

## Exactly Solvable Model of Monomer-Monomer Reactions on a Two-Dimensional Random Catalytic Substrate

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We study the equilibrium properties of a monomer-monomer  $A + B \rightarrow \emptyset$  reaction on a two-dimensional substrate containing randomly placed catalytic bonds. Interacting  $A$  and  $B$  species undergo continuous exchanges with particle reservoirs and react as soon as a pair of unlike particles appears on sites connected by a catalytic bond. For *annealed* disorder in the placement of catalytic bonds the model is mapped onto a general spin  $S = 1$  model and solved exactly for the pressure in a particular case. At equal activities of the two species a second order phase transition is revealed.

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Catalytically activated reactions (CARs) involve particles that react only in the presence of another agent—a catalyst—and remain chemically inactive otherwise. These processes are widespread in nature and used in a variety of technological and industrial applications [1].

The work of Ziff, Gulari, and Barshad [2] on the “monomer-dimer” model, introduced as an idealized description of the important process of CO oxidation on a catalytic surface, as well as subsequent studies of a simpler “monomer-monomer” reaction model [3,4], has been an important step in the understanding of CARs properties by revealing several remarkable features [2]. On a two-dimensional (2D) substrate, upon lowering the CO adsorption rate the system undergoes a first-order phase transition from a CO saturated inactive phase into a reactive steady state, followed by a continuous transition into an O<sub>2</sub>-saturated inactive phase, which belongs to the same universality class as directed percolation and the Reggeon field theory [5]. The monomer-monomer model exhibits a first-order transition from a phase saturated with one species to one saturated with the other; allowing desorption of one species leads to a continuous transition that also belongs to the directed percolation universality class [4]. For these two models, different aspects of the dynamics of the adsorbed phase have been investigated [2–7], confirming an essentially collective behavior. In contrast, the equilibrium properties of CARs are much less studied, and the understanding of the equilibrium state remains rather limited. Recently, an exact solution for a one-dimensional monomer-monomer model has been presented [8], but for the physically important 2D substrates no exact solutions are known as yet.

Realistic substrates are typically disordered and the actual catalyst is an assembly of mobile or localized catalytic sites or islands [1]. Theoretical studies that have addressed the behavior of CARs on disordered substrates have been focused so far on the effect of site-dependent adsorption and desorption rates because natu-

ral catalysts are, in general, energetically heterogeneous [9,10]. In contrast, the behavior on inert substrates that are decorated by catalytic particles has been much less studied; this case is relevant for artificially designed catalysts [11]. With the exception of a few exactly solvable 1D models of  $A + A \rightarrow \emptyset$  reactions [12] and a Smoluchowski-type analysis of  $d$ -dimensional CARs [13], for random spatial distributions of the catalyst only phenomenological generalizations of the mean-field “law of mass action” have been proposed so far [1]. Consequently, an exact analytical solution of (albeit idealized) models involving a 2D random catalytic substrate is very desirable since it provides valuable insight into the effects of disorder on the CARs properties.

In the following we present such an *exactly solvable* model of a monomer-monomer  $A + B \rightarrow \emptyset$  reaction on a 2D inhomogeneous, catalytic substrate and study the equilibrium properties of the two-species adsorbate. The substrate contains randomly placed catalytic bonds of mean density  $q$ , which connect neighboring adsorption sites. The interacting  $A$  and  $B$  (monomer) species undergo continuous exchanges with corresponding adjacent gaseous reservoirs. A reaction  $A + B \rightarrow \emptyset$  takes place instantaneously if  $A$  and  $B$  particles occupy adsorption sites connected by a catalytic bond. We find that for the case of *annealed* disorder in the placement of the catalytic bonds the reaction model under study can be mapped onto the general spin  $S = 1$  (GS1) model [14]. This allows us to exploit the large number of results obtained for the GS1 model [14] in order to elucidate the equilibrium properties of the monomer-monomer reaction on random catalytic substrates [15]. Here we concentrate on a particular case in which the model reduces to an exactly solvable Blume-Emery-Griffiths (BEG) model [16,17] and derive an exact expression for the disorder-averaged equilibrium pressure of the two-species adsorbate. We show that at equal partial vapor pressures of the  $A$  and  $B$  species this system exhibits a second-order phase transition which

reflects a spontaneous symmetry breaking with large fluctuations and progressive coverage of the entire substrate by either one of the species.

