New Concept for Magnetization Switching by Ultrafast Acoustic Pulses

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It is shown theoretically that a single acoustic pulse, a few picoseconds long, can reverse magnetization in a magnetostrictive material Terfenol-D. Following giant magnetoelastic changes of free energy density, the magnetization vector is ejected from a local in-plane energy minimum and decays into another minimum. For an acoustic pulse duration significantly shorter than magnetization precession period $\tau_{ac} \ll T_{prec}$, the switching threshold is determined by the *acoustic pulse area*, i.e., pulse integral in the time domain, similar to coherent phenomena in optics. Simulation results are summarized in a magneto-acoustic switching diagram and discussed in the context of all-optical magnetization switching by circularly polarized light pulses.

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Searching for new possibilities of ultrafast magnetization switching is motivated by the continuously growing demand for faster data recording technologies, which are based on the reversal of individual magnetic bits at the nanoscale. Among the different mechanisms of magnetization switching, the acoustically driven switching at ultrafast time scales remains largely unexplored.

The time-resolved observation of magnetization dynamics in ferromagnetic nickel induced by femtosecond laser pulses revealed the phenomenon of ultrafast demagnetization in nickel [1]. The ultrafast drop of magnetization on a subpicosecond time scale was caused by transient elevation of the electron temperature close to the Curie point. The subsequent dynamics of magnetization recovery on a time scale exceeding tens of picoseconds could be adequately reproduced by Landau-Lifshitz-Gilbert (LLG) equations [2,3] describing damped precession of a magnetization vector in the presence of temperature-dependent magnetocrystalline anisotropy [4].

The most recent experiments combining picosecond acoustics [5] with ultrafast magnetooptics showed that the magnetocrystalline anisotropy can also be changed by picosecond acoustic strain pulses to a ferromagnetic semiconductor GaMnAs [6,7] or nickel [8], thus triggering the magnetization precession without heating the sample. However, due to the relatively small magnetostrictive coefficient in GaMnAs ($\Lambda \sim 10^{-5}$ at cryogenic temperature [9]) and nickel ($\Lambda \sim 3 \times 10^{-5}$ at room temperature [10]), the resulting transient change in the magnetization direction appeared to be too small and the magnetization vector returned to its initial direction; i.e., magnetization switching (reversal) did not occur.

The first experimental demonstration of ultrafast *nonthermal* magnetization switching was reported in 1998 by Back and co-workers [11], who used ultrashort pulses of a magnetic field induced by relativistic electron bunches to switch magnetization in Co/Pt film. Most recently, a spectacular observation of all-optical magnetization switching in GdFeCo using circularly polarized light pulses [12,13] raised a lot of questions about physically unclear switching mechanisms suggesting that not only transient overheating of electrons but also the dynamics of lattice temperature may be necessary to explain the underlying physics [14]. Not only the minimum amount of deposited heat [14] but also a minimum amount of "circularity" was necessary to obtain switching [15], at least within a certain range of optical excitation [16].

In this Letter, we theoretically investigate the interaction of ultrashort acoustic pulses with Terfenol-D $(Tb_xDy_{1-x})Fe_2$, the rare-earth compound famous through its giant magnetostrictive coefficient $\Lambda \sim 10^{-3}$ [10], and demonstrate the possibility of ultrafast magnetoacoustic switching. Moreover, the results are discussed within the framework of recently observed all-optical magnetization switching by single circularly polarized femtosecond laser pulses, which is undoubtedly accompanied by the generation of picosecond pulses of coherent acoustic phonons [5].

