Magnetic behavior of a mixed Ising ferrimagnetic model in an oscillating magnetic field

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The magnetic behavior of a mixed Ising ferrimagnetic system on a square lattice, in which the two interpenetrating square sublattices have spins σ ($\pm 1/2$) and spins S ($\pm 1,0$), in the presence of an oscillating magnetic field, has been studied with Monte Carlo techniques. The model includes nearest- and next-nearest-neighbor interactions, a crystal field, and the oscillating external field. By studying the hysteretic response of this model to an oscillating field, we found that it qualitatively reproduces the increasing of the coercive field at the compensation temperature observed in real ferrimagnets, a crucial feature for magneto-optical applications. This behavior is basically independent of the frequency of the field and the size of the system. The magnetic response of the system is related to a dynamical transition from a paramagnetic to a ferromagnetic phase and to the different temperature dependence of the relaxation times of both sublattices.

INTRODUCTION

The behavior of ferrimagnetic compounds in the presence of oscillatory fields has long been used for technological applications, such as high-density magneto-optical recording,[1] but little is known about the mechanisms responsible for this behavior. In a ferrimagnet, the different temperature dependencies of the sublattice magnetizations raise the possibility of the appearance of compensation temperatures: temperatures below the critical point, where the total magnetization is zero.[2,3] It has been shown experimentally that the coercive field is very strong at the compensation point favoring the creation of small, stable, magnetic domains.[4] This temperature dependence of the coercivity near the compensation point can be applied to writing and erasing in high-density magneto-optical recording media, where the temperature changes are achieved by local heating the films by a focused laser beam. It has been shown that magneto-optic thin films with compensation temperatures higher than room temperatures can attain a direct overwrite capability.[5] As far as we know there have been only very few crude attempts to reproduce theoretically the increase of the coercivity near the compensation point using mean-field approaches.[6] Recently new classes of magnets have been synthesized with molecular organic chemistry techniques.[7] Biocompatible, organic materials, optically transparent, with spontaneous moments at room temperature are not far from reality. Ferrimagnetic ordering seems to play a fundamental role in some of these materials. Ferrimagnetic compounds, called Prussian blue analogs, with a critical temperature of 240 K have been reported.[8] Organometallic compounds as the amorphous $V(\mathrm{TCNE})_x y(\mathrm{solvent})$ where TCNE is tetracyanoethylene are believed to have ferrimagnetic structure and ordering temperatures as high as 400.[9] Some of these compounds have compensation temperatures near 30 K.[10] Most of these compounds have been synthesized by assembling molecular building blocks of different magnetic moments in such a way that adjacent magnetic moments are antiparallel.[7] Since real ferrimagnets have extremely complicated structures, mixed Ising models have been introduced as simple systems that can show ferrimagnetic behavior[11–13] and may show compensation points when their Hamiltonian includes second-neighbor interactions.[14] In this paper, we present a Monte Carlo study of a mixed Ising spin system, where spins that can take the values $\pm 1/2$ and spins that can take the values $\pm 1,0$, are nearest neighbors on a two-dimensional square lattice and interact antiferromagnetically. Spins of the same type are next-nearest neighbors. We analyze the magnetic response of this system in the presence of an oscillating magnetic field. From these studies, we determine the dynamic order parameter, the coercive field and their variation with temperature, frequency, amplitude of the applied field, and size of the system. The results reproduce the rapid increase of the coercivity at the compensation temperature. The dynamical order parameter calculations suggest that the model exhibits a phase transition between a paramagnetic and a ferromagnetic region. A similar result was observed by a mean-field study of a simpler version of this model.[15] Mean-field approaches and Monte Carlo simulations indicate the presence of a dynamical phase transition in a kinetic Ising model.[16,17] However the distinctive behavior of the coercive field at the compensation temperature in ferrimagnets seems to be related to the different relaxation times of the sublattices.

THE MIXED ISING MODEL

Our model consists of two interpenetrating square sublattices. One sublattice has spins σ that can take two values $\pm 1/2$; the other sublattice has spins S that can take three values, $\pm 1,0$. Each S spin has only σ spins as nearest neighbors and vice versa.

