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# Phase-change magnetic memory: Rewritable ferromagnetism by laser quenching of chemical disorder in Fe<sub>60</sub>Al<sub>40</sub> alloys

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## TEASER

In systems that store data magnetically, the information can be corrupted by external magnetic fields and by thermal fluctuations. One possible approach, which could solve both of these problems, is altering reversibly the magnetization magnitude instead of its direction. The authors propose such an alternative magnetic memory upon the basis of chemical order-disorder transformations in Fe-Al alloys. It is found out that room-temperature ferromagnetism in Fe<sub>60</sub>Al<sub>40</sub> thin-film ( $\approx 40$  nm) samples is recoverable after rapid resolidification of the alloy, which follows its irradiation by a short laser pulse with intensity sufficient for melting.

## ABSTRACT

High-intensity laser irradiation can effectively couple to practically all kinds of materials, causing modification of their physical properties due to laser-induced phase transformations. This gives rise to a broad field of laser micro- and nanofabrication with its various technological applications. We demonstrate that, in an Fe<sub>60</sub>Al<sub>40</sub> (FeAl) thin-film ( $\sim 40$  nm) alloy, a short laser pulse is capable of (re)writing the ferromagnetism observed at room temperature (RT). The energy of the pulse generating a ferromagnetic region has to be so high to provide a temperature rise up to melting of the FeAl layer, while the ferromagnetic state can be erased by various kinds of lower-intensity thermal treatment. This cycling of RT ferromagnetism can be explained in terms of the chemical order (B2) – disorder (A2) phase transition in the FeAl crystal lattice, which is affected by laser-induced melting and rapid resolidification. Our finding has implications for the development of a magnetic memory technology that would use the reversibility of the modulus of the magnetization vector instead of its direction. This promises to circumvent the problem of the superparamagnetic limit for magnetic data storage density.

## INTRODUCTION

Interaction of strong laser radiation with solids and their surfaces is a long-standing research problem [1] that has given rise to intense studies of ultrafast phase transformations in the context of microfabrication [2, 3]. In combination with a small object (e.g., a nanoparticle [4, 5] or the tip of a scanning probe microscope [6, 7]) placed near a solid surface subject to patterning, laser light can act as

a source of the surface modifications produced through the optical near-field enhancement at the nanoscale (<100 nm) [8]. Various works that address the direct laser-assisted patterning of small features contribute to the field of thermal nanolithography [9, 10, 11]. A particular kind of phase changes that holds promise for laser nanostructuring of solid surfaces is crystallization/vitrification (or chemical ordering/disordering) of phase-change alloys [12, 13, 14]. If a phase transformation of this kind is accompanied by a reversible change of physical properties of the system, e.g., optical reflectivity or electrical resistivity, it can serve as a basis for manufacturing of next-generation memory devices that might have a relatively easy scaling path to reach densities exceeding those offered by current and future recording technologies. For example, information bits in conventional magnetic recording are limited in their sizes by thermal upsets of the magnetizations that occur when the bit sizes are below the superparamagnetic threshold [15]. An alternative approach to the magnetic storage technologies is to cycle (generate and delete) regions whose magnetic properties are different from those of a surrounding matrix [16, 17, 18, 19]. In such patterned media, the bits of binary information are “larger magnetization” and “smaller magnetization” to be still distinguishable from each other under spontaneous changes of magnetization directions, [17, 20] as having different levels of the stochastic magnetic signal [21]. Recently, it has been demonstrated [17] that crystalline  $\text{Fe}_{40}\text{Ni}_{38}\text{Mo}_4\text{B}_{18}$  films can be vitrified and then again crystallized by laser irradiation. Although all obtained states, with different chemical order, are ferromagnetic in such alloys, the detected difference in magnetic response is sufficient (~5 %) to be distinguishable with a magnetic sensor placed close to the film surface. Another phase-change magnetic material was uncovered by *Song et al.* [18] who demonstrated a nearly two-fold reversible change of the saturation magnetization in  $\text{Fe}_7\text{Ge}_{15}\text{Sb}_{33}\text{Te}_{45}$  thin films induced by short-pulse laser irradiation. Even a much higher contrast in the magnetic properties, from ferrimagnetic to antiferromagnetic state, has been achieved and attributed to the order-disorder phase transition in the  $\text{FeMnP}_{0.75}\text{Si}_{0.25}$  compound synthesized in the bulk form [19].

Here we consider a class of materials, that is, binary thin-film alloys with chemical order-disorder transition, that can be of interest in view of phase-change magnetic memory. We demonstrate that room-temperature (RT) ferromagnetism in a thin-film ( $\approx 40$  nm)  $\text{Fe}_{60}\text{Al}_{40}$  (FeAl) alloy is rewriteable and reveal the physical conditions required for writing and erasing ferromagnetic regions within the film. A short laser pulse with energy sufficient for transient melting of an FeAl film, which initially has nearly zero magnetic response at RT, is capable of producing the ferromagnetism within the laser spot [22]. This ferromagnetic state can be deleted by applying different types of lower-intensity (below melting) thermal treatments, e.g., standard annealing, high-density electric current, or a train of laser pulses, and then recovered by subsequent high-intensity laser irradiation. The material selected for probing thermally induced phase transformations,  $\text{Fe}_{60}\text{Al}_{40}$ , is well established [23-35] to exhibit the chemical reversible order-disorder (B2-A2) phase transition, which occurs via atomic jumps between adjacent Fe and Al-rich planes through the vacancy diffusion mechanism [23, 24, 25]. It has been previously demonstrated experimentally [26-34] and supported by first principles calculations [35, 36, 37] that the ordered B2 state (Fig. 1a) in  $\text{Fe}_{1-c}\text{Al}_c$  ( $c > 0.3$ ) alloys is non(ferro)magnetic [38]. Transformation of the chemically ordered B2 into disordered A2 state – with increasing the number of nearest Fe-Fe neighbors (Fig. 1b) – provides the enhancement of ferromagnetism [26-35] due to bigger overlapping of electron  $d$  orbitals between the Fe atoms occupying centers and vertexes of the body-centered cubic (bcc) FeAl lattice [34, 39]. It has been also argued [28] that the magnetic percolation between Fe atoms in the  $\text{Fe}_{60}\text{Al}_{40}$  alloy leads to the emergence of long-range ferromagnetic order. This picture is consistent with vast evidence that the ferromagnetism in FeAl is highly influenced by structural defects such as antiphase boundaries produced at high densities by plastic deformation [26] or quenching [32]. Previously, the disordering and, thus, the onset of ferromagnetism were achieved through various kinds

of treatments, e.g., cold working [26, 27], ball milling [28, 29, 30], and ion irradiation [24, 25]. The produced ferromagnetism can be suppressed (chemical ordering) with standard thermal annealing in a temperature range of 400–1000 K [29, 31]. In our work, we show that the ferromagnetism in FeAl is also rewritable with short-pulse laser irradiation. This finding is promising for applications utilizing compact and inexpensive laser sources for magnetic recording technologies.

