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# Collective dynamics of interacting Ising spins: Exact results for the Bethe lattice

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We study the low temperature dynamics in films made of molecular magnets, i. e. crystals composed of molecules having large electronic spin  $S$  in their ground state. The electronic spin dynamics is mediated by coupling to a nuclear spin bath; this coupling allows transitions for a small fraction of electronic spins between their two energy minima,  $S^z = \pm S$ , under resonant conditions when the change of the Zeeman energy in magnetic dipolar field of other electronic spins is compensated by interaction with nuclear spins. Transitions of resonant spins can result in opening or closing resonances in their neighbors leading to the collective dynamics at sufficiently large density  $P_0$  of resonant spins. We formulate and solve the equivalent dynamic percolation problem for the Bethe lattice (BL) of spins interacting with  $z$  neighbors and find that depending on the density of resonant spins  $P_0$  and the number of neighbors  $z$  the system has either one ( $2 < z < 6$ ) or two ( $z \geq 6$ ) kinetic transitions at  $P_0 = P_{c1} \approx e^{-1/3}/(3z)$  and  $P_0 = P_{c2} \approx e^{-1}/z$ . The former transition is continuous and associated with the formation of an infinite cluster of coupled resonant spins similarly to the static percolation transition occurring at  $P_0 \approx 1/z$ . The latter transition,  $z > 5$ , is discontinuous and associated with the instantaneous increase in the density of resonant spins from the small value  $\sim 1/z$  to near unity. Experimental implications of our results are discussed.

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## I. INTRODUCTION

Percolation theory describes the flow in heterogeneous media. It is successfully applied to a variety of physical, chemical, biological and even social processes ranging from the hopping conductivity in doped semiconductors<sup>1</sup> to the evolution of large genetic networks and self-organized criticality<sup>2,3</sup>. For instance, the low temperature conductivity of hopping insulators can be modeled by the equivalent network of resistances replacing elementary electron hopping events<sup>1</sup>. The percolation theory finds the optimum set of interconnected open channels characterized by resistances not exceeding a certain maximum and forming an infinite cluster to guide electrons through the sample.

Open percolation channels are usually treated as stationary in time. It is relatively easy to study the static case numerically. Moreover there exist analytical solutions including percolation on the Bethe lattice (BL) and in low-dimensional systems<sup>4,5,6,7,8</sup>. In all cases the percolation kinetic transition is found to be continuous. System parameters including, for instance, diffusion coefficient, correlation radius and average size of percolating clusters all show scaling behavior near the critical point similarly to the one for second order phase transitions.

The model of static percolation is often an approximation. Flowing particles interact with each other and their motion can open or close percolation channels leading to, for instance, conductivity noise<sup>9</sup>. The significance

of fluctuations for the cooperative dynamics of quantum defects, spins in spin glasses, protons in ionic conductors and electrons in hopping insulators was pointed out in Refs.<sup>10,11,12,13,14,15</sup>. In a more refined analysis of percolation one should include the possibility that the flows through open channels can open or close other channels. These dynamic correlations can affect kinetic transitions in a fundamental way and lead to the cooperative dynamics, e. g. percolation may occur at smaller density of open channels and become discontinuous. In this paper we suggest a simple model of dynamic percolation for the low-temperature kinetics of the system of interacting magnetic molecules and obtain an exact description of the kinetic transitions on BL. According to the arguments of Refs.<sup>16,17</sup> the use of BL is more justified in our case than for the static percolation because of the similarity in the phase space structure for Bethe lattice and many-body system of interacting spins. In both cases the size of the phase space grows exponentially with the system size ( $2^N$  for  $N$  spins and  $z(z-1)^{N-1}$  for the number of nodes in BL with the coordination number  $z$  and  $N$  shells of the tree).

In the present work we study the dynamic percolation model on the Bethe lattice and then project our results on the electronic spin dynamics in films made of magnetic molecules such as  $Mn_{12}$ ,  $Fe_8$ , etc. These molecules are usually composed of transition metals, forming the magnetic core, coupled to various organic ligands<sup>18</sup>. They possess large electronic spin  $S$  in the ground state (both

$Mn_{12}$  and  $Fe_8$  molecules are characterized by the ground state “central spin”  $S = 10$ ). At low temperatures only the two lowest states of each molecule, characterized by the spin projections  $S_i^z = \pm S$  to the easy axis  $z$ , are occupied and each molecule can be modeled as a two-level system with two states corresponding to the Ising pseudo-spin 1/2. The tunneling gap  $2\Delta_o$  between the two lowest spin states in zero external transverse field is tiny,  $\sim 10^{-11} - 10^{-7}$  K. It is much smaller than other relevant parameters including the strength of the dipolar interaction between the nearest-neighbor spins,  $U_D \sim 0.1$  K, and the half-width of the distributions of the nuclear biases,  $E_o \sim 10^{-2} - 10^{-3}$  K. Under these conditions only spins exposed to a total longitudinal bias (external field plus internal dipolar demagnetization field) smaller than  $E_o$  can efficiently transfer between their two states due to energy exchange with the nuclear spin bath<sup>20,21,22</sup>. These spins are the resonant, or “open”, ones. At the same time, since  $U_o \geq E_o$ , each spin flip affects all its neighbors and may create new resonances (or destroy existing ones, see Sec. II). The associated collective dynamics of spins is studied within the dynamic percolation model in Secs. III, IV (see Fig. 1).