The Hamiltonian of the model is given by

$$\mathcal{H} = -J_1 \sum_{\langle nn \rangle} \sigma_i S_j - J_2 \sum_{\langle nnn \rangle} \sigma_i \sigma_k + D \sum_j S_j^2 - H(t) \left(\sum_i \sigma_i + \sum_j S_j \right), \quad (1)$$

where the J's are exchange interaction parameters, D is the crystal field, and H is an oscillating magnetic field of the form

$$H(t)=H_0\cos(\omega t), \tag{2}$$

where ω is the frequency of the external field, its period is given by $\Theta=2\pi/\omega$. The J's, D, and H_0 are all in energy units. We choose $J_1=-1$ such that the coupling between nearest neighbors is antiferromagnetic.

Previous results with Monte Carlo and transfer-matrix techniques have shown that the J_1-D model (J_2 and H are equal to zero) does not have a compensation temperature. These studies show that a compensation temperature is induced by the presence of the next-nearest-neighbor (nnn) ferromagnetic interaction, J_2, between the $\pm 1/2$ spins. The minimum strength of the $J_2>0$ interaction for a compensation point to appear depends on the other parameters of the Hamiltonian.[14]

MONTE CARLO CALCULATIONS

We use standard importance sampling techniques to simulate the model described by Eq. (1) on a $L\times L$ square lattice with periodic boundary conditions. Configurations are generated by randomly choosing spins on the lattice and flipping them one at a time according to a heat bath algorithm. In each complete sweep through the lattice $L\times L$ sites are visited. Each Monte Carlo step per spin is associated with a time interval τ_S such that the frequency of the external field can be written as

$$\omega=\frac{2\pi}{(N)\tau_S}, \tag{3}$$

where N is the number of Monte Carlo steps per spin necessary to cover an entire cycle of the field. To perform the simulations, we arbitrarily choose τ_S to be one, such that $\Theta=N$. Our program calculates the sublattice magnetizations per site at the time t defined as

$$M_1(t)=\frac{2}{L^2}\sum_j S_j(t), \quad M_2(t)=\frac{2}{L^2}\sum_i \sigma_i(t) \tag{4}$$

and the total magnetization per spin at the time t, $M(t)=\frac{1}{2}[M_1(t)+M_2(t)]$. The averages are taken over all configurations, the sums over j are over all sites with S spins, and the sums over i are over all sites with σ spins. Each sum has $L^2/2$ terms.

The compensation temperature is defined as the temperature below the critical, $T_{\rm comp}<T_{\rm crit}$, where the two sublattice magnetizations cancel each other such that the total magnetization is zero, i.e.,

$$|M_1(T_{\rm comp})|=|M_2(T_{\rm comp})| \tag{5}$$

and

$$\mathrm{sign}[M_1(T_{\rm comp})]=-\mathrm{sign}[M_2(T_{\rm comp})]. \tag{6}$$

To characterize the time behavior, we calculate the dynamical order parameter Q defined as

$$Q=\frac{2\pi}{\omega}\oint M(t)dt. \tag{7}$$

The closed integral implies that the integral is performed over a cycle of the external magnetic field.

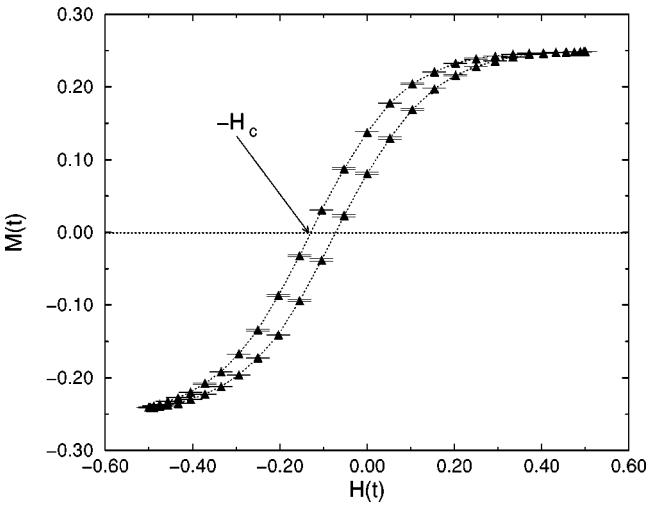

FIG. 1. Hysteresis loop ($J_2=6$, $D=-1.9$, $k_BT=0.5$, $\omega=\pi/30$). The coercive field is indicated.

