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Probing the energy barriers in nonuniform magnetization states of circular dots by broadband ferromagnetic resonance

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The time evolution of the ferromagnetic resonance output signal in the arrays of permalloy circular dots of submicron sizes was measured near the critical fields of the vortex nucleation and annihilation. Surprisingly short times of the transition from the quasiuniform to the vortex magnetization state (several milliseconds) were detected. The observed effects are explained by overcoming the field dependent energy barriers in the process of vortex core nucleation. The energy barrier values found from the time dependences of the ferromagnetic resonance peak intensities were compared with the ones calculated within the rigid vortex model. The rigid vortex model overestimates the nucleation barriers and a more adequate magnetization reversal model is needed. There is a strong dependence of the stable, metastable energy minima and energy barriers on the magnetic field and dot geometrical parameters.

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The problem of overcoming energy barriers by thermal activation is one of the longstanding fundamental problems in physics and physical chemistry. The corresponding relaxation times vary from nanoseconds to many years. The first description of the problem was made by van't Hoff and Arrhenius at the end of 19th century, who related a rate constant of a chemical reaction to the activation energy of the process. Then, the van't Hoff–Arrhenius equation was derived[1] from statistical thermodynamics within the transition state theory.[2] In the field of magnetism overcoming an energy barrier was described by Néel[3] for a particular case of uniform magnetization rotation in a uniaxial single domain particle. Then, the Néel's problem was solved rigorously by Brown.[4] A review of thermal relaxation is given by Aharoni[5] and simulation aspects are reviewed in Ref. 6. Calculation methods still need to be developed to find the energy barriers in an inhomogeneous magnetization state, where the magnetization reversal modes are essentially nonuniform, but such methods are necessary to calculate the magnetization switching times, switching fields, and thermostability of different magnetization states.

A typical stable, inhomogeneous magnetization configuration in submicron flat particles (dots) in a zero field is a vortex state (VS).[7] Applying an in-plane magnetic field H, the vortex can be expelled from the dot at an annihilation field H_{an}, forming a single-domain (SD) saturated state.[8] When decreasing the field starting from the SD state, a magnetic nonuniformity appears in the dot at a nucleation field H_{n}. This nonuniformity, called the C state (CS), can be described as a magnetization distribution created by a fictitious vortex centered outside the dot. With decreasing H the vortex approaches the dot, a vortex core forms at the dot edge, and propagates to the dot center,[8] i.e., the magnetization reversal can be represented as a vortex motion through the dot. But it is unclear how the reversal process occurs in time when the external field is changed. The reversal is inevitably related to overcoming energy barriers existing between different magnetization configurations: SD, VS, and CS.[9] The field dependence of these barriers allows one to predict the temperature dependence of $H_{\mathrm{an}}(T)$, $H_{\mathrm{n}}(T)$.[10]

In this Rapid Communication we present the measurements of the magnetization dynamics of permalloy (Py) circular dot arrays by applying an in-plane magnetic field H. The excited eigenfrequencies are determined by the magnetic energy minima. Probing the time evolution of the dot array high-frequency response we are able to make conclusions about changing the dot occupation numbers in different energy minima. Overcoming the energy barrier by an individual dot is essentially a random process, and its parameters can be found from many repeated experiments with the same dot,[11–13] whereas measurements of an integral signal of the dot array allow one to probe the magnetization dynamics in a statistically averaged dot magnetization state. The results of overbarrier jumps can be detected and magnetization relaxation parameters are estimated after one measurement.

The time of the transition of a dot between minimal energy states is determined by an energy barrier ΔE separating these states. The magnetic energy of a dot having the shape of a circular cylinder of radius R and thickness L depends on its magnetization distribution $\mathbf{M}(\mathbf{r})$, external field H, and values of R,L.[7] The magnetization of a thin dot $\mathbf{M}(\boldsymbol{\rho})$, where $\boldsymbol{\rho}$ is the in-plane radius vector, can be represented by the rigid vortex model (RVM) as a displaced vortex, $\mathbf{M}(\boldsymbol{\rho},H) = \mathbf{M}(\boldsymbol{\rho} - \mathbf{X})$, with the core center located in the point $\mathbf{X}(H)$.[7,8] Both $\mathbf{M}(\boldsymbol{\rho},H)$ and the magnetic energy $E(\mathbf{s})$ accounting for the exchange, magnetostatic, and Zeeman energy within the RVM are functions of only one parameter, the vortex core position $\mathbf{s} = \mathbf{X}/R$. This allows one to simplify essentially the problem of energy barrier description by reducing it to a one "reaction" coordinate s approach. The vortex core penetrating to the dot ($s \approx 1$) costs a finite energy and the corresponding energy barrier can be found.

