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Chemical order relaxation in a substitutional solid alloy around the critical temperature

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The relaxation rate for phase transformations depends on how the system is far from its thermodynamic equilibrium. If, for example, temperature in the system approaches to the phase transition point, the relaxation rate becomes close to zero. We theoretically investigate the laser-induced kinetics of chemical order-disorder transformations (CODTs) in substitutional solid alloys at temperatures T satisfying the condition that $|T - T_c| \ll T_c$, where T_c is the CODT critical temperature. In these studies, we use the equation obtained by Metiu, Kitahara, and Ross, that is, $d\eta/dt = -(\Gamma/2k_BT)\partial F/\partial\eta$, where $0 < \eta < 1$ is the chemical order parameter, Γ is the frequency of atomic jumps, k_B is the Boltzmann constant, and F is the free energy of the system, which we combine with the Landau theory for second-order phase transitions. If the two lowest terms are retained only in a Taylor expansion of F, the $\eta(t)$ dependence can be found analytically. Such an approach is assumed to be valid for sufficiently small $\eta < 0.5$. As an example, we simulate CODTs in the thin-film (40-nm thickness) binary alloy of $\mathrm{Fe}_x\mathrm{Al}_{1-x}$ $(x = 0.6)$. Our simulations show that both extensive chemical ordering and disordering in the *solid* alloy are feasible under short-pulse laser irradiation, at least, at a nanosecond time scale. This finding can be useful for improving the properties of functional alloyed materials and for extension of their potential applications.

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I. INTRODUCTION

Since the 1970s, after a study by Ovshinsky [1], materials that exhibit reversible changes in their physical properties such as electrical resistivity [1], optical reflectivity [2,3], or magnitude of magnetization [4,5] due to thermally induced changes in their crystalline structure, so-called phase-change materials, have been attracted a steady interest in view of their applications in memory [2–8] and computing [8–10] devices. For such materials, it is crucial that their relaxation to the thermodynamic equilibrium occurs rapidly enough. A one type of phase-change materials, namely chalcogenide glasses, exhibits fast structural order (crystalline)–disorder (amorphous) transitions with the critical temperature T_m that corresponds to the phase transition between the liquid and solid states [2–7]. It was demonstrated, e.g., Refs. [11–15], that the phase changes result from applying heating pulses with duration in a broad range of time scales, from the nanosecond to subpicosecond regimes. A motivation for these studies is to clarify how the concept of phase-change memory and specific phase-change materials proposed are applicable to data recording and storage [16,17].

As phase-change materials, one can consider alloys and compounds whose properties depend on the chemical order in their atomic lattice [18–20]. To extend their functionality [1–17], it is an important point to address the kinetics of chemical order-disorder transformations (CODTs) [21–31] by emphasizing the CODT relaxation rate [32–35]. It has been found experimentally [32,33] that a single laser pulse of even a subpicosecond duration provides formation of the disordered state in a melted film of the $\mathrm{Fe}_{0.6}\mathrm{Al}_{0.4}$ alloy, which persists in the alloy upon resolidification and cooling down to room temperatures. It is also striking that the disordered state is able to transform into the ordered one with short-pulse laser irradiation of lower intensity [32]. More recently, it was shown by simulations [35], based on the microscopic approach to the CODT kinetics, that the fast relaxation to the disordered state is feasible without melting if the CODT critical temperature (T_c) is significantly lower than the melting point (T_m). As there can be no a need to cycle melting and resolidification of the alloy for its reversible transformations, such phase-change materials can have an enhanced endurance, which would be useful for their applications. In our current CODT studies, we employ a phenomenological macroscopic approach upon the basis of the Metiu, Kitahara, and Ross (MKR) equation for temporal evolution of the chemical order parameter $0 < \eta < 1$ [25,27], which is combined with the Landau theory for second-order phase transitions [21]. From the viewpoint of practical applications [1–17], it is challengeable to consider such alloys in which a change in η mediates the modification of their physical properties. For example, there previously were taken significant efforts [36–53] and a consensus has been reached in understanding the phenomenon of disorder-induced magnetism occurring in intermetallic alloys such as $\mathrm{Fe}_x\mathrm{Al}_{1-x}$ $(x \sim 0.6)$. We show that there is a not so broad range of temperatures T around T_c, within of which the relaxation of η in the $\mathrm{Fe}_{0.6}\mathrm{Al}_{0.4}$ alloy towards both the disorder ($\eta \rightarrow 0$) and, at least, partial order ($\eta \sim 0.5$) is rapid enough, at least, in the nanosecond regime. As the relationship between η and magnetism in $\mathrm{Fe}_x\mathrm{Al}_{1-x}$ alloys was established both theoretically [51] and experimentally [52], experimental observations of fast and reversible transitions between the magnetic states under CODTs would be realistic [32].