RESULTS

The value of Q is calculated by averaging its values over 100 cycles of the external field, once the system is in its stationary state. Most of the measurements were done for a $L=40$ lattice. Lattices of different sizes were used to study the finite-size effects. In Fig. 1, we show a hysteresis loop, $M(t)$ vs $H(t)$, for a particular combination of parameters in the Hamiltonian. The coercive field H_c is defined as the minimum value of the external field needed for the total magnetization to go to zero, as is indicated in the figure. In Fig. 2 and Fig. 3 we show the coercive field vs the temperature for oscillating fields of several amplitudes, H_0. In the same figures, we also plot the total magnetization for the equivalent system subject to a constant field of magnitude H_0. Notice that the compensation temperature, defined as the point where the total magnetization is zero, previous verification that Eqs. (5) and (6) are satisfied, increases with the magnetic field, whereas the temperature at which the magne-

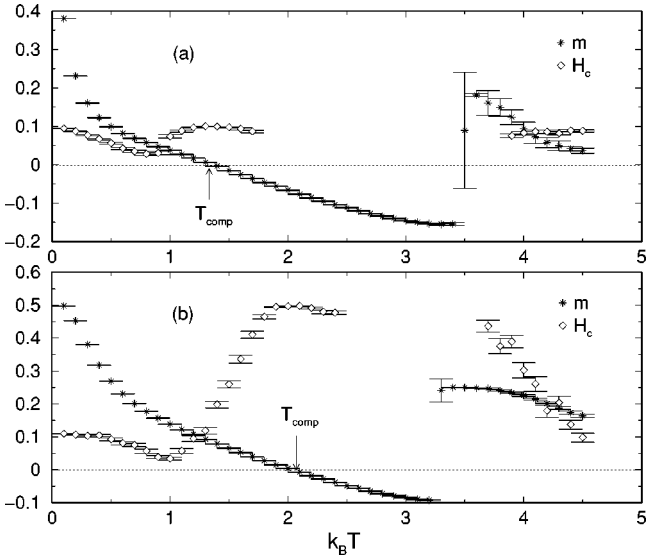

FIG. 2. Coercive field and magnetization vs temperature. ($J_2=6$, $D=-1.9$, $\omega=\pi/100$.) (a) $H_0=0.1$. (b) $H_0=0.5$. Notice the discontinuity in the coercive field.

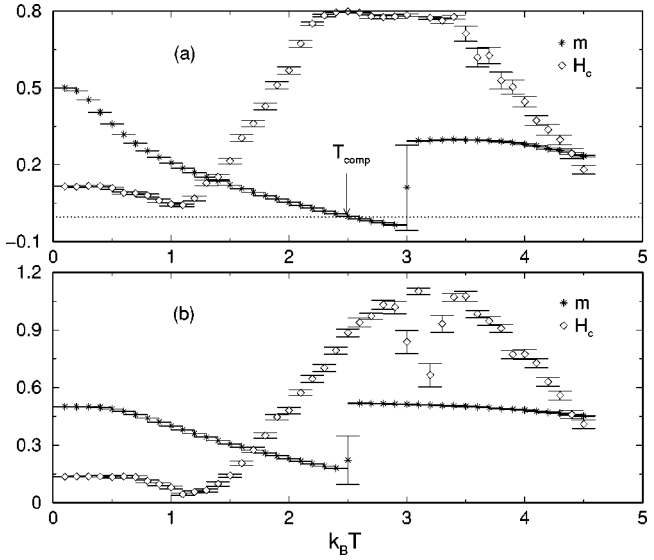

FIG. 3. Coercive field and magnetization vs temperature. ($J_2=6$, $D=-1.9$, $\omega=\pi/100$.) (a) $H_0=0.8$. (b) $H_0=2.1$. For this choice of parameters there is no compensation point for $H_0>1$.

tization becomes discontinuous does the opposite. At a certain field, which amplitude depends on the parameters of the Hamiltonian, both temperatures become equal and for any field of larger amplitude, there is no more compensation point, as can be seen in Fig. 3(b). From the figures, it is clear that the coercive field increases in the vicinity of the compensation temperature where it reaches its maximum. These results are summarized in Fig. 4. As expected, the maximum value of the coercive field at the compensation temperature is given by H_0.

