Collective behavior in the kinetics and equilibrium of solid-state photoreaction
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There is current interest in developing photoactive materials that deform on illumination. The strategy is to develop new photoactive molecules in solution, and then to incorporate these in the solid-state either by crystalization or by inserting them into polymers. This letter shows that the kinetics and the nature of the photo-induced phase transitions are profoundly different in single molecules (solution) and in the solid state using a lattice spin model. In solution, where the molecules act independently, the photoreaction follows first-order kinetics. However, in the solid state where the photoactive molecules interact with each other and therefore behave collectively during reaction, photoreactions follow the sigmoidal kinetics of nucleation and growth as in a first-order phase transition. Further, we find that the exact nature of the photo-induced strain has a critical effect on the kinetics, equilibrium, and microstructure formation. These predictions agree qualitatively with experimental observations, and provide insights for the development of new photoactive materials.

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Some molecules can absorb light of a certain wavelength and change their shape, dissociate, or combine to form new molecules. These photoreactions (photo-isomerization, dimerization, etc.) enable applications including optical grating [1], image storage [2], surface alignment [3], wettability and permeability modulation [4,5], and photomechanical actuation [6–12]. Photomechanical actuation is particularly attractive because it can be effected at a distance, different frequencies can be used to actuate different modes or control an object, and corrosion-free lightweight fiber optic cables can deliver significant power over long distances. This has motivated the development of a number of new photoactive solids [10–15]. Still, promising molecules in solution do not always lead to effective photoactive solids, and this motivates the current work.

When embedded in a solid, the molecules cannot react independently, but interact collectively through the solid matrix. Despite the extensive studies on single-molecule photoreaction [16–18], studies on solid-state photoreaction are still based on either simple first-order reaction kinetics, or mean-field approaches such as the Johnson–Mehl–Avrami–Kolmogorov (JMAK) model [19–22] and the Finke–Watzky model [23]. The coarse-grained nature of these models limits their capability to explore the more detailed microstructure inside the material, and thus they do not fully explain various observations, such as the sigmoidal kinetics reported by recent experimental evidence [24–26], and the formation of specific domain patterns in the solid during the reaction [27–29].

In this letter, we address the above challenges in solid-state photoreaction by proposing a driven Ising lattice spin model with both nearest-neighbor and long-range intermolecular interactions where the evolution is driven by light-induced photoreaction in addition to the usual thermal relaxation. The light illumination pumps photon energy into the system while the thermal fluctuation drives a relaxation, and the system eventually reaches equilibrium or the photo-stationary state. Similar kinds of external driving have been studied in nonequilibrium friction and shear in driven Ising systems [30–32].

We start by describing the energy landscape of azobenzene (a representative photoactive molecule) at the single-molecule level inferred from first principles calculations with the corresponding reaction paths illustrated in Fig. 1a [16–18]. The electronic-ground-state energy landscape has a double-well structure, with two minima corresponding to the trans- and cis-isomers, trans being lower energy than cis. Thermal fluctuations exist between these two states following the usual rate kinetics. Also existing is a metastable electronic excited state that is accessed when the trans-isomer absorbs a photon. The molecule relaxes back to either the trans- or cis-state, with a probability determined by the ground-state energy landscape.

We incorporate the above single-molecule framework into a solid of many photoactive molecules by adopting a 2D periodic square lattice of \( n \times n \) elements extended periodically (analogous to spins in an Ising model), each taking a value \( S_i = +1 \) (cis) or \(-1\) (trans) (Fig. 1b).
We consider the following Hamiltonian for our Ising model:

$$H = -\frac{1}{2} \sum_{\langle \alpha, \beta \rangle} S_\alpha S_\beta + h \sum_{\alpha = 1}^{n^2} S_\alpha + EW(S_\alpha). \quad (1)$$

The first term represents the nearest-neighbor interaction that describes the weak intermolecular force such as the dispersion force between molecules ($\langle \alpha, \beta \rangle$ denotes all pairs of nearest neighbors, and $1/2$ accounts for the interchange of $\alpha$ and $\beta$). We take $J$ to be positive so that neighboring elements prefer the same state. This also gives rise to the interfacial energy between different phases. The second term describes the energetic preference of the $\text{trans}$-state with $h$ being positive. The third term describes the long-range elastic interaction. During a solid-state photoconversion, the shape change of molecules may induce non-negligible stress and strain fields in the solid. The transformation strain or photostrain, defined as the spontaneous strain of the material under no external stress, in one location affects stress field globally and interacts with a photo-strain at a distal location. Hence, photoconversion induces a long-range elastic interaction [33,34]. $EW(S_\alpha)$ in (1) is the total elastic energy in the system, where $E$ is the plane strain Young's modulus of the solid and $W(S_\alpha)$ ($\alpha = 1, ..., n^2$) is the dimensionless elastic energy that depends on $S_\alpha$ as described later.

