ULTRA-THIN MAGNETIC FILMS AND THE STRUCTURAL GLASS TRANSITION: A MODELLING ANALOGY

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In this work we study a two dimensional Ising model for ultra-thin magnetic films which presents competition between short range ferromagnetic interactions and long-range antiferromagnetic dipolar interactions. We present evidence that the dynamical and thermodynamical properties of the model allow for an alternative interpretation, in terms of glass forming liquids. In particular, the existence of a first order phase transition between a low temperature crystal–like ordered phase a high temperature liquid–like disordered phase, which can be supercooled below the melting point, together with a drastic slowing down after a quench to low temperatures suggest that these materials could present a phenomenology similar to that observed in glass forming liquids.

1. Introduction

The physics of glass forming liquids and structural glasses in general, appears today as a great challenge in statistical mechanics and chemical physics. Despite the huge effort devoted to the field and the enormous improvements obtained in the comprehension of these complex systems, due both to theoretical and experimental studies, there are still many open questions concerning their phenomenology.

Most of the theoretical knowledge in the field resides today in two different approaches. On one side, there are different phenomenological and first principles microscopic theories (for a recent review see Ref.[1]). Among the last ones,

perhaps the most successful one is the *mode coupling* theory [1], in the sense of having more experimentally verified predictions. However, up to now no one of the existing theories can account for a complete description of the observed phenomenology. On the other hand, the constant improvement in the computational capacity allowed the implementation of very accurate molecular dynamics simulations. Most of these simulations are based on small binary systems of particles interacting through *Lennard–Jones* like potentials. While the existing microscopic theoretical approaches seem to be very limited due to the complexity of the analytical treatment, the numerical approach is limited by the small number of particles that can be considered and the extremely small time span that a simulation can cover, specially for modelling systems that have an astonishing slow relaxation dynamics [2].

In a completely different scheme, the statistical physics community has been looking, since many decades ago, for a simple lattice model able to catch (independently of the degree of accuracy of its microscopic description) those few relevant ingredients which are responsible of the rich dynamical and thermodynamical phenomenology of these materials.

There is a general consensus in the community in the sense that a relevant element in the description of structural glasses is the appearance of frustration at the level of microscopic interactions between molecules. And, unlike what happens in many other complex systems, as for instance spin glasses, this frustration is not due to the existence of randomness, but to the emergence of competition between attractive and repulsive interactions acting on each particle.

Among the many different approaches presented in the literature in order to introduce a lattice model for structural glasses, we want to mention in first place that presented by Shore, Holzer and Sethna [3], since, as will become clear in short, it is intimately related to the scope of our paper. They consider the magnetic Ising system on a square and on a cubic regular lattices, with ferromagnetic interactions between nearest–neighbors plus antiferromagnetic interactions between next–nearest neighbors spins, and without taking into account any kind of randomness in the Hamiltonian of the model. For the two dimensional case, they actually found a relatively simple dynamical and thermodynamical behavior, which is far from being *glassy*. But the situation was completely different for the three dimensional case, where at very low temperatures they found a drastic slowing down of the relaxation, ruled by a logarithmic domain growth law, proper of glassy systems. This simple model, without imposed disorder and with competition, showed to be able to reproduce at least partially and qualitatively the complex phenomenology of a glassy processes. Nevertheless, its main limitation was the existence of a second order phase transition between the high temperature disordered phase (analog to a liquid state) and the ferromagnetic ordered phase (analog to a crystalline state). This continuous transition without coexistence of phases can not give account of the process of supercooling a liquid, which is intimately related to the process of

forming a structural glass.