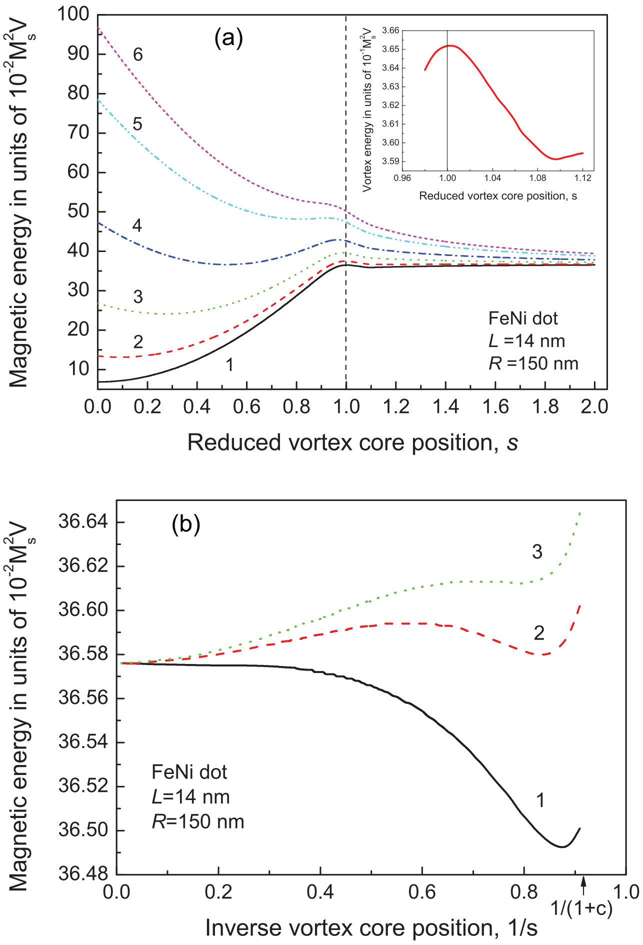

FIG. 1. (Color online) Magnetic energy of the dot vs vortex core position s $[W(s)+h]$ for different in-plane magnetic fields $h = H/M_s$: (a) $h = 0$ (1), $h = h_n = 0.066$ (2), $h = 0.20$ (3), $h = h_0 = 0.405$ (4), $h = h_{an} = 0.717$ (5), and $h = 0.90$ (6); (b) $h = h_n = 0.066$ (1), $h = 0.0745$ (2), and $h = h_C = 0.078$ (3). Inset: The energy barrier at the dot border at $H = 0$. The Py dot sizes are $R = 150$ nm, $L = 14$ nm, and the saturation magnetization is $M_s = 800$ G. The reduced vortex core radius is $c = R_c/R = 0.09$.

The dependences of the reduced energy $W(s) = E(s)/M_s^2V$ on s calculated within the RVM, where V is the dot volume, are plotted in Fig. 1 for different values of H. There is only a single energy minimum at $H \geqslant H_{an}$ (Fig. 1) corresponding to the SD state ($s = \infty$). The VS energy minimum appears at $H = H_{an}$ and becomes the lowest one at $H < H_0$.[7] $E_{SD} = E_{VS}$ at $H = H_0$ and the SD state is metastable at the fields $H_n < H < H_0$. A new additional metastable state (CS, $1/s \approx 0.9$) appears at $H = H_C$ slightly above H_n ($H_n < H_C < H_0$) and exists down to $H = 0$, resulting in a bistability of the dot magnetization states VS and CS at $H < H_n$. This strongly nonuniform, metastable C state was overlooked in Refs. 8 and 14, where only a quasiuniform C state with $1/s \ll 1$ was considered. The SD state corresponds to the $W(s)$ local minimum at $H > H_n$ and the local maximum at $H \leqslant H_n$, and there is transition over a small energy barrier of the SD state to a new CS with $1/s \approx 0.9$ (see Fig. 1) at $H \geqslant H_n$. The transitions from the metastable CS to the ground VS start at $H < H_n$. The dependence of the energy $W(s)$ near $s \approx 1$ (CS $\rightarrow$ VS energy barrier) at $H = 0$ is shown in the inset to Fig. 1, i.e., there are three stable states within the field interval $0 < H < H_{an}$ of interest, the VS, CS, and SD states. When decreasing H, the magnetization evolution for a typical L (10–50 nm) goes, starting from the saturated SD state through CS to VS. The situation is very different for ultrathin [1 monolayer (ML)] dots,[15] where the transition from the metastable SD to stable VS was detected at $H = 0$ and no intermediate CS minima appear.

