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## Influence of long-range interactions on the switching behavior of particles in an array of ferromagnetic nanostructures

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### Abstract

The interaction of Co/Pt nanodots is studied by means of the anomalous Hall-effect. On purpose the system of four dots is driven into a state of instability by applying a magnetic field perpendicular to the easy axis of magnetization. The dots become susceptible to thermal excitation and correlated switching is observed over large distances. The experimental finding is supported by theoretical simulations that reveal the importance of correlations and their influence at large length scales. This unexpected result sheds light on the general behavior of ensembles of systems with small energy barriers like superparamagnetic ensembles or switching of high density bit pattern media around the switching field.

 Online supplementary data available from [stacks.iop.org/NJP/16/083012/mmedia](http://stacks.iop.org/NJP/16/083012/mmedia)



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Keywords: magnetism, interaction, patterned media, magnetization dynamics, nanoparticle, nanodot

## 1. Introduction

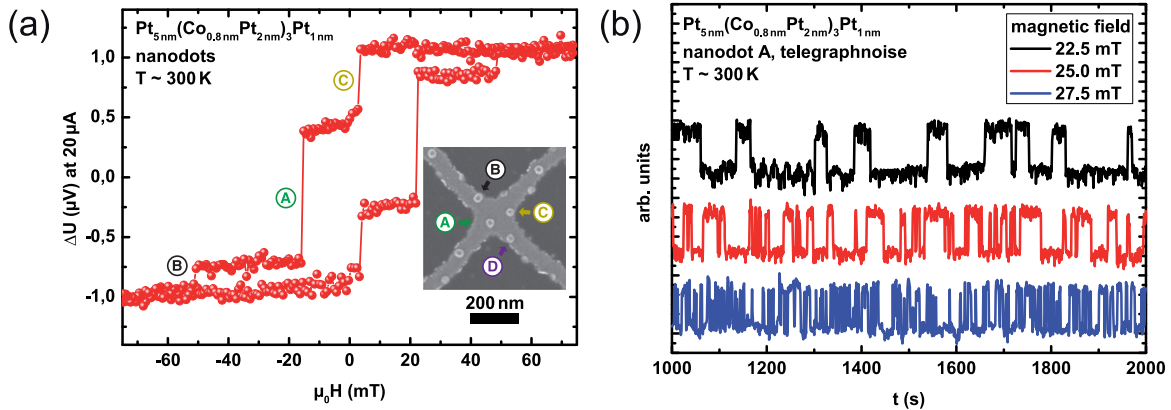
Magnetization reversal of ferromagnetic nanodots and their switching field distributions are relevant issues in fundamental nano-magnetism as well as in research on new concepts for magnetic storage [1–11]. Many basic studies, e.g. on superparamagnetism, are utilizing large ensembles of particles, in which the separation of particles is random and thus the behavior defined by distributions of distances and corresponding interactions. In storage media, on the other hand, the ultimate goal is to go for the highest packing density, which requires a minimal distance in between dots or grains [2–4, 12, 13]. As rule of thumb, one can assume in any such nanodot or nanoparticle assembly, that magnetostatic interactions are of minor importance when the separation of nanostructures is larger than the diameter of the particle [14–19]<sup>5</sup>. This is in general true and is abundantly proven in the state of remanence. The situation changes dramatically, however, in the case of thermally induced switching, in the vicinity of magnetization reversal or in arrangements of artificial structures that exhibit frustration (artificial spin ice [20–26]). In the latter situations the system becomes unstable against temporal changes in residual magnetic fields created in the surrounding. In particular it means that magnetic properties of nanodots in ensembles, e.g. either superparamagnetic features or switching field distribution in arrays of ferromagnetic dots, are strongly dependent on the magnetic environment [9].

In this paper we demonstrate that many-particle interactions are crucial for the magnetic behavior and the switching of particles as they can have a substantial impact on a surprisingly large length scale. To study this most relevant effect, we have devised an experimental pathway to artificially destabilize the magnetic state of particles by means of applying a magnetic field perpendicular to the easy axis of magnetization. We furthermore take full advantage of our experimental approach, which enables us to detect the response of every particle in a small ensemble individually, so that the magnetic behavior of each particle can be exactly compared to its magnetic surroundings at every point within the time window of our experimental set-up. With the experiments we prove unambiguously that interaction-induced correlated switching of magnetic particles occurs on length scales of up to three times the particle diameter, in contrast to the previous generally held belief.