To be more specific, one may be interested in the correlation function  $C(t) = N^{-1} \sum_i \langle S_i^z(t) S_i^z(0) \rangle$  where  $N$  is the total number of spins, and the average is taken over initial conditions, and evolution histories. An interesting quantity to investigate in the infinite time limit would be the fraction of spin which never change their magnetization  $W_*$ . This fraction can be easily seen to result in the finite value of  $C(t \rightarrow \infty)$  since dynamically frozen spins contribute unity to the sum above. The other question concerns spin diffusion and the possibility to transfer energy and magnetization over large distances in which case one should be looking for the onset of percolating clusters of mobile spins.

The effect of dynamic and percolation transitions on spin relaxation in films of magnetic molecules, based on examples of  $Mn_{12}$  and  $Fe_8$ , is discussed in Sec. V. Our goal here is to investigate the case of completely demagnetized samples at relatively high temperatures  $k_B T > U_D$  to avoid the “dipolar ordering” effects and related sample-geometry dependent demagnetization fields. In this limit the resonant spins are nearly uniformly and randomly distributed in the system bulk contrary to the case of strongly polarized samples where resonant spins form spatially ordered structures, the “resonant surfaces”, and initial conditions play an important role in the system kinetics<sup>22</sup>.

We predict the reduction of the relaxation rate by many orders of magnitude in the vicinity of the transition point. At the same time the abrupt change of the relaxation rate is smeared out by its continuous dependence on the total longitudinal bias, which may obscure the experimental observation of the transition point.

## II. COLLECTIVE DYNAMICS OF INTERACTING SPINS

Since the model of spin dynamics is already formulated in great detail in Refs.<sup>20,21,22</sup>, here we briefly outline its main features and then introduce its Bethe Lattice version. As it was mentioned in the Introduction, the weakness of spin tunneling in molecular magnets allows an accurate representation of the system in terms of Ising pseudospins 1/2 coupled to each other by the long-range magnetic dipolar interaction  $U_{ij} S_i^z S_j^z$  with

$$U_{ij} = U_D a^3 \frac{1 - 3n_{ij}^2}{R_{ij}^3}, \quad n_{ij} = \frac{\mathbf{R}_{ij}}{R_{ij}}, \quad (1)$$

where  $U_D$  is the strength of the dipole-dipole interaction of spins;  $a$  is the lattice constant; and  $\mathbf{R}_{ij}$  is the vector connecting molecules  $i$  and  $j$ . For the sake of simplicity we limit our consideration to the case of 2D square lattice. In this model each spin  $\mathbf{S}_i$  is subjected to the “molecular” field of all other spins

$$U_i = \sum_j U_{ij} S_j^z \quad (2)$$

and the external magnetic field  $\mu h$ , where  $\mu$  is the related magnetic moment.

The spin dynamics is associated with the spin tunneling between states  $S^z = \pm 1/2$ . The transition of each spin between these two states requires them to acquire or release the longitudinal energy Eq. (1) to some thermal bath because of the energy conservation. The coupling of spins to phonons usually responsible for low-temperature dynamics in dielectrics is extremely weak and can be neglected. The relaxation takes place due to the interaction of the central electronic spins with the nuclear spin bath<sup>20,21,22</sup>. Since in the limit  $\Delta_o \ll E_o$  only spins inside the “resonance window”  $2E_o$  are allowed to tunnel, the approximate constraint  $|\mu h + U_i| < E_o$  determines the subspace of “resonant spins”. In this form the constraint is most suitable for the dynamic percolation model studied below in Sections III and IV. More accurate dependence of the relaxation time on energy is considered in Section V.

The only spins that are allowed to tunnel in any given configuration are the resonant ones. Transitions of resonant spins can change the status of their neighbors from non-resonant to resonant and vice versa by changing their local bias from being larger than  $E_o$  to being smaller (see Fig. 1. At the same time in the new configuration the transitions of neighbors, diffused into the resonance window, can push the former resonant spins out of resonance. Depending on the density of resonant spins  $P_0 \sim E_o/U_D$  there exist two possibilities. If  $P_0$  is very small (i.e., the distance between the resonant spins is large), the transitions of resonant spins can essentially affect only their local environment and spins in the sample can be separated into a small “mobile” and large “immobile” groups. The mobile group consists of percolating

