we depict typical time series of the HS fraction under irradiation.

We find a sudden change in the saturated values. For $a\leq 0.003$, the system stays in the LS state, while it jumps to the HS state for $a\geq 0.004$. Excitation processes under photo-irradiation have been extensively studied experimentally. For example, experimental results for the SC material $[\mathrm{Fe(pic)_3}]$ indicate (somewhat smeared) threshold behavior.[25] In Fig. 2, we show the dependence of the saturated value of the order parameter M on a. Because of the cooperative interaction, the HS fraction remains low when the irradiation is weak. On the other hand, it approaches unity when a exceeds a threshold.

We also found a sudden change in M as a function of time. We depict an example of the transition process for $L=50$ in Fig. 3(a), which shows a sharp change around $t=5000$ Monte Carlo steps per site (MCSS). The HS fraction stays at an intermediate value ($M\simeq -0.2$) for a while, and then it rapidly increases to the saturated value. This sudden

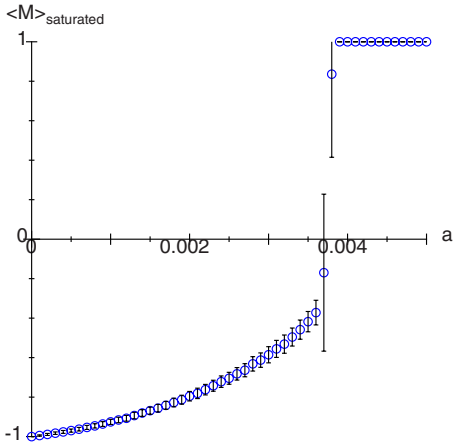

FIG. 2. (Color online) Saturated value of the order parameter M, proportional to the HF fraction, shown vs the irradiation intensity a.

change is reminiscent of a transformation via single-droplet nucleation from the metastable phase to the equilibrium phase. However, no visible clusters are observed. In Fig. 3(b) we depict a configuration at $t=6000$ MCSS, near the starting point of the sudden change. This absence of cluster structures is the same characteristic found previously in simulations of hysteresis in this model.[18]

IV. CRITICAL PROPERTIES OF SWITCHING PHENOMENA

A. Size dependence of the transition time

Next, we study the system-size dependence of the transition time. Here we define the transition time τ as the time at which the average of σ_i reaches 0.5, corresponding to a HS fraction of 0.75, starting from the perfect LS state ($\sigma_i=-1$). We perform $N_{\text{sample}}(\geq 200)$ independent runs for each parameter set and average the results. In Fig. 4, we depict the size dependences of τ for $a \in [0.00365, 0.00400]$, obtained by averaging over the samples. Here, we plot the data vs $L^{2/3}$ because it is expected that the transition time diverges as $L^{2/3}$ at the spinodal point (see below). We plot the transition times for various values of a as functions of $N^{1/3}=L^{2/3}$ in Fig. 4. At

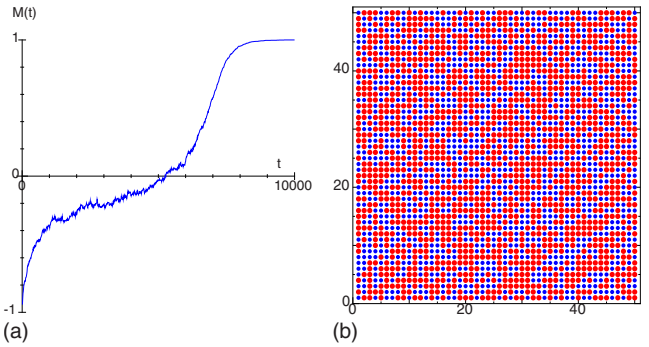

FIG. 3. (Color online) (a) An example of the time evolution of the order parameter $M(t)$ under irradiation with $a=0.004$. Time is given in MCSS. (b) Spin configuration at $t=6000$ MCSS during the process of photo-irradiation shown in (a).

