Elastic interaction model for hysteresis in molecular magnets

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The spin transition compounds are molecular magnets, switchable between two states in thermodynamic competition: the diamagnetic low spin state (LS) and the paramagnetic high spin state (HS). When the elastic interactions in these systems are stronger than a threshold value, the abrupt transition is accompanied by complex hysteresis processes like thermal or pressure hysteresis [1, 2], which makes them suitable for a wide range of applications.

We model here the hysteretic behavior in spin transition compounds, considering the molecules situated in a bi-dimensional hexagonal lattice and interacting by the way of elastic connecting springs [3], that stand for both long range and short range interactions (fig. 1). Even if the present model uses less parameters, it is able to simulate all the main experimental curves, and predict the clustering and nucleation phenomena.
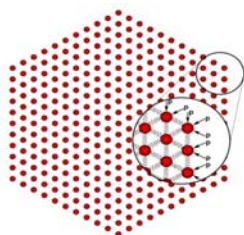

Fig. 1 The hexagonal network of particles connected with springs.

At high temperature, the molecules are in the HS state; by lowering the temperature they pass to LS state, if able to exceed the elastic force from the closest springs. The switch of individual molecules is checked randomly by a Monte Carlo procedure. When a molecule changes its state, the new equilibrium positions of all molecules are calculated. The transition of a molecule from a state to another corresponds to a change of its volume (HS molecules have a bigger volume) and modifies the global interactions in the system and finally the position of each molecule.

The transition probability for a molecule to pass from one state to another depends on the temperature and on the pressures acting on it, according to:

$$P^i_{HS \to LS} = \frac{1}{\tau} \exp\left(-\frac{E_A - \kappa p_i}{T}\right)$$

$$P^i_{LS \to HS} = \frac{1}{\tau} \exp\left(-\frac{\Delta H - T\Delta S}{T}\right) \exp\left(-\frac{E_A + \kappa p_i}{T}\right)$$

where ΔH is the enthalpy variation, ΔS is the entropy variation, E_A is the activation energy, T the temperature, p_i the pressure acting on i molecule.

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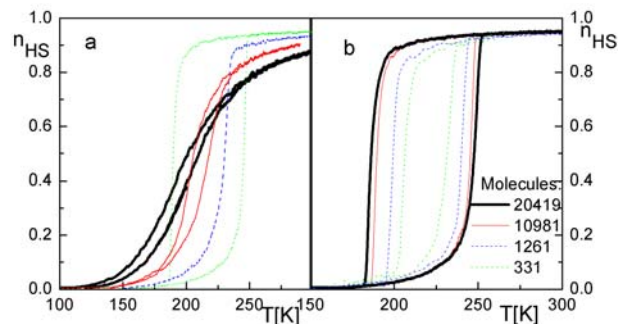E-mail: cristian.enachescu@uaic.ro

Fig.2 Thermal hysteresis for different interactions (a) and system sizes.(b)

In Fig. 2a we present how the interactions are influencing the width and shape of the hysteresis: smooth transition showing no hysteresis in absence of interactions and steeper hysteresis for increasing interactions, similar to experimental curves presented elsewhere. The effect of the system size is presented in fig. 2b and show a behavior in good agreement with very recent experimental data on spin crossover nanoparticles (width increasing for larger system sizes).
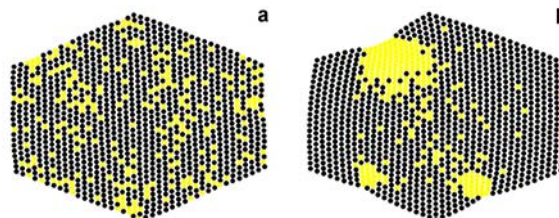

Fig. 3 Snapshots of spin configurations when around 10% of molecules are in the LS state showing the difference between the behavior observed for weak (a) and strong (b) interactions

For moderate values of the spring constant, some clusters form, but on average the HS and LS molecules are distributed randomly in the sample during the relaxation process (Fig. 3a). If the spring constant is high enough then the clusters are bigger and develop faster throughout the sample leading to fluctuations, and nucleation and growth phenomenon (Fig 3b). An infinite avalanche process is then started leading to non-random distributions of HS and LS complexes.

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