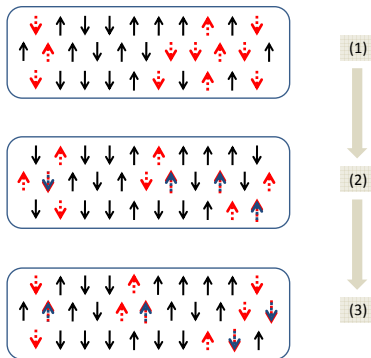


FIG. 1: Evolution of “percolating” (or “open”) spins affecting their neighbors in 2D lattice. When some of the open spins (shown by dashed red arrows) make transitions (shaded arrows for steps (2) and (3)) they switch the state of the neighboring spins between “open” and “close”.

spins, capable of making transitions in the course of system evolution, while the immobile group consists of non-percolating spins which cannot flip in spite of the field fluctuations caused by mobile spins. If the density  $P_0$  is large, the resonant spin transitions create or destroy resonances around them leading to the collective dynamics involving practically all spins after sufficiently long time.

In what follows we investigate the kinetic transition between these two regimes. The study of realistic 2D lattice of spins, coupled by the dipolar interaction, can not be done exactly analytically and requires numerical simulations. However, we can solve a similar problem on the Bethe lattice with random interactions between the neighbors. We assume that spins  $1/2$  occupy all sites of the Bethe lattice and each spin interacts with all its  $z$  neighbors. The interaction constant  $U_{ij}$  between two neighboring spins is assumed to be the Gaussian random variable with the zero average and dispersion  $U_D$ . This assumption is approximate - dipolar interactions in a real system involve not only nearest neighbors but also spins separated from the given one by more than one lattice constant. The long range nature of dipolar interactions can not be neglected in three-dimensions, but in the two-dimensional system the lattice sum of  $1/r^3$  is finite and the nearest neighbor approximation is justified. It is clear that the model we study and the real problem differ from each other; however they can belong to the same universality class because the phase-space structure for interacting Ising spins is similar to that for the Bethe lattice. The phase space grows exponentially with the number  $N$  of spins as in many-body problem<sup>16,17</sup>, which permits us to neglect “close loops” for different evolution paths of the system in its phase space thus making it similar to the Bethe lattice.

As mentioned above, we restrict our considerations to the limit of low temperatures, where each molecule can still be modeled as a two-level system, but we assume  $k_B T > U_D$  to avoid the dipolar ordering and, consequently, any long-range statistical correlations be-

tween spins. Indeed, such correlations should affect the kinetic transition. This assumption is not restrictive since, for instance, in  $Fe_8$  and  $Mn_{12}$  the regime of pure ground-state tunneling can be reached at temperatures  $T \lesssim 0.4 K$  and  $T \lesssim 0.8 K$  respectively which is larger than  $U_D$  in both systems<sup>19</sup>. We also consider only completely depolarized system here.

### III. DYNAMIC PERCOLATION PROBLEM ON BETHE LATTICE

In the Bethe lattice the spin is resonant (open) if its local bias (or Zeeman splitting)  $U_i = \sum_j U_{ij} S_j$ , Eq. (2), is smaller than  $E_o$ . Since the dipolar fields distribution in the depolarized limit is similar to the Gaussian one (see Fig.4 below), the fraction of resonant spins (i.e., the probability that the spin is open) is determined by  $P_0 \approx 2E_o/(U_D\sqrt{2\pi z}) \ll 1$ . We expect (and this will be confirmed by the final result) that the collective dynamics can take place if each spin has approximately one resonant neighbor (cf. Ref.<sup>7</sup>), which leads to the estimate for the transition point as  $P_{0c} \sim 1/z$ . This happens at  $E_o \sim U_D\sqrt{\pi/(2z)}$ . Near the transition point one can approximately ignore correlations between different configurations of neighbors corresponding to open states of the given spin. Indeed, if the spin is open in the given configuration the probability that it will be open again after  $k < z$  turns of neighbors can be estimated as  $P_k \sim 2E_o/(U_D\sqrt{2\pi k})$ . The number of neighbor transitions  $Z$  needed to bring the given spin back to the open state can be estimated setting  $\sum_{k=1}^Z P_k \approx 1$ . One can see that at  $E_o \sim U_D\sqrt{\pi/(2z)}$  the next opening of the given spin can be expected, on average, after turns of around  $z/2$  neighbors so the new resonant environment is fully different from the previous one.

Under above assumptions one can formulate the following model of dynamic percolation on the BL, see Fig. 1. Consider a lattice occupied by spins  $1/2$  having random projection  $S^z = \pm 1/2$ . Assume that in any given configuration of spins the rules for deciding about the spin states are the same: the probability that a given spin is open (i.e., “dynamic”) in each of the  $2^z$  configurations of neighbors is  $P_0$ . In other words, the dynamics of the model is fixed by having a table for each spin with  $2^z$  open and close states (the fraction of closed states is  $W_0 = 1 - P_0$ ). Correlations between open states of neighbors in different configurations are ignored. Only open spins are allowed to change their states. As time evolves, all open spins have equal chances to make a transition. The spin overturn affects all its  $z$  neighbors in a way that they have to change “status” according to their tables. Our main goal is to study the cooperative dynamics of spins, characterized by the fraction  $P_* = 1 - W_*$ , which were involved into dynamics at some stage (if the spin ever flips in the course of evolution we call it “percolating”) and to find out when an infinite percolating cluster is formed by those percolating spins.