## 2. Fabrication

Using nano-scale fabrication methods, the ferromagnetic dots are carved out from a planar Pt/Co/Pt trilayer ( $\text{Pt}_{5\text{ nm}}(\text{Co}_{0.8\text{ nm}}\text{Pt}_{2\text{ nm}})_3\text{Pt}_{1\text{ nm}}$ ) by ion milling utilizing a shadow mask built from  $\text{SiO}_2$  particles. Details of the multilayer fabrication utilizing dc magnetron sputtering and ion assisted sputtering have been published previously [27, 28]. The so-resulting ferromagnetic dots are then aligned with a Hall-cross that is created in the Pt layer underneath the magnetic

<sup>5</sup> For films with in-plane magnetization it was demonstrated that interaction becomes vanishing small above a separation in the range of dot diameter [15, 16]. In the case of ultrathin films with perpendicular magnetization the interaction range is apparently even smaller [17–19].



**Figure 1.** AHE response of a system of four nanodots in fields of different orientation (at 300 K). (a) Magnetic hysteresis of four nanodots obtained via AHE at room temperature. The field is oriented parallel to the easy axis of magnetization. The SEM image (inset) shows the four dots on the Hall-cross. Three clear jumps can be assigned to the reversal of individual dots: dot A switches at 20 mT while the smaller jumps at 5 and 50 mT are caused by reversal of dots C and B, respectively. A signal from dot D cannot be seen at room temperature. The diameter of the dots A, C, and D are about 36 nm while dot B has a diameter of about 40 nm. (b) AHE signal versus time (telegraph noise) for different fields applied within the film plane. The magnitude of the field was chosen to destabilize dot A to study thermal effects. The higher the field the higher is the frequency of switching.

film using electron beam lithography with negative resist. Further details of the fabrication process are described in the supplementary material.

It is well known that the magnetic anisotropy of such multilayer ferromagnets can be tuned via the thickness of the Co and/or Pt films [28–33]. For the investigations presented here, we specifically fabricated dots with weak perpendicular magnetic anisotropy. To achieve this, we start with films that exhibit a canted magnetization orientation, in which the interface-induced perpendicular anisotropy and the magnetostatic energy are nearly canceling each other out [33–38]. The structuring into dots (diameter  $d=36$  nm) causes then a reduction of the magnetostatic self-energy, which shifts the effective anisotropy  $K_{l,eff}$  into the range of a perpendicular easy axis of magnetization.

### 3. Results and discussion

To probe the magnetization state and behavior of individual dots, the anomalous Hall-effect is used, which is directly proportional to the out-of-plane component of magnetization. As in a conventional Hall-type measurements, a current is applied and a cross voltage is measured. The normal Hall effect is negligibly small in the Co/Pt material system and only the magnetization-dependent anomalous contribution is seen in measurements [39–41], thus giving access to the magnetization state in individual nanoparticles. Figure 1(a) displays the magnetization behavior of the system obtained in a single out-of-plane field sweep at room temperature for a system that consists of four dots on the Hall-cross (see inset of figure 1(a)). The assignment of hysteresis loop features to the individual ferromagnetic dots can be made by applying the current across adjacent leads and measuring the anomalous Hall voltage across the opposite leads as