When increasing the field from $H = 0$ the ground VS becomes metastable at $H > H_0$ (curve 4 in Fig. 1), but there is no transition to the stable SD ($s = \infty$) state up to $H = H_{an}$ due to a considerable energy barrier at $s < 1$, which decreases with increasing H and disappears at H_{an}. The field calculated by the RVM, $H_n = 0.066M_s \approx 53$ Oe, is in good agreement with the field extracted from the hysteresis loop, $H_n \approx 50$ Oe, which is measured by the Kerr effect. The calculated $H_{an} = 0.717M_s \approx 570$ Oe is higher than the experimental value, ~410 Oe. Possible sample defects should decrease H_n and increase H_{an} in comparison to the ideal, defect-free critical field values. This allows us to speculate that the defects do not contribute essentially to the dot magnetization reversal.[8]

If the dot is in the SD state in a field $H > H_n$ and we abruptly change the field to $H = 0$, then the dot transits initially to the CS (overcoming an energy barrier[7,14]), and eventually to the ground VS with $s = 0$. The transition time is determined by the energy barriers ΔE_{ij} between the SD state and CS, and then between CS and VS, where $i, j = \{\text{SD, CS, VS}\} \equiv \{1, 2, 3\}$. According to the Arrhenius–van't Hoff law the transition time (relaxation time) from state i to state j, τ_{ij}, is

$$\tau_{ij} = \frac{1}{\nu_0} \exp\left(\frac{\Delta E_{ij}}{k_B T}\right), \tag{1}$$

where k_B is the Boltzmann constant, T is temperature ($T = 293$ K in our case), and ν_0 is an attempt frequency. If the relaxation times τ_{ij} are found experimentally, then the corresponding energy barriers ΔE_{ij} can be calculated.

To check these ideas we consider a two-dimensional (2D) array of uncoupled ferromagnetic dots. The number of the dots in the array is $N > 10^6$ and the number of the dots in the i state is N_i. There are transitions of the dots from one metastable state to the other one, changing H. Accounting for the fact that interdot coupling is negligibly small, we assume that these transitions occur randomly and independently in each dot. Initially, the dot array stays a long time at the field $H > H_{an}$. Therefore, all the dots are in the SD state ($i = 1$), and $N_1 = N$, $s_1 = \infty$. At the field that decreases down to $H \geqslant H_0$ we still have $N_1 = N$, $s_1 = \infty$, because the SD state is the ground state. If we instantly change the field to 0, then in the initial moment $t = 0$ the occupation numbers preserve their values: $N_1 = N$, $s_1 = \infty$. Then, due to the instability of the SD state at $H = 0$ (see Fig. 1), the SD state dots transit to CS ($i = 2$), and then to VS ($i = 3$), where $s_3 = 0$.

The system of differential equations (the master equations) describing the time evolution of the occupation numbers N_i

after the field switch-off at $t = 0$ can be written in the form[16,17]

$$\frac{dN_i}{dt} = -(N_i - N_{i\infty})\sum_{j\neq i}\frac{1}{\tau_{ij}} + \sum_{j\neq i}(N_j - N_{j\infty})\frac{1}{\tau_{ji}}, \quad (2)$$