II. THE CALCULATION

In a substitutional binary alloy, the relaxation of $\eta = n_1 – n_2$, where $n_{1(2)}$ is the probability for an atom of the component 1 (2) to occupy a site r in the atomic lattice, is governed by the MKR equation [25] (see also discussions in Ref. [27]), that is,

$$\frac{d\eta}{dt} = -\frac{\Gamma}{2k_BT}\frac{\partial F}{\partial \eta}, \tag{1}$$

where Γ is the frequency of atomic jumps, which provide ordering of atoms within unit cells of the atomic lattice, k_B is the Boltzmann constant, and F is the free energy. Within a frame of the Landau theory [21], the increment of $F(\eta = 0, T = T_c)$ can be written in the form of a Taylor series, that is,

$$\begin{aligned}\Delta F &\equiv F(\eta, T) - F(0, T_c)\\ &= \tfrac{1}{2}A(T - T_c)\eta^2 + \tfrac{1}{4}B\eta^4 + \tfrac{1}{6}C\eta^6 \dots,\end{aligned} \tag{2}$$

where the first three nonzero coefficients are as follows:

$$\frac{1}{2}A = \frac{1}{2!}\frac{\partial^3 F}{\partial \eta^2 \partial T}\bigg|_{\eta=0, T=T_c}, \tag{3a}$$

$$\frac{1}{4}B = \frac{1}{4!}\frac{\partial^4 F}{\partial \eta^4}\bigg|_{\eta=0}, \tag{3b}$$

$$\frac{1}{6}C = \frac{1}{6!}\frac{\partial^6 F}{\partial \eta^6}\bigg|_{\eta=0}. \tag{3c}$$

In Eq. (2), the coefficients at η, η^3, η^5, ... have to be equal to zero because ΔF does not change upon a change in the sign of η. To find coefficients at η^2, η^4, η^6, ..., we have used the expression for F obtained in the self-consistent field approximation [22,23]:

$$\begin{aligned}F &= \frac{1}{2}\sum_{r,r'} V(r - r')n(r)n(r') + k_BT\sum_r [n(r)\ln n(r)\\ &\quad + (1 - n(r))\ln(1 - n(r))],\end{aligned} \tag{4}$$

where the first term is the internal energy with $V(r–r')$ being the interatomic potential between atoms located in sites r and r', while the second one is the configurational entropy. The Fourier transform of $V(r–r')$ at $2\pi/a$, where a is the lattice constant, is $–k_BT_c/x(1–x)$ [22,23] to define T_c in Eq. (2). In Eq. (4), the quantity of $n \equiv n_{1(2)}$ relates to η as

$$n = x + \eta(1 - x), \tag{5}$$

where x is the content of the component 1 (2) in the alloy. To find the coefficients in Eq. (2) with the help of Eq. (4), we use that an increment of F is

$$\begin{aligned}\Delta F &\equiv \sum_{r'}\frac{\delta F}{\delta n(r')}\delta n(r') = \sum_r\sum_{r'} V(r - r')n(r)\delta n(r')\\ &\quad + k_BT\sum_{r'}\ln\frac{n(r')}{1 - n(r')}\delta n(r'),\end{aligned} \tag{6}$$

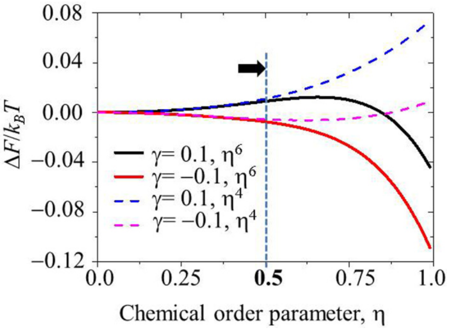

FIG. 1. $\Delta F/k_BT$ versus η at $\gamma = (T–T_c)/T = \pm 0.1$ with the three (solid black and red curves) and two (dashed blue and magenta curves) terms which are left in Taylor expansion in Eq. (2). If $\eta < 0.5$, it is assumed that the two terms can only be left.

