

## Local Control of Ultrafast Dynamics of Magnetic Nanoparticles

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(Received 21 October 2008; revised manuscript received 2 December 2008; published 3 February 2009)

Using the local control theory we derive analytical expressions for magnetic field pulses that steer the magnetization of a monodomain magnetic nanoparticle to a predefined state. Finite-temperature full numerical simulations confirm the analytical results and show that a magnetization switching or freezing is achievable within few precessional periods and that the scheme is exploitable for fast thermal switching.

DOI: 10.1103/PhysRevLett.102.057204

PACS numbers: 75.10.Hk, 75.40.Mg, 75.60.Jk, 82.50.Nd

**Introduction.**—A fast magnetization reversal of magnetic nanoparticles is of a key importance for the realization of high-rate magnetic recording [1,2]. Several techniques are currently envisaged for the magnetization switching such as the laser-induced spin dynamics [3] based on the inverse Faraday effect [4,5], the reversal triggered by external static or alternating magnetic fields [6–12] or by a spin-torque acting on the magnetization due to a passing spin-polarized electric current [13,14]. Transverse magnetic field pulses are also efficient for a swift reversal [15–20], and if finely tuned in duration [2,21] can even lead to a quasiballistic switching. A further fundamental issue, addressed here is how to steer the magnetic dynamics to a desirable state by external fields. Generally, a number of control schemes have been established mainly in quantum chemistry [22–25]. Particularly interesting is the local control theory (LCT) [24,25] in which the control fields are constructed from the response of the system offering thus a physical interpretation of the control mechanism. We adopt the idea of LCT to steer the magnetization dynamics of nanoparticle by transverse magnetic pulses. We obtain transparent analytical expressions for the control pulses that allow a fast switching or a quasi “freezing” at a predefined magnetization state. For the scheme to be applicable, the field durations have to be shorter than the field-free precessional period but no special pulse-duration tuning is required; the field strengths are to be determined according to the analytical expressions provided here. In our control strategy the magnetization dynamics proceeds via sudden impulsive kicks guiding the magnetization towards a predefined direction; the pulses are intervened by field-free magnetization precessions and relaxation. A similar mechanism has recently been realized experimentally [12] using spin-polarized picosecond current pulses resulting in a spin-transfer-torque-driven stroboscopic dynamics. The robustness of the predictions we demonstrate with finite-temperature full numerical calculations and for different types of anisotropy fields. We confirm the analytical results and un-

cover the potential of this scheme for fast thermal switching that can be the basis for fast thermal sensors.

**Theory.**—We consider a nanoparticle with a size such that it displays a long-range magnetic order and is in a single domain remanent state. Examples are Fe<sub>50</sub>Pt<sub>50</sub> [2,26] or Fe<sub>70</sub>Pt<sub>30</sub> [2,27] nanoparticles which possess, respectively, a uniaxial or a cubic anisotropy. Following the Landau-Lifshitz-Gilbert (LLG) approach we model the dynamics of the magnetization direction by the classical evolution of a unit vector  $\mathbf{S}$ . The particle’s magnetic moment at saturation  $\mu_S$  is assumed time invariant. The system energy derives from  $\mathcal{H} = \mathcal{H}_A + \mathcal{H}_F$ , where  $\mathcal{H}_A$  and  $\mathcal{H}_F = -\mathbf{S} \cdot \mathbf{b}_0(t)$  stand, respectively, for the anisotropy and the Zeeman energy of  $\mathbf{S}$  in the external field  $\mathbf{b}_0(t)$ . For a particular type of anisotropy described by  $f_A(\mathbf{S})$  we write  $\mathcal{H}_A = -Df_A(\mathbf{S})$  with  $D$  being the anisotropy constant.  $\mathbf{S}(t)$  develops according to LLG equation [28] as  $\frac{\partial \mathbf{S}}{\partial t} = -\frac{\gamma}{(1+\alpha^2)} \mathbf{S} \times [\mathbf{B}_e(t) + \alpha(\mathbf{S} \times \mathbf{B}_e(t))]$ , where  $\mathbf{B}_e(t) = -[1/(\mu_S)] \partial \mathcal{H} / \partial \mathbf{S}$  is the effective field,  $\gamma$  is the gyromagnetic ratio and  $\alpha$  is the Gilbert damping parameter. In spherical coordinates where the  $z$  axis is along the easy axis we specify  $\mathbf{S}$  by the azimuthal ( $\phi$ ) and polar ( $\theta$ ) angles and cast the LLG equation as [2,29]

