Power-law relaxation decay in two-dimensional arrays of magnetic dots interacting by long-range dipole-dipole interactions

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(Received 30 May 2001; published 25 October 2001)

The magnetic relaxation of two-dimensional arrays of dipolar coupled magnetic dots has been measured and simulated. Arrays (50×50) with perpendicular magnetized Co dots ($2\ \mu m\times2\ \mu m$) were patterned using a high resolution Ga^+ focused ion beam irradiation. Magnetic domain pattern and time relaxation of the dot arrays were investigated using Faraday magneto-optical microscopy. For arrays designed with high irradiation doses ($\geqslant0.5$ nC/cm), the magnetic relaxation of the array proceeds by the magnetization reversal of individual dots and follows a power-law time decay. The long-range character of the dipolar interaction is found to be responsible for magnetic relaxation with a power-law decay. Monte Carlo simulations, based on a modified Ising Hamiltonian, reproduce this time dependence, and show that the power law is not a consequence of either the finite size or the boundary of the arrays, and it is independent of the shape of dots as well.

DOI: 10.1103/PhysRevB.64.184440 PACS number(s): 75.75.+a

I. INTRODUCTION

Systems of reduced dimensions such as small particles and thin films have experienced much interest over the last decades. One intriguing aspect of reducing the size of magnetic systems to microscales or nanoscales, emerges from the influence of the dipolar interaction on their magnetic properties. The long-range character of the dipolar interaction induces drastic limitations in the design of future nano-size devices or in the realm of high-density magnetic recording.

A conventional approach to magnetization reversal studies of assemblies of small magnetic particles, takes into account effects of disorder, size and/or orientation distributions, but treats particles as being independent. The most simple models for interpreting magnetic aftereffect or magnetization reversal dynamics in thin films consider that the magnetic layer is composed of independent activation volumes. In these two above cases several types of time relaxation dependencies are predicted, for example logarithmic or exponential like decays.[1] Other studies, mostly based on numerical calculations, that include the dipolar interaction, have reported interesting outcomes for the magnetization dynamics.[2–6] However, to date, there is a lack of convincing *experimental* works on the role played by the dipolar interaction on the magnetic relaxation phenomenon.

In the present work, we have studied the influence of the long-range dipole-dipole interaction on the magnetic relaxation of two-dimensional arrays of perpendicularly magnetized Co dots, using magneto-optical microscopy and Monte Carlo simulations. We mainly focus our attention on the non-equilibrium magnetic properties for Co dots coupled only by dipolar interaction. However, by focused ion beam (FIB) patterning, the exchange interaction between dots may be controlled and varied continously.[5] Almost like a toy model, two-dimensional arrays of micron size Co dots, fabricated by the FIB technique, stands as an ideal system to study the effect of the dipolar interaction both experimentally and by numerical simulations. FIB line patterning allows to define an array of dots of identical geometry, each dot behaving as a single magnetic entity. Hence, unlike most previous experimental studies of magnetic relaxation on assemblies of small particles,[1] in our system the magnetic dots have exactly the same size, shape and magnetic properties, and form a regular two-dimensional array.

II. EXPERIMENTAL DETAILS

The virgin media for these experiments was a highly homogeneous Pt(3.4 nm)/Co(1.4 nm)/Pt(4.5 nm) layer structure, epitaxially grown on a $Al_2O_3(0001)$ crystal, using magnetron and rf sputtering. Arrays of 50×50 square dots were patterned on this Co film using high resolution FIB.[7] The $2\ \mu m\times2\ \mu m$ square dots were defined from the virgin film by sweeping a 20 nm wide (at FWHM) Ga^+ spot in two orthogonal directions. Like the virgin media, it has been shown that the dots exhibit a high perpendicular magnetic anisotropy.