It is interesting to notice the asymmetric behavior of the coercive field around the compensation point. In the low-temperature region, $T<T_{\rm comp}$, the coercive field decreases with increasing T until it reaches a minimum, after which it grows rapidly reaching its maximum at $T_{\rm comp}$, when $T>T_{\rm comp}$ the coercive field decreases. Notice that for small values of H_0, there is a range of temperatures for which the

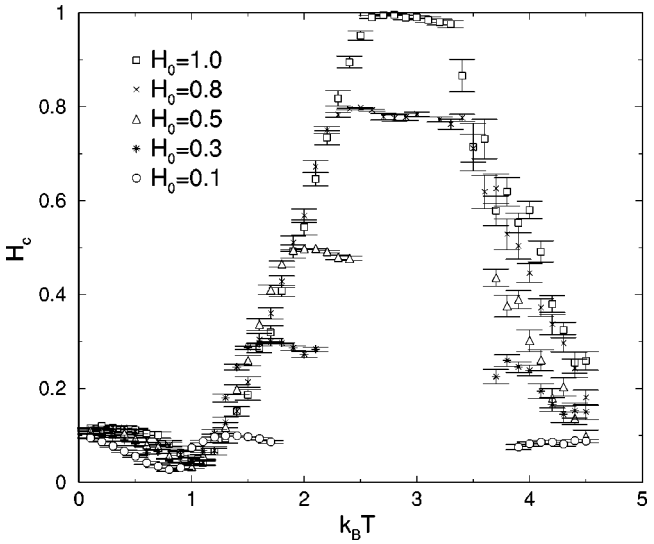

FIG. 4. Coercive field vs temperature ($J_2=6$, $D=-1.9$, $\omega=\pi/100$). The maximum value of H_c is given by H_0.

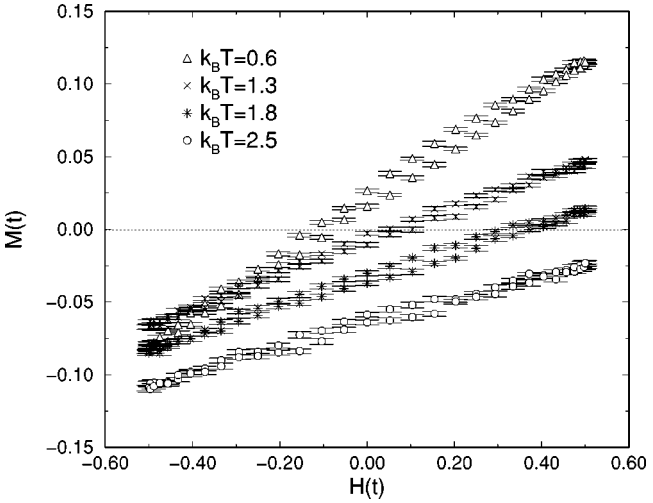

FIG. 5. Hysteresis loop ($J_2=6$, $D=-1.9$, $\omega=\pi/100$, $H_0=0.5$). Notice that for high temperatures there is no coercive field (see Fig. 6).

coercive field is not defined (see Fig. 4). This behavior of the coercive field has been observed experimentally.[18] This result can be understood by looking at Fig. 5, where it is shown how the hysteresis loop changes with the temperature. As the temperature increases the loop moves in such a way that the coercive field increases until it reaches its maximum, after which, if the temperature keeps increasing, the loop stays below (or above) the $M=0$ axis without crossing it, meaning that the applied field is not strong enough to flip the spins. If we look at Fig. 6, where we plot the coercive field and the dynamical order parameter vs the temperature, we see that there is a dynamical phase transition between a paramagnetic region $Q\approx 0$ and a ferromagnetic region $Q\neq 0$, the region where the coercive field is not defined is well into the ferromagnetic phase where the magnetization does not change sign.

By changing the size of the system and studying the behavior of the coercive field, see Fig. 7, we notice that there are finite size effects, particularly evident for small systems

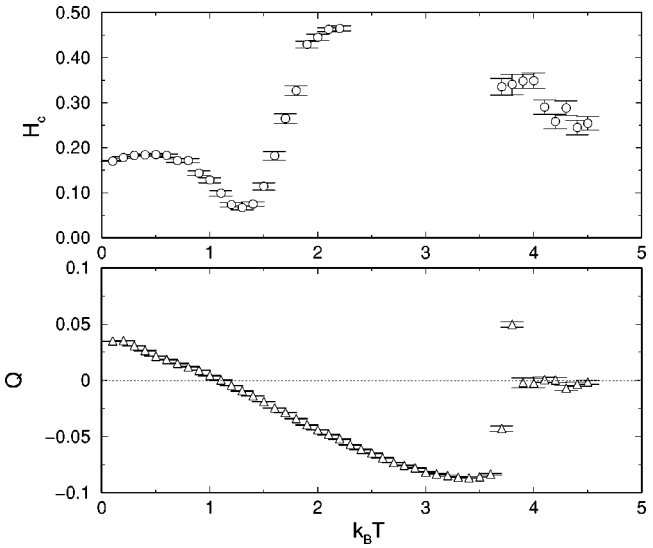

FIG. 6. Coercive field and dynamical order parameter vs temperature ($\omega=\pi/30$, $H_0=0.5$, $J_2=6$, $D=-1.9$).