$$\begin{aligned} (1 + \alpha^2) \frac{d\phi}{dt} &= \frac{1}{\sin\theta} \frac{\partial \mathcal{H}}{\partial \theta} - \frac{\alpha}{\sin^2\theta} \frac{\partial \mathcal{H}}{\partial \phi}, \\ (1 + \alpha^2) \frac{d\theta}{dt} &= -\frac{1}{\sin\theta} \frac{\partial \mathcal{H}}{\partial \phi} - \alpha \frac{\partial \mathcal{H}}{\partial \theta}. \end{aligned} \quad (1)$$

Hereafter the time is measured in units of the field-free precessional period  $T^{\text{pre}}c$  and the energy  $\mathcal{H}$  in units of  $\mu_S B_A$  where  $B_A = 2D/\mu_S$  is the maximum uniaxial anisotropy field. E.g., for Fe<sub>50</sub>Pt<sub>50</sub> we have  $T^{\text{pre}}c = 5$  ps, the maximum anisotropy field is  $\sim 7$  T and the magnetic moment per nanoparticle is around  $22\,000 \mu_B$  [26]. The field-free solution of (1) is known; e.g., for a uniaxial anisotropy and starting from the angles  $\phi_f(t = \bar{t}_0)$  and  $\theta_f(t = \bar{t}_0)$  one finds (e.g., [30])

$$\phi_f(t) = \phi_f(\bar{t}_0) \pm \frac{t - \bar{t}_0}{1 + \alpha^2} \pm \frac{1}{\alpha} \ln \left| \frac{\cos \theta_f(\bar{t}_0) (1 + \sqrt{1 + \tan^2 \theta_f(\bar{t}_0) e^{-[2\alpha(t - \bar{t}_0)/(1 + \alpha^2)]}})}{1 + \cos \theta_f(\bar{t}_0)} \right|, \quad \tan \theta_f(t) = \tan \theta_f(\bar{t}_0) e^{-(\alpha/(1 + \alpha^2))(t - \bar{t}_0)}. \quad (2)$$

“+” (“−”) refers to  $0 < \theta < \pi/2$  ( $\pi/2 < \theta < \pi$ ).

To control the dynamics we apply along the  $x$  and  $y$  axis two magnetic field pulses  $\mathbf{b}_x$  and  $\mathbf{b}_y$  of durations  $2\varepsilon$  and shapes  $f(t)$  centered at some moment  $t = t_0$ . Their relative strengths are given by the mock angle  $\phi_0$ , with  $\tan \phi_0 = |b_y|/|b_x|$ ; the total fields strength is  $|f|b_0/(2\varepsilon)$ . Hence  $\mathbf{b}_0(t) = \mathbf{b}_x + \mathbf{b}_y$  is

$$\mathbf{b}_0(t) = \begin{cases} \frac{f(t)b_0}{2\varepsilon} (\cos \phi_0 \mathbf{e}_x + \sin \phi_0 \mathbf{e}_y), & t_0 - \varepsilon < t < t_0 + \varepsilon \\ 0, & \text{elsewhere.} \end{cases} \quad (3)$$