## METHODS

*Sample preparation and characterization.*— Polycrystalline films of Fe<sub>60</sub>Al<sub>40</sub> composition and thickness of 40 nm were prepared by magnetron sputtering from an Fe<sub>60</sub>Al<sub>40</sub> target onto thermally oxidized Si(100) substrates. Magnetron sputtering was performed at RT in an Ar<sup>+</sup> atmosphere of 1.8 x 10<sup>-3</sup> mbar. In order to get the initial ideally nonmagnetic state (B2 ordered phase), the sputtered samples were thermally annealed at 1 x 10<sup>-5</sup> mbar in a vacuum annealing furnace in a temperature range of 700–800 K. After annealing, the grain diameter increased up to ≈15 nm, which was determined with the Scherrer equation from grazing incidence X-Ray diffraction (GIXRD) patterns. In laser irradiation of the post-annealed samples, we use intense incident fluence (up to ~1 J/cm<sup>2</sup>) laser beams with both nanosecond and sub-picosecond durations. In these experiments, the 3<sup>rd</sup> harmonics (λ=355 nm) of a nanosecond (≈5 ns) Q-switched Nd:YAG laser (EKSPILA) as well as the fundamental line (λ=1030 nm) of an Amplitude Systèmes femtosecond laser system with pulse duration of 500 fs were employed. Magnetic properties of the as-prepared, thermally annealed and laser-irradiated samples were probed with longitudinal magneto-optic Kerr effect (MOKE). To observe the MOKE, the probing beam from a laser diode module, operating at wavelength radiation of λ=670 nm with a power of 5.5 mW, was focused on the film surface.

*Simulation.* – The kinetics of laser-induced heating and cooling is studied with a conventional two-temperature model (TTM) [40] enhanced with computational treatment of melting and resolidification occurring under nonequilibrium conditions of superheating/undercooling, as well as the heat transfer from the film to the substrate. The TTM provides a continuum-level description of the laser excitation and subsequent relaxation of the conduction-band electrons. The equations of the TTM model are solved using the backward (implicit) Euler algorithm based on the block tridiagonal matrix inversion. The simulations are performed for a computational system consisting of 40 nm Fe<sub>60</sub>Al<sub>40</sub> film on a 2-μm-thick silicon substrate covered by a 150-nm-thick SiO<sub>2</sub> layer. A detailed description of the model is given in Refs. [41, 42], and below we only outline the parameters relevant for the current setup.

The irradiation of the target by a 500 fs laser pulse is represented through a source term added to the TTM equation for the electron temperature. The source term accounts for the excitation of the conduction band electrons by a laser pulse with a Gaussian temporal profile and reproduces the exponential attenuation of laser intensity with depth under the surface (Beer–Lambert law), with the optical absorption depth of 19 nm assumed to be the same as for pure Fe at the laser wavelength of 1030 nm [43]. Since the laser spot size used in the experimental setup is much larger than the heat diffusion length during the time of melting and resolidification, ~1 μm, we assume the one-dimensional heat transfer along the direction normal to the sample surface.

Since the available temperature dependencies of electron heat capacity and electron-phonon coupling accounting for the contribution from the thermal excitation from the electron states below the Fermi level are mostly limited to pure metals [43], the dependences calculated for α-Fe similarly to those calculated for some other pure metals in Ref. [44] are used in the calculations performed for the Fe<sub>60</sub>Al<sub>40</sub> alloy. Similarly to Ref. [41], the temperature dependence of the electron thermal conductivity is approximated by the Drude model relationship,  $K_e(T_e, T_l) = v^2 C_e(T_e) \tau_e(T_e, T_l) / 3$ , where  $C_e(T_e)$  is the electron heat capacity,  $v^2$  is the mean square velocity of the electrons contributing to the electron heat

conductivity, approximated in this work as the Fermi velocity squared,  $v_F^2$ , and  $\tau_e(T_e, T_l)$  is the total electron scattering time defined by the electron-electron scattering rate,  $1/\tau_{e-e} = AT_e^2$ , and the electron-phonon scattering rate,  $1/\tau_{e-ph} = BT_l$ , so that  $1/\tau_e = AT_e^2 + BT_l$ . The value of the coefficient  $A$  ( $8.9 \times 10^5 \text{ K}^2 \text{ s}^{-1}$ ) is estimated within the free electron model [45]. The coefficient  $B$  is described as a function of the lattice temperature, so that the experimental temperature dependences of the thermal conductivity of  $\text{Fe}_{60}\text{Al}_{40}$  alloy [46, 47] under the conditions of electron-phonon equilibrium are reproduced in the simulations.

A single “melting” temperature of the  $\text{Fe}_{60}\text{Al}_{40}$  alloy,  $T_m = 1662 \text{ K}$ , approximated as an average of solidus and liquidus temperatures [48], is used to simplify the description of melting and resolidification processes in the model. The temperature dependence of the velocity of the liquid-crystal interface is described by the Wilson–Frenkel expression [49] with parameters taken for Fe (100) interface from Ref. [50]. Since pure Fe has different melting temperature than the  $\text{Fe}_{60}\text{Al}_{40}$  alloy, the diffusion activation energy and the heat of fusion are renormalized based on the  $T_m$  of the alloy to ensure the same velocity of the interface at the same values of relative undercooling  $T/T_m$ :

$$V_l = V_0 \cdot \exp\left(-\tilde{Q} \cdot T_m / T\right) \left[1 - \exp\left(-\tilde{H}_f \cdot (T_m / T - 1)\right)\right],$$

where  $V_0$  is a pre-factor equal to 2200 m/s,  $\tilde{Q} = Q^{Fe} / k_B T_m^{Fe} = 1.23$  is the reduced diffusion activation energy, and  $\tilde{H}_f = H_f^{Fe} / k_B T_m^{Fe} = 0.884$  is the reduced heat of fusion. To account for fast homogeneous melting of superheated solid, the regions of the target where the lattice temperature exceeds  $1.3T_m$  are set to undergo instantaneous melting. If liquid is undercooled below  $0.7T_m$ , the onset of homogenous solidification is assumed, and liquid transforms into a solid state. The lattice heat capacity of the alloy is assumed to be equal to  $3R$  and is calculated based on the lattice parameter  $a = 0.29 \text{ nm}$  of  $\text{Fe}_{60}\text{Al}_{40}$  B2 structure, e.g. Ref. [28], which gives  $3.4 \times 10^6 \text{ J/m}^3 \text{ K}$ . The heat of fusion is estimated as a weighted average of values for Al and Fe based on the composition of the alloy, yielding  $1.7 \times 10^9 \text{ J/m}^3$ .