Since then, many other attempts have been done in order to improve that first intent. In a series of papers, Lipowski and co–workers [4] considered the same model introduced by Shore *et al.* plus a four spin plaquette ferromagnetic term in the Hamiltonian. This model captures most of the complex dynamics of the original model, and presents a first order phase transition, as desired. Nevertheless, its ground state is infinitely degenerate, a fact that can be hardly associated to the crystalline ordering of a solid. Later on, Cavagna, Giardina and Grigera [5] considered a two dimensional model with a two terms Hamiltonian: the previously described four spin ferromagnetic plaquette plus a five term ferromagnetic plaquette. This is actually an excellent model which describes most of the expected features of a two–dimensional structural glass. Another interesting approach [6] considered a system with nearest–neighbors coupling and antiferromagnetic coulomb interactions, which also proved to be an adequate model for describing three dimensional glass forming liquids. Nevertheless, all these systems, besides their great value as statistical mechanics prototypes for modelling a structural glass without imposing disorder in the Hamiltonian, are not really inspired by any physical realization. In this work instead, we will present a model which has been vastly analyzed during the last ten years and is considered to be the proper tool for describing the physics of real ultra thin magnetic films, but instead of paying attention to the magnetic behavior, we will show that the model, and consequently perhaps ultra thin films, present evidence of displaying a structural glass–like state.

2. A metal on metal ultra thin film model

The physics of ultra–thin magnetic films has deserved a great interest during the last years, not only because of their multiple technological applications, such as data storage and catalysis, but also because their study has opened novel and nontrivial questions related to the role of microscopic competitive interactions in the overall behavior of a system. In particular, under suitable thermal and magnetic conditions, ultra thin magnetic films form unusual complex patterns of magnetization [7,8]. And precisely, most of the potential technological applications of these materials reside in the ability of controlling these patterns, both in time and space, with a high degree of accuracy. For instance, in the case of data storage the stabilization of very small metastable magnetic domains could eventually increase the compression obtained nowadays in recording devices.

The model we will analyze in this paper has been used mainly in the study of metal films on metal substrates, as for example Fe on Cu [9] or Co on Au [10]. In these cases, an adequate theoretical description of the system must take into account, at least, a three terms Heisenberg Hamiltonian, including: *i)* the usual ferromagnetic exchange interactions between nearest–neighbors spins, *ii)* the dipole–dipole interactions which, despite their considerable small strength (when compared with the exchange interactions) become relevant due to their long range, and finally *iii)* a sur-

face anisotropy term that takes into account the magnetic influence of the substrate on the spins of the film. The anisotropy induced by the dipolar interaction tends to align the spins parallel to the film, but, as the thickness of the film is reduced (usually around approximately five monolayers) the surface anisotropy overcomes the anisotropy of the dipolar interaction and the system suffers a reorientation transition at which the spins suddenly align perpendicular to the plane defined by the proper film. Under these particular conditions, the physics of the material can be appropriately described by replacing the Heisenberg spin variables by the much simpler Ising magnetic moments located at the nodes of a square lattice, and the Hamiltonian takes the form:

$$H = -\delta \sum_{\langle i,j \rangle} S_i S_j + \sum_{(i,j)} \frac{S_i S_j}{r_{ij}^3} \tag{1}$$

where $S_i = \pm 1$ and δ is the ratio between the exchange J_0 and dipolar J_d interactions strengths ($\delta = J_0/J_d$). Here the first term represents the ferromagnetic exchange interaction and the sum runs only over nearest–neighbors spins, while the second one represents the dipole–dipole interaction once the spins have aligned perpendicular to the plane. In this last case, the sum runs over all pairs of spins of the lattice and r_{ij} is the distance, measured in crystal units, between the sites i and j. Then, the system is ruled only by two variables: the usual temperature T and the parameter δ, which depends on the composition and preparation of the sample. We will restrict ourselves to consider the case $J_d > 0$, in such a way that $\delta > 0$ corresponds to ferromagnetic exchange interactions and $\delta < 0$ to antiferromagnetic ones. Note that the model introduced by Shore *et al.* [3] can be considered as truncated version of the model defined by Hamiltonian (1).