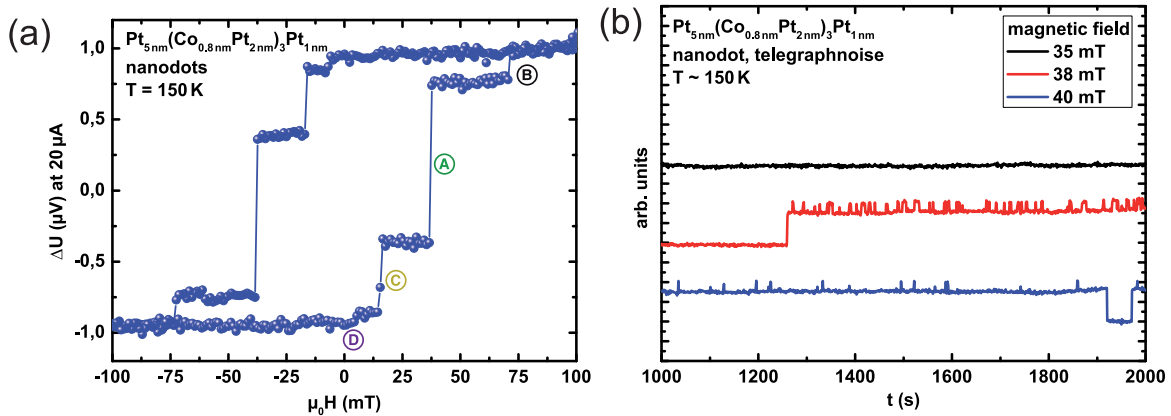
demonstrated in [42] or via the signal height by means of comparing the measured relative strengths with the calculated sensitivities for the conventional geometry [43–45]. The results of the latter analysis for the system shown in figure 1 reveal the following: the first reversal at about 5 mT corresponds to the switching of dot C, the second reversal at about 20 mT is produced by dot A and the last switching at about 50 mT corresponds to the reversal of dot ‘B’<sup>6</sup>.

For the purpose of having access to the thermally activated magnetization dynamics of these Co/Pt nanodots, an in-plane field is applied to modify and reduce the barrier height separating oppositely magnetized perpendicular states [46]. By tuning the field strength appropriately, the instability due to thermal agitation of the dot magnetization can be observed. As the dots reveal different coercive fields the temporal behavior of an individual dot can be separately studied via an appropriately tuned in-plane field. Figure 1(b) displays the telegraph noise obtained from dot A for three different field values. It is evident that the field lowers the barrier and the frequency of switching grows with increasing field strength. From the field dependent measurements the magnetic anisotropy of dot A is determined, which we have used to estimate the values of the anisotropy of the other dots (see supplementary material).

At  $T = 150$  K, the fourth dot also becomes visible in the hysteresis curves that are displayed in figure 2(a). There are again the three clear jumps, which can be associated with the dots that cause the room temperature signal. Their coercive fields are slightly increased in comparison to the room temperature measurement. The signal of dot D appears at very small fields. In relation to the low temperature results it becomes evident that at room temperature the switching of dot D is not observed since it is superparamagnetic and its small signal is partially masked by the larger signal of dot C. Although the hysteresis loops at both temperatures are very similar, the investigations of the telegraph noise by magnetic in-plane field reveal characteristic differences (figure 2(b)). The plots show the temporal signal for field strengths, for which telegraph noise sets in. The fields are higher than at room temperature since the magnetization states are more robust against thermal activation, which can also be seen from the fact that the coercivities are increased. At the lowest in-plane field strength (35 mT), no switching event is observed. Upon increasing the field strength to 38 mT, the switching that corresponds to the one shown in figure 1(b) sets in (same scales of ordinates). However, a second switching is visible in the experiment (figure 2(b)) that is faster and has a smaller signal height. At 40 mT the frequency of the switching event with smaller amplitude is decreasing again. The latter is discussed at the end of the paper. The telegraph noise also reveals that effectively three voltage levels are involved, which are called upper, medial and lower level in the following. The medial voltage level represents the state of lowest energy of the four dots as it is predominantly occupied in time which results from a detailed analysis of frequency distribution (see supplementary material).