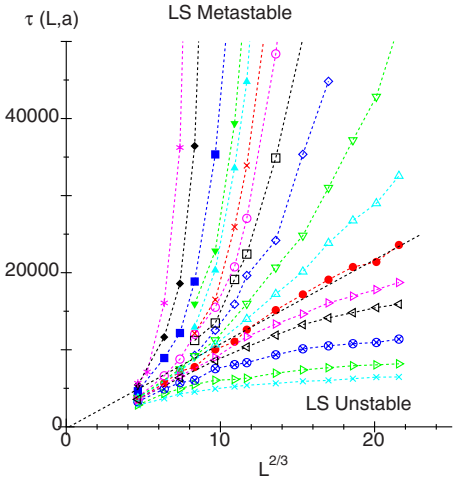

FIG. 4. (Color online) Dependence of the transition time τ on system size L and irradiation intensity a. Shown as τ vs $L^{2/3}$ for $a=0.00365$, 0.00370, 0.00375, 0.00377, 0.00378, 0.00379, 0.00380, 0.00381, 0.00382, 0.00383, 0.00384, 0.00385, 0.00386, 0.00387, 0.00390, 0.00395, and 0.00400 from above to below. Straight-line behavior consistent with Eq. (6) is seen for $a=0.00385 \approx a_{\text{SP}}$. Labels indicate the regions where the LS state is metastable and unstable, respectively.

$a=0.00385$ they follow a straight line, consistent with the properties of the mean-field spinodal point Eq. (6).

We found the sudden change in the saturated value of M around $a_{\text{SP}} \approx 0.0385$ in Fig. 2. Below this value, the transition times shown in Fig. 4 increase rapidly with L, while above it, they saturate for large L. As shown in Fig. 1, for $a < a_{\text{SP}}$ the times of the jump are widely distributed. This is a sign of stochastic nucleation, although we do not observe any clusters in Fig. 3(b). For $a > a_{\text{SP}}$, the transformation process becomes nearly deterministic. In Table I, we list typical examples.

It should be noted that the transition time increases with the system size. This system-size dependence is *qualitatively* different from the behavior in systems with short-range interactions.[21] In the latter case, the transition time is determined by the probability of creating a critical *localized, compact* droplet, whose free energy is *independent* of the system size. Droplets thus nucleate in a Poisson process of rate $\propto L^d$, where d is the spatial dimensionality of the system (here, $d=2$). Thus, the transition time decreases with system size in the SD regime, while it is size independent in the MD regime.

Compared to this dependence in short-range systems, the increase in the transition time in the present system with the system size is notable. The divergence when $L \to \infty$ indicates that the present threshold phenomenon is not just a crossover as in short-range systems, but a true critical phenomenon. Thus we will study its critical properties by analyzing the size dependence of the transition time.

The simple van Hove dynamics in the mean-field approximation implies that the waiting time before a jump over the free-energy barrier for $a < a_{\text{SP}}$ has the form

$$\tau \sim \exp(cN), \tag{4}$$

where $N=L^d$ is the volume of the system and c is positive. The present case appears similar. Indeed, the present thresh-

TABLE I. List of average and standard deviation of the distribution of the transition time τ.

a	L	N_{sample}	$\langle\tau\rangle$	$\sqrt{\langle\tau^2\rangle-\langle\tau\rangle^2}$
0.00375	20	200	12174.045	8189.569
0.00375	40	200	105239.460	96223.716
0.00375	60	400	4608237.905	4343268.886
0.00385	10	750	3575.267	1997.806
0.00385	20	700	7193.613	3700.245
0.00385	30	700	10045.030	4813.200
0.00385	40	700	12608.614	5804.244
0.00385	50	750	15142.640	6938.876
0.00385	60	1000	17180.127	7243.917
0.00385	70	1000	19097.047	7628.018
0.00385	80	500	20736.824	8399.472
0.00385	90	500	21392.036	7663.977
0.00385	100	1050	23596.121	8676.333
0.00400	20	200	4281.680	1321.136
0.00400	40	200	5394.570	1111.389
0.00400	60	200	5905.740	945.039
0.00400	80	600	6245.863	727.554
0.00400	100	350	6475.271	591.310