where τ_{ij} are given by Eq. (1), and $N_{i\infty}$ is the equilibrium number of the dots in the i state at $t \to \infty$. We assume that $N_{1\infty} = N_{2\infty} = 0$, $N_{3\infty} = N$, i.e., after the field switch-off all the dots transit to VS ($s = 0$) during a long enough time. On the basis of the RVM (see Fig. 1) we can consider that the transition times from the ground VS τ_{31}, τ_{32} are essentially higher than all other transition times. Accounting for the time hierarchy $\tau_{31} > \tau_{32} > \tau_{23} \gg \tau_{21} > \tau_{12}$, we can solve the system of equations (2) and find the time evolution of the occupation numbers: $N_1 \cong N[e^{-t/\tau_{23}}\tau_{12}/\tau_{21} + (1 - \tau_{12}/\tau_{21})e^{-t/\tau_{12}}]$, $N_2 \cong N[e^{-t/\tau_{23}} - e^{-t/\tau_{12}}]$, $N_3 \cong N[1 - e^{-t/\tau_{23}}]$. $N_1(t)$ decreases exponentially with elapsing time, initially following the "rapid" exponent $\exp(-t/\tau_{12})$, then following the "slow" exponent $\exp(-t/\tau_{23})$. $N_2(t)$ increases, passes through a maximum at $t \sim \tau_{12}$, and then decreases with the same rate as N_1. Finally, there is an accumulation of dots in the VS with the rate determined also by the "slow" $\exp(-t/\tau_{23})$. The dependences $\tau_{12,21}(H)$ can be calculated easily, but they are of minor importance for our experiment, where the time evolution of the spectra is determined by the relaxation time $\tau_{23}(H)$.

To measure the relaxation times we developed a setup allowing one to switch the magnetic field from 0 to 400 Oe and back during $t \sim 1$ ms, i.e., we were able to study the transient dynamics having the characteristic times $\tau_{ij} \geqslant 1$ ms. The setup consists of two coaxial solenoids, inserted one into the other. One of the solenoids operates in a stationary regime with a maximum field ~800 Oe (well above $H_{\rm an} = 410$ Oe). The second solenoid operates in a pulse regime with a pulse rise time ~1 ms. Its magnetic field is controlled by the Honeywell Linear Output Magnetic Field Sensor SS495 and it can be directed parallel or antiparallel to the field created by the first solenoid for a time $\Delta t = 0$–15 s.

Switching of the dot array magnetization is accomplished in the following way. Initially all the dots are saturated (SD state) in the field of a stationary solenoid $H > H_{\rm an}$. Then, the field is reduced to $H = 290$ Oe $\approx H_0$ (see Fig. 1), keeping the dots in the SD state. Then, after a 10 s delay, the total H is reduced to zero by the pulse magnet field for a time Δt. According to the solutions $N_i(t)$, a part of the dots transits to CS ($s \approx 1.1$) or VS ($s = 0$) (see Fig. 1) during the time $t = \Delta t$. After switching off the pulse field and establishing the field H_0 again (see Fig. 1), the dots that were transited to the VS state stay in the same state, but their cores are shifted from the positions $s = 0$ ($H = 0$) to $s = 0.53$ ($H = H_0$). All the CS dots transit to the SD state because the CS minimum disappears. Therefore, after switching off the pulse magnetic field, the occupation numbers are

$$N_1 \cong Ne^{-\Delta t/\tau_{23}}, \quad N_2 = 0, \quad N_3 \cong N(1 - e^{-\Delta t/\tau_{23}}). \quad (3)$$

To investigate the time evolution of the particle numbers in the SD (N_1) and V (N_3) states we used the Vector Network Analyzer ZVA-8. A square array 5×5 mm^2 of circular Py dots with radii $R = 150$ nm and thickness $L = 14$ nm prepared by electron-beam lithography was placed over a coplanar waveguide. The dot edge-to-edge distance was large