The problem under consideration has common features and differences with the bootstrap percolation problem<sup>25,26</sup> used as a mathematically idealized model for such phenomena as nucleation and growth applied in the study of crack formation, magnetic alloys, hydrogen mixtures, and computer storage arrays. It has been extensively studied both for the  $2D$  lattice and BL. The bootstrap percolation problem can be formulated in terms of spins  $1/2$  placed in all lattice sites. The spin is open and allowed to flip if the number of neighbors with  $S^z = +1/2$  exceeds the predefined number  $0 < n \leq z$ . The cooperative dynamics is determined by the initial density  $P_0$  of spins with positive projections and in the most investigated case of  $n = 2$  this dynamics vanishes in a discontinuous manner at sufficiently small  $P_0$ ; for the  $2D$  lattice the threshold density  $P_{0c}$  approaches zero with increasing the system size. In spite of similarities, our model differs qualitatively because the bootstrap percolation results in the irreversible evolution of spin configuration towards the increase in the density of spins with positive projection, while in our case the density of open spins practically does not fluctuate and we study a reversible equilibrium dynamics. One consequence of this difference is vanishing  $P_{0c}$  in the bootstrap percolation problem in the  $2D$  lattice while in our case there is no cooperative dynamics at sufficiently small density of open spins  $P_0 < 2^{-z}/z$  ( $z = 4$  in  $2D$ ) because non-percolating spins form an infinite cluster blocking such dynamics, cf. Ref.<sup>1</sup>.

The exact description of the dynamic percolation for BL can be obtained similarly to Refs.<sup>5,25</sup> (see Fig. 2). First, we calculate the density  $P_*$  of percolating spins. Note that percolating spins can be in the close state for some time. Thus percolating spins include open spins and all other spins which can enter an open state at some point in time. In what follows we show that the density  $P_*$  can undergo a sharp raise (see Fig. 3) with increasing the density of open spins  $P_0$  above some critical value  $P_{c2}$ . This discontinuous transition happens after the formation of the infinite percolating cluster at  $P_0 = P_{c1}$  which promotes the cooperative dynamics.

#### IV. KINETICS TRANSITIONS IN BETHE LATTICE

Consider the probability  $W_* = 1 - P_*$  that a given spin is non-percolating, i.e. it is never involved into dynamics despite some of its  $z$  neighbors making transitions on BL, see Fig. 2. This probability depends on the states of all its neighbors, which can be treated as un-correlated due to the specific properties of BL. Each neighbor of the selected non-percolating spin is characterized by the conditional probability  $W_e > W_*$  that it is also not percolating (see Fig. 2). All neighbors are in identical situation by construction.

The probability  $W_*$  that the spin is not percolating can be determined by considering different local environ-

ments distinguished by the number of percolating neighbors  $k = 0, 1, \dots, z$ , see Fig. 2. There are  $z!/(k!(z-k)!)$  independent ways to be surrounded by  $k$  percolating spins. In each of them the selected spin will experience all  $2^k$  possible states in the course of evolution (assuming that percolating spins flip an unlimited number of times; this is definitely true before the infinite cluster is formed) and in each state it has to remain closed. The corresponding probability is  $(W_0)^{2^k}$  (recall that  $W_0 = 1 - P_0$ ). Summing up all the probabilities for different  $k$  we get

$$W_* = \sum_{k=0}^z \frac{z!(1 - W_e)^k W_e^{z-k}}{k!(z-k)!} W_0^{2^k}. \quad (3)$$

The probability  $W_e$  for neighbors of the given non-percolating spin to be non-percolating as well is defined similarly to Eq. (3) (see Fig. 2). One should consider only the remaining  $z - 1$  neighbors characterized by the same probability  $W_e$  to be non-percolating. Accordingly we obtain the self-consistent equation for the probability  $W_e$  in the form

$$W_e = \sum_{k=0}^{z-1} \frac{(z-1)!(1 - W_e)^k W_e^{z-k-1}}{k!(z-k-1)!} W_0^{2^k}. \quad (4)$$

In Fig. 3 the solution of Eqs. (3), (4) for the density of percolating spins  $P_* = 1 - W_*$  is shown for BL with  $z = 4, 5, 6$  and  $7$  neighbors. One can see that for  $z = 6$  and  $z = 7$  there exists the discontinuous transition at  $P_0 = P_{c2}$  (label  $P_{c1}$  is reserved for the formation of the infinite percolating cluster, see below) where the density of percolating spins  $P_*$  jumps from the small value  $P_* \sim 1/z$  almost to unity  $P_* \sim 1 - (W_0)^{2^z}$ . At  $P_0 > P_{c2}$  the vast majority of spins belong to the infinite percolating cluster.