Switching to a new time variable  $\tau(t) = \frac{t - (t_0 + \varepsilon) + 2\varepsilon}{2\varepsilon}$  we derive for the equation of motion

$$\begin{aligned} \frac{1}{2\varepsilon} \frac{d\phi}{d\tau} &= p \left[ \frac{1}{\sin \theta} \frac{\partial \mathcal{H}_A}{\partial \theta} - \frac{\alpha}{\sin^2 \theta} \frac{\partial \mathcal{H}_A}{\partial \phi} \right] - \frac{pb_0 f(t(\tau))}{2\varepsilon} \\ &\quad \times \left[ \frac{\cos \theta}{\sin \theta} \cos \delta \phi + \alpha \frac{\sin \delta \phi}{\sin \theta} \right], \\ \frac{1}{2\varepsilon} \frac{d\theta}{d\tau} &= p \left[ -\frac{1}{\sin \theta} \frac{\partial \mathcal{H}_A}{\partial \phi} - \alpha \frac{\partial \mathcal{H}_A}{\partial \theta} \right] + \frac{pb_0 f(t(\tau))}{2\varepsilon} \\ &\quad \times [-\sin \delta \phi + \alpha \cos \theta \cos \delta \phi], \end{aligned} \quad (4)$$

where  $\delta \phi = \phi - \phi_0$  and  $p = 1/(1 + \alpha^2)$ . If the magnetic pulses are shorter than the precessional period then from Eq. (4) we infer for the angles stroboscopic evolution from before  $[\phi(t^-), \theta(t^-)]$  to after  $[\phi(t^+), \theta(t^+)]$  the pulses the relation (we introduced  $t^- := t_0 - \varepsilon$ ,  $t^+ := t_0 + \varepsilon$ )

$$\begin{aligned} \frac{d\phi}{d\tau} &= -\frac{1}{\sin \theta} \frac{b_0 f(t_0)}{1 + \alpha^2} [\cos \theta \cos \delta \phi + \alpha \sin \delta \phi], \\ \frac{d\theta}{d\tau} &= \frac{b_0 f(t_0)}{1 + \alpha^2} [-\sin \delta \phi + \alpha \cos \theta \cos \delta \phi], \end{aligned} \quad (5)$$

which is valid up to terms of the order  $(\varepsilon/T^{\text{prec}})^2$ . After the pulse, i.e., for  $t > t^+$  the dynamics is governed by Eq. (2) with the initial conditions  $\phi_f = \phi(t^+)$ ,  $\theta_f = \theta(t^+)$ . This procedure is repeated accordingly.

*Controlled switching.*—As we are interested in switching we require in the spirit of local control theory that

$$\theta(t^+) > \theta(t^-) \quad \forall t^+, t^-. \quad (6)$$

As inferred from Eq. (5), this condition is fulfilled if  $\delta \phi = \phi - \phi_0 = 3\pi/2$ . If a sequence of the pulses (3) each centered at the times  $t_{0,i}$  is applied then  $\mathbf{S}(t)$  evolves as

$$\phi(t_i^+) = \phi(t_i^-) + \alpha \ln \left| \frac{\tan(\frac{\theta(t_i^-)}{2}) + \frac{1}{2} \frac{b_0 f(t_{0,i})}{1 + \alpha^2}}{\tan(\frac{\theta(t_i^-)}{2})} \right|, \quad (7)$$

$$\theta(t_i^+) = \theta(t_i^-) + \frac{b_0 f(t_{0,i})}{1 + \alpha^2},$$

where  $t_i^\pm = t_{0,i} \pm \varepsilon$ .