The phenomenological expression for free energy density $F(\vec{M}) = F_k + F_{me} + F_d + F_z$ for (110) thin films of $Tb_{0.27}Dy_{0.73}Fe_2$ epitaxially grown on sapphire reads [17]

$$F_k = K_1(\alpha_x^2\alpha_y^2 + \alpha_x^2\alpha_z^2 + \alpha_z^2\alpha_y^2) + K_2(\alpha_x^2\alpha_y^2\alpha_z^2), \quad (1)$$

$$F_{me} = b_1(\alpha_x^2 e_{xx} + \alpha_y^2 e_{yy} + \alpha_z^2 e_{zz}) + b_2(\alpha_x\alpha_y e_{xy} + \alpha_x\alpha_z e_{xz} + \alpha_y\alpha_z e_{yz}), \quad (2)$$

$$F_d = \frac{\mu_0}{2}(M_s \cos\theta)^2. \quad (3)$$

In Eqs. (1)–(3), F_k, F_{me}, and F_d denote the magnetocrystalline anisotropy and magnetoelastic and magnetostatic terms, respectively, and the Zeeman contribution $F_z = -\mu_0\vec{H}_{ext}\cdot\vec{M}$ is disregarded throughout this Letter, as we consider the case of zero external magnetic field $\vec{H}_{ext} = 0$. Terfenol-D is characterized by magnetocrystalline anisotropy coefficients $K_1 = -0.87\ \mathrm{J/cm^3}$ and $K_2 = 2.35\ \mathrm{J/cm^3}$ [17] and magnetoelastic coefficients

$b_1 = -(3/2)\Lambda_{100}(c_{11} - c_{12}) = -80$ J/cm^3 and $b_2 = -3\Lambda_{111}c_{44} = -85$ J/cm^3, which depend on the components of the linear elastic tensor $c_{11} = 141$ GPa, $c_{12} = 64.8$ GPa, and $c_{44} = 21$ GPa [10]; α_x, α_y, and α_z are direction cosines of the magnetization vector $\vec{M} = M_s(\alpha_x, \alpha_y, \alpha_z)$ in the crystallographic coordinate system (x, y, z) and saturation magnetization $\mu_0 M_s = 0.945$ T; e_{ij} is the strain tensor.

Epitaxial growth of a thin Terfenol-D film in the (110) direction on a lattice-mismatched sapphire substrate induces the built-in static strain described by the following tensor:

$$e_{\text{stat}} = \begin{pmatrix} 0 & e_{xy} & 0 \\ e_{xy} & 0 & 0 \\ 0 & 0 & -\frac{2c_{12}}{c_{11}} e_{xy} \end{pmatrix}, \tag{4}$$

which is determined by a single strain component $e_{xy} = -0.55\%$ [18] in a crystallographic coordinate frame (x, y, z).

In the rotated frame (x', y', z'), the direction of magnetization is determined by two angles: θ (out-of-plane angle) and ϕ (in-plane angle); see Fig. 1(a). The competition of different contributions in the total free energy density results in four local in-plane energy minima corresponding to four different magnetization directions 1, 2, 3, and 4 in Fig. 1(b). The explicit dependence of the magnetoelastic term F_{me} on both the strain components and magnetization direction and large values of magnetoelastic coupling coefficients b_1 and b_2 suggest that application of external strain will shift the minima of free energy minima and, therefore, change the magnetization direction. Indeed, the application of time-independent uniaxial strain η in the direction normal to the surface of a thin Terfenol-D film results in the in-plane shift of all four energy minima by the angle $\Delta\phi$, as illustrated in Fig. 1(c) for $\eta = -0.3\%$ (film compression) and $\eta = 0.9\%$ (film tension).

The action of a time-dependent uniaxial strain (acoustic pulse) $\eta(t)$ can be described by adding the following time-dependent strain tensor $e_{\text{dyn}}(t)$ in a crystallographic coordinate frame:

$$e_{\text{dyn}}(t) = \frac{1}{2}\begin{pmatrix} \eta(t) & -\eta(t) & 0 \\ -\eta(t) & \eta(t) & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{5}$$

where the rotation of coordinates system by 45° from (x', y', z') into (x, y, z) leads to a factor of $1/2$ and generates the nondiagonal terms.