The heat capacity of Si and  $\text{SiO}_2$  (silica) are assumed to be equal to  $1.66 \times 10^6$  and  $1.63 \times 10^6 \text{ J/m}^3 \text{ K}$ , respectively [51], while the thermal conductivities are fitted to the experimental temperature dependences provided in Ref. [52]. Since the value of interfacial thermal conductance depends not only on the materials in the contact but also on the method of the film deposition, it cannot be reliably evaluated without direct experimental measurements. Therefore, the thermal conductance of the  $\text{FeAl/SiO}_2$  interface is assumed to be  $10^8 \text{ W/m}^2 \text{ K}$ , which is a typical value measured for metal-oxide interfaces in experiments [52]. The value of the thermal conductance of the  $\text{SiO}_2/\text{Si}$  interface is taken as  $2.5 \times 10^9 \text{ W/m}^2 \text{ K}$  [53].

## RESULTS

Experimentally, we study  $\text{Fe}_{60}\text{Al}_{40}$  films sputtered at RT onto thermally oxidized Si (100) substrates ( $\text{SiO}_2/\text{Si}$ ) with sizes of  $10 \times 10 \text{ mm}^2$ . As-prepared samples are thermally annealed at  $T=773 \text{ K}$  (TA) for their successive treatment by laser irradiation. Figure 2a shows magnetization curves obtained by measuring the MOKE at RT in a TA- $\text{Fe}_{60}\text{Al}_{40}$  film [31] and after irradiation of this sample by a 5-ns pulse delivered to the sample surface by a multimode laser beam [54] with incident fluence close to the ablation threshold ( $\sim 0.5 \text{ J/cm}^2$ ). The ablation induced by the nanosecond (ns) laser with the multimode beam was detected with an optical microscope as the appearance of multiple damaged locations within a spot with diameter of a few tens of micrometers on the film surface.

With our MOKE setup, we observe a nearly zero MOKE response (signal-to-noise ratio is about 1:1) from the TA- $\text{Fe}_{60}\text{Al}_{40}$  film in applied magnetic field up to  $\mu_0 H \sim 100 \text{ mT}$  (inset in Fig. 2a). We see, however, that the TA- $\text{Fe}_{60}\text{Al}_{40}$  film irradiated by laser exhibits a strong MOKE response, comparable to that from a

film of pure  $\alpha$ -Fe ( $\approx 0.1^\circ$  in Kerr rotation), while the hysteresis loop measured is typical for structurally homogeneous ferromagnetic materials. We also see that the MOKE response after irradiation by the laser is close to that after irradiation by 25 keV Ne<sup>+</sup> ions [31]. The saturation magnetization of the sample irradiated by the ion beam was measured to be  $\approx 0.98$  T [31], which is nearly twice lower than the saturation magnetization in pure  $\alpha$ -Fe. The strongly ferromagnetic state obtained can be suppressed, in particular, by repeating the TA procedure [38]. In Fig. 2b we demonstrate cycling of the RT MOKE response, which reflects an extent of the chemical order, under successive TA procedures and laser irradiation. In the experiments, we produced ferromagnetic regions in a TA-Fe<sub>60</sub>Al<sub>40</sub> sample, which were erased repeating the TA procedure and then were recovered by laser. Alternatively, a high-density electric current passing through a  $\mu\text{m}$ -scale paramagnetic Fe<sub>60</sub>Al<sub>40</sub> stripe [55] or a train of lower-intensity laser pulses [56] can be used for erasing ferromagnetic regions produced by single high-intensity laser pulses, as shown schematically in Fig. 2c.

Having analyzed the microstructure of our samples with GIXRD, we confirm the relationship between the magnetic order and structural transformation in the FeAl system. Figure 2d shows GIXRD scans taken at a 1-degree grazing angle of the incident x-ray beam for the TA-Fe<sub>60</sub>Al<sub>40</sub> sample and after its subsequent ns-laser irradiation in the regime that provides the enhancement of ferromagnetism (Fig. 2a-b). In order to collect GIXRD patterns of laser-irradiated samples, we scanned the whole sample surface by the laser beam. In addition, we show a GIXRD scan for a similar TA-Fe<sub>60</sub>Al<sub>40</sub> sample which was irradiated by Ne<sup>+</sup>-ions for chemical disordering of the alloy and thus for achieving the ferromagnetism induced by the ion beam [31]. We see that strong laser irradiation induces a transformation that is similar to that occurring under ion irradiation, i.e., chemical disordering of the crystal structure [31]. Indeed, both kinds of treatments give reduction the (100) reflection from the superstructure (shown in the inset in greater detail), which indicates the reversible (Fig. 2b) transformation of the nearly nonmagnetic, structurally ordered, B2 state into the ferromagnetic, structurally disordered, A2 phase.

Interestingly, we find that the ferromagnetism in a TA-Fe<sub>60</sub>Al<sub>40</sub> sample can be induced even with femtosecond (fs) laser. Figure 3 (a) shows distribution of the MOKE response along a location on the surface of a TA-Fe<sub>60</sub>Al<sub>40</sub> film irradiated by a  $\sim 500$ -fs pulse with an incident fluence above the ablation threshold, that is,  $\sim 0.4$  J/cm<sup>2</sup> for this pulse duration. In order to retrieve the distribution of the MOKE response, the light beam from the fs-laser system is focused onto the sample surface with a 30-cm-focal-length lens. The magnetization in the irradiated zone is probed with a MOKE beam focused onto the spot with diameter of  $\sim 30$   $\mu\text{m}$ . In order to explain the obtained MOKE distribution, Figure 3(b) depicts the fs-laser beam profile approximated as  $\exp(-2\rho^2/D^2)$ , where  $\rho$  is the distance from the laser spot center and  $D \approx 200$   $\mu\text{m}$  is the laser spot diameter. The melted and ablated zones as well as the corresponding MOKE response are shown at the bottom of Fig. 3 (b). We see that the MOKE response (and thus magnetization) is maximal at the periphery of the ablated region, while the measured hysteresis loop is close to that observed after irradiation by a ns-laser pulse (Fig. 2a). The MOKE intensity quickly decays with increasing distance from its edge. Thus, the patterned ferromagnetic region is ring-shaped and adjacent to the ablated zone, suggesting that melting of the FeAl material is needed for the formation of ferromagnetic state upon resolidification and cooling down to RT. This conclusion correlates with the results of simulations of laser-induced phase transformations in metallic targets (e.g., Ref. [57]), which predict the melting fluence threshold to be several times lower than the threshold for spallation/ablation.