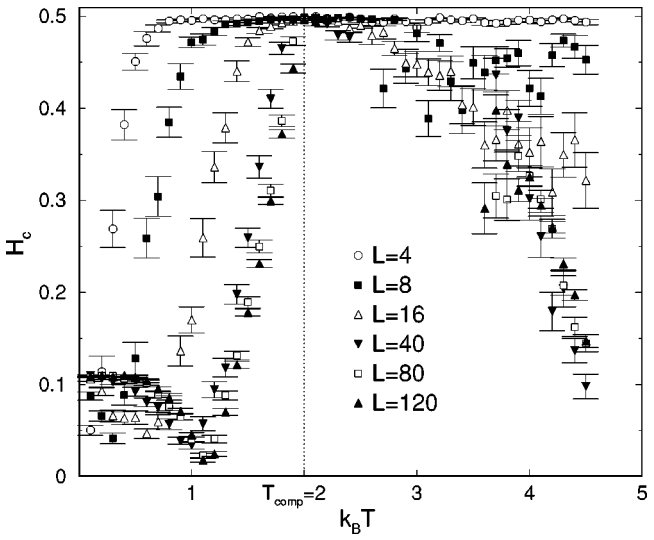

FIG. 7. Coercive field vs temperature for different lattice sizes ($\omega=\pi/100$, $H_0=0.5$, $J_2=6$, $D=-1.9$).

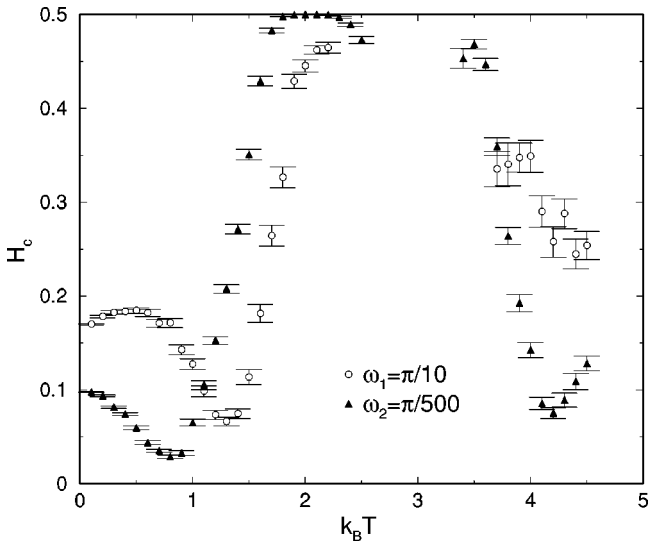

FIG. 9. Coercive field vs temperature ($H_0=0.5$, $J_2=6$, $D=-1.9$).

($L<20$). However, for larger systems, the location of the peak of the coercive field around the compensation temperature seems to be independent of the size of the system. For small systems ($L<20$), the peak of the coercive field appears before the system reaches its compensation temperature. Also, the coercive field seems to decrease more rapidly for the larger systems.

In Fig. 8, we present some results that show the dependence of the coercive field with the size of the system. These results agree qualitatively with the experimental behavior of magnetic films and nanostructured Fe and Ni samples[19] for which the coercitivity depends on the average size of the grain. The size dependence of the coercive field is very similar to the size dependence of the switching field of a kinetic Ising model (field at which magnetization reversal is thermally induced on experimental time scales for given temperatures and system sizes), which behavior has been shown to be strongly dependent on the modes by which the system decays.[20,21]

Next, we explore how the results depend on the frequency. In Fig. 9, we show how the coercive field vs the temperature changes for different values of the frequency of the external field. We found a quite different response to the frequency of the magnetic field depending on the dynamical phase of the system. In the paramagnetic phase ($Q\approx 0$, see Fig. 6), the coercive field is larger for systems driven by fields with higher frequency, but in the ferromagnetic phase ($Q\neq 0$), just the opposite happens, as can been seen in Fig. 9. This behavior is related to the temperature dependence of the relaxation time in the different regions.[21,22] In the ferromagnetic phase we must take into account the relaxation time of both sublattices the σ and the S, whereas in the paramagnetic phase only the σ one is relevant because the S lattice follows the field with almost no delay.[15] We also notice in Fig. 9, that when the field has a high frequency, the maximum value of the coercive field (that occurs at the compensation temperature) does not reach the field amplitude, i.e., the coercive field does not reach its saturation value.