Inserting the total strain $e(t) = e_{\text{stat}} + e_{\text{dyn}}(t)$ in Eq. (2) generates explicit time dependence of free energy $F(t)$, which drives the magnetoacoustic dynamics described by the LLG equation [8]

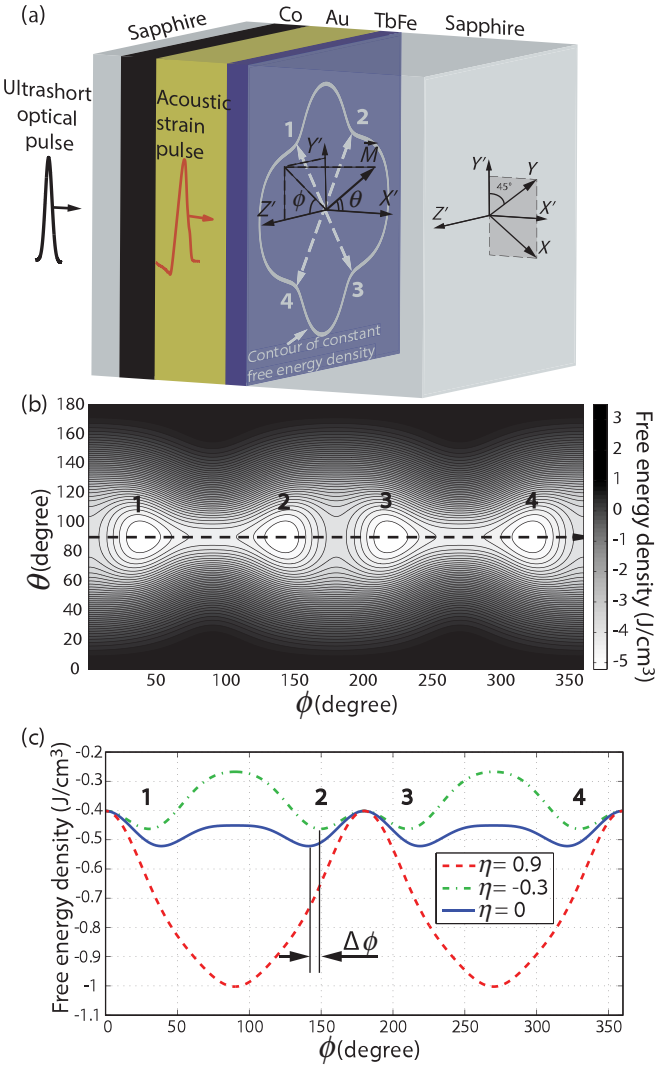

FIG. 1 (color online). Ultrafast optical excitation of a hybrid sapphire–Terfenol-D–gold–cobalt–sapphire multilayer structure generates an ultrashort unipolar acoustic pulse, which results in application of the time-dependent strain to a thin film of Terfenol-D (see the Supplemental Material [20] for details). Free energy density in a (110) film of Terfenol-D grown on sapphire (a) possesses four in-plane energy minima (b). Application of a *static* uniaxial compressive ($\eta = -0.3\%$) or tensile ($\eta = 0.9\%$) strain in the direction perpendicular to the film leads to the in-plane shift $\Delta\phi$ of all four energy minima (c).

$$\frac{d\vec{M}}{dt} = -\frac{\gamma}{1+\alpha^2}\left[\mu_0 \vec{M} \times \vec{H}_{\text{eff}} - \frac{\alpha}{M_s}\vec{M} \times (\vec{M} \times \vec{H}_{\text{eff}})\right], \tag{6}$$

where the first term describes the torque driving the precession of the magnetization vector around the effective time-dependent magnetic field $H_{\text{eff}}(t)$,

$$\vec{H}_{\text{eff}}(t) = -\frac{1}{\mu_0}\frac{dF(t)}{d\vec{M}}, \tag{7}$$

and the second term describes precession damping according to the phenomenological Gilbert damping parameter $\alpha = 0.1$ [19]; γ is the gyromagnetic ratio.