The result of uniform ion beam irradiation is to modify the structural and magnetic properties of the ultrathin film. As under irradiation with He^+ ions, it has been demonstrated that the coercivity and anisotropy of Pt/Co/Pt films can be progressively reduced with increasing the He^+ irradiation fluence.[8] For intermediate fluences, the film may even become paramagnetic. At high fluences the film etching becomes predominant. Thus, by Ga^+ FIB patterning we were able to design dot arrays with varying the exchange coupling through the irradiated FIB lines separating the dots. Then, the modified exchange interaction has to be compared to the dipolar coupling.

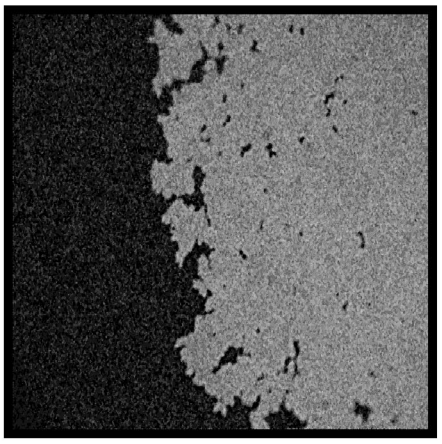
In our samples the lineic fluence in lines has been varied from 0.001 to 5 nC/cm (or from 2.08×10^{12} to 1.04

$\times 10^{16}$ ions/cm^2). Lines irradiated under low doses (<0.05 nC/cm) are still magnetic but show a reduced coercivity as compared to that of the virgin layer. In that case, the dots are coupled by both the exchange and dipolar interactions. For intermediate lineic doses ($0.1-0.25$ nC/cm), the central part of the lines becomes paramagnetic. At higher lineic fluences ($\geqslant 0.5$ nC/cm), the lines become completely etched, which means that the dots are now magnetically coupled by only the dipolar interaction.[5,7]

Static and dynamic magnetic properties of individual dots, and of the arrays as a whole, were measured using high resolution (~ 0.4 μm) polar magneto-optical microscopy.[9] Images were recorded using a cooled CCD camera and a green LED light source. The magnetic domain patterns were extracted by processing the difference between an image acquired under a saturating magnetic field H_s and the remanant image state, after applying a field H for a certain time. Hence, to follow the time evolution of the domain pattern from the saturated state, magnetic field of fixed amplitude H, lower than the coercive field was applied during a time t and images were recorded in zero field. The normalized relaxation curves were extracted from the magneto-optical images by adding ''up'' or ''down'' saturated magnetic dots. Since the nucleation appears at low field at dot boundaries, because of the reduction in coercivity by irradiation in lines, domain walls propagate rapidly in dots as soon as the field exceeds their switching field. Then, nearly all dots may be treated as giant Ising macrospins. Only in a few defected dots (about 1%), domain walls remain pinned inside the dots, thus they are counted as reversed for a threshold occupancy overheading 0.5. Magneto-optic microscopy is especially suitable for dynamic measurements, due to its fast acquisition time and to its availability to investigate the magnetization reversal both at local or large scales.

Figure 1 shows the typical magnetic domain patterns for samples patterned under different fluences. For low fluences, less than 0.001 nC/cm, the relaxation takes place by a rapid domain wall motion from rare nucleation events, similarly to the virgin film [Fig. 1(a)]. For an intermediate fluence of about 0.1 nC/cm, the reversal occurs once again by domain wall propagation across the entire dot array, but here the domain walls are delimited by FIB patterned lines [Fig. 1(b)]. In this case, the irradiation is large enough to locally reduce the exchange interaction across the FIB lines, but not to cancel out it. Hence, the magnetic image shows two large domains separated by a domain wall composed of small portions of straight FIB lines. For fluences larger than 0.5 nC/cm, the irradiated lines have been completely etched and the system behaves like an array of discrete, magnetized square dots only coupled by dipolar interactions [Fig. 1(c)].