old behavior is essentially the same as the sudden phase change near a mean-field spinodal point, and we expect that the parameter dependence is the same as that near the spinodal point of the Ising ferromagnet with weak long-range interactions (the Husimi-Temperley model), with the irradiation intensity a playing the role of the applied magnetic field. Thus, the transition time for $a > a_{\text{SP}}$ should diverge as[26]

$$\tau \propto (a - a_{\text{SP}})^{-1/2}, \tag{5}$$

independent of L. At the spinodal point, the transition time diverges with the system size N as[27–29]

$$\tau \propto N^{1/3}. \tag{6}$$

These dependences have been obtained by several methods for the spinodal phenomenon in the Husimi-Temperley model,[27–29] most recently by a master-equation analysis.[30]

B. Finite-size scaling of the transition time

Equations (4)–(6) can be combined into a scaling form,[29,30]

$$\tau \propto L^{d/3} f[L^{2d/3}(a - a_{\text{SP}})], \tag{7}$$

where the scaling function $f(x)$ has the following asymptotic dependences on the scaling variable $x = L^{4/3}(a - a_{\text{SP}})$

$$f(x) \sim \begin{cases} \exp(|x|^{3/2}) & \text{for } x \ll -1 \\ x^0 & \text{for } |x| \ll 1 \\ x^{-1/2} & \text{for } x \gg 1 \end{cases}. \tag{8}$$

In Fig. 5, we plot the scaling function $f(x) = \tau / L^{2/3}$ on a logarithmic scale vs $|x|^{3/2}$. All the points collapse onto a scaling

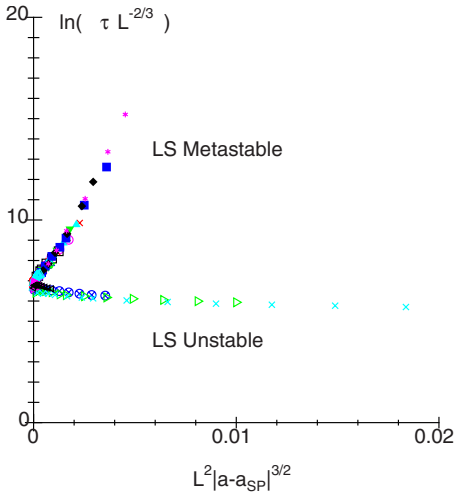

FIG. 5. (Color online) Plot of the scaling function for the transition time, shown as $\ln f(x)$ vs $|x|^{3/2} = L^2|a - a_{\text{SP}}|^{3/2}$. We adopt $a_{\text{SP}} = 0.00385$. The symbols denote data for the same values of a, and the labels have the same meanings as in Fig. 4.

form, which shows the asymptotic behavior given in Eq. (8). Thus we confirm that the models with long-range interactions mediated by local elastic distortions due to the spin dependence of the size of unit cell exhibit mean-field spinodal phenomena, in which the metastable state has an extremely long lifetime compared with short-range models.

As mentioned above, the size dependence of the standard deviation of the transition time is very different above and below the threshold as listed in Table I. Above a_{SP}, the transition-time distribution is narrowly localized, and the standard deviation decreases with system size. By contrast, far below a_{SP}, the distribution is exponential indicating a Poisson process, so that the standard deviation approximately equals the average. This difference can be depicted in a scaling plot. In Fig. 6 we show the ratio of the standard deviation to the average of the transition time vs the scaling variable x. The results collapse quite well onto a scaling function.