In contradiction to the behavior at room temperature the signal heights are at variance with the signals obtained in the hysteresis loops. The hysteresis loop (figure 2(a)) shows jump heights of  $(1.13 \pm 0.03) \mu\text{V}$ ,  $(0.22 \pm 0.03) \mu\text{V}$ ,  $(0.46 \pm 0.03) \mu\text{V}$ , and  $(0.08 \pm 0.03) \mu\text{V}$  for dots A, B, C, and D, respectively. The signal heights in the telegraph noise are  $(0.83 \pm 0.05) \mu\text{V}$  for the big jump and  $(0.25 \pm 0.05) \mu\text{V}$  for the smaller jump. While the smaller jump height is close to the value found for dot B the large jump signal cannot be explained by any of the individual dot

<sup>6</sup> The slight asymmetry seen in the hysteresis (figure 1) is due to the stochastic influence of temperature on the reversal at fields about the coercive field.

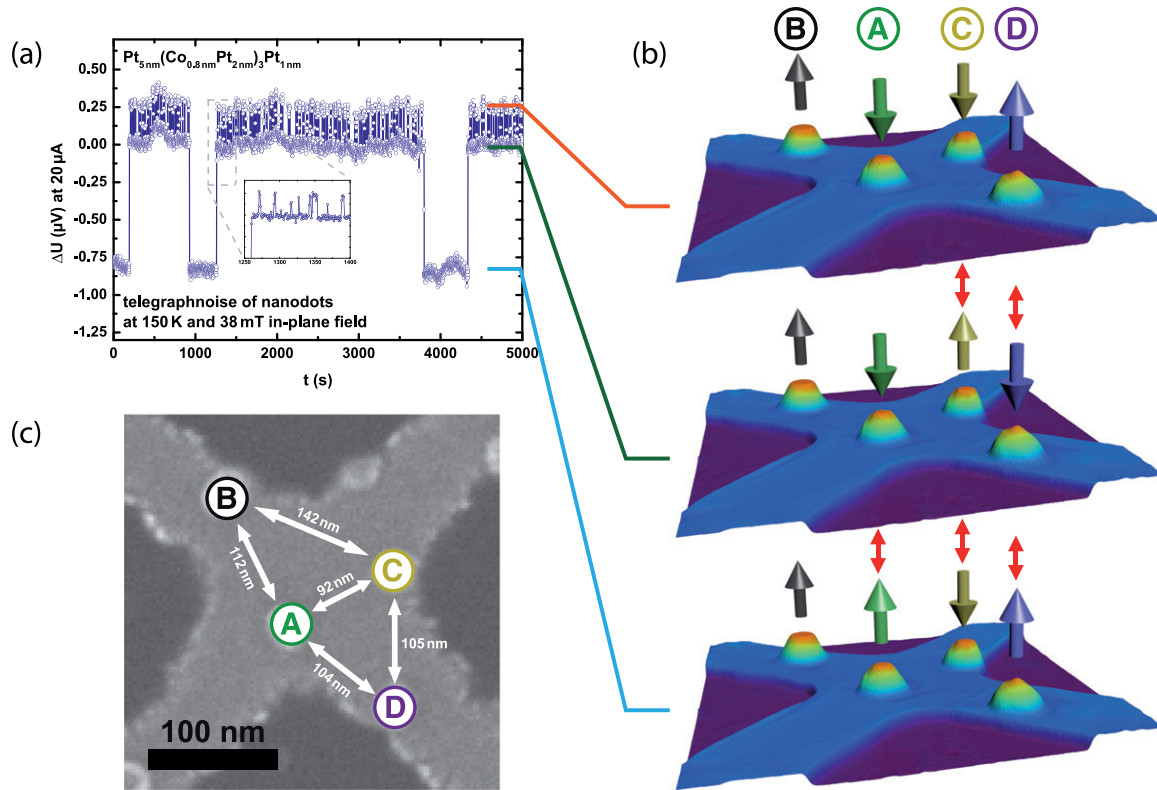


**Figure 2.** AHE response of a system of four nanodots in fields of different orientation (150 K). (a) Magnetic hysteresis obtained via AHE at 150 K for the same dot assembly as in figure 1. The field is oriented parallel to the easy axis of magnetization. A slight background signal has been subtracted. In addition to the three large jumps a small signal is found at low fields. The latter signal is due to the switching of dot D. In comparison to the measurement at room temperature the coercive fields are higher. (b) Telegraph noise observed at 150 K for different fields applied within the film plane. The ordinate and abscissa have the same scales as in figure 1(b). The field strengths are tuned to values that initiate thermal switching of dot A. At 35 mT there is no switching observable while at 38 mT switching between three levels are obtained (lower, medial and upper level). At fields above 40 mT the frequency of switching signature becomes less visible.