In the high fluence regime, when only the dipolar interaction is present, the demagnetized equilibrium domain structure exhibits large checkerboard regions,[7] i.e., magnetic dots tend to be surrounded by antiparalelly coupled first-nearest neighbors. The occurrence of a checkerboard pattern is consistent with the coupling of dots by the dipolar interaction due to its positive prefactor [see the Hamiltonian's below, Eqs. (1) and (2)]. We can also obtain the value of the dipolar field acting on each dot from measured hysteresis loops of the system as a whole. It is typically equal to few ten's of Oe for our dot arrays.[7]



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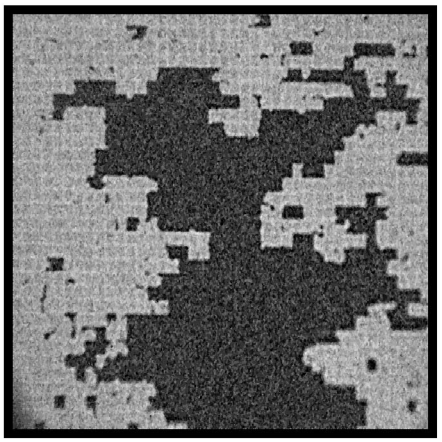

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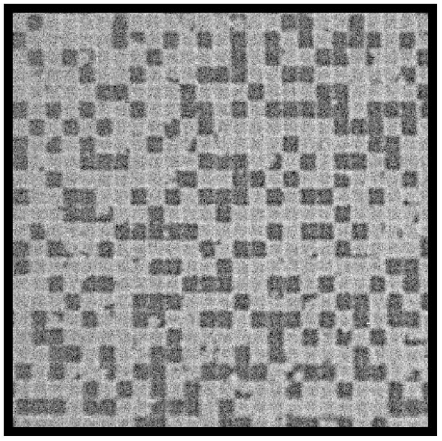

(*c*)

FIG. 1. Typical magnetic domain patterns for samples irradiated with different fluences, (a) low fluence (<0.001 nC/cm): large exchange interaction, (b) intermediate fluence ($0.1-0.25$ nC/cm): competition between exchange and dipolar intraction and (c) high fluence ($\geqslant 0.5$ nC/cm): only dipolar interaction.

Two distinct types of time dependences of the magnetic

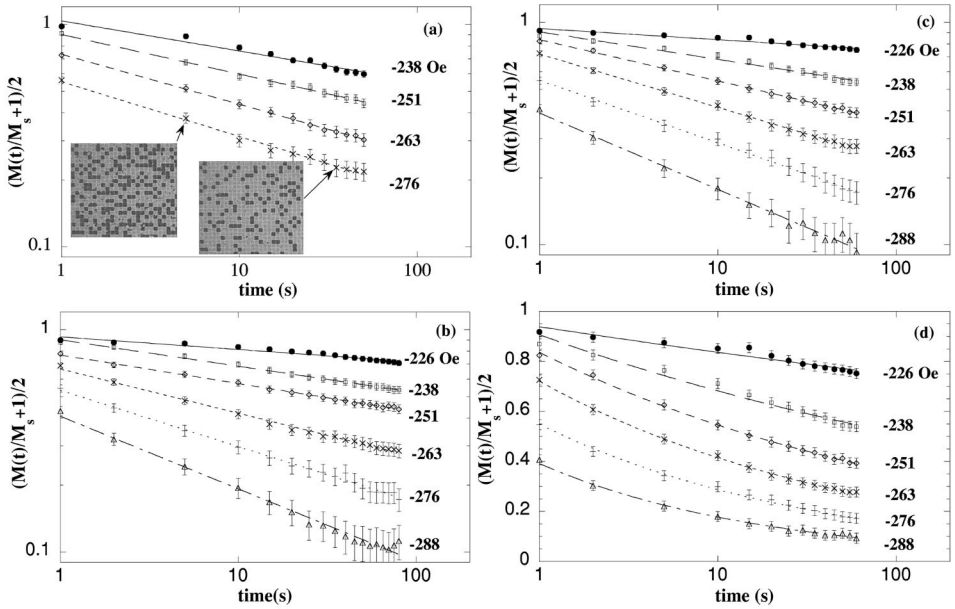

FIG. 2. Magnetic relaxation measurements at different applied magnetic field for samples irradiated with high fluences (0.5, 2.5, and 5 nC/cm) where dots (50 $\times$50) interact only by dipole-dipole interaction. The power-law decay can be observed in (a), (b), and (c), the data are plotted in a log-log scale. The same data of (c) plotted in a linear-log scale (d) shows that the relaxation does not follow a logarithm decay. The inset in (a) exhibits magnetic domain patterns during the magnetic relaxation at the central area of the array (25$\times$25 dots) at $t=5$ and 50 s. The continuos lines are mean square fittings.