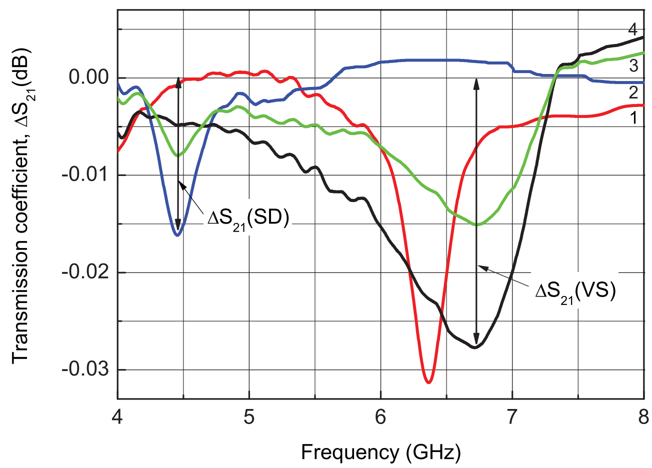

FIG. 2. (Color online) The transmission coefficient $\Delta S_{21}(\omega)$ through the coplanar waveguide covered by the dot array at different values of the in-plane magnetic field H: (1) $H = 580$ Oe $> H_{\rm an}$; (2) $H = 290$ Oe $= H_0$, the field is decreased from $H_{\rm an}$; (3) $H = 290$ Oe, the field is increased after a waiting time $\Delta t = 5$ ms at $H = 0$; (4) $H = 290$ Oe, the field is increased after a waiting time $\Delta t = 8$ s at $H = 0$.

enough (300 nm) to exclude the influence of the interdot magnetostatic interaction. The coplanar line was sputtered on a polycor substrate of thickness 0.5 mm, the width of the central waveguide was 200 μm, and the gaps were of 100 μm. The microwave field across the sample was nonuniform, similar to the excitation field used in the experimental setups in Refs. 18 and 19. We measured the microwave transmission coefficient S_{21} in the frequency range $\omega/2\pi = 4$–8 GHz at different values of H. The dependences $\Delta S_{21}(\omega)$ are shown in Fig. 2. The transmission coefficient S_{21} for the saturated dot array ($H = 580$ Oe $> H_{\rm an}$) has a resonance maximum at $\omega/2\pi = 6.4$ GHz (see curve 1 in Fig. 2). At a field decreasing to $H = 290$ Oe $\approx H_0$, the SD peak frequency decreases in accordance with the Kittel equation down to 4.45 GHz. If the field is increased from 0 up to 290 Oe after a waiting time of $\Delta t = 5$ ms at $H = 0$ (curve 3 in Fig. 2), then, except for the SD peak, an additional resonance peak (6.7 GHz) appears. This peak corresponds to the azimuthal spin waves excited in the VS dots.[18–20] When applying our measurement technique we observe curve 2 in Fig. 2 at $\Delta t = 0$. The number of the dots in the SD state decreases with a time Δt increase, and the value of ΔS_{21}(SD) also decreases (see curve 3 in Fig. 2). We denote as ΔS_{21}(SD) the decrease of the transmission coefficient in this case. Simultaneously, with a decreased number of the dots in the SD state, the dots in the VS appear. These dots create absorption peaks at the resonance frequency of the VS magnetostatic oscillations (6.7 GHz). We denote as ΔS_{21}(VS) the increase of the transmission coefficient for the VS. For a large enough waiting time $\Delta t \gg \tau_{23}$ the SD peak disappears [ΔS_{21}(SD) $\to 0$ at $\Delta t \to \infty$], and there is only a magnetostatic spin wave resonance in the VS [curve 4 in Fig. 2, ΔS_{12}(VS) $\to$ max at $t \to \infty$].

We express the time evolution of ΔS_{21}(VS) and ΔS_{21}(SD) by the occupation numbers $N_1(t)$, $N_3(t)$ given by Eq. (3). We denote the maximal resonance decrease of the transmission

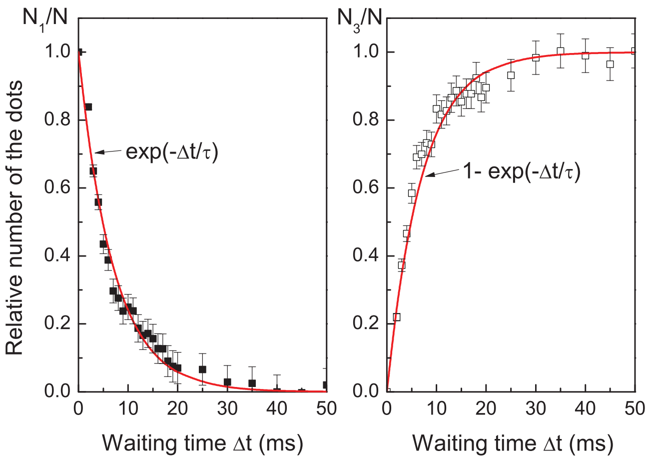

FIG. 3. (Color online) Dependences of the dot numbers in the SD state (N_1) and VS (N_3) on the waiting time Δt after the field decrease to 0 from the initial SD state at $H = 290$ Oe $\approx H_0$ (squares) measured by the change in the resonance transmission coefficients. Left panel: SD resonance; right panel: VS resonance. The solid lines are the dependences calculated by Eq. (4) assuming $\tau_{23} = 7.0$ ms.