switching transitions obtained in the hysteresis loops. Even though the identification of the smaller signal seems to be straightforward, there are serious arguments against the switching of dot B. From the hysteresis curves it is evident that dot B has the highest coercivity and anisotropy of the four dots (see supplementary material). The latter means that assuming single particle switching the frequency should be the lowest from all particles. The switching between the medial and upper level, however, appears with a high repetition rate which obviously cannot be attributed to switching of dot B in a single switching scenario.

A measurement on an extended time interval is shown in figure 3(a) for an applied field of 38 mT. The temporal evolution reveals the already mentioned three voltage levels that are involved in the switching (inset). A detailed analysis of the width of the signal in the different levels can be found in the supplementary materials. Obviously, the system exhibits fast fluctuations in between the higher voltage levels (medial and upper). The total separation of the lower and upper voltage level is  $(1.08 \pm 0.05) \mu\text{V}$ . The value is apparently close to the signal of dot A in the hysteresis. When the value obtained in the hysteresis loop is corrected for the tilting of magnetization in the in-plane field, one could expect a signal height of  $1.05 \mu\text{V}$  for the reversal of dot A. The good agreement between the corrected and telegraph signal indicates that the net change between the lower and upper voltage level is caused exclusively by the reversal of dot A. Hence, we may conclude that the plain analysis of the experimentally measured voltage levels indicate that dot A is a key player in the observed thermal switching while other signals cannot be attributed to single dot switching.

Next, it is instructive to focus on the time scales of the switching events in figure 3(a). Utilizing the anisotropy that has been determined (see supplementary material) the effective



**Figure 3.** Telegraph noise and the corresponding states of the system consisting of four nanodots. (a) Telegraph noise obtained in an in-plane field of 38 mT over an expanded time period. It becomes evident that the signal reveals switching between three different levels. Detailed analysis of the jump heights leads to a scenario that is sketched in (b). (c) SEM micrograph of the cross. The separation of the dots are indicated.

barrier heights for single particle switching in the in-plane field can be estimated for dot A:  $\Delta E_A(38 \text{ mT}) \approx 37 k_B T$ <sup>7</sup>. The plain result is that dot A should not switch on the time scale of the experiment (at 150 K) while dots C ( $\Delta E_C(38 \text{ mT}) \approx k_B T$ ) and D ( $\Delta E_D(38 \text{ mT}) \ll k_B T$ ) should switch with a frequency that is too fast to be resolved within our experiment (assuming an attempt frequency in the range of  $10^{11}$  Hz). This means that experimental result and expected behavior (based on single particle potentials) disagree completely, as values for dots C and D are too large and those for dot A are too small. So an agreement cannot be achieved via a variation of the attempt frequency. Even more important, the inset in figure 3(a) demonstrates that the transition from the bottom level always ends in the medial level, which is definitely produced by a simultaneous switching of more than one dot. Hence, switching of single dots cannot explain the finding and correlated switching has to be assumed.

To get better insight into the highly complex energy landscape of the multi-dots system, simulations in the framework of Landau–Lifshitz–Gilbert spin dynamics have been performed. The dots have been treated as macroscopic dipoles. Exactly the same arrangement of four dots