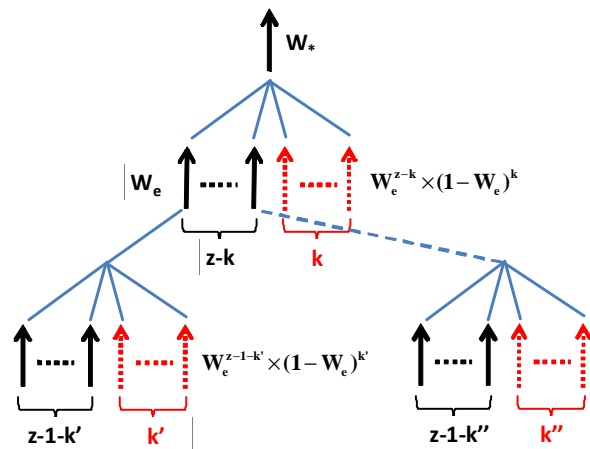


FIG. 2: Configurations of neighboring spins in the Bethe lattice and their probabilities. Red dotted arrows show percolating spins and black solid arrows show non-percolating spins. Blue lines connect neighboring spins.

The physics behind the jump of the percolating spin density at the transition point  $P_{c2}$  can be illustrated in

the limit of large number of neighbors  $z \gg 1$  when Eq. (4) can be simplified by taking advantage of  $P_e, P_0 \ll 1$  and expanding  $(1 - P_0)^{2^k} \approx 1 - 2^k P_0$  (recall that  $P_e = 1 - W_e$ )

$$P_e \approx P_0(1 + P_e)^{z-1} \approx P_0 \exp(zP_e). \quad (5)$$

This equation has a solution only for  $P_0 < P_{c2} \approx e^{-1}/z$ , which agrees with the numerical solutions for  $P_{c2}$  at  $z \geq 7$ . In our opinion the discontinuous transition is caused by an avalanche-type growth of the percolating site number in the vicinity of other percolating sites. For instance, if the density of percolating sites  $P_*$  reaches the value  $1/z$  then approximately each site has one open neighbor. Its probability to become percolating increases by the factor of 2 which leads to the formation of new open sites. This process leads to the jump in the density of percolating spins to near unity.

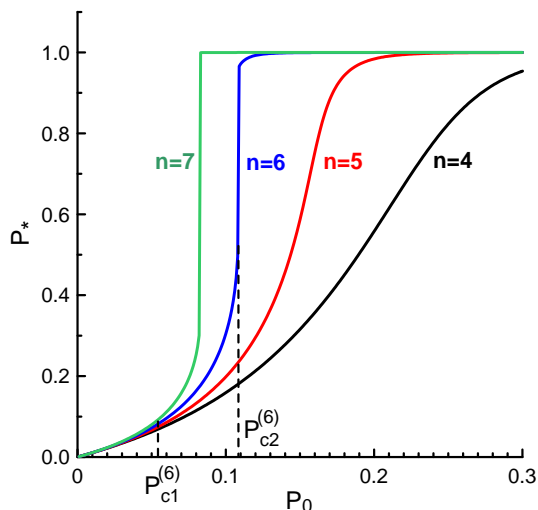


FIG. 3: Dependence of the percolating sites density  $P_*$  on the density of open spins  $P_0$  for  $z = 4, 5, 6$  and  $7$  neighbors. There is a discontinuous change in  $P_*$  for  $z > 5$  where its density instantaneously jumps to  $\approx 1$ . Transition points  $P_{c1} \approx 0.0546$ ,  $P_{c2} \approx 0.1085$  are shown for  $z = 6$ .

The discontinuous transition was found in the bootstrap percolation problem on Bethe lattice<sup>25</sup>, but at effective parameter  $P_0 = 1/(z-1)^2$ . It is interesting that in spite of the absence of kinetic transition in the bootstrap percolation problem in the infinite  $2D$  lattice, the transition in the finite system is essentially discontinuous<sup>26</sup>. This gives us some hope that the discontinuous dynamic percolation transition may take place in our problem applied to realistic lattices of finite dimensions ( $d > 1$ ) similarly to a Bethe lattice.