relaxation were observed; they correspond to the characteristic limiting domain patterns described above (Fig. 1). For samples irradiated at low and intermediate fluences, the in-field magnetized state is initiated by a few nucleation centers allowing a rapid growth of domains by wall propagation. In this case, the magnetic relaxation follows a quasi-exponential time decay described by the Labrune-Fattuzo model[10] as in the virgin continuous films.[9–11]

The discreteness produced by lines irradiated under high fluence, i.e., small individual entities (square dots) coupled by the long-range dipole-dipole interaction, gives rise to a quite unusual time dependence for the magnetic relaxation of the full array—a power-law decay. This type of behavior is shown clearly in Fig. 2 on a plot of $[M(t)/M_s+1]/2$ in a log-log scale, for three different samples, irradiated with FIB fluences of 0.5, 2.5, and 5 nC/cm. After plotting the same data using different axis scales no doubt remains about the validity of such a power-law over two decades of time. For example, Fig. 2(d) replots data for the 5 nC/cm sample on a linear-log scale. It is obvious that the relaxation does not follow a logarithmic decay as it is usually observed for an ensemble of small independent magnetic particles.[1] Indeed, the result is even more convincing for curves with larger slope, because for a small exponent γ the power-law decay can be approximated to $t^{-\gamma}\approx 1-\gamma\ln(t)+1/2\gamma[\ln(t)]^2+\cdots$, so that the relaxation can be associated to a logarithmic decay.

Concerning the value of the exponent γ, which in some sense provides the relaxation rate, we find experimentally that γ increases quite linearly with the applied field (see Fig. 3). Furthermore, the same dependence on field is rather expected since the coercive field, 285 Oe does not vary much for all studied samples. The fact that the dynamics becomes field independent indicates that irradiation doses larger than 0.5 nC/cm are sufficient to destroy the exchange interaction between the dots.

III. MONTE CARLO SIMULATIONS

In order to gain some insight into the role played by the dipolar interaction on the magnetic relaxation of a two-dimensional array of magnetic dots with perpendicular anisotropy, we have performed Monte Carlo (MC) simulations based on a modified Ising Hamiltonian. Including the long-range dipole-dipole interactions and an external applied magnetic field, the Hamiltonian takes the simple form

$$\mathcal{H}=-J\sum_{\langle i,j\rangle}\sigma_i\cdot\sigma_j+\frac{\mu}{a^3}\sum_{i,j}\frac{\sigma_i\cdot\sigma_j}{r_{ij}^3}-H\mu\sum_i\sigma_i. \qquad (1)$$

The variable σ_i assumes the value ± 1 on a site i of a two-dimensional square lattice, where $+1$(-1) denotes the magnetic moments pointing up (down) along an axis perpen-