We also note that the pronounced MOKE response is also achievable with a single fs pulse irradiation below the ablation threshold (Fig. 3c). In the multi-shot regime – with a pulse repetition rate of  $10^4$  Hz and with the number of pulses increasing to  $10^2$  and  $10^4$ , the MOKE response further enhances. However, its increase is limited by ablation of the FeAl film. The ablation thresholds are established through optical microscopy measurements and are indicated by the arrows in Fig. 3c. As a number of shots increases, the ablation threshold shifts toward the lower values of laser energy,

suggesting that the ablation of the thin FeAl film exhibits the so-called incubation effect that has been reported, for instance in Ref. [58]. It is interesting to note that a train of  $10^4$  fs pulses initiates ablation at a pulse energy that is still insufficient to induce a nonzero MOKE response with a single pulse and is, presumably, below the melting threshold with a single pulse. This difference between the results obtained in the single- and multi-shot regimes indicates the cumulative effect of multiple shots on the onset of melting and ablation.

## DISCUSSION

The experimental results described above suggest that short-pulse laser irradiation produces chemical disordering in the FeAl atomic lattice above the critical temperature  $T_c$  for the order-disorder transition. According to the Fe-Al phase diagram [48] that shows the ordered B2 phase to be the equilibrium state of  $\text{Fe}_{60}\text{Al}_{40}$  alloy below  $T_c=1563$  K for the A2-B2 transformation, which is only 5% below the solidus temperature of the alloy [48] (a single “melting temperature”,  $T_m = 1662$  K, defined here as average between the solidus and liquidus temperatures of 1642 K and 1682 K, respectively, and is used in this paper to simplify the discussion and modeling). The disordering is observed in the regions that are likely to undergo transient melting followed by resolidification, and the disordered state in these regions manifests itself by exhibiting ferromagnetism after cooling to room temperature. This observation, however, leads to a question of why does the disordered state (and thus ferromagnetism) persists in the sample, while thermal annealing at a much lower  $T=773$  K readily produces chemical ordering (Fig. 2). The formation of chemically disordered structures by rapid solidification by itself is not surprising and has been considered theoretically [59] and observed experimentally [60, 61, 62] even for alloys with  $T_c > T_m$ , where a strong driving force for ordering exist at any level of undercooling. For  $\text{Fe}_{60}\text{Al}_{40}$ , where  $T_c < T_m$ , the chemical disorder can be readily trapped by the advancing solidification front, and the transformation to the ordered phase can be expected to occur through the diffusional atomic rearrangements in the solid state.

In order to reveal the physical conditions leading to the generation and survival of the chemically disordered state in the irradiated film, we perform a series of continuum-level simulations of melting and resolidification of a 40-nm-thick FeAl film deposited on a  $\text{SiO}_2(150 \text{ nm})/\text{Si}$  substrate and irradiated with a 500 fs laser pulse. These simulations allow us to evaluate the cumulative atomic diffusion length in the FeAl alloy after its resolidification. The computational model is described in the section “Methods”, and the simulation conditions mimic the ones of experiments used for collection of data presented in Fig. 3.

The surface temperature profiles shown in Fig. 4a-c for three values of the absorbed laser fluence clearly demonstrate the strong effect of melting on the thermal history of the surface region of the film after the solidification and, therefore, on the extent of atomic rearrangements that can take place during the cooling of the solidified surface. The variation of the intensity of color in the colored areas under the temperature profiles provides a visual representation of the temperature dependence of the atomic mobility in the solid state. In the simulations where complete or partial melting takes place, Figs. 4a, b, the reordering starts only after the resolidification and cooling down below  $T_c$ , if  $T_c$  is lower than the temperature reached at the end of resolidification (Fig. 4b). It is notable that the conditions of partial melting (Fig. 4b) provide more time for the active high-temperature diffusion after the end of resolidification as compared to the complete melting (Fig. 4a), the liquid is undercooled to a stronger level before the homogeneous crystallization occurs, and the sharp temperature spike caused by the release of the latent heat of melting is followed by a rapid cooling. At the low fluence,  $F_{abs} = 14 \text{ mJ/cm}^2$ , Fig. 4c, a brief ( $\sim 10$  ps) superheating of the surface above  $T_m$  is too momentary for any significant

propagation of the melting front, which is limited to less than 0.5 nm or a half of a cell in the spatial discretization in the model. Thus, we consider this fluence to be just below the melting threshold (see Fig. 4d). As shown below through the analysis of cumulative diffusion length, the exposure of the solid film to high temperatures provides an opportunity for substantial atomic rearrangements, even though the diffusion is activated only during a few hundreds of picoseconds (Fig. 4c).

Since the A2-B2 transition occurs via atomic mixing between Fe- and Al-dominated planes proceeding through the vacancy diffusion mechanism [23, 24, 25], we evaluate the cumulative mean square displacement of Fe and Al atoms in the course of cooling after the end of resolidification as

$$\Delta^2 = 6 \int_{t_1}^{t_2} D[T_s(t)] dt, \quad (1)$$

where  $D(T_s) = D_0 \exp(-Q/RT_s)$  is the atomic diffusivity in the alloy with the pre-factor  $D_0 \approx 1.1 \times 10^{-3} \text{ m}^2/\text{s}$  [22] and activation energy  $Q = 241 \text{ kJ/mol}$  [23, 24, 25],  $R = 8.314 \text{ J K}^{-1} \text{ mol}^{-1}$  the universal gas constant,  $t_1$  is the time of complete resolidification of the film (indicated by the arrows for two values of  $F_{\text{abs}}$  in Fig. 4a and Fig. 4b), while  $t_2$  is the total duration of the simulation chosen to ensure a negligible change of  $\Delta^2$  upon further integration beyond  $t_2$ . Since the thermal energy redistribution within the 40-nm-thick metal film is rapid and takes  $\sim 100 \text{ ps}$ , the temperature of the surface of the film,  $T_s$ , is used in the evaluation of the atomic diffusion length in the film. Note that Eq. (1) with parameters evaluated for the equilibrium vacancy concentrations provide a lower-end estimation of the atomic diffusion, as the rapid resolidification proceeding under conditions of strong undercooling below the melting temperature can generate a strong supersaturation of the film with vacancies [42].