coefficient ΔS_{21} in the SD state as ΔS_{21}^0(SD), and in the VS state as ΔS_{21}^0(VS) (curves 2 and 4 in Fig. 2, correspondingly at $\Delta t = 0$ and $\Delta t = \max$) and write

$$N_1/N = \Delta S_{21}(\mathrm{SD})/\Delta S_{21}^0(\mathrm{SD}),$$
$$N_3/N = \Delta S_{21}(\mathrm{VS})/\Delta S_{21}^0(\mathrm{VS}). \quad (4)$$

The changes in the maximal transmission coefficient at the resonance in the field $H = 290$ Oe were $\Delta S_{21}^0(\mathrm{SD}) = 0.016$ dB, $\Delta S_{21}^0(\mathrm{VS}) = 0.027$ dB, correspondingly. In the field 580 Oe the transmission coefficient was 0.031 dB, independent on the waiting time Δt. The dependences ΔS_{21}(SD), N_1 and ΔS_{21}(VS), N_3 on time are plotted in Fig. 3. The best agreement with experimental data is obtained for the relaxation time $\tau_{23} = 7.0$ ms. This relaxation time, according to Eq. (1), corresponds to the energy barrier between the CS and VS of $\Delta E_{23}(0) = 16 k_B T$ assuming a typical $\nu_0 \sim 10^9$ Hz.[12] The bias field can be reduced to a finite value $H < H_\mathrm{n}$ and the dependence of the barrier height $\Delta E_{23}(H)$ on H can be measured. The results of the measurements of the relaxation time $\tau_{23}(H)$ and the CS $\to$ VS energy barrier $\Delta E_{23}(H)$ within the interval -84 Oe $\leqslant H \leqslant 48$ Oe $< H_\mathrm{n}$ are shown in Fig. 4. The experimentally obtained barriers are essentially lower than the barriers calculated within the RVM at $s \approx 1$. Another RVM prediction is the linear dependence of the energy barrier near the dot border, $\Delta W(H)$, on the magnetic field H [yielding $\Delta W(H_n) \approx 0.0082$]. This prediction fails because we measured rather the exponential field dependence of the CS $\to$ VS barrier $\Delta E(H)$ (Fig. 4).

To determine the switching time from the VS to SD state, the dot array initially was magnetized by applying a bias magnetic field, which was increased from $H = 0$ to $H = H_0$. Then, the pulse field creating the total field $H > H_\mathrm{an}$ transferred the dot array to the SD state and kept it there during a time Δt. In this case, the relaxation time was lower than the setup resolution time of $\sim$1 ms. The short time of the VS $\to$ SD state transition is explained by the transition of the

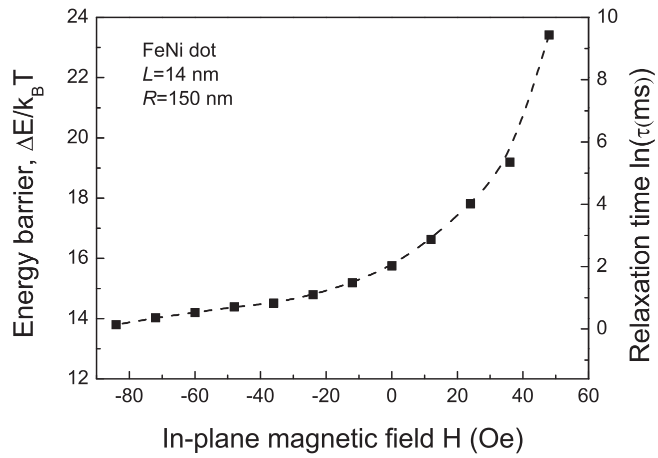

FIG. 4. The measured magnetization relaxation time and reduced barrier height (squares) of the Py dot array vs in-plane bias field H for the CS $\to$ VS transition.

vortex with $s = 0.53$ (see Fig. 1, $H = 0.405 M_s$) to $s = \infty$ occurring along the curve, where $H = H_\mathrm{an}$ and the absence of the energy barrier within the RVM. The equilibrium value of s gradually increases with H, increasing up to a maximal value $s \approx 0.85$ at $H = H_\mathrm{an} = 0.717 M_s$, and the vortex energy minimum disappears at $H \geqslant H_\mathrm{an}$.