We consider a 2D regular lattice of  $N$  adsorption sites (Fig. 1), which is in contact with the mixed vapor phase of  $A$  and  $B$  particles. The  $A$  and  $B$  particles can adsorb onto *vacant* sites and can desorb back to the reservoir. They are characterized by chemical potentials  $\mu_A$  and  $\mu_B$  maintained at constant values and measured relative to the binding energy of an occupied site, so that  $\mu_{A,B} > 0$  corresponds to a preference for adsorption. The  $A$  and  $B$  particles have hard cores prohibiting double occupancy of sites and nearest-neighbor (NN) attractive  $A-A$ ,  $B-B$ , and  $A-B$  interactions of strengths  $J_A$ ,  $J_B$ , and  $J_{AB}$ , respectively. The occupation of the  $i$ th site is described by a pair  $c_i$  of Boolean variables  $n_i$  and  $m_i$  such that

$$c_i \equiv (n_i, m_i) = \begin{cases} (1, 0), & \text{site } i \text{ occupied by } A, \\ (0, 1), & \text{site } i \text{ occupied by } B, \\ (0, 0), & \text{site } i \text{ empty,} \\ (1, 1), & \text{excluded by hard cores.} \end{cases} \quad (1)$$

We assign to some of the lattice bonds (solid lines in Fig. 1) “catalytic” properties such that if an  $A$  and a  $B$  particle occupy simultaneously NN sites connected by such a catalytic bond, they instantaneously react and desorb, and the product ( $AB$ ) leaves the system;  $A$  and  $B$  particles occupying NN sites not connected by a catalytic bond harmlessly coexist, and we assume that the reverse process of a simultaneous adsorption of an  $A$  and a  $B$  on a catalytic bond has an extremely low probability and can be neglected. The “catalytic” character of the lattice bonds is described by Boolean variables  $\zeta_{\langle ij \rangle}$ , where  $\langle ij \rangle$  denotes a pair of neighboring sites  $i$  and  $j$ ,

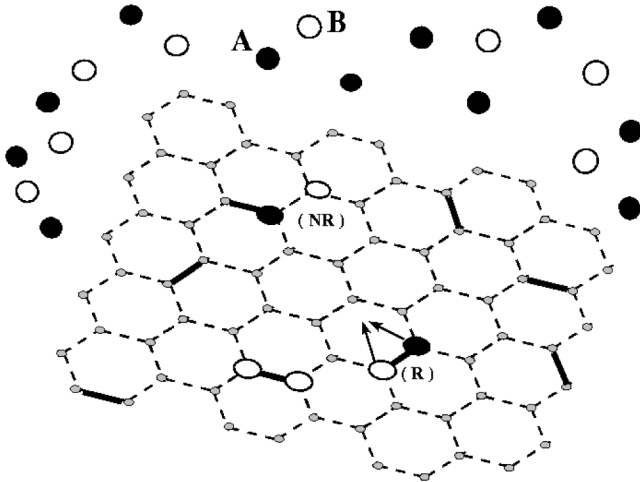


FIG. 1. A 2D lattice of adsorption sites (small gray circles) in contact with a mixed vapor phase. Black and white circles denote  $A$  and  $B$  particles, respectively. The solid lines denote “catalytic bonds.” (R): configuration in which an instantaneous reaction takes place ( $\nearrow$ ) upon which the reactants leave the system. (NR): NN pair of  $A$  and  $B$  that do not react since the sites are not connected by a catalytic bond.

$$\zeta_{\langle ij \rangle} = \begin{cases} 1, & \langle ij \rangle \text{ is a catalytic bond,} \\ 0, & \text{otherwise,} \end{cases} \quad (2)$$

and we take  $\{\zeta_{\langle ij \rangle}\}$  as independent, identically distributed random variables with the distribution

$$\rho(\zeta) = q\delta(\zeta - 1) + (1 - q)\delta(\zeta). \quad (3)$$

The probability  $q$  that a given bond is catalytic equals the mean density of the catalytic bonds. The two limiting cases,  $q = 0$  and  $q = 1$ , correspond to an *inert* substrate and to a *homogeneous catalytic* one, respectively.