In 1995 MacIsaac and coauthors [11] presented the first study of the thermodynamics of the model (for a complete review of the subject see [12]). Concerning the order of the ground state, it is antiferromagnetic when $\delta < \delta_a \approx 0.425$, but when $\delta > \delta_a$ the system orders forming *stripes* whose width h depends on the value of δ. In particular, h increases as δ increases and, surprisingly, the ferromagnetic state is always metastable respect to a striped one. In other words, irrespectively of the strength of the dipole–dipole interactions, the frustration induced by the antiferromagnetic term avoids the usual ferromagnetic ordering. In the same paper [11] they also presented a phase diagram of the model, obtained through Monte Carlo simulations on a relatively small system of $N = 16 \times 16$ spins. They observed that, for fixed values of δ, the system suffers an order–disorder phase transition between a low temperature striped phase and a high temperature tetragonal phase. The last one consists of extended magnetic domains characterized by predominantly square corners, which induces a four fold rotational symmetry (as can be clearly observed in numerical simulations of the structure factor). The existence of this tetragonal phase has been recently verified experimentally in a fcc Fe on Cu(100) films [7,8] and had already been predicted by Abanov *et al.* by means of a continuous approximation [13].

In Fig. 1 we present a detail of the phase diagram obtained in [14] corresponding to the intermediate values of δ. Here $h1$ and $h2$ indicates the regions where the ground states have widths $h = 1$ and $h = 2$, respectively, and AF indicates the antiferromagnetic phase. The gray region indicates the presence of metastable states. The lines (diamonds) separating the low temperature phases are all first order ones, and TP indicates the existence of a triple point (as will be become clear in the next section).

3. Super cooled tetragonal liquid state

In this section we will present some recent and preliminary evidence that strongly suggests that metal on metal ultra thin magnetic films could have a glassy transition.

Supercooled glass forming liquids have the property of getting trapped into a liquid metastable state (with respect to the crystalline state), when cooled below the melting temperature under suitable conditions. But, if the cooling is done suddenly enough at sufficiently low temperatures, the characteristic relaxation time attains macroscopic scales, the supercooled state is structurally arrested and the material behaves as a solid without any pattern of long range order. Under these conditions the systems becomes a *glass*. Then, a basic ingredient for a glass transition model is the existence of a first order phase transition between a high temperature liquid–like disordered phase and a low temperature crystal–like ordered phase.

We will now show that the order-disorder phase transition observed in the model described by Hamiltonian (1), is actually a weak first order one, at least for intermediate values of δ (though some analytical approximate results [16] suggest that this could be valid for any finite value of δ).

In Fig. 2 we plot the specific heat C_L as a function of the temperature T for $\delta = 3$, and three different system sizes. We can clearly identify two peaks. The low temperature one increases with the size L and coincides with the temperature at which the tetragonal phase appears. Then it is associated with the stripe-tetragonal transition. The second broader peak does not manifest any dependence on the system size and indicates the continuous decay of the tetragonal phase into the paramagnetic phase. We also present some snapshots of typical configurations of the system for different temperatures.

What about the order of the transition? One way of determining it is through the analysis of the finite size scaling behavior of different moments of the energy, like the specific heat

$$C = \frac{1}{NT^2}\left(\langle H^2\rangle - \langle H\rangle^2\right) \tag{2}$$

and the Binder fourth order cumulant

$$V_L \equiv 1 - \frac{\langle H^4\rangle}{3\langle H^2\rangle^2}\,, \tag{3}$$

which permit to distinguish between continuous and discontinuous transitions. In a first order phase transition the specific heat presents a maximum at a pseudo

critical temperature $T_c^1(L)$ and the Binder cumulant a minimum at another different pseudo critical temperature $T_c^2(L)$. Those temperatures present the following finite size scaling behavior $T_c^1(L) \sim T_c + AL^{-2}$ and $T_c^2(L) \sim T_c + BL^{-2}$, with $B > A$, T_c being the transition temperature of the infinite system [15]. In Fig. 3 we plot T_c^1 and T_c^2 *vs.* $1/L^2$ for $\delta = 2$, identifying clearly the finite size scaling behavior expected in a first order phase transition [16]. Moreover, numerical simulations of the energy histogram around the transition point for $\delta = 2$ show a two peak structure, proper of this kind of transitions [16].

Next we will show that it is possible to get a supercooled metastable state below the melting temperature, another significant feature of glass forming liquids. In Fig. 4 we plot (full circles) the average internal energy per particle $u(T)$ as a function of the temperature in a quasi-static cooling from a high temperature. Each point corresponds to an average over many different initial conditions and sequences of random numbers. In the same plot (empty triangles), we also display the result of a quasi–static heating from the ground state. We clearly observe the emergence of hysteresis, proper of a first order phase transition. From these energy curves we obtained the free energy per spin on cooling and heating, finding that both curves intersect at $T_m = 0.805 \pm 0.005$, a temperature that we identify with the *melting point*.