The realization of this LCT scheme is then as follows: Starting from a known (e.g., equilibrium) state  $\phi = \phi(0)$ ;  $\theta = \theta(0)$  we apply at  $t = t_{0,1}$  the first fields  $b_x$  and  $b_y$  (3) with strengths such that  $\phi_0 = \phi(0) - 3\pi/2$  (cf. Fig. 1). Equation (7) delivers the tilt angles  $\theta(t_1^+)$  and  $\phi(t_1^+)$ . During a time lag (dark time)  $\tau_1$  the propagation proceeds according to Eq. (2) with the initial values  $\phi_f(\bar{t}_0) = \phi(t_1^+)$  and  $\theta_f(\bar{t}_0) = \theta(t_1^+)$ . At  $t = t_{0,2}$  we apply a second pulse with  $b_x$  and  $b_y$  such that  $\phi_0 = \phi_f(t_1^+ + \tau_1) - 3\pi/2$ . From Eq. (7) we deduce that after the second pulse  $\theta(t_2^+) = \theta_f(t_1^+ + \tau_1) + \frac{b_0 f(t_{0,2})}{1 + \alpha^2}$ . This procedure is repeated until we achieve the state with  $\theta = \pi/2$ . As clear from (7) the tilt angle is always increased upon the pulse with an amount that goes linearly with the fields strength  $b_0$ . On the other hand, the variation of  $\phi$  with  $b_0$  is only logarithmic, in fact if the time delay between the pulses is only a fraction of the precessional period,  $\phi$  is hardly changed.

*Freezing.*—The scheme allows also for the stabilization of the magnetization around a desirable  $\theta_f$ : At first, starting from a given state we apply the control scheme and achieve  $\theta_i$  at some time  $t_i$ . During a field-free period  $\tau$  the angle  $\theta_i$  develops to  $\theta_f(t_i + \tau)$ . To compensate for this change we apply a pulse (centered at  $t_{0,i}$ ) which shifts the angle to  $\theta^+ = \theta_f(t_i + \tau) + \frac{b_0 f(t_{0,i})}{1 + \alpha^2}$ . To stabilize the magnetization we choose  $b_0$  such that  $\theta^+ = \theta_i$ . The procedure is then repeated during the stabilization time. To minimize the adjustment of  $b_0$  between consequent pulses the repetition rate should be large.

*Numerical results and illustrations.*—Figure 1 shows the magnetization reversal according to our zero temperatures ( $T = 0$ ) analytical scheme and in the damping regime appropriate for magnetic nanoparticles. Figure 1 confirms our analysis and the physical picture drawn above. However, the following issues need to be clarified for

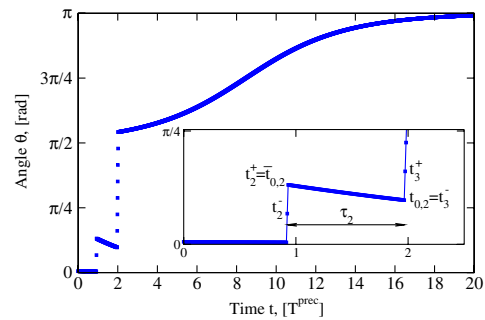


FIG. 1 (color online). Evolution of  $\theta(t)$  according to the proposed control scheme and for  $\phi(t = 0) = \pi/180 = \theta(t = 0)$ ,  $\phi_0 = \arctan(b_y/b_x) = 2\pi/3$ ,  $\alpha = 0.05$ ,  $f = 1$ ,  $b_0 = 0.2$ . Inset shows the short-time behavior (pulses are off for  $\theta > \pi/2$ ).

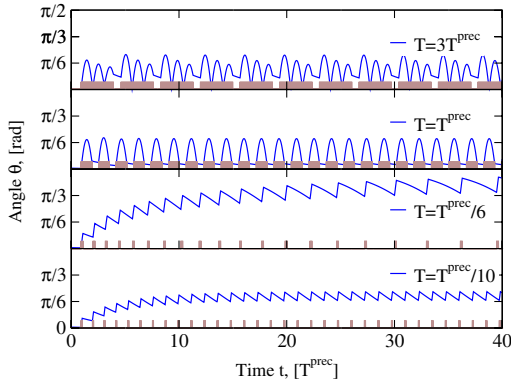


FIG. 2 (color online).  $\theta(t)$  for different pulse durations (solid rectangles).  $T^{\text{prec}}$  is the precessional period and  $b_0 = 0.3$ ,  $\alpha = 0.05$ .