The interpretation of the experiment strongly depends on whether there is a second metastable state (CS) of the circular Py dot at $H = 0$. The RVM predicts a metastable CS $H \leqslant H_C$ and two transitions decreasing the field from $H > H_\mathrm{n}$ down to $H = 0$: (1) SD $\to$ CS and (2) CS $\to$ VS over the energy barrier or the vortex core penetration to the dot. If there would be only one stable VS at $H = 0$, then the energy barriers disappear, decreasing H below H_n, and the magnetization evolution occurs starting from finite $s \gg 1$ to $s = 0$, according to the Landau-Lifshitz-Gilbert (LLG) equation of magnetization motion,[5] due to the precession and damping terms. This equation allows one to estimate a thermalization time $\tau_0 \approx 1/\alpha\omega_0 \approx 30$ ns, where α is the damping constant, and $\omega_0/2\pi \approx 480$ MHz is the vortex gyrotropic frequency.[7] Then, during the time (3–5)$\tau_0 \approx$ 100–150 ns, the dot reaches the equilibrium VS ($s = 0$ if $H = 0$). But in our experiment this relaxation time is equal to several ms, i.e., it is at least 10^4 times longer. Such a discrepancy can be explained by the existence of an energy barrier between the CS and VS at the dot border and/or random local energy barriers (due to variable defects) along the path of the vortex core to the dot center via the dot border. In real samples there are other interactions which are related to the presence of defects and may influence essentially the energy barriers,[21] i.e., τ has an extrinsic, sample dependent contribution, which cannot be calculated within a general approach. To calculate τ we need to assume some specific defect distribution in the sample and introduce corresponding pinning potentials. But we believe that the contribution of the dot border to the resulting nucleation barrier is dominating and is mainly responsible for the measured values of τ of about 7 ms.

The RVM predicts a metastable CS and high energy barrier for the CS $\to$ VS transition for the given dot geometrical parameters. The calculated value of the barrier related to the symmetry breaking vortex core nucleation is $\Delta W \approx 0.0060$

or $\Delta E \approx 93k_BT$. According to Eq. (1) there should be no transitions over such a high barrier during a reasonable measurement time. But a change of the dot excitation spectra means that such transitions indeed occur in our experiments during surprisingly short times (several ms), whereas times of $\sim$10–10^3 s were expected.[9,12,15] A real reversal mode can be essentially different from the calculated one within the RVM, especially if the core position s is close to 1 because the vortex can be strongly deformed near the dot border. Micromagnetic simulations showed that the vortex core is elongated perpendicularly to the direction of the bias field H. The RVM assumes a vortex displacement saving the core shape and neglects this deformation. Therefore, it cannot describe properly the magnetization reversal mode decreasing H near H_n. An analytic theory allowing one to extend the dependence of the energy $E(s)$ in a one-dimensional configuration space to the point $s = 1$ is missing because details of the deformed core magnetization are unknown.[22] Therefore, a more adequate model is needed to describe the nucleation energy barrier.

In summary, the low-field relaxation times of the array of Py dots from the SD state to the VS were measured by probing the dot spin excitation spectra. The relaxation times are surprisingly short ($\sim$7 ms at $H = 0$). They were interpreted as a consequence of the energy barriers related to the vortex core penetrations to the dots. The dependence of the energy barriers on the in-plane field was extracted from the broadband ferromagnetic resonance experiments. The measured barriers are essentially smaller than the barriers predicted by the RVM. The measured switching time from the VS to SD state is also quite short, but it is not in contradiction with the RVM. The developed method of measuring the time dependent intensities of the ferromagnetic resonance peaks in the different metastable and ground states is applicable to explore the transient dynamics in any magnetic system.

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