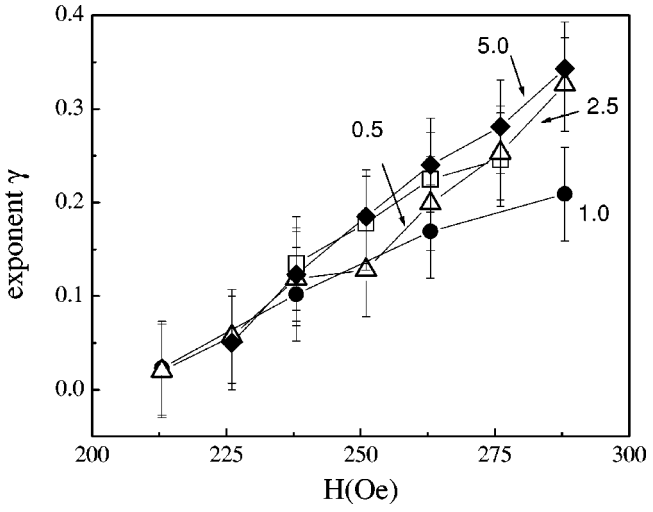

FIG. 3. Field dependence of the exponent γ obtained from magnetic relaxation measurements for samples irradiated with high doses (0.5, 1.0, 2.5, and 5 nC/cm).

dicular to the array plane. A remarkable distinction between the two first terms, exchange and dipolar interactions, is related to the range of interaction, the first is of short-range and the second is of long-range interaction (J is the exchange constant coupling). Another distinction is related to their opposite sign which causes competition between them: the exchange interaction favors a parallel alignment of magnetic moments, whereas the dipole-dipole interaction leads to their antiparallel alignment. The third term describes the interaction between the magnetic moments (μ) and the applied magnetic field (H). The distance between the moments r_{ij} is measured in units of the lattice constant a. In our model, we simulate the magnetic relaxation of dots coupled only by dipolar interaction, this situation corresponds to samples submitted to high irradiation doses. Thus, in the absence of exchange interaction, the Hamiltonian becomes

$$\mathcal{H}' = \sum_{i,j} \frac{\sigma_i \cdot \sigma_j}{r_{ij}^3} - h \sum_i \sigma_i . \tag{2}$$

For the sake of simplicity one normalizes the Hamiltonian by μ^2/a^3, and all constants become adimensional. The normalized field h is the only remaining parameter of the model.

In MC simulations we have used the single-spin-flip Metropolis dynamics formalism.[12] To simulate the magnetic relaxation one followed the same protocol than that used in the measurements. Starting with all moments aligned, the total magnetization relaxes with a time dependence determined by the temperature and the opposite applied magnetic field. The lattice is composed by $L \times L$ magnetic moments with L $=32$, 64, or 128. The time unit is one MC step per site (MCS), i.e., after $L \times L$ random choices and tests on the spins configurations. Although there is no obvious relationship between the simulation time (MC step) and the actual time, some insights (or qualitative comparison) on the influence of h on the time dependence of the magnetic relaxation can be achieved. We have used open and periodic boundary conditions. The former is a reflection of the experimental situation based on the construction of the finite size of the dot arrays. The periodic boundary conditions were employed using the Ewald summation technique. Furthermore, we included in the model an energy barrier which corresponds to the coercive field of a single magnetic dot.[3,4]

A simple picture such as a double well separated by an energy barrier (E_B) can be used to take into account the magnetic anisotropy (and coercivity) of a single dot. Each minimum corresponds to the energy of a given magnetic moment at site i pointing up or down, and the barrier corresponds to the energy needed to reverse the magnetic moment. An applied field and/or the interaction among moments produces an asymmetry in the double well. The energy minima are then shifted from the symmetric configuration by $\Delta E/2$, where ΔE is the energy difference between two minima and is given by the Hamiltonian [Eq. (2)]. Hence the energy difference between each minimum and the top of the barrier is $E_B \pm \Delta E/2$. In the framework of the MC method relevant to our problem, the probabilities of hoping from one minimum to the other can be described as follows : for $\Delta E > 0$ the probability of a moment flip, p_i is given by $p_i = \exp[-(E_B + \Delta E/2)/T]$, else for $\Delta E \leqslant 0$, $p_i = \min\{1, \exp[-(E_B - \Delta E/2)/T]\}$ (T is here the reduced temperature). This algorithm satisfies the detailed balance condition required by the MC method.[3]