Finally, let us describe the behavior of the system when it is suddenly quenched into a very low temperature. In Fig. 5 we plot the time evolution of the internal energy per spin along a single Monte Carlo run for $\delta = 2$, $L = 32$ and $T = 0.2$. We observe that the system is stuck into a disorder configuration, with a very slow relaxation rate (almost logarithmic, as can be observed in the inset). The dashed line indicates the energy per spin of the ground state, and we see that the system is magnetically arrested in this out of equilibrium disordered state. We also present, at different times, snapshots of the corresponding pattern of magnetization, where one can recognize and almost tetragonal phase. This behavior, characterized by a slow relaxation of a disordered liquid–like phase well below the melting temperature, which can neither be associated with nucleation nor with coarsening, can be clearly interpreted in terms of the appearance of a glassy phase.

4. Conclusions

In this paper we have revisited the two dimensional Ising model with competing nearest–neighbors ferromagnetic interactions and long range antiferromagnetic dipole–dipole interactions from a new point of view. Instead of concentrating on the magnetic properties of the model, we have investigated a possible characterization of the tetragonal phase as a lattice version of a liquid that can give place to a glass state a very low temperatures.

It has been well established in our simulations that the order–disorder temperature driven phase transition between the tetragonal and striped phases is a weak discontinuous one, as revealed by the scaling law of the pseudo critical temperatures

obtained from the specific heat and the fourth order Binder cumulant. Analitical results on a continuous version of the model give further support to this conclusions [16]. We have also shown, by simulating a quasi–static cooling from infinite temperature, that the tetragonal phase can be supercooled well below the melting temperature. Furthermore, when the system is suddenly cooled down into a low enough final temperature, it gets stuck in a glass like state, characterized by an extremely slow relaxation process.

It is important here to mention that previous papers had already reported the existence of some glassy like phenomena, as for instance, aging [17,18] and logarithmic domain growth law [19], but all of them were connected to the existence of metastable stripe states, which it is well known, modify the landscape of the free energy function. Instead, the results presented in this paper refer to a different and novel observation, namely, the existence of a first order phase transition between the tetragonal (liquid) and the striped phases (crystal) and the supercooling of the former in a long standing metastable state well below the melting temperature, indicating the emergence of a two dimensional glass.

Finally, let us stress that the present model, unlike all the other lattice models cited in this papers, did not arise as a statistical mechanics toy model able to catch the main features of a glass forming liquid. On the contrary, the model is widely accepted to be the proper one for describing the physics of real metal on metal ultra thin magnetic films. In other words, our results strongly suggest that these materials present a glass transition, a fact that would have many relevant technological consequences. As much as we know, this is the first example of a physical realization of a two dimensional magnetic system without imposed disorder that can be considered as a glass forming liquid.

Nevertheless, this point requires further investigation. In that sense, a careful study of the behavior of the relaxation time as the temperature decreases, as well as an adequate characterization of the nucleation process, will not only be a clear confirmation of the existence of a glass state but will also bring light into its nature. Works along these lines are in progress and will be published elsewhere. Experimental checks of our predictions will be also very helpful.