As shown in Fig. 4e, the dependence of the effective atomic diffusion length ( $\Delta$ ) on the fluence exhibits a sharp drop from its maximum value of 0.25 nm reached at  $F_{\text{abs}} \approx 32 \text{ mJ/cm}^2$  down to  $\sim 0.07 \text{ nm}$  upon the complete melting of the FeAl film. This drop in  $\Delta$  can be explained by strong undercooling of the melt prior to the complete resolidification [41]. When the film is only partially melted, the solidification proceeds through the propagation of solidification front from the solid part of the film, and the temperature stays at a relatively high level of  $\sim 0.94T_m$  until the end of the resolidification (see Fig. 4b). The films melted to the full depth, on the other hand, resolidify through the rapid nucleation and growth of new crystallites activated when the molten metal cools down to  $\sim 0.77T_m$ . The release of the heat of melting upon the solidification leads to a transient spike in the temperature of the film, *e.g.*, up to  $T(t_1) \approx 0.86T_m$  at  $F_{\text{abs}} \approx 35 \text{ mJ/cm}^2$  (Fig. 4a). The diffusional reordering of the random solid solution generated by the solidification starts at this maximum temperature, but the film continues to cool, and the temperature rapidly drops down to the level where the diffusional reordering ceases. Indeed, from the value of  $\Delta < 0.1 \text{ nm}$  calculated for  $F_{\text{abs}}$  above the threshold for complete melting to the full depth of the film, Fig. 4e, we can estimate that only  $\approx 16\%$  of atoms should change their positions per laser pulse, which is the upper bound for the amount of the reordered B2 phase. Therefore, the A2 phase and thus ferromagnetism can be expected to persist in the alloy after its cooling to RT. Note that a strong temperature dependence of the thermodynamic driving force for the A2 to B2 ordering phase transformation does not allow us to quantitatively link the magnitude of  $\Delta$  to the degree of the phase transformation produced by the laser irradiation. Nevertheless, it provides a useful measure of the extent of the laser-induced atomic rearrangements in the solidified film, as a sufficiently large magnitude of  $\Delta$  is a necessary (although not sufficient) condition for the transformation to the chemically ordered state.

The relatively low value of  $\Delta$  produced upon irradiation by a single laser pulse below the melting threshold suggests that such pulse is unlikely to induce complete A2 to B2 phase transformation.



Multiple laser pulses with the energy in each pulse not sufficient to induce melting and thus chemical disordering in the film can provide a sufficient cumulative time of the thermal spikes for the generation of a substantial amount of the B2 phase. The laser-induced reordering and, thus, erasing of the ferromagnetism has been demonstrated in our further experiments performed with magnetic microscopy imaging based on x-ray magnetic circular dichroism (XMCD) collected *in situ* [56]. The performed experiments indicated that even a single femtosecond pulse with intensity below  $T_c$  was able to lower the XMCD contrast obtained with a higher intensity (above  $T_c$ ) pulse, while the magnetic signal was totally suppressed with  $\sim 10^4$  pulses to be recovered a subsequent higher-intensity pulse. These results are compatible with the cumulative mean square displacement of Fe and Al atoms evaluated using Eq. (1). Alternatively, the reordering under standard thermal annealing at moderate temperatures, e.g., at  $T = 773$  K, can be achieved within  $t \sim a^2/D \approx 1.0$  s, where  $D \sim 0.1$  nm<sup>2</sup>/s at this  $T$  [23, 24].

## CONCLUSIONS

We find that thin ( $\sim 40$  nm) Fe<sub>60</sub>Al<sub>40</sub> films exhibit nonvolatile ferromagnetism at RT after irradiation by nanosecond or femtosecond laser pulses. This ferromagnetism can be erased under longer heat treatments at moderate temperatures (below melting), e.g., by standard thermal annealing, high-density electric current, irradiation by a longer laser pulse or a train of short laser pulses, all of which can lead to reordering of the atomic structure. After such treatment, the chemically disordered state (and thus RT ferromagnetism) can be regenerated through irradiation by a subsequent high-energy laser pulse. The simulations of laser interaction with FeAl films suggest that the ferromagnetic state is produced by rapid melting and resolidification of the FeAl film that traps the metastable chemically disordered state. We envision that cycling of the ferromagnetism in Fe<sub>60</sub>Al<sub>40</sub> alloys could be utilized in the design of rewritable devices for long-time magnetic storage of information, e.g., in cryptography [63], ID cards [17] and archiving [64]. Using this approach in combination with near-field patterning techniques [3, 4, 5, 6, 7, 8] for rewriting of nanoscale ( $< 100$  nm) magnets would have an advantage over conventional magnetic memories, since the problem of thermal fluctuations of magnetic moments below the superparamagnetic threshold [15] can be circumvented if one reads out, for instance, the magnetic permeability [17] rather than the orientation of magnetization in an information bit.

## ACKNOWLEDGMENTS

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## CAPTIONS TO FIGURES

Figure 1. FeAl structure: (a) Ordered B2 and disordered A2 structures in the body-centered cubic (bcc) FeAl lattice. The B2–A2 transition can proceed through atomic jumps between Fe and Al-rich planes via the vacancy diffusion mechanism (vacancies are indicated by the dashed boxes). (b) The bcc unit cell in FeAl. In the A2 phase, an Fe atom appearing in the cube center has Fe neighbors at a closer distance of  $a\sqrt{3}/2$ , where  $a$  is the lattice constant. This atomic configuration provides bigger overlapping of electron  $d$  orbitals, which is responsible for the enhancement of ferromagnetism.

Figure 2. Laser-induced ferromagnetism: (a) MOKE hysteresis loops taken at RT of a TA-Fe<sub>60</sub>Al<sub>40</sub> sample and after its irradiation by a 5-ns laser pulse with an incident fluence close to the ablation threshold,  $\sim 0.5$  J/cm<sup>2</sup>. For comparison, we show the MOKE response of the same sample irradiated by 25 keV Ne<sup>+</sup> ions [31]. The inset depicts the MOKE response of a TA-Fe<sub>60</sub>Al<sub>40</sub> film in applied fields up to  $\mu_0 H \sim 100$  mT (b) Cycling of the MOKE response under successive application of thermal annealing and laser irradiation. (c) Schematic of laser writing of strongly ferromagnetic regions with a high-fluence laser pulse as well as of their suppression with thermal treatments of different kinds at moderate temperatures, e.g., thermal annealing in a furnace or with a train of lower-intensity laser pulses [55]. (d) GIXRD scans of the TA-Fe<sub>60</sub>Al<sub>40</sub> sample and after its irradiation with a ns-laser pulse near the ablation threshold, as well as after its irradiation by 25 keV Ne<sup>+</sup> ions [31]. The inset shows laser- and ion-induced degradation of the superstructure in the ordered B2 phase. The X-ray data is collected after ns-laser irradiation that produces the transformation through the whole sample area by scanning over it in the multi-shot regime.