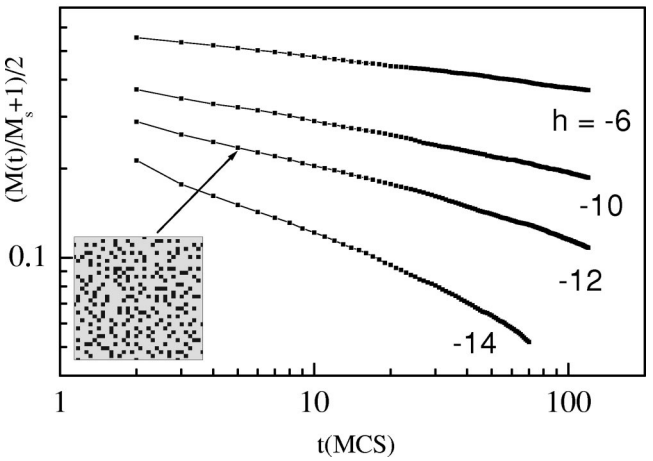

FIG. 4. An example of magnetic relaxation calculated through Monte Carlo simulations. One used open boundary conditions. Simulations were done for 64×64 dots, $T=1$, $E_B=10$, and varying the reduced applied magnetic field h. The rounding for higher fields is due to finite size effects. In the dipolar interaction the dots interact with all other dots. The inset shows the magnetic domain pattern recorded at 5 s for the central part of the array (32×32).

The time dependence of the magnetic relaxation was simulated in a lattice of 64×64 moments following the same protocol used in the experiments. Our main result is that the magnetic relaxation follows a power-law decay as proved on a log-log scale (Fig. 4). We used normalized parameters, temperature $T=1$ and energy barrier $E_B=10$, and the magnetization is averaged over 25 samples (runs). Notice that the parameter values are not unique, since any temperature well below the order-disorder critical temperature and field around the coercive field, represented by E_B, yield the same calculated time dependence. In a similar picture, a power-law time decay of the magnetic relaxation has been recently reported using MC simulations applied to perpendicular magnetized thin films.[6] In this case, a crossover from exponential to power-law decay has been evidenced depending on the ratio between exchange (short-range) and dipolar (long-range) interactions. Hence for arrays of dots, MC simulations lead to a magnetization time decay following a power-law decay $M(t)/M_s = -1 + 2\ t^{-\gamma}$, where $\gamma(T,h)$ is temperature and field dependent.

IV. DISCUSSIONS

In the light of MC simulations it is useful to discuss some possible implications on the magnetic relaxation induced by the boundary and the finite size of arrays and the range of the dipolar interaction. Such insights might provide some helpful suggestions for further experimental studies.

In relation to boundary and finite size effects, one notice that the size of the dot arrays (50×50) is relatively small

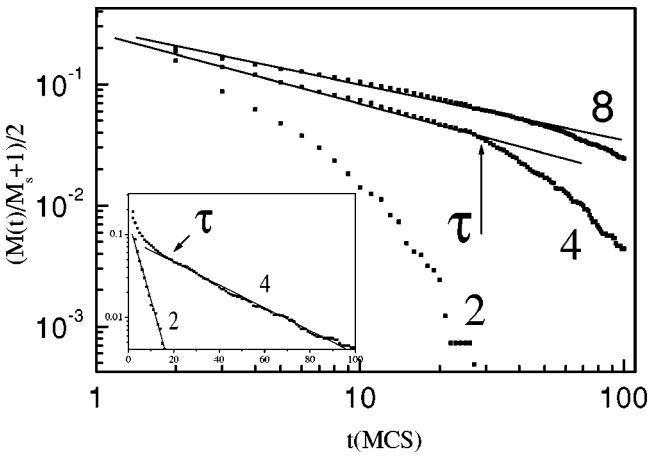

FIG. 5. Simulated magnetic relaxation varying the length of interaction on the dipolar interaction. One uses the same parameters of the Fig. 4 and $h=-10$. Taking into account in the dipolar interaction 2, 4, or 8 shells of neighbors, the effect of the range of interactions on the magnetic relaxation is clearly evidenced. For two shells of neighbors (short-range) the magnetic relaxation follows an exponential decay (see inset with a linear-log plot). Taking more neighbors (long-range), the power-law decay emerges and remains valid up to a certain time τ (see arrow). This time increases with the range of interaction.