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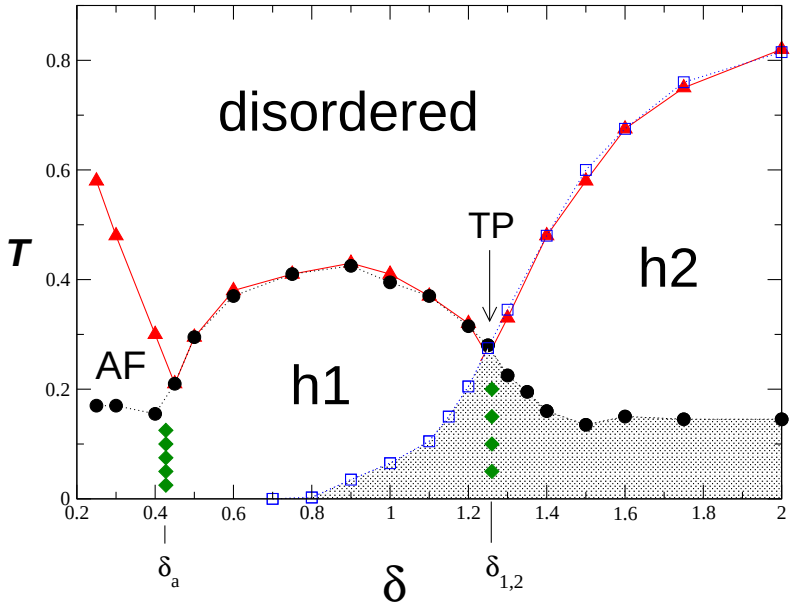

Figure 1. The phase diagram for intermediate values of δ obtained in $L = 24$ lattices. Triangles: critical temperature obtained from the maximum in the specific heat; circles: stability line of the $h1$ stripe phase; open squares: stability line of the $h2$ stripe phase; diamonds: first order transition lines between low temperature ordered phases. TP indicates a triple point.

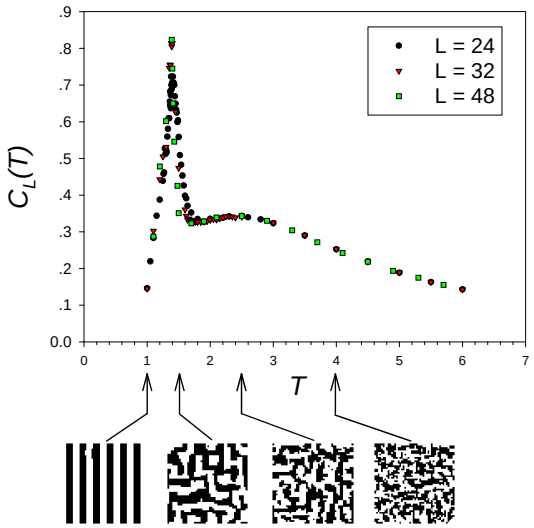

Figure 2. Specific heat *vs.* T for $\delta = 3$ (corresponding to an $h = 4$ ground state) and three different system sizes. Some typical equilibrium configuration at the indicated temperatures for $L = 48$ are shown below. Note the sequence of transitions $h4 \rightarrow$ tetragonal $\rightarrow$ paramagnetic.

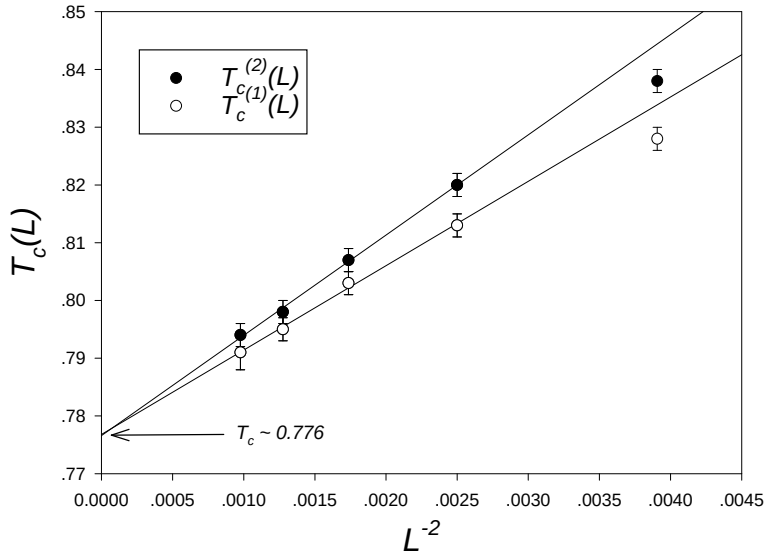

Figure 3. Pseudo critical temperatures T_c^1 (maximum of the specific heat) and T_c^2 (minimum of the Binder cumulant) vs. L^{-2} for $\delta = 2$.