Figure 3. Correlation of MOKE response with transient melting of FeAl: (a) Distribution of the MOKE response over the spot on the sample surface irradiated by a  $\sim 500$  fs laser pulse with energy above the threshold for ablation in a single pulse irradiation. The inset depicts an optical image of the irradiated spot with ablated zone (dark spot) and surrounding ring-shaped ferromagnetic zone outlined by the dashed circle. (b) The Gaussian profile of the laser beam with indication of the ablation zone and the FeAl ring with the modified properties. A strong MOKE response and ferromagnetic hysteresis is detectable at RT inside the ring, while the signal is absent or at least weakly ferromagnetic [38] (slightly tilted blue line) at a film location close to the ring. (c) Dependence of the MOKE response on the pulse energy for different number of pulses: 1,  $10^2$ , and  $10^4$ . The arrows indicate the thresholds for ablation.

Figure 4. Simulation results for a 40 nm Fe<sub>60</sub>Al<sub>40</sub> film on a SiO<sub>2</sub>(150 nm)/Si substrate irradiated by a 500 fs laser pulse: (a-c) Time evolution of the surface temperature,  $T_s$ , shown for three values of absorbed fluence,  $F_{\text{abs}}$ , that correspond to complete melting ( $F_{\text{abs}} = 35$  mJ/cm<sup>2</sup>), partial melting ( $F_{\text{abs}} = 32.5$  mJ/cm<sup>2</sup>), and no melting ( $F_{\text{abs}} = 14$  mJ/cm<sup>2</sup>) of the Fe<sub>60</sub>Al<sub>40</sub> film. The surface temperature is normalized to the melting temperature  $T_m = 1660$  K. The colored areas under the curves correspond to solid state of the film surface, and the intensity of the color schematically represents the temperature dependence of atomic mobility. The arrows mark the times of complete solidification of the film. (d) Dependence of the maximum melting depth in the irradiated FeAl film on the absorbed fluence,  $F_{\text{abs}}$ . (e) The cumulative atomic diffusion length,  $\Delta$ , in the solid film during the laser-induced temperature spike. In the melted alloy, the temperature decreases substantially below  $T_m$  (undercooling) before the resolidification of the film, leading to the sharp drops of atomic diffusion length at the melting threshold and after complete melting.

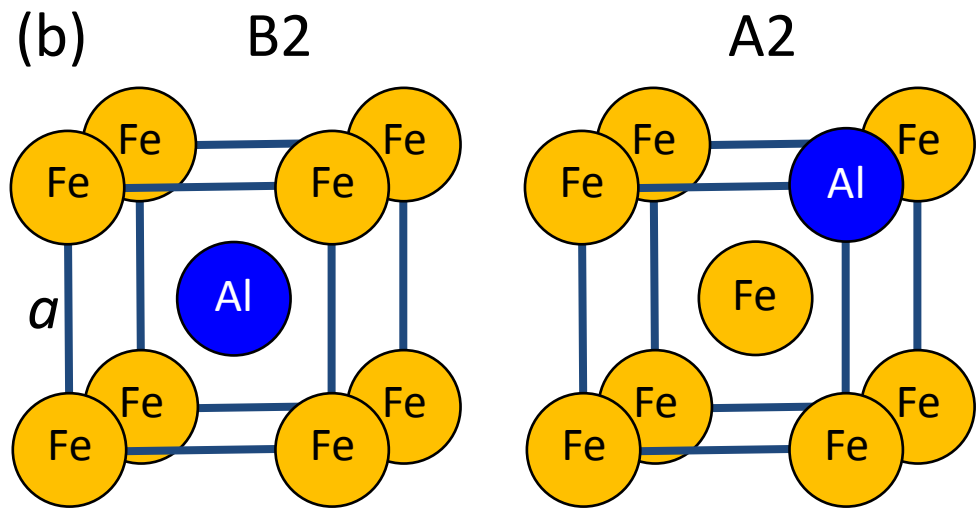
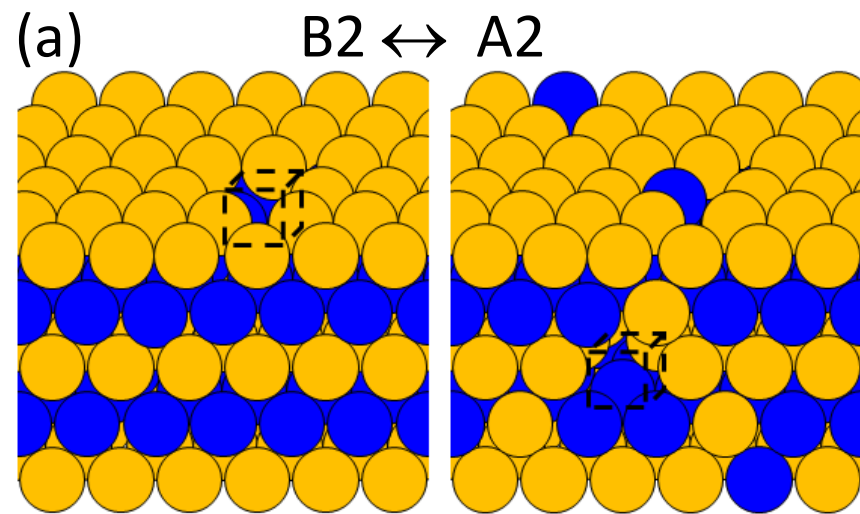


Figure 1 Polushkin et al.