The condition of instantaneous reaction  $A + B \rightarrow \emptyset$  together with negligible simultaneous adsorption of an  $A$  and a  $B$  on a catalytic bond is formally equivalent to allowing a NN  $A-B$  repulsive interaction of strength  $\lambda \gg 1$ , followed by the limit  $\lambda \rightarrow \infty$ , for  $A-B$  pairs connected by catalytic bonds. Hence, in thermal equilibrium and for a given configuration  $\zeta \equiv \{\zeta_{\langle ij \rangle}\}$ , the partition function of such a two-species adsorbate is

$$Z_N(\zeta) = \lim_{\lambda \rightarrow \infty} \sum_{\{c_k\}} \exp[-\beta \mathcal{H}_\lambda(\zeta)], \quad (4)$$

where  $\beta^{-1} = k_B T$  is the thermal energy, while the Hamiltonian  $\mathcal{H}_\lambda(\zeta) = H_\lambda(\zeta) + H_0$  naturally separates into a disorder-dependent part,

$$H_\lambda(\zeta) = \lambda \sum_{\langle ij \rangle} \zeta_{\langle ij \rangle} (n_i m_j + n_j m_i), \quad (5)$$

where the summation extends over all pairs  $\langle ij \rangle$ , and a disorder-independent contribution

$$H_0 = - \sum_{\langle ij \rangle} [J_A n_i n_j + J_B m_i m_j + J_{AB} (n_i m_j + n_j m_i)] - \sum_{i=1}^N (\mu_A n_i + \mu_B m_i). \quad (6)$$

In what follows we shall focus on situations in which the disorder in the placement of the catalytic bonds is *annealed*. In this case the thermodynamics of the system is given by the disorder-averaged pressure (in units of the lattice cell area),

$$P \equiv P(T, \mu_A, \mu_B) = \frac{1}{\beta} \lim_{N \rightarrow \infty} \frac{1}{N} \ln \langle Z_N(\zeta) \rangle_\zeta, \quad (7)$$

where  $\langle \cdots \rangle_\zeta$  denotes the average over all possible realizations  $\{\zeta_{\langle ij \rangle}\}$ . Once  $P$  is known all other thermodynamic quantities of interest can be obtained by differentiating  $P$  with respect to  $\mu_A$ ,  $\mu_B$ , or  $T$ .

Averaging  $Z_N(\zeta)$  in Eq. (4) is straightforward,

$$\begin{aligned} \langle Z_N(\zeta) \rangle_\zeta &= \sum_{\{c_k\}} e^{-\beta H_0} \lim_{\lambda \rightarrow \infty} \langle e^{-\beta H_\lambda(\zeta)} \rangle_\zeta \\ &= \sum_{\{c_k\}} e^{-\beta H_0} \prod_{\langle ij \rangle} \lim_{\lambda \rightarrow \infty} [q e^{-\lambda \beta (n_i m_j + n_j m_i)} + 1 - q] \\ &= \sum_{\{c_k\}} e^{-\beta H_0} \prod_{\langle ij \rangle} (1 - q)^{n_i m_j + n_j m_i} = \sum_{\{c_k\}} e^{-\beta H_e}, \end{aligned} \quad (8)$$

and yields the “effective” Hamiltonian

$$\mathcal{H}_e = - \sum_{\langle ij \rangle} \{ [J_{AB} + \beta^{-1} \ln(1-q)](n_i m_j + n_j m_i) + J_A n_i n_j + J_B m_i m_j \} - \sum_{i=1}^N (\mu_A n_i + \mu_B m_i). \quad (9)$$

Introducing the “spin” variables  $\sigma_i \in \{0, \pm 1\}$ ,

$$\sigma_i = \begin{cases} +(-)1, & \text{site } i \text{ occupied by } A \text{ (} B \text{),} \\ 0, & \text{site } i \text{ empty,} \end{cases} \quad (10)$$

such that  $n_i = (\sigma_i + \sigma_i^2)/2$  and  $m_i = (-\sigma_i + \sigma_i^2)/2$ ,  $\mathcal{H}_e$  can be cast into the form of the classical Hamiltonian of the general spin  $S = 1$  model [14],

$$\mathcal{H}_e = -J \sum_{\langle ij \rangle} \sigma_i \sigma_j - K \sum_{\langle ij \rangle} \sigma_i^2 \sigma_j^2 - C \sum_{\langle ij \rangle} (\sigma_i \sigma_j^2 + \sigma_j \sigma_i^2) - H \sum_{i=1}^N \sigma_i + \Delta \sum_{i=1}^N \sigma_i^2 \quad (11)$$

with coupling constants

$$J = \frac{J_A + J_B - 2J_{AB}}{4} - \frac{\ln(1-q)}{2\beta}, \quad (12a)$$

$$K = \frac{J_A + J_B - 2J_{AB}}{4} + \frac{\ln(1-q)}{2\beta}, \quad (12b)$$

$$C = \frac{J_A - J_B}{4}, \quad H = \frac{\mu_A - \mu_B}{2}, \quad \Delta = -\frac{\mu_A + \mu_B}{2}. \quad (12c)$$