If the system is initially prepared in minimum 2, the application of instantaneous steplike strain (see the Supplemental Material [20]) results in damped precession of the magnetization vector towards the new minimum $2'$ with a precession period $T_{\text{prec}} \simeq 25$ ps; see Fig. 2. However, if the strain is turned off after some time, the situation equivalent to the application of a rectangular acoustic pulse of finite duration τ_{ac}, the precession trajectory will decay back into minimum 2. Such a magnetoacoustic precession trajectory induced by the action of a picosecond acoustic pulse with $\tau_{ac} = 3$ ps and strain amplitude $\eta_{\text{ac}} = 3 \times 10^{-3}$ is shown in Fig. 2 and can be explained analytically.

Near the beginning of the pulse, when the system is at one of the four in-plane ($\theta = \pi/2$) energy minima, the LLG equation (6) is dominated by the magnetization precession around the effective field $\vec{H}_{\text{eff}}(t)$, which is pointing in the direction of the new minimum. Therefore, the magnetization moves out of plane (θ changes) according to (see the Supplemental Material [20])

$$\frac{d\phi}{dt} = 0, \qquad \frac{d\theta}{dt} = \gamma \frac{(b_2 + 2b_1)}{2M_s(1+\alpha^2)} \alpha'_y \alpha'_z \eta(t). \quad (8)$$

The integration of Eq. (8) for an ultrashort acoustic pulse $\eta(t)$ obeying the condition $\tau_{\text{ac}} \ll T_{\text{prec}}$ approximates well the acoustic *out-of-plane deflection* of the magnetization vector by the angle

$$\Delta\theta_{\text{ac}} \simeq \gamma \frac{(b_2 + 2b_1)}{2M_s(1+\alpha^2)} \alpha'_y \alpha'_z \int \eta(t)dt. \quad (9)$$

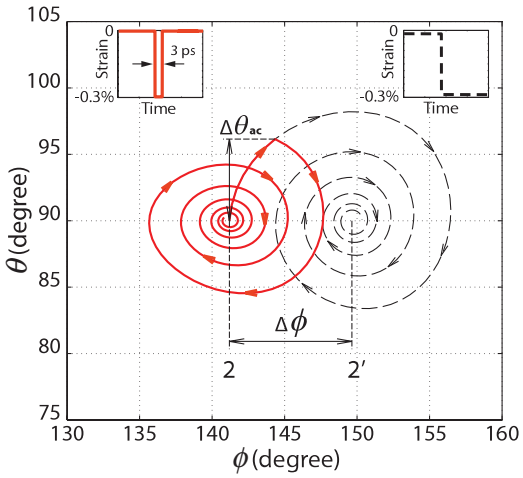

FIG. 2 (color online). Starting from energy minimum 2, a *dynamic* steplike strain induces a damped magnetization precession around the shifted minimum $2'$ (dashed line). An ultrashort acoustic pulse results in the out-of-plane deflection $\Delta\theta_{\text{ac}}$ followed by damped precession around the initial minimum 2 (continuous line).

The product of directional cosines $\alpha'_y \alpha'_z$ in Eq. (9) equals 0.48 for energy minima 1 and 3 and -0.48 for the two other minima, respectively. Therefore, depending on the initial condition, the same acoustic pulse will eject the magnetization vector out of the sample plane in opposite directions. Equation (9) clarifies the microscopic physical model beyond the time-dependent magnetic torque $|\vec{M} \times \vec{H}_{\text{eff}}|$ introduced by Kim and co-workers [8] and shows that the prefactor in Eq. (9) is dominated by the ratio of magnetoelastic coupling coefficients b_1 and b_2 (which are both proportional to the respective coefficients of magnetostrictive tensor Λ) to saturation magnetization M_s.

Similar to polarization dynamics in coherent optics, the acoustic rotation angle $\Delta\theta_{\text{ac}}$ of the magnetization vector appears to be proportional to the *acoustic pulse area* $\int \eta(t)dt$ for arbitrary acoustic pulses obeying $\tau_{\text{ac}} \ll T_{\text{prec}} \simeq 25$ ps. It suggests that the so-called bipolar acoustic pulses generated at free metal-air interfaces [5] are particularly inefficient in magnetoacoustics, since positive and negative parts in a bipolar pulse cancel each other giving a zero acoustic pulse area. The multilayer structure in Fig. 1(a) was designed to generate both compressional and tensile unipolar picosecond acoustic pulses with an amplitude up to 1.0% [21,22], which are better suited for experimental investigations in coherent magnetoacoustics.