this procedure to be of practical interest. (i) Do we need a precise tuning of the pulses durations, (ii) will thermal fluctuations invalidate our findings, and (iii) how effective is this scheme when applied to other type of anisotropy fields. To address these points we implemented a finite-temperature full numerical realization [31] of the present control scheme (cf. [1,2], and references therein for an overview on numerical micromagnetic methods), i.e., the analytical expressions deliver the appropriate input parameters for the numerics. The damping parameter is chosen according to experimental findings [2]. For the simulation presented here we use square-shaped pulses, i.e.,  $f(t) = 1$  for  $t_0 - \varepsilon < t < t_0 + \varepsilon$ . Basically the same conclusions are valid for other pulse shapes, e.g., Gaussian pulses [32]. Figure 2 demonstrates the evolution sensitivity of the angle  $\theta$  when pulses with different durations are applied. It also shows the range of validity of our scheme. As inferred from Fig. 2 a fine tuning of the pulse duration is not mandatory as long as it is smaller than  $T^{\text{prec}}$ . The strength  $b_0$  determines the value of the tilt angle [as follows from Eq. (7)]. The insensitivity to the pulse duration is favorable for practical applications, however the generation of magnetic pulses shorter than  $T^{\text{prec}}$  might be a challenge; the light-induced generation of subpicosecond shaped magnetic pulses [33] may circumvent this problem. As for the role of the magnetization dynamics during the

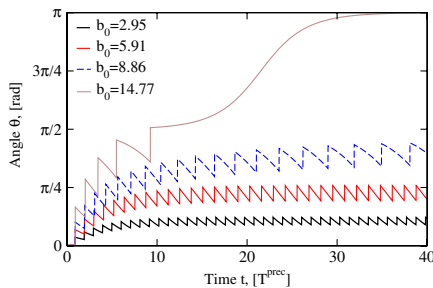


FIG. 3 (color online). Tilt angle  $\theta(t)$  within the present local control scheme for different fields strengths  $b_0$ . Other parameters:  $\alpha = 0.05$ ,  $T_0 = 0$  K. (Pulses are off when  $\theta > \pi/2$ ).

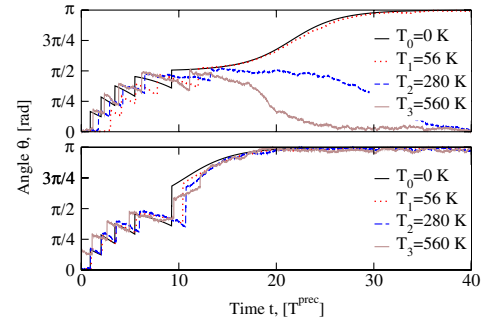


FIG. 4 (color online). Temperature-dependent controlled evolution of the angle  $\theta(t)$  ( $\alpha = 0.05$ ,  $b_0 = 14.77$ ). The pulses are applied if  $\theta < \pi/2$  only (top panel) or throughout (below).

pulses our simulations (cf. Fig. 3) confirm qualitatively the analytical predictions. According to Eq. (7) a minimal fields strength  $b_0$  is required for switching, for  $b_0$  determines  $\theta(t^+)$ . To realize the stabilization scheme one tunes  $b_0$  to steer the magnetization to a nonequilibrium  $\theta_t$  (cf. Fig. 3) and keep it there (as long as  $\mathbf{b}_0$  is on).

Figure 4 proves the robustness of the scheme to thermal fluctuations. Here we highlight a special feature of the temperature-dependent magnetization dynamics: To achieve switching, the pulses have to be applied even if  $\theta_t > \pi/2$ , since due to thermal excitations the magnetization may swing back to the original state. This effect is avoided by applying the pulses even if  $\theta > \pi/2$  (Fig. 4, lower panel). Generally, we observe that thermal fluctuations have little influence on the effect of the pulses (i.e., on the dynamics during and right after the pulses), in contrast to continuous fields [31]. The field-free precessional motion between the pulses is generally modified at  $T > 0$ .