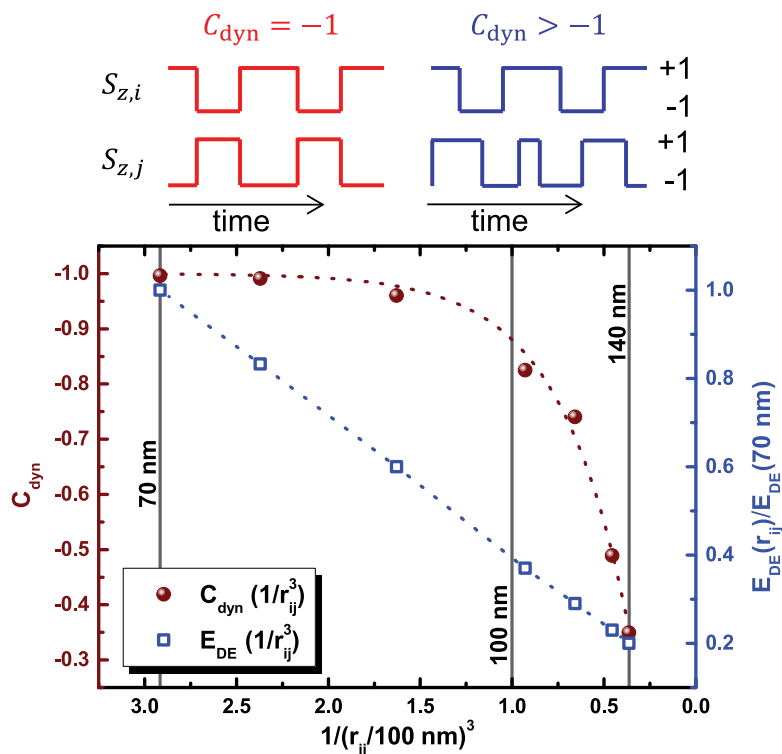
<sup>7</sup> Calculated under the assumption of a uniaxial potential and macrospin behavior. In this case the energy barrier for an in-plane field is given by  $\Delta E(H) = KV(1 - H/H_K)^2$  with  $\mu_0 H_K = 2K/M_S$ . The determined values for the anisotropies of the dots can be found in the supplementary material.

has been modeled and the dynamics of the switching behavior analyzed. In accordance with the experimental results it has been found that the dynamical behavior of the four dots is very different from that expected due to the energy barriers determined from the temperature dependent coercive fields of individual magnetic particles. The predominantly populated state is the one which minimizes the dipolar coupling and corresponds to the middle panel of figure 3(b). The calculations, however, revealed that this state is temporally degenerated with respect to the orientation of the dots D and C. In other words, on the timescale at which the dots A and B remain stable, their D and C counterparts perform many switching events between configurations of upper and middle panels of figure 3(b). This finding raises the exciting question of whether this instability may explain the deviation of the energy barriers from those of individual, non-coupled dots.

Generally, to answer this question one has to know the time dependent evolution of the complete energy landscape. The latter is a tremendous task, because the energy landscape is nine-dimensional (the magnetization of each macroscopic dipole  $i$  can be described by two spherical coordinates  $(\theta_i, \varphi_i)$  plus time). The best way to solve this problem is to analyze the dynamical correlation function  $C_{\text{dyn}} = 1/T \int_{-\infty}^{\infty} dt S_{z,i}(t)S_{z,j}(t+s)$  between pairs of magnetic moments  $i$  and  $j$ , where  $S_z$  is the  $z$ -component of magnetization of moment  $i$  and  $j$  at times  $t$  and  $t+s$ , respectively. This function gives the information whether a given state is still correlated after a delay time  $s$ . Hence,  $|C_{\text{dyn}}| \rightarrow 1$  corresponds to the correlated, in-phase switching, while  $C_{\text{dyn}} \rightarrow 0$  to stochastic noise. In the simulations this function was evaluated for vanishing delay  $s = 0$  and variable center-to-center distances  $r_{ij}$  to check for the degree of correlation for simultaneous switching. The dependence of the function  $C_{\text{dyn}}(r_{ij}^3)$  on dot separation is given in figure 4<sup>8</sup>. The distance is normalized with respect to a separation of 100 nm, which is comparable to the scales in the experiment (see figure 3(c)). In figure 4 the spheres give the time-averaged dynamical correlation, while the open squares show the decrease of the strength of dipolar interaction with  $1/r_{ij}^3$  (dotted curves as guide to the eye). The plot clearly shows that the dynamical correlation  $C_{\text{dyn}}$  decreases much slower than the static dipolar coupling. The plot clearly shows that the dynamical correlation decreases much slower than the static dipolar coupling. For the separation of dots C and D (105 nm) the correlation function is still more than 0.8 and thus correlated switching will be found. Hence, these two dots are switching in-phase and the stability of the anti-parallel configuration is increased. Even the farther separated dots ( $r_{ij} > 4d \approx 140$  nm) show strong correlations (see figure 4) which is responsible for the occasional switching of dot A although its anisotropy energy is much larger than the dipolar energy. The physical reason for the long range phenomenon is the minimization of the time-averaged or -integrated total potential energy of all dots. This manifests in the many-body dynamical correlations and prevents the magnetic moments from the dephasing. In other words the many-body dynamical effects, described here experimentally and theoretically, correspond to a minimization of a dynamical quantity—the spin dynamical version of the action—rather than to a mere minimization of single particle energies in a static viewpoint for individual dots.