When using rectangular unipolar acoustic pulses, the deflection angle $\Delta\theta_{\text{ac}}$ is proportional to the product $\eta_{\text{ac}}\tau_{\text{ac}}$ and thus can be increased by using larger strain amplitude η_{ac} or somewhat longer pulse duration $\tau_{\text{ac}} \ll T_{\text{prec}}$.

Figure 3 shows that both compressional [$\eta_{\text{ac}} < 0$, Fig. 3(a)] and tensile [$\eta_{\text{ac}} > 0$, Fig. 3(b)] rectangular unipolar acoustic pulses with $\tau_{\text{ac}} = 3$ ps are capable of switching the initial magnetization direction into the new minimum, which represents the main result of this Letter. For example, a 3-ps-long compressional pulse with strain amplitude 1.6% switches the magnetization from minimum 2 into minimum 4 [$2 \rightarrow 4$; see Fig. 3(a)]. Application of a subsequent identical compressional pulse brings the magnetization back into minimum 2 [$4 \rightarrow 2$], suggesting that a pulse train will result in clockwise rotation of the magnetization vector. In contrast, a sequence of 3-ps-long tensile strain pulses will periodically switch the magnetization between minima 2 and 1 [$2 \rightarrow 1$ and $1 \rightarrow 2$; see Fig. 3(b)]. Therefore, the results of our simulations suggest that a clean experimental demonstration of magnetoacoustic switching would necessarily imply a single-shot experiment where the magnetic system is prepared in the same state before the action of the subsequent acoustic pulse.

The typical deflection angle required for switching is about 20°, and the different threshold switching amplitudes and pathways for tensile and compressive pulses are caused by different heights of the potential barrier between the neighboring energy minima. The more general

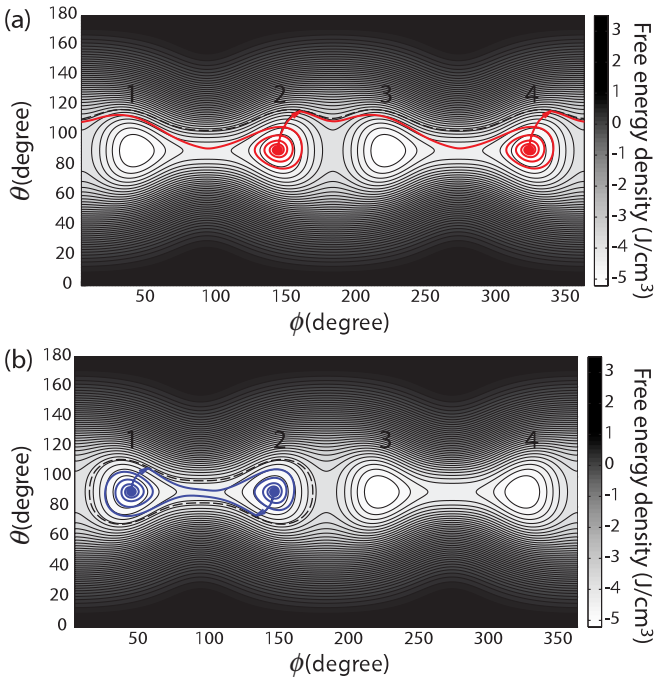

FIG. 3 (color online). After application of a large-amplitude 3-ps-long acoustic pulse, the magnetization vector initially prepared in state 2 will decay (switch) into one of the neighboring energy minima. (a) A sequence of compressional pulses with $\eta_{ac} = -1.6\%$ induces the clockwise rotation of the magnetization vector: $2 \rightarrow 4$, $4 \rightarrow 2$, etc. (b) A sequence of tensile pulses with $\eta_{ac} = 0.9\%$ results in repetitive switching between two adjacent energy minima: $2 \rightarrow 1$, $1 \rightarrow 2$, etc. Dashed contours show the magnetization trajectories after excitation by a single pulse in the absence of damping.