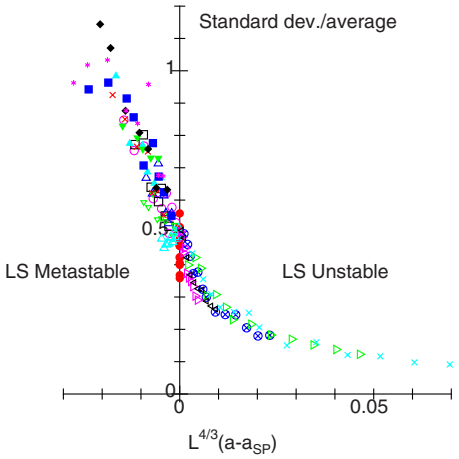

FIG. 6. (Color online) Scaling plot of the ratio of the standard deviation and the mean of the transition time with $x = L^{4/3}(a - a_{\text{SP}})$ and $a_{\text{SP}} = 0.00385$. The symbols denote data for the same values of a, and the labels have the same meanings as in Fig. 4.

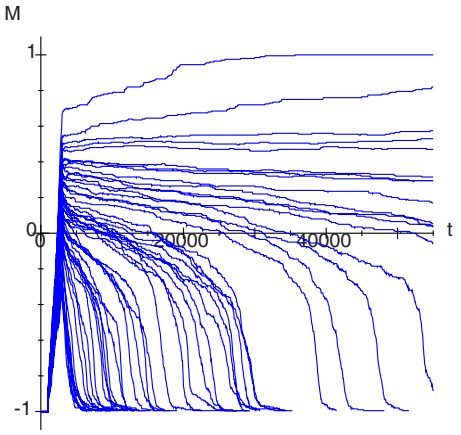

FIG. 7. (Color online) Time series of M for the model with $k=100$ subjected to a short irradiation pulse (50 samples). During the first 1000 MCSS, the system evolves without irradiation, after which an irradiation pulse with $a=0.002$ is applied for 2500 MCSS. After the pulse, the system again evolves without irradiation. A threshold value of M is seen, beyond which the system continues to evolve toward the HS phase after the end of the pulse.

C. Pulsed photoexcitation

So far, we have studied only the complete LS-to-HS switching process induced by photo-irradiation at a low temperature. However, if we stop the irradiation at an intermediate stage of the excitation process, we can observe further relaxation of the order parameter. The nature of this relaxation reflects the stability structure of the order parameter.[13] We found that for the case of $k_1=50$, all samples relaxed back to the LS state, even from a nearly fully saturated HS configuration (not shown). Thus we conclude that the HS state is unstable in the absence of irradiation in this case. By contrast, with a stronger coupling constant, $k_1=100$, we found a threshold value of the HS fraction around $M \simeq 0.4 (f_{\mathrm{HS}} \simeq 0.7)$ at a low temperature of $T=0.05$, as depicted in Fig. 7. Above this threshold, the HS fraction continues to increase after a has been returned to zero, while it decreases again below the threshold. This behavior indicates that the system has a bistable structure at this temperature for sufficiently strong elastic interactions.

We expect that a separation of trajectories as depicted in Fig. 7 can be observed in real experiments.

V. SUMMARY AND DISCUSSION

In this paper we have studied threshold phenomena in the excitation processes induced by photo-irradiation of a model spin-crossover material in which the size difference between the HS and LS states causes an effective long-range interaction among the molecules. We considered the dependences of the transition time on the system size and the irradiation strength and found that the lifetime of the metastable LS state increases exponentially with the number of molecules. This behavior contrasts with the case of short-range interaction models, in which local nucleation takes place, leading to an L^{-d} decrease of the lifetime with the system size. The observed finite-size dependences are explained by the existence of a sharp spinodal transition characteristic of a dynamic mean-field model. Thus, we have now confirmed that spin-crossover materials with elastic interactions show mean-field type behavior in both their static[18] and dynamic properties. We believe that this truly critical nonequilibrium phenomenon should be experimentally observable in many elastically coupled physical systems, including in SC materials exposed to photo-irradiation.[25]

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