The Hamiltonian in (1) describes a variety of materials: (i) a solution, uncrosslinked polymer melt, or diluted-crosslinked polymer network when $J/kT = E/kT = 0$, (ii) moderately crosslinked polymer when $J/kT \neq 0$ and $E/kT = 0$, and (iii) crystals of densely packed photoactive molecules when $J/kT$ small and $E/kT$ large.

To model photo-isomerization using this Hamiltonian, we adopt a Monte Carlo algorithm, where in each step we conduct a single-flip trial following one of two processes. In a photoactivation process, we flip a random element to the cis-state with a probability of $\min[1, \exp(-\Delta H/kT)]$ if it is initially trans, and do nothing otherwise. This corresponds to the reaction paths at the single-molecule energy landscape illustrated in Fig. 1a. In a thermal relaxation process, we follow the standard Metropolis importance sampling by flipping a random element with a probability of $\min[1, \exp(-\Delta H/kT)]$ [35,36]. $\Delta H$ in both cases denotes the change of total Hamiltonian upon flipping. We define the dimensionless light intensity $I$ as the ratio between the photoactivation and the thermal relaxation steps. For example, if we conduct photoactivation for $N$ steps and thermal relaxation for 1 step, then $I = N$.

With a time scale assigned to each step, the single-flip process of the Monte Carlo simulation can be interpreted kinetically by the Glauber kinetics of the Ising model [36,37], which allows us to explore the reaction kinetics. We record the evolution of $\text{cis}$-ratio with reaction time represented by the increasing MC (Monte Carlo) steps per element (the accumulated number of steps divided by $n^2$).

We conduct our simulations on a $100 \times 100$ periodic lattice ($n = 100$), and an initial condition of 0% $\text{cis}$-ratio. The total MC steps per element are 2000, large enough for the reaction to reach photo-stationary. The photo-stationary $\text{cis}$-ratio is calculated as the average of the last MC step per element ($10^4$ steps). While the results presented here are in two dimensions to manage the computational complexity, we expect to see qualitatively similar results in three dimensions.

We start without the elastic interaction ($E = 0$), and study the difference between photoreactions in solution ($J/kT = 0$) and moderately cross-linked solid ($J/kT > 0$) with fixed $h/kT = 1$ that seeks to drive the material to the $\text{trans}$-state. Fig. 2a plots the photo-stationary $\text{cis}$-ratio as a function of light intensity $I$. The photo-stationary state in solution ($J/kT = 0$) shows first-order reaction kinetics. When $I = 0$, the photo-stationary $\text{cis}$-ratio is finite due to thermal fluctuation since $J/kT = 0$, but is much below 50% due to the positive $h/kT$. The $\text{cis}$-ratio increases monotonically with $I$, linearly at small $I$, and approaches 100% exponentially at large $I$. The material remains completely disordered through the reaction as shown in Fig. 2b and Movie 1. The forward reaction rate which has contributions from both the photo activation and the thermal relaxation can be expressed as $(1 - \phi_{\text{cis}}) \exp(-2h/kT)(I + 1)\tau^{-1}$ where $\tau$ is the time constant of each step. The backward reaction rate is governed by thermal relaxation alone and is expressed as $\phi_{\text{cis}}\tau^{-1}$. Equilibrating the two rates leads to the photo-stationary $\text{cis}$-ratio $\phi_{\text{cis}} = 1 - [1 + (I + 1) \exp(-2h/kT)]^{-1}$. This analytical solution is plotted in Fig. 2a as the solid line, showing good agreement with the simulation.

In contrast to solution, the photo-stationary $\text{cis}$-ratio in solid ($J/kT > 0$) shows a clear discontinuity (Fig. 2a) indicating a first-order phase transition. This is accompanied by the nucleation and growth as shown in Fig. 2b and Movie 2. Recall that the system starts with 0% $\text{cis}$-ratio. When the incoming light intensity is small, the molecular interaction suppresses almost all forward reaction by either photo activation or thermal relaxation because neighboring elements energetically favor the same state. Indeed, the lack of any initial $\text{cis}$ creates a high energy barrier for nucleation to promote trans-to-$\text{cis}$ isomerization. Once the light intensity is large enough to overcome this energy barrier, $\text{cis}$-nuclei form from a sufficient amount of $\text{cis}$-molecules. Subsequently, the nuclei keep growing with an auto-catalytic mechanism, until it reaches about 100% $\text{cis}$-ratio. The molecular interaction between $\text{cis}$-molecules further suppresses almost all backward reaction to trans.