Thus, in the case of annealed disorder, the  $A + B \rightarrow \emptyset$  reaction model under study can be mapped exactly onto the GSI model, which has been extensively analyzed [14]. The accumulated knowledge on its critical behavior, phase diagrams, as well as low- and high-temperature expansions [14], can be straightforwardly used to elucidate the equilibrium properties of the present CAR model for general values of  $\mu_A$ ,  $\mu_B$ ,  $J_A$ ,  $J_B$ ,  $J_{AB}$ , and  $q$ , as well as for different types of embedding lattices [15].

In the remaining part of this Letter we focus on the symmetric case  $\mu_A = \mu_B$  and  $J_A = J_B$ , implying  $C = H = 0$  so that the model reduces to the original BEG model [16]. Additionally, we set  $J_{AB} = 0$  and consider a honeycomb lattice and a particular relation between  $K$  and  $J$ , for which the 2D BEG model, and hence the monomer-monomer reaction model under study, can be solved *exactly* [17,18].

Following Ref. [18], in the subspace  $e^{-\beta K} = \cosh(\beta J)$  the partition function of the 2D BEG model on the honeycomb lattice may be expressed in terms of the partition function of a zero-field Ising model on the honeycomb lattice, which is known in closed form [19]. In this subspace the 2D BEG model exhibits an Ising-type phase

transition with a line of critical points obeying  $\tanh(\beta J) = (2 + e^{\beta \Delta})/2\sqrt{3}$  [17]. For our model,  $e^{-\beta K} = \cosh(\beta J)$  implies

$$J_A = J_B = \beta^{-1} \ln(1+q). \quad (13)$$

In the subspace defined by Eq. (13) the disorder-averaged pressure [Eq. (7)] is given *exactly* by

$$\beta P = \frac{1}{2}(1 + 2e^{\beta \mu}) - \frac{3}{2} \cosh(\beta J) - f(J), \quad (14)$$

where  $\mu = \mu_A = \mu_B$ ,  $\beta J = \tanh^{-1}[2q/(2 + e^{-\beta \mu})]$ , and  $f(J)$  denotes the known free energy of a spin-1/2 Ising model with NN interaction  $J$  on a honeycomb lattice (see Ref. [19]). Since in this particular case the exact expression for the average “magnetization”  $M_0 = \langle \sigma_i \rangle$  is also known [20], the average densities of  $A$  and  $B$  species are straightforwardly calculated as  $\langle n_i \rangle = (M_0 + \langle \sigma_i^2 \rangle)/2$  and  $\langle m_i \rangle = (-M_0 + \langle \sigma_i^2 \rangle)/2$ , where  $\langle \sigma_i^2 \rangle = z \partial_z (\beta P)$  and  $z = e^{\beta \mu}$ . Furthermore, the line of critical points as a function of  $q$  [within the subspace defined by Eq. (13)] where a continuous transition takes place is given by

$$\beta \mu_c = -\ln[2(q\sqrt{3} - 1)]. \quad (15)$$

We now emphasize several features of these results. (i) For  $\mu$  below its critical value [Eq. (15)] we find  $\langle n_i \rangle = \langle m_i \rangle \geq 0$  (see also Fig. 2). Upon exceeding  $\mu_c$  (by increasing the vapor pressure in the reservoirs) one of the densities (with equal probability) decreases sharply but *continuously* to zero while the other one rapidly attains