The cooperative dynamics in the ensemble of interacting spins arises in the presence of the infinite cluster of percolating sites. Such cluster obviously exists at  $P_0 > P_{c2}$ , but it may be formed at smaller density  $P_0$  of open sites. Indeed, the probability to have a percolating site near another percolating site is about a factor of 2 larger than in the absence of percolating neighbors because of the doubling in the number of explored configura-

tions. The corresponding critical point  $P_0 = P_{c1}$  where the infinite cluster of percolating spins is formed can also be found exactly. It can be shown (see Appendix) that the formation of the infinite cluster is determined by the equation

$$(z-1)(1 + F_0 - 2F_1) = 1 + (z-1)^2[F_2(1 - F_0) + F_1^2 + F_0 - 2F_1], \quad (6)$$

where  $F_m$ , see Eq. (13) in the Appendix, is the probability of finding a non-percolating spin surrounded by  $z$  neighbors where  $m = 0, 1, 2$  of them are *definitely* percolating,  $2 - m$  neighbors are *definitely* non-percolating and the remaining group of  $z - 2$  neighbors can contain both percolating and non-percolating spins. For small  $P_0$  we have  $F_m \approx 1 - 2^m P_0 \cdot (m(1 - W_e) + W_e)^{z-2}$ . The numerical analysis of the transition point  $P_{c1}$  corresponding to the formation of an infinite percolating cluster confirms that  $P_{c1} < P_{c2}$ . In the  $z \gg 1$  limit one finds that the infinite cluster is formed at  $P_0 > P_{c1} \approx e^{-1/3}/(3z) < P_{c2}$  in agreement with the numerical solution of Eq. (6).

## V. DISCUSSION. HOW CAN THE KINETIC TRANSITION BE OBSERVED IN MOLECULAR MAGNETS?

Below we consider the kinetic transition in a 2D square lattice of spins representing magnetic molecules using the results obtained for the Bethe lattice. This consideration should be applicable to the recently synthesized two-dimensional crystals of  $Mn_{12}$  molecules<sup>27</sup>. Assume that the easy axis is perpendicular to the sample plane. Since only spins inside the resonance window can change their state, the density of resonant spins in the longitudinal magnetic field  $h$  can be estimated as

$$P_0 \approx 2g(\mu h)E_o, \quad (7)$$

where  $g(E)$  is the probability density for the Zeeman splitting  $E$  (longitudinal bias) originated from the spin-spin interaction (see Fig. 4) and  $\mu$  is the magnetic moment of the molecule.

To study the kinetic transition using the previous results for the Bethe lattice one has to introduce the number of neighbors parameter  $z$ . We assume that the number of neighboring spins can be estimated as the number of spins whose resonance can be affected by the transition of the given spin. For the crude estimate we count neighboring spins as those coupled to the given spin by the interaction exceeding the width of the resonant window  $2E_o$ . In the limit  $E_o \ll U_D$  one can estimate this number in a quasi-continuum approach as

$$z \approx \pi(U_D/2E_o)^{2/3}. \quad (8)$$

According to the solution for the Bethe lattice (see Fig. 3) the sharp change of transition rate for almost all spins should take place near the point  $P_0 = P_{c2}$ . Assuming

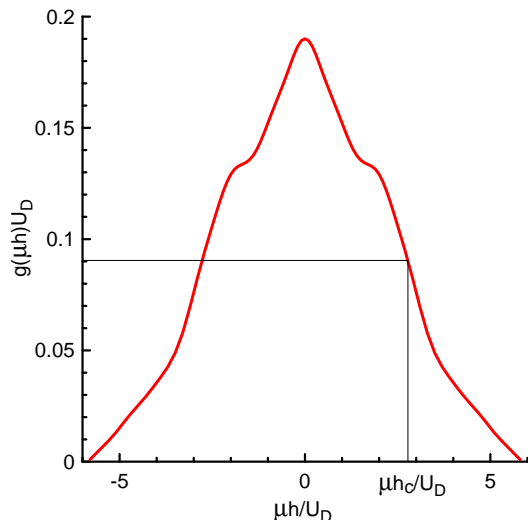


FIG. 4: Rescaled probability density for the spin energy splitting on a model square lattice of magnetic molecules. The predicted transition point is shown by solid lines.

$z \gg 1$  the crude estimate for the transition point can be made using the approximate relationship  $P_{c2} \approx e^{-1}/z$  (see Sec. IV). Accordingly we get

$$E_c \approx \frac{U_D}{2(\pi e g(\mu h) U_D)^3}. \quad (9)$$

The distribution of the dipolar bias energies  $g(E)$  in a sample can be easily calculated, see Fig. 4. The minimum threshold is realized at zero field (maximum density of resonances) where  $g(0) \approx 0.19/U_D$  so that we get  $E_{c2}(0) \approx 0.12U_D$ .

Using the above analysis of the kinetic transition one can approximately characterize the spin relaxation for different molecular magnets. In  $Fe_8$  one has  $U_D \approx 130$  mK and  $E_o \approx 6$  mK<sup>23</sup> so even in the absence of the external field the system is in the localization regime. The situation is different in  $Mn_{12}$ , where  $U_D \approx 70$  mK and  $E_o \approx 80$  mK<sup>24</sup> and the collective spin dynamics exists at zero magnetic field according to the theory, Eq. (9). However, application of the external longitudinal magnetic field can reduce the resonance probability and result in the localization. Using Eq. (9) one can estimate the value of the external field corresponding to the transition point from

$$g(\mu h_c) U_D \approx \frac{1}{\pi e} \left( \frac{U_D}{2E_o} \right)^{1/3} \approx 0.09 \quad (10)$$

and the transition in  $Mn_{12}$  takes place at the field  $h_c \approx 2.8U_D/\mu \sim 0.2$  T (see Fig.4). One can also expect that the transition rate should decrease near the transition point.