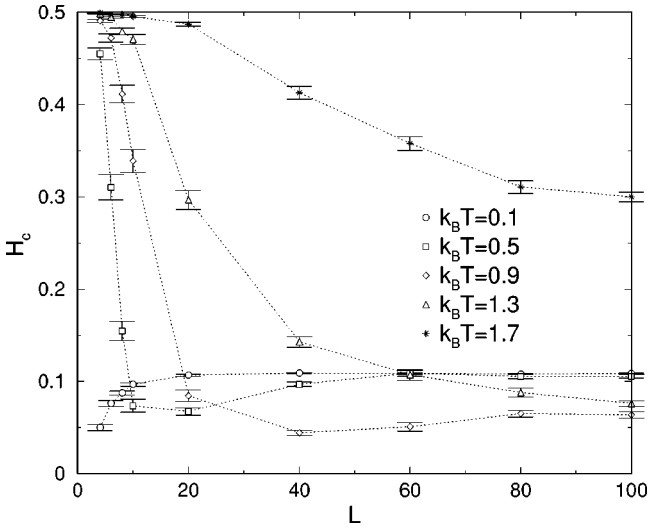

FIG. 8. Coercive field vs lattice size ($\omega=\pi/100$, $H_0=0.5$, $J_2=6$, $D=-1.9$). The lines are guides for the eye.

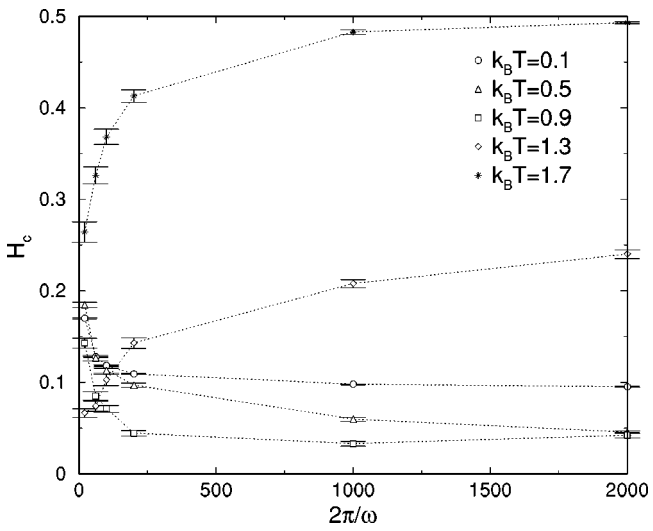

FIG. 10. Coercive field vs inverse frequency ($H_0=0.5$, $J_2=6$, $D=-1.9$). The lines are guides for the eye.

In Fig. 10, we show the behavior of the coercive field vs the inverse frequency for the different values of the temperature. If the field has a long period, the coercive field seems to reach a value that is independent of the frequency and depends on the temperature. Again we expect that this behavior is related to the temperature dependence of the relaxation times in the different phases.

V. CONCLUSIONS

We have applied a Monte Carlo algorithm to the study of the magnetic response of a mixed Ising ferrimagnetic model to an oscillating magnetic field. We found that this model gives very good qualitative agreement with the magnetic behavior of real ferrimagnets. It shows a rapid increase of the coercive field at the compensation temperature, a crucial feature that makes ferrimagnetic compounds extremely useful for thermo-optical applications. It also reproduces qualitatively the dependence of the coercitivity with the size of the sample observed experimentally. The results show the existence of a dynamical phase transition in which the mean-period averaged magnetization Q changes from $Q \approx 0$ to $Q \neq 0$. Work in progress indicates that, as recent studies show is also the case for the kinetic Ising model,[21,22] some aspects of the hysteretic response as its dependence on the frequency and amplitude of the oscillating field, depends on the metastable decay mode. The behavior in the different regimes can be explained by the nucleation mechanism (i.e., single-droplet or multidroplet) by which the system decays.[23]

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