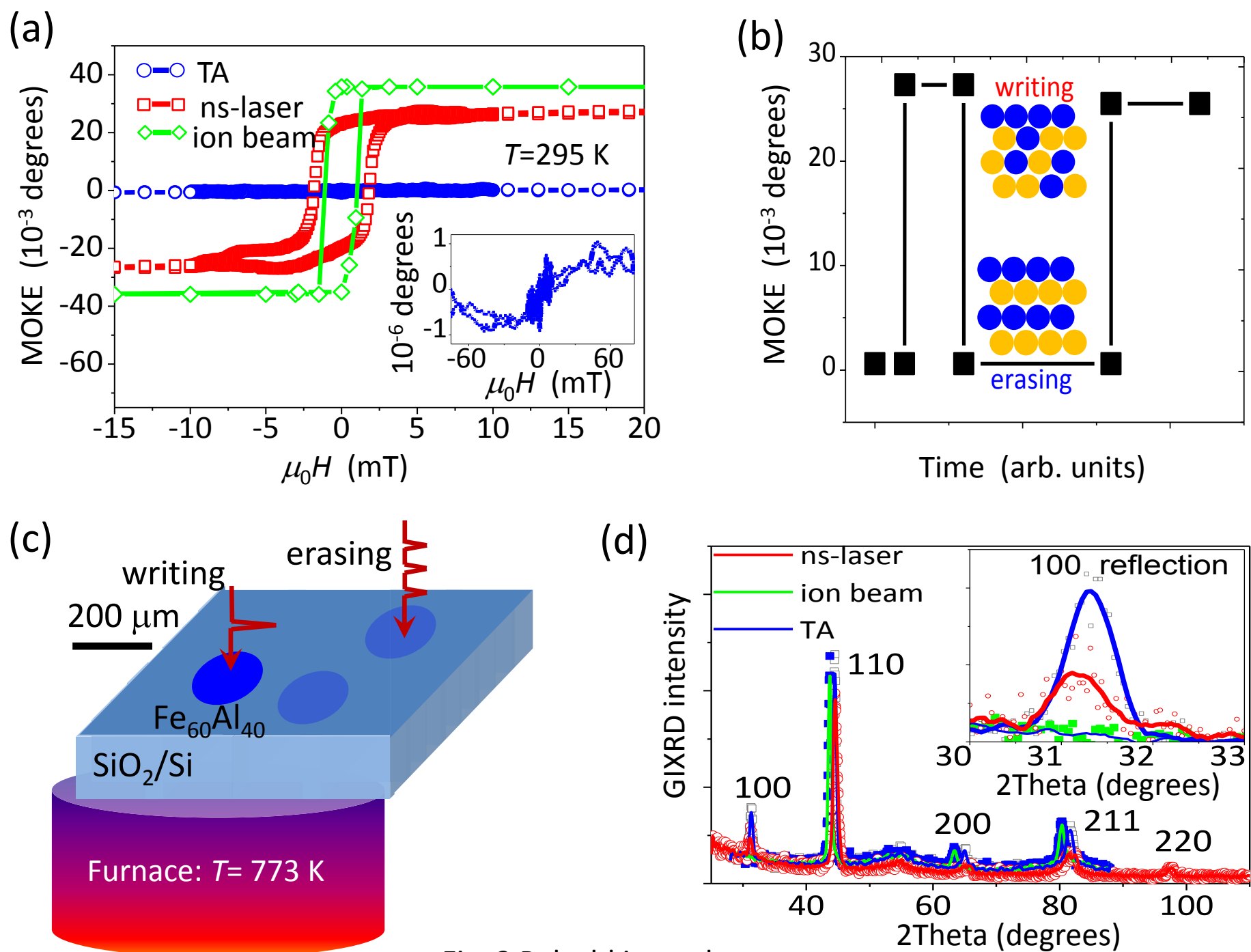


Fig. 2 Polushkin et al.

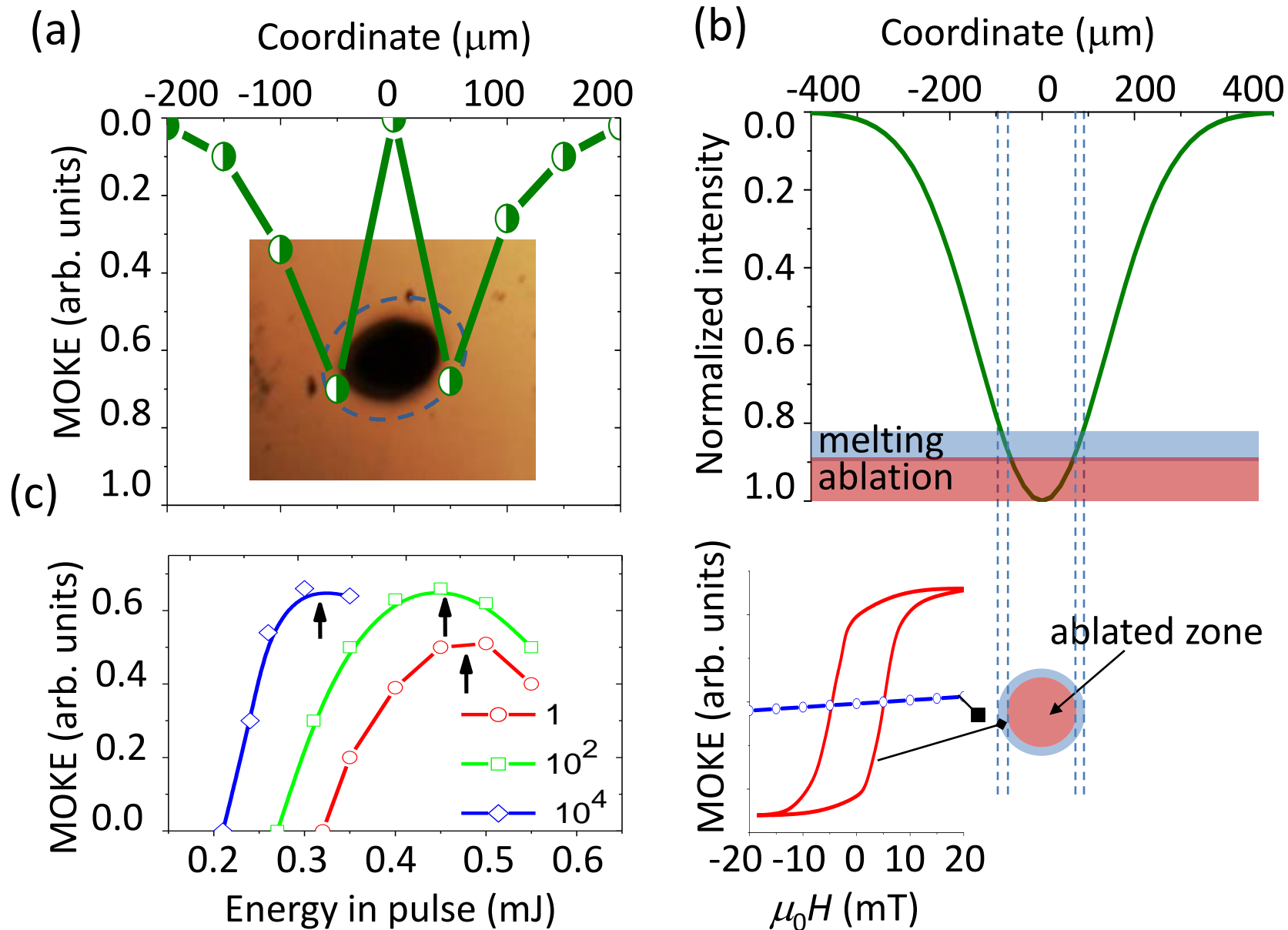


Fig. 3 Polushkin et al.