Although theory predicts the absence of cooperative dynamics at  $h > h_c$  the reality is more complicated because spins which do not belong to the transition window yet have small but finite transition rate. Indeed, spins

with the Zeeman energy  $E > E_o$  can make transitions; however their transition rate is becoming exponentially small<sup>20,21,22</sup>

$$\tau^{-1} \propto \exp(-|E|/E_o), \quad (11)$$

because such transition require simultaneous flips of large number of nuclear spins. Then, we predict the exponentially small transition rate which can be estimated following the standard percolation theory approach<sup>1</sup> as the rate Eq. (11) at the Zeeman splitting  $E$  equal to the threshold value  $E_c$ , Eq. (9)

$$\tau^{-1} \propto \exp(-E_c/E_o) \approx \exp(-(g(\mu h_c)/g(\mu h))^3). \quad (12)$$

This dependence is illustrated in Fig. 5 to show the expected strong change in the rate of spin relaxation at large fields. Unfortunately, the dynamics slowing down takes place at fields  $h \approx 2h_c$ , where the density of resonant spins is already very small (see Fig. 4) and the predicted reduction of the relaxation rate by many orders of magnitude is more difficult to study. The dynamic transition itself becomes a crossover because the transition rate is a continuous non-vanishing function of energy, Eq. (11).

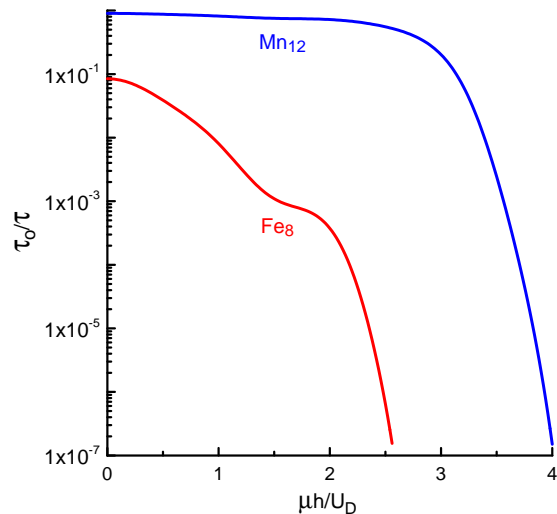


FIG. 5: Dependence of the relaxation rate  $\tau^{-1}$  on the magnetic field near the transition point calculated using the exponential approach Eq. (12).  $\tau_o$  is the characteristic relaxation time.

In addition to the application of the longitudinal magnetic field one can also affect the transition by diluting  $Fe_8$  magnets. The reduction of the concentration of  $Fe_8$  molecules to  $x \ll 1$  will amount for changing  $U_D/E_o$  ratio, and thus provide a knob for determining the transition point experimentally. Assuming that the effective constant for magnetic dipolar interaction scales proportionally to the  $x^{3/2}$  one can estimate that at  $x \leq 0.25$  the collective dynamics in  $Fe_8$  films will take place.

## VI. CONCLUSION

We considered the dynamic percolation problem on the Bethe lattice of spins  $1/2$ . Dynamics was introduced through the open spins capable to change their states and affect the status of neighboring spins switching them between open and closed states. The problem was solved exactly. We found two kinetic transitions including the continuous transition associated with the formation of the infinite percolating cluster and the discontinuous transition associated with the avalanche-type growth in the number of percolating sites. This model approximately describes the low-temperature dynamics of molecular magnets stimulated by their interaction with the nuclear spin bath. In this model open spins are those having the small Zeeman splitting compared to their hyperfine interaction. The sharp kinetic transition is predicted for the  $2D$  lattice of magnetic molecules, however the abrupt change of the relaxation rate is smeared out by its continuous dependence on the total longitudinal bias Eq. (11), which may obscure the experimental observation of the transition point.

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## VII. APPENDIX

Consider the probability  $P(N)$  that two spins separated by  $N$  sites are connected through percolating sites. It is clear that if this probability decreases with  $N$  slower than the inverse number of paths of length  $N$  starting at the given site,  $(z-1)^{-N}$ , then the infinite cluster of percolating sites is formed. This criterion is also applicable to the static percolation problem on BL where the connection probability is given by  $P_0^N$ . Accordingly, the infinite cluster is formed at  $P_0 \geq 1/(z-1)$  in full agreement with the exact solution<sup>7</sup>.