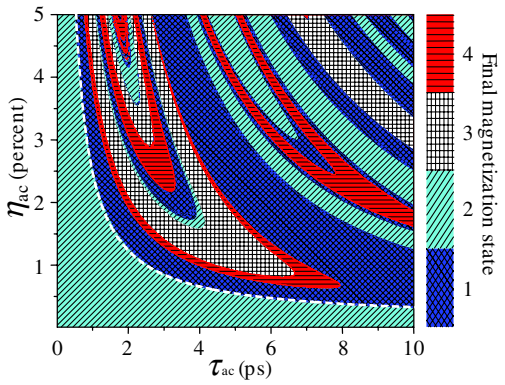

FIG. 4 (color online). Diagram of magnetoacoustic switching. Depending on the duration τ_{ac} and amplitude η_{ac} of a unipolar tensile acoustic pulse, the magnetization vector initially prepared in state 2 undergoes the transition $2 \rightarrow 1$ (diagonal mesh), $2 \rightarrow 4$ (horizontal lines), $2 \rightarrow 3$ (horizontal mesh), or $2 \rightarrow 2$ (no switching, diagonal lines). The dashed white curve shows the contour of constant acoustic pulse area $\eta_{ac}\tau_{ac} = 2.6$ (see the Supplemental Material [20] for details).

phase diagram for magnetoacoustic switching is shown in Fig. 4, where the boundaries between different switching zones generally follow the $1/\tau_{ac}$ dependence, in agreement with the assumption that primarily the amplitude of out-of-plane acoustic deflection $\Delta\theta_{ac} \sim \eta_{ac}\tau_{ac} = \text{const}$ determines the switching pathway. Similar analysis for the acoustic shear pulses leads to the same conclusions, in particular, with respect to the acoustic pulse area and dependence of the switching amplitude on the acoustic pulse duration.

It is quite remarkable that the threshold fluence for all-optical magnetization switching in a rare-earth compound GdFeCo induced by a single circularly polarized pump pulse almost does not depend on the optical pulse duration [14]. Longer optical excitation leads to longer acoustic pulses [23], which are generated by thermoelastic expansion of laser-heated GdFeCo. The analysis of the thermoelastic generation mechanism by longer optical pulses suggests that the absorbed laser fluence is proportional to the product of acoustic pulse amplitude by its duration, i.e., to the acoustic pulse area. Therefore, the observed constant threshold fluence for all-optical magnetization switching in GdFeCo could be explained by the magnetoelastic mechanism. From the magnetoacoustic point of view, a significantly lower magnetostriction coefficient $\Lambda \sim 10^{-5}$ in GdFeCo [24] is balanced by a much lower room temperature saturation magnetization $\mu_0 M_s \simeq 8$ mT close to the compensation point [12], thus giving the same order-of-magnitude prefactor Λ/M_s in Eq. (9). The observed increase of switching fluence in GdFeCo with temperature [15] is consistent with the decrease of Λ/M_s in rare-earth compounds for higher temperatures [17]. The recent experiments in $Fe_{100-x}Tb_x$ alloy films [25] showed that all-optical switching could be observed only in samples characterized by a small remanent magnetization, and the measurements in rare-earth orthoferrite $(SmPr)FeO_3$ [26] revealed an important role of coherent low-amplitude spin precession. Therefore, despite being far away from quantitative modeling, all these arguments corroborate the hypothesis [21] that even a low-amplitude magnetoacoustic precession could play an important role in the dynamics of all-optical magnetization switching in strongly absorbing samples. Moreover, possible magnetoelastic contributions of helically polarized acoustic shear pulses excited by circularly polarized electromagnetic pulses [27,28] may solve the puzzle of an undefined long-lived reservoir for angular momentum as highlighted in the most recent systematic investigations [14,15].

In summary, in this Letter, we have theoretically predicted a new mechanism of ultrafast *nonthermal* magnetoacoustic switching in Terfenol-D. This phenomenon may open the door to ultrafast magnetic recording not relying on heating the magnetic material close the Curie point.

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