We now show that the trans-to-$\text{cis}$ transformation with increasing illumination in the solid is the manifestation of a first-order phase transition induced by external field. As noted in the case of solution, the single-molecule forward reaction rate is $(1 - \phi_{\text{cis}}) \exp(-2h/kT)(I + 1)\tau^{-1}$ which may be rewritten as

$$\phi_{\text{cis}} \exp\left(-\frac{2h - kT \log(I + 1)}{kT}\right) \tau^{-1}. \quad (2)$$

Therefore, the light-driven forward reaction is equivalent to adding an additional external potential $-((1/2) kT \log(I + 1)$ to the system. With this, the photoreaction-incorporated Hamiltonian is

$$H = -\frac{1}{2} \sum_{\langle \alpha, \beta \rangle} S_\alpha S_\beta + (h - (1/2) kT \log(I + 1)) \sum_{\alpha = 1}^{n^2} S_\alpha + EW(S_\alpha). \quad (3)$$

We conduct the same Monte Carlo simulation using this modified Hamiltonian with only thermal relaxation steps. The results open
Fig. 2. Comparison between photoreactions in solution and solid, with fixed $h/kT = 1$. (a) The photo-stationary cis-ratio vs. light intensity. Filled circles/squares: original Hamiltonian (1). Open circles/squares: Photoreaction-incorporated Hamiltonian (3). Black solid line: analytical solution $\phi_{\text{cis}} = 1 - [1 + (I + 1) \exp(-2h/kT)]^{-1}$. (b) Typical microstructure during photoreaction. Black square: cis. White square: trans. Also see Movies 1&2. (c, d) cis-ratio vs. time with fixed light intensity as $I = 7$ on logarithmic time (c) and logarithmic cis-ratio (d) scale.

dots in Fig. 2a) overlap perfectly with those from the original algorithm. Further, note that (3) is the classical Ising Hamiltonian when $E = 0$, and this undergoes a first-order phase transition induced by external field below the critical temperature which in our case is rescaled by illumination.

To further probe the effect of nucleation and growth, we compare the reaction kinetics. Fig. 2c and d plot the evolution using logarithmic scale on time in 2c and on cis-ratio in 2d. In solution, the cis-ratio grows following the first-order kinetics with a constant relaxation, reaching a finite photo-stationary plateau. In contrast, in solid, we observe initial fast exponential relaxation to a metastable state (inset of Fig. 2d), followed by a significantly slower sigmoidal kinetics of nucleation and growth. We attribute this slow kinetics to the lack of cis-nuclei initially in the system. Due to the single-flip nature of the simulation, forming an effective nucleus takes a long time. Once large enough cis-nuclei form, the system quickly goes through the auto-catalytic reaction towards the final photo-stationary state with nearly 100% cis-ratio. This sigmoidal kinetics in solid-state photoreaction has been experimentally observed recently in various material systems [24–26].

In addition to the sigmoidal kinetics, recent experiments on solid-state photo-dimerization also show that the reaction kinetics depends significantly on reaction history, while the photo-stationary state only depends on the final light intensity [26]. We are unaware of any existing model that is capable of capturing this path-dependent reaction kinetics. Fig. 3a shows the reaction kinetics (trans-ratio vs. time) under various light intensities and reaction histories. In one group of simulation (solid lines in Fig. 3a), we first apply a light with intensity $I = +\infty$ until reaching 5% trans-ratio, and then apply a weak light. In the other group (dashed lines in Fig. 3a), we directly apply the corresponding weak light. For each different weak light intensity, it is observed that both reaction paths (strong-weak vs. directly weak) lead to the same photo-stationary state. The photo-stationary trans-ratio only depends on the final weak light intensity. However, the reaction kinetics greatly depends on the reaction history. For instance, comparing the two curves at $I = 1/5$, the photo-stationary state requires much longer time under a weak light alone compared to the strong-weak illumination. In the strong-weak illumination, one has both phases in coexistence and therefore does not need an initial nucleation event. Under weak illumination alone, the need for the initial nucleation leads to slower kinetics.