<sup>8</sup> The calculation was performed assuming a damping constant  $\alpha = 0.2$  to achieve reasonable computation times as the convergence of the numerical simulation is low when going towards the damping parameter of Co ( $\alpha = 0.3$ ). The investigations of the correlation on damping (performed at lower damping parameters) reveal that the distance over which correlation is found increases with  $\alpha$ . Hence the effect is expected to be even more important.





**Figure 4.** Dynamical correlation-function  $C_{\text{dyn}}$  and dipolar energy  $E_{\text{DE}}$  as a function of dot separation. Comparison between the two-dot dynamical correlation-function  $C_{\text{dyn}}$  (spheres) and the normalized dipolar energy  $E_{\text{DE}}$  between to dipoles (open squares, normalized to the value at 70 nm) as a function of  $1/r_{ij}^3$  normalized to 100 nm. It is evident that the correlation-function decreases more slowly than the dipolar energy, responsible for the antiparallel correlation. The functional form of  $C_{\text{dyn}}(1/r_{ij}^3)$  is independent of the damping parameter  $\alpha$  (here  $\alpha=0.2$  is taken). Dotted curves are a guide to the eye.

With the result of the theoretical analysis it becomes clear that the fast switching between the middle and upper voltage level is a correlated switching of dots C and D. From the hysteresis measurement in figure 2(a) a signal height of  $(0.32 \pm 0.06) \mu\text{V}$  has to be expected, taking the field induced canting into consideration. A complete analysis of all transitions between all possible states reveals that the scenario proposed in figure 3(c) is the one that comes closest to the experimental observed value concerning level separation<sup>9</sup>. Both our theoretical and experimental results prove unambiguously that it is crucial to know very accurately the magnetic behavior of the dots in the surrounding on a scale that is far beyond the separation of nearest neighbors, to properly describe thermal switching or switching field distributions.

Finally, we would like to comment on the frequency decrease of switching when changing from 38 mT to 40 mT in figure 2(b). The latter is an indication for some asymmetry in our experimental set-up, in particular a small misalignment of the external field, which favors one over the other state. The effect of the misaligned field is also seen in the room temperature

<sup>9</sup> From the experimental point of view a fast switching of dot D in the lower state cannot be ruled out.

measurements (figure 1(b)) where the switching between the two states with same energy is slightly asymmetric in time.

#### 4. Conclusion

In conclusion it is shown that magnetostatic interactions can play an important role in the dynamic reversal process of magnetic dots even when the system is mainly determined by anisotropy barriers. The observed switching behavior demonstrates that correlations determine the switching. Dynamical processes have been observed that are in contradiction to a single particle scenario. The correlations can cause erroneous interpretations when only single particles are studied, neglecting the temporal behavior of the surroundings. The spin-dynamical-simulations and the calculation of the dynamical correlation function prove that dynamic correlations open up new reversal paths through the multidimensional energy landscape. The most intriguing theoretical outcome is that dynamical correlations are farther reaching than static interactions due to long range dipolar fields. It follows that correlations come into play when a magnetic system is close to the state of thermal instability. The experiments demonstrate that the interactions cannot be disregarded when thermally assisted switching is the focus of interest, which has wide ranging implications for the fundamentals of superparamagnetism as well as technical applications, such as heat or thermally assisted magnetic recording [1].

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