compared to an assembly of magnetic particles in a form of a fine powder, where similar experiments on magnetic relaxation have previously been performed.[1] In order to verify whether the small size of the dot arrays could produce a particular time dependence we have performed MC simulations varying the system size. Thus, using open boundary conditions for arrays as small as 8×8 and as large as 128×128 we have obtained a power-law decay for the magnetic relaxation. To insure that no finite size effect bias the calculated time dependence, we have used periodic boundary conditions with the Ewald summation technique. In principle, this should reproduce the behavior of quite large systems. Using the Ewald technique with a large enough number of images (say 10–20 lateral images) and for the same parameters used in Fig. 4, the magnetic relaxation remains a power-law. Hence, MC simulations indicate that the power law decay measured in our samples is not an artificial outcome resulting from the boundaries or the size of arrays.

The long-range dipole-dipole interaction is the fundamental ingredient which gives rise to a power-law decay of the magnetic relaxation. It is quite natural to inquire how the *range* of dipolar interaction should affect the time dependence observed in experiments and simulations. In order to elucidate this question we have performed MC simulations for 64×64 dots using open boundary conditions with the range for the dipole-dipole interaction varying from first-nearest neighbors up to all dots. The essential result found is that the power-law persists up to a given time τ, and soon after the magnetic relaxation falls out to an exponential decay. It can be evidenced, for instance, for a range of interaction which includes 4 rings or shells of neighbors (see Fig. 5). At this range the relaxation changes from power-law to exponential around 20 MCS. The same behavior occurs at larger ranges, but accompanied with a larger time τ. It can be seen in the same Fig. for the range of interaction taking 8 shells of neighbors. On the other hand, for the dipolar interaction limited to only two shells of neighbors, the magnetic relaxation follows an exponential decay (see inset of Fig. 5). This suggests that, in experimental systems, the dots ''see'' (i.e., interact via dipole-dipole interaction) many other dots, at least for a range of interaction beyond 4 shells of neighbors. Bearing in mind that in the experimental situation of interacting dots placed many lattice steps away, it is a natural outcome to consider that the shape of dots, or even local details in their shape, does not play a dominant role in the dynamic magnetic properties of the entire array. The changing of time dependence is a direct manifestation of the occurence of a characteristic length. In our problem this characteristic length is nothing more than the magnetic correlation length among dots, which is a direct consequence of the long-range dipolar interaction. If the correlation length is large enough to enclose a large number of moments but smaller than the system size, finite size effects become apparent and the time dependence changes from power-law to exponential decay after some time (see, for instance, Ref. 13). Preliminary results show that the correlation length is temperature and magnetic field dependent.[14]

From the measured hysteresis loops, one are able to estimate the value of the dipolar field on the dots. This is only possible for one or two particular simple configurations (remember the dipolar field on any dot depends on the arrangement of all its neighbors), but the result gives some insights into the strength of the dipole-dipole interaction. For this estimation, we have considered the reversal field of the first and last magnetic dots to reverse their magnetization. The hysteresis loops of an ensemble of similar noninteracting dots will be very square, giving a well defined switching field, like in a continuous film. In real cases, the switching field can vary from dot to dot, often as a consequence of the different nucleation fields. The spread of the switching fields for an array of noninteracting dots is then expected and found to be much larger than that obtained for the continuous film.[15] In conterpart, the patterning technique favors the domain nucleation in low field at the edges of the dots. Thus, the spread in switching fields is not influenced here by the probability of nucleation *within* a dot. After nucleation at edges the wall propagation occurs when the dipolar field at the central zone of the dots just exceeds the propagation field. So, in the considered arrays, the spread in switching fields is mainly due to the dipolar fields experienced by each individual dot under the influence of all its neighbors. Hence the first dot reverses at a switching field $H_s=H_c-|H_d|$, while the last dot reverses at $H_s=H_c+|H_d|$ and the coercive field of the whole dot array is the same as that of the continuous film.[7] For arrays irradiated at high fluences ($\geqslant0.5$ nC/cm), the first and last dots to reverse their magnetization were observed directly from magnetooptical microscopy during hysteresis measurements, and in the present case one deduces a dipolar field H_d equal to 63 Oe. The same behavior in hysteresis loops has been observed for one-dimensional arrays of microwires interacting only by dipole-