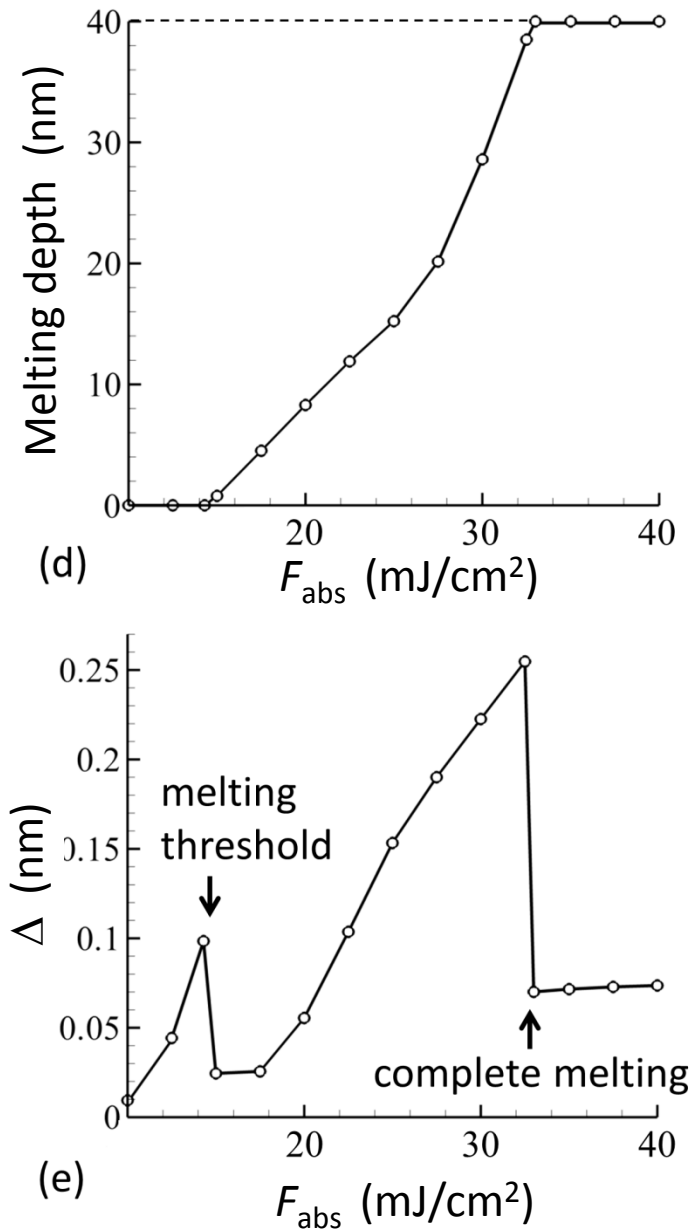
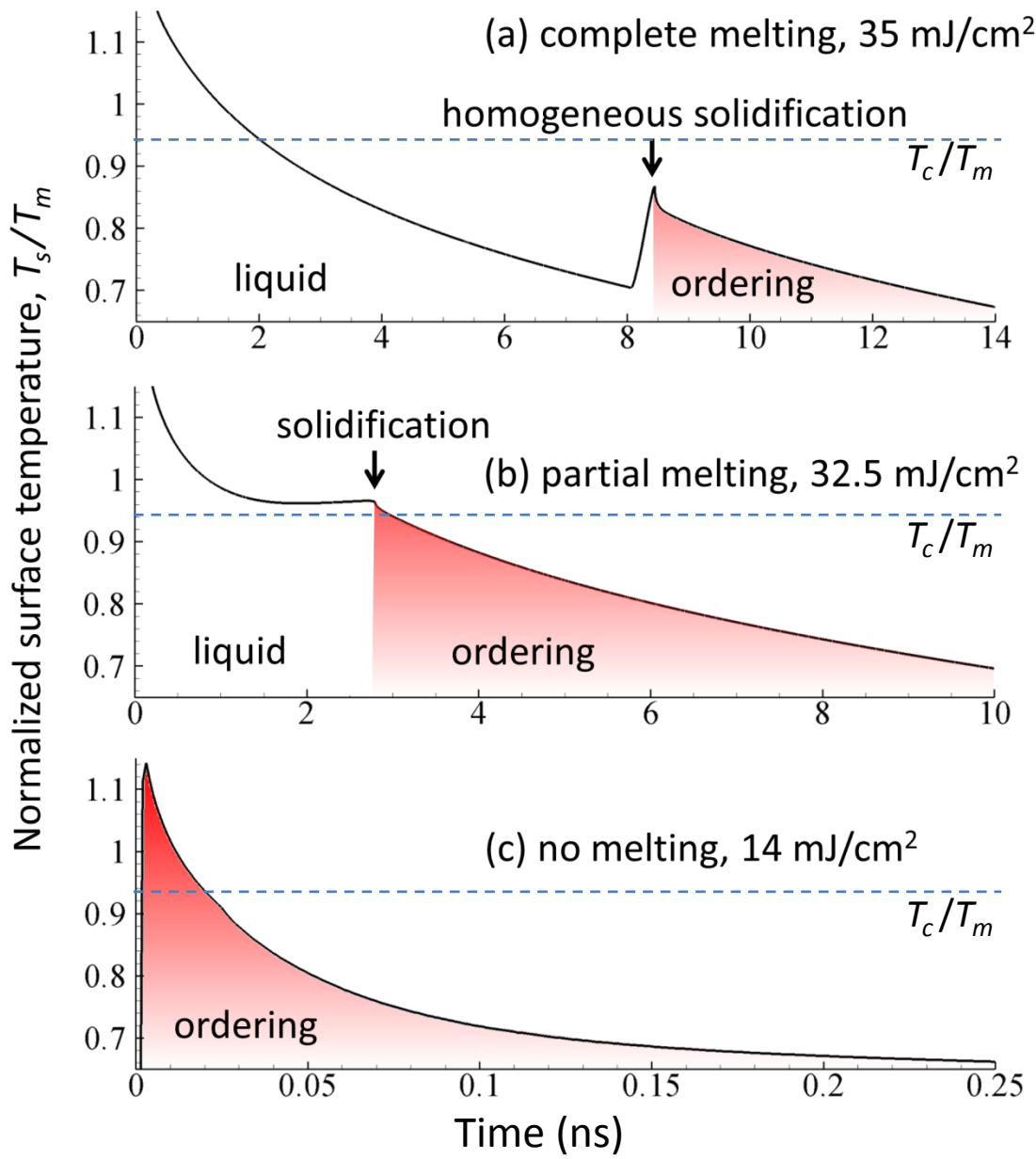


Fig. 4 Polushkin et al.