The possibility of field-assisted stabilization (freezing) can be exploited for fast field-assisted thermal switching: Starting at  $T \approx 0$  we utilize our scheme to drive the magnetization to a state  $\theta_t \lesssim \pi/2$  (as shown in Fig. 5) and then freeze it there. At low temperatures switching does not occur irrespective of the waiting time (inset of Fig. 5). When the temperature increases however, the thermal fluctuations increase but cannot lead to a reversal in absence of the field, as demonstrated by the inset of

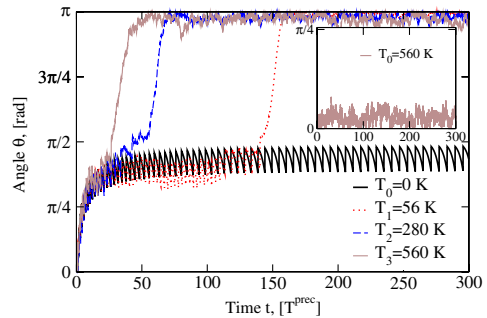


FIG. 5 (color online). Thermal-assisted controlled switching in the presence of short pulses with an amplitude  $b_0 = 8.86$ . Inset shows switching is not possible for  $b_0 = 0$ .

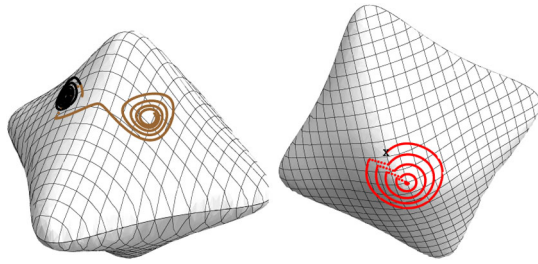


FIG. 6 (color online). Polar diagram of the energy surface for a cubic anisotropy with magnetization trajectories. Left panel is a top view on the energy surface: For  $b_0 = 0$  (dark trajectory); for a  $b_0 = 2.06$  control field (light trajectory). Trajectories start at  $\phi(t=0) = 1.9\pi$ ,  $\theta(t=0) = \pi/3.8$ . Right panel is a bottom view at the energy surface: freezing field is  $b_0 = 0.59$  and the magnetization is initially at the position marked (X). In both cases  $\alpha = 0.05$ .

Fig. 5. The presence of the fields assists a fast magnetization reversal, a behavior that cannot be realized with static fields, since a magnetization freezing is necessary. In practice, the reversal process may be functionalized as a fast thermal sensor to monitor swiftly a temperature increase.

The question of to what extent the present scheme is applicable to another anisotropy type we address by studying the magnetization control of  $\text{Fe}_{70}\text{Pt}_{30}$ -nanoparticles which possesses cubic anisotropy [27,34]. For a cubic anisotropy the field-free ground-state energy landscape contains several minima [35]. By switching we mean then a magnetization transfer between these minima and not necessarily a change from a parallel to an antiparallel state. Figure 6 demonstrates the applicability of our control proposal. Starting from a state close to an energy minimum the magnetization precesses and relaxes in a field-free manner to the ground state. When the magnetic pulse is applied according to our LCT the magnetization is transferred almost directly to the next energy minimum in the positive energy semisphere. With the freezing scheme outlined above it is even possible to stabilize the magnetization on top of the barrier (Fig. 6).

*Summary.*—A sequence of two perpendicular magnetic pulses, each with a duration less than the precessional period is capable of increasing monotonically the magnetization tilt angle as to achieve a predefined state within tens of picoseconds. The method is exploitable for fast field-assisted thermal switching.

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