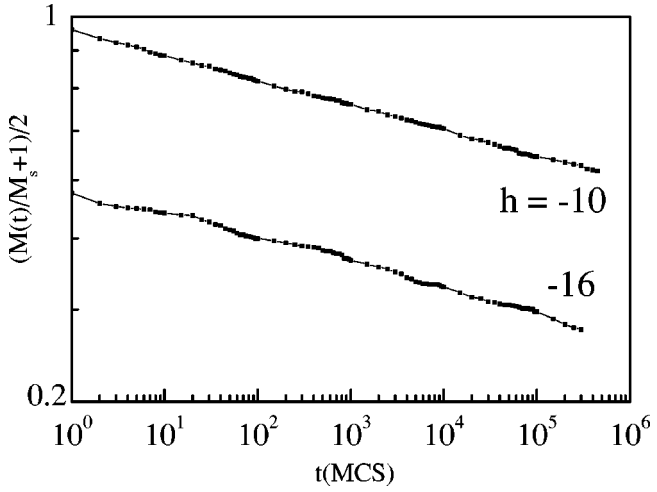

FIG. 6. Simulated magnetic relaxation with periodic boundary conditions (Ewald summation technique). The size of the array is 32×32 dots and one used 16 lateral images, $T=0.5$, $E_B=10$, and $h=-10$ or -16. Notice that the power law remains valid for 5 to 6 time decades.

dipole interaction.[16] A last remark concerns the dipolar field inhomogeneity inside the dots. Calculations of the dipolar field with same geometrical and magnetic parameters as in our samples have shown that the dipolar field can be considered as homogenous over an extended central zone of the dots but is far larger at their edges.[17] Therefore the magnetization reversal of the dots starts at their edges, and depending on the value of the dipolar field at their central zone a rapid domain wall propagation is observed.[7] Thus, the dipolar fields generated by all neighbors, and not only by first-nearest neighbors, contribute to the dynamics of magnetization reversal of the full array of dots.

As a final discussion it is interesting to remark that in the 1980's a large number of works have been devoted to non-equilibrium properties of disordered magnetic systems such as spin glasses. In spin glasses the magnetic relaxation has been found to follow a stretched exponential decay over many decades of time, but may be approximated by a power-law decay if the relaxation is measured over a relatively short period.[18] Although our measurements are plotted for only two decades of time due to experimental limitations, from simulations the power-law time decay is found to remains valid at least for five or six decades in MC simulations. Therefore, both experimental and MC simulations (Fig. 6) state that the power-law decay is the correct time dependence for a $2D$-array of magnetic dots interacting by dipole-dipole interaction.

In conclusion, we have studied the influence of the dipolar interaction on the magnetic relaxation of a two-dimensional array of magnetic dots, both by magneto-optics and Monte Carlo simulations. We have shown through Monte Carlo appropriate simulations, that the long-range character of the dipolar interaction is responsible for the power-law decay on the magnetic relaxation. This is the first experimental observation of such a time decay for an assembly of identical small magnetic elements. Monte Carlo simulations suggest that the observed time dependence found in experiments is not related to the finite size of arrays nor to boundary effects, and is independent on the shape of dots.

ACKNOWLEDGMENTS

L.C.S. is very grateful to Paulo Murilo C. de Oliveira for enlightening discussions on long-range correlated systems. The stay at Orsay of L.C.S. is financially supported by CNPq/Brazil. We also acknowledge the support of the SUB-MAGDEV European TMR network which provided a grant funding for R.H.

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