Each site of the line connecting spins 0 and  $N+1$  has  $z-2$  neighbors which do not belong to the line. It is convenient for the rest of the discussion to introduce a short notation for the probability of finding a non-percolating spin (or  $n$ -spin) when  $m=0,1,2$  of them are *definitely* percolating,  $2-m$  neighbors are *definitely* non-percolating and the remaining group of  $z-2$  neighbors can contain both percolating and non-percolating spins.

$$F_m = \sum_{k=0}^{z-2} \frac{(z-2)!(1-W_e)^k W_e^{z-k-2}}{k!(z-k-2)!} W_0^{2k+m}, \quad (13)$$

cf. Eqs. (3) and (4). Indeed,  $F_m$  are all we need to know to calculate the probability that a given line spin is non-percolating when the states of its line neighbors are

known. Since  $W_e$  is known as a function of  $W_0$ , we can concentrate on properties of the line spins alone. Note, that this decomposition of probabilities Eq. (13) is possible due to the Bethe lattice factorization around non-percolating spins. In other words different realizations for spins belonging to branches of BL separated by  $n$ -spins are independent of each other. Since the states of the end spins (0 and  $N+1$ ) can not change scaling properties of the long line connecting them we select them to be  $n$ -spins to simplify the derivation of  $P(N)$ . We then consider  $P(N)$  as complementary to the probability of having an arbitrary line decomposition in terms of  $p$ - and  $n$ -spins with at least one  $n$ -spin among  $N > 0$ , i.e.

$$1 = \sum_{\{n\}} \rho_{n,p}(\{n\}), \quad (14)$$

where in each sequence  $\{n\}$  all  $N+2$  spins are divided into  $2s+1$  segments of alternating  $n$ - and  $p$ -spins as  $\{n\} = (n_1, p_1, n_2, \dots, p_s, n_{s+1})$  with  $n_i, p_i \geq 1$  ( $n_i$  is used for  $n$ -spins and  $p_i$  for  $p$ -spins). The probability of a particular sequence is denoted as  $\rho_{n,p}[n_1, p_1, \dots, p_s, n_{s+1}]$ . Clearly,  $n_1 + p_1 + \dots + p_s + n_{s+1} = N+2$ .

Due to factorization provided by  $n$ -spins (no information can be exchanged between the neighbors of non-percolating spins and thus all branches of BL around them are statistically independent) we have

$$\rho_{n,p} = F_0^N, \quad (\text{for } s=0 \text{ or } n_1 = N+2), \quad (15)$$

$$\rho_{n,p} = \rho_e(n_1)\rho_e(n_{s+1}) \prod_{j=2}^{s-1} \rho_n(n_j) \prod_{j=1}^s P(p_j), \quad (\text{for } s > 0)$$

Here  $\rho_n(m)$  is the probability of having a cluster of  $m$   $n$ -spins in a row, and,  $\rho_e(m)$  is a similar quantity for the first and last groups of  $n$ -spins. These probabilities can be immediately computed by counting how many  $n$ -spins have  $m$   $p$ -spins as their neighbors (recall that the end spins (0 and  $N+1$ ) are assumed to be non-percolating)

$$\begin{aligned} \rho_n(1) &= F_2, & \rho_n(m > 1) &= F_1^2 F_0^{m-2}, \\ \rho_e(1) &= 1, & \rho_e(m > 1) &= F_1 F_0^{m-2}. \end{aligned} \quad (16)$$

The self-consistent equation (14) for  $P(N)$  is solved using generating functional  $P(x) = \sum_{N=1}^{\infty} x^N P(N)$  (and similarly defined  $\rho_e(x) = \sum_{N=1}^{\infty} x^N \rho_e(N)$  and  $\rho_n(x) = \sum_{N=1}^{\infty} x^N \rho_n(N)$ ) which transforms (14) into

$$\frac{x}{1-x} = \frac{F_0 x}{1-F_0 x} + \frac{\rho_e^2(x) P(x)}{x^2} \sum_{k=0}^{\infty} [\rho_n(x) P(x)]^k, \quad (17)$$

with  $\rho_e(x) = x + F_1 x^2 / (1 - F_0 x)$  and  $\rho_n(x) = F_2 x + F_1^2 x^2 / (1 - F_0 x)$ . After elementary algebra we find

$$P(x) = \frac{x^3(1-F_0)}{(1-x)(1-F_0 x)\rho_e^2(x) + x^3(1-F_0)\rho_n(x)}, \quad (18)$$

which can be further simplified to

$$P(x) = \frac{x(1-F_0)}{(1-x)(1-F_0 x + 2F_1 x) + x^2[F_1^2 + F_2(1-F_0)]}.$$



The infinite cluster is formed if  $P(N)$  decreases with  $N$  slower than  $1/(z-1)^N$ . This means that the threshold is determined by the divergence of  $P(x)$  at  $x = (z-1)$  which is only possible if the denominator in Eq. (18) is zero. Thus the formation of the infinite cluster is determined by the equation

$$(z-1)(1+F_0-2F_1) = 1 + (z-1)^2[F_2(1-F_0) + F_1^2 + F_0 - 2F_1], \quad (19)$$

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