We then slightly increase $J/kT$ while keeping other conditions the same, and the results are shown in Fig. 3b. It is observed that all the photo-stationary states reach close to either 0% or 100% trans-ratio due to the stronger interaction. Further a bifurcation is observed for the intermediate illumination $I = 1/5$ where the photo-stationary state is nearly all trans for strong-weak illumination but nearly all cis for weak illumination. This is again due to the stronger intermolecular interaction that suppresses nucleation.

We now turn to the case with elastic interactions ($E > 0$). Each element $\alpha$ located at $x^\alpha$ is assigned with a photo-strain $\varepsilon_{\text{ps}}^{\alpha}$. We choose two specific types of photo-strain, along with their corresponding dimensionless total elastic energies (assuming zero average stress) $\bar{W}(S_{\alpha})$ as (see Supplementary Information for the
Shear:

\[
\epsilon_{ij}^{\text{trans}} = S_n \text{diag} \left( \begin{array}{cc} 1 & -1 \\ \end{array} \right), \\
W(S_n) = \frac{1}{2n^2} \sum_{k=0}^{n} \frac{(k_1^2 - k_2^2)^2}{k^4} \left| \hat{S}(k) \right|^2.
\]

Volumetric:

\[
\epsilon_{ij}^{\text{trans}} = S_n \text{diag} \left( \begin{array}{cc} 1 & 1 \\ \end{array} \right), \\
W(S_n) = \frac{n^2}{2} \left( \left( \langle S_n^2 \rangle \right)^2 - \langle S_n \rangle^2 \right).
\]

\[\hat{S}(k) = \sum_{a=1}^{n^2} S_n \exp(-i(k \cdot x^a)), \]

where \(k\) is the coordinate in the Fourier space.

**Fig. 3.** Photo-reaction kinetics under different reaction histories and intermolecular interactions \(J/kT\). \(hJ = 0.2\) is fixed in both figures. (a) \(J/kT = 0.4\). (b) \(J/kT = 0.45\). All simulations start from 100% Photoreaction kinetics under different reaction histories and intermolecular interactions Fig. 3. R. Bai, Y.S. Teh and K. Bhattacharya Extreme Mechanics Letters 43 (2021) 101160

\[\epsilon^+ - \epsilon^- = \frac{1}{2} (a \otimes n + n \otimes a). \]

With shear photo-strain, neighboring domains with different shear can coexist with zero stress if the boundary between them has a normal \(n = (\pm 1,1)^T\). Therefore, the formation of stripe or tweed domains can ameliorate the elastic energy as in first-order phase transition in martensites [40–42]. This allows for first-order kinetics for small \(E/kT\) and small \(J/kT\) and a smaller critical illumination at large \(E/kT\). In contrast, with volumetric photo-strain, there are no vectors \(a\) and \(n\) that enable us to solve (7). Therefore, the elastic energy cannot be ameliorated and this impedes nucleation when \(E/kT > 0\). In both cases, the nearest-neighbor interaction \(J\) acts as the effective interfacial energy between neighboring domains. A larger \(J/kT\) thus induces a larger length scale of the initial domain size, leading to the coarsening of domains. This collective behavior of domain pattern formation in solids has been observed experimentally in azobenzene self-assembly monolayers [28,29]. The results here are also consistent with the observation in molecular crystals of photo-active molecules: some so-called photo-salient crystals shatter when illuminated [43] while others undergo photo-actuation without shattering [44]. The difference has been shown to be consistent with the ability of the photo-strain to satisfy the Hadamard jump condition [45].

In addition to the long-range elastic interaction, many photo-active molecules interact under the long-range electrostatic...
dipole–dipole interaction. For example, an azobenzene isomer can be modified with additional functional groups to carry a permanent dipole moment [29]. The energy due to the electrostatic dipole–dipole interaction can be calculated following the same methodology proposed in our current model. In that case, the domain formation is expected to depend additionally on the distribution of dipole moments.

In summary, we have postulated an Ising-like lattice with Monte Carlo simulation to explore the collective behavior in solid-state photoreaction. Unlike photoreactions in solution following a first-order kinetics, photoreaction in solid shows a feature analogous to first-order phase transition: a sigmoidal kinetics of nucleation and growth. This leads to a bifurcation in the photo-stationary state. The photo-strain upon reaction significantly affects the kinetics, equilibrium, and formation of microstructure through the long-range elastic interaction. These predictions agree qualitatively well with recent experimental results. This study is hoped to help the future design and optimization of solid-based photomechanical systems.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.eml.2020.101160.

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