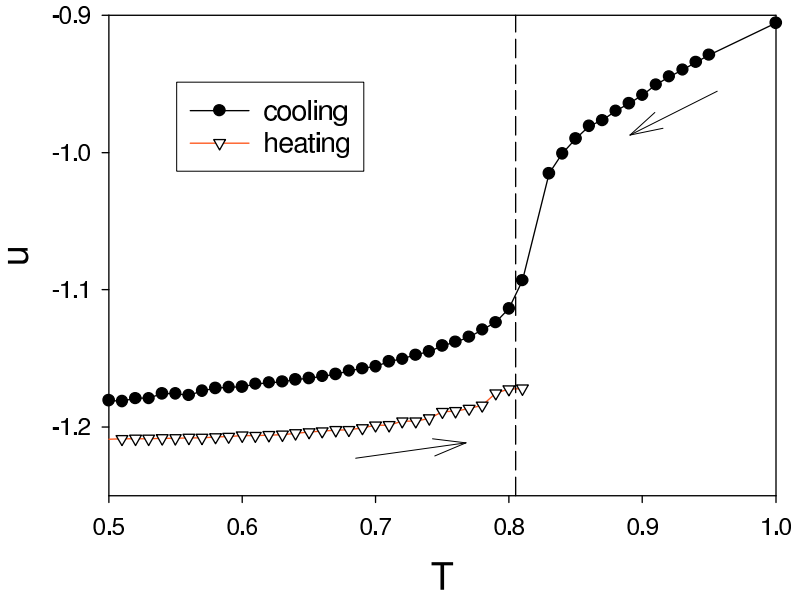

Figure 4. Internal energy per spin $u(T)$ obtained by quasi–static cooling from infinite temperature and quasi–static heating from the ground state.

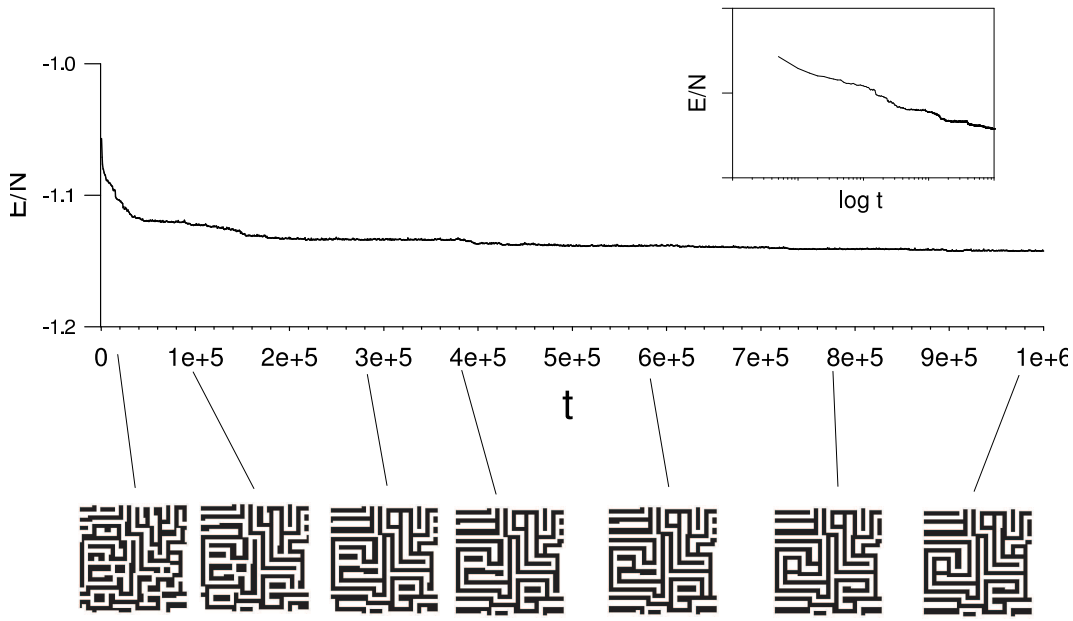

Figure 5. Time evolution of the energy per spin in a single MC run. Snapshots of the spin configurations are shown below the figure. The inset presents the same results for the time evolution of the energy per spin in a log-normal plot.