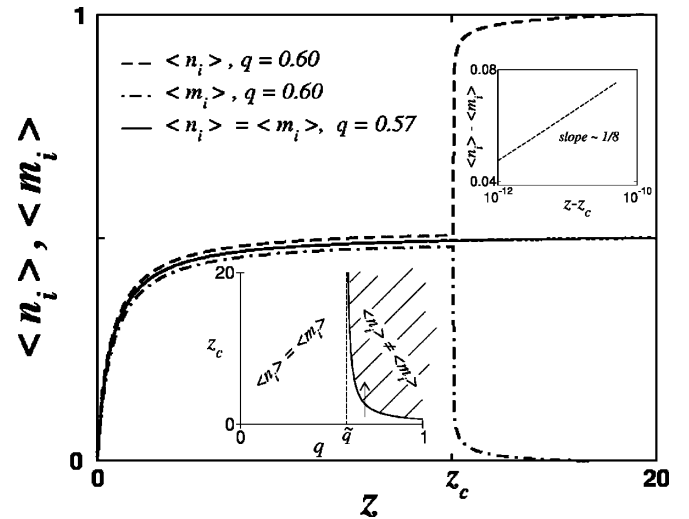


FIG. 2. Average density of  $A$  particles,  $\langle n_i \rangle$ , and of  $B$  particles,  $\langle m_i \rangle$ , as function of their common fugacity  $z$  above ( $q = 0.60$ ) and below ( $q = 0.57$ ) the threshold value  $\tilde{q} = 1/\sqrt{3}$  for the concentration of catalytic segments. For clarity, the curves corresponding to  $q = 0.60$  have been symmetrically shifted up ( $\langle n_i \rangle$ ) and down ( $\langle m_i \rangle$ ), respectively. The upper inset (logarithmic scale) illustrates the scaling behavior  $\langle n_i \rangle - \langle m_i \rangle \sim (z - z_c)^{1/8}$  for  $z \rightarrow z_c$ . The lower inset shows the critical line  $z_c(q)$  [Eq. (15)]; the arrow indicates a path of increasing  $z$  at fixed  $q$  which crosses the transition line.

unity. This reveals a spontaneous symmetry breaking and implies that the substrate becomes poisoned; i.e., most of it is covered by either one of the species. If the chemical potentials  $\mu_A$  and  $\mu_B$  differ slightly, the transition to the poisoned state is smeared out but remains detectable. (ii) The transition can occur only if the mean density  $q$  of catalytic bonds  $q$  is sufficiently high, such that  $q > \tilde{q} = 1/\sqrt{3} \approx 0.577$ . For  $q < \tilde{q}$ ,  $\langle n_i \rangle = \langle m_i \rangle$  for all  $\beta\mu$ , and both tend to  $1/2$  as  $\beta\mu \rightarrow \infty$ . (iii)  $\mu_c \geq 0$  for  $q \in [1/\sqrt{3}, \sqrt{3}/2]$ , which means that in this range of  $q$  values the transition occurs in situations in which adsorption on the substrate is favored. For  $q \in (\sqrt{3}/2, 1]$ ,  $\mu_c < 0$ , and hence the transition takes place for the case that desorption into the reservoir is favored. (iv) There occur large scale critical fluctuations of the densities of adsorbed  $A$  and  $B$  particles upon approaching  $z_c = \exp(\beta\mu_c)$  from above or below (by varying the vapor pressure in the reservoirs), and the compressibility of the adsorbed phase diverges as  $|z - z_c|^{-7/4}$  for  $z \rightarrow z_c$  and  $\mu_A = \mu_B$ , and as  $|\mu_A - \mu_B|^{-14/15}$  for  $\mu_A \rightarrow \mu_B$  and  $z = z_c$  [21]. (v) An analysis of the model on a Bethe lattice (coordination number  $\gamma = 3$ ) [15] shows that the case  $e^{\beta K} \cosh(\beta J) = 1$  is not singular, i.e., the transition line discussed above persists for  $e^{\beta K} \cosh(\beta J) \leq 1$  and for  $1 < e^{\beta K} \cosh(\beta J) < 2$ .

In conclusion, this study presents an *exactly solvable* model of a monomer-monomer  $A + B \rightarrow \emptyset$  reaction on a 2D random catalytic substrate. This exact solution has been obtained via a mapping of the partition function of the two-species adsorbate onto the partition function of a general spin  $S = 1$  model and by noticing that for certain relations between the corresponding coupling constants the latter reduces to an exactly solvable 2D BEG model [17]. In this case we have determined the annealed disorder-averaged equilibrium pressure of the two-species adsorbate and have shown that the system under study exhibits a second-order (robust in parameter space) 2D Ising-like phase transition if the mean density of the catalyst is sufficiently large.

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