where $\delta F/\delta n(r')$ is the variational derivative of F. Thus, one obtains

$$\frac{\partial F}{\partial n} \equiv \frac{\delta F}{\delta n(r')} = \sum_r V(r - r')n(r) + k_BT\ln\frac{n(r')}{1 - n(r')}, \tag{7}$$

$$\frac{\partial^2 F}{\partial n^2} \equiv \frac{\delta^2 F}{\delta n^2(r)} = V(r - r') + \frac{k_BT}{n(r')(1 - n(r'))}, \tag{8}$$

$$\frac{\partial^3 F}{\partial n^3} = k_BT\frac{2n - 1}{n^2(1 - n)^2}, \tag{9}$$

$$\frac{\partial^4 F}{\partial n^4} = 2k_BT\frac{1 - 3n(1 - n)}{n^3(1 - n)^3}, \tag{10}$$

$$\frac{\partial^5 F}{\partial n^5} = 6k_BT\frac{(2n - 1)(2n^2 - 2n + 1)}{n^4(1 - n)^4}, \tag{11}$$

$$\frac{\partial^6 F}{\partial n^6} = 24k_BT\frac{5n^4 - 6n^3 + 2n^2 - 3n + 1}{n^5(1 - n)^5}. \tag{12}$$

As a result, for the first three coefficients in Eq. (2), we have that

$$A = \frac{1 - x}{x}k_B, \tag{13}$$

$$B = \frac{1}{3}(1 - x)k_BT\frac{1 - 3x(1 - x)}{x^3}, \tag{14}$$

$$C = \frac{1}{5}(1 - x)k_BT\frac{5x^4 - 6x^3 + 2x^2 - 3x + 1}{x^5}. \tag{15}$$

Figure 1 shows the free energy change $\Delta F/k_BT$ as a function of η in a substitutional binary alloy with $x = 0.6$. In this plot, the solid and dashed curves are such dependences at $\gamma \equiv (T–T_c)/T = \pm 0.1$ with the three (up to η^6) and two lowest terms (up to η^4), respectively, which are left in Taylor expansion in Eq. (2). Since a conspicuous difference between the solid and dashed curves appear to be at $\eta > 0.5$, as indicated by the arrow in Fig. 1, one can tentatively leave the first two terms only in Taylor expansion, at least, for $\eta < 0.5$. Substituting such a cut Taylor series into the right-hand part of Eq. (1), we obtain the following equation for $\eta(t)$:

$$\frac{d\eta}{dt} + \alpha(t)\eta + \beta(t)\eta^3 = 0, \tag{16}$$

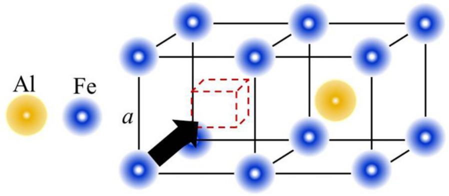

FIG. 2. An elementary diffusion jump into an antistructure site of a bcc lattice, which is occupied by a vacancy. Events of such a kind lead to a stronger chemical disorder in the alloy.

where $a(t)=\Gamma(t)A\gamma(t)/2k_B$ and $\beta(t)=\Gamma(t)B(t)/2k_BT(t)$ are time-dependent coefficients in view of a dependence of T on t. As for $\Gamma(t)$, this quantity can be written as

$$\Gamma(t)=D(t)xc_v(t)Z/a^2,$$

where $c_v(t)$ is the vacancy concentration, $D(t)=D_0\exp(-E_m/k_BT(t))$ is the atomic diffusivity, E_m is the activation energy for atomic migration, and Z is the number of nearest neighbors in the atomic lattice. Figure 2 illustrates an elementary diffusion jump into an antistructure site in a body-centered cubic (bcc) lattice ($Z=8$), which is occupied by a vacancy [54,55]. Events of such a kind lead to a stronger chemical disorder in the alloy. The vacancy concentration obeys the relaxation equation of the Bloch-type [56], that is,

$$\frac{\mathrm{d}c_v}{\mathrm{d}t}=\frac{c_{eq}(T)-c_v}{\tau}, \tag{17}$$

where $c_{eq}(T)=\exp(s_v/k_B)\exp(-E_v/k_BT)$, s_v and E_v are the entropy and energy for vacancy formation, $\tau=L^2/D$, and L is the crystalline grain diameter or film thickness. By physical meaning, the quantity of $c_{eq}(T)$ is the vacancy concentration at the equilibrium; while the quantity of τ is the characteristic time of vacancy life between its formation and annihilation at crystallite or film boundaries. The equilibrium state, i.e., $\mathrm{d}c_v/\mathrm{d}t=0$, is achievable by approaching exponentially $c_v(t)$ to $c_{eq}(T)$ under isothermal conditions for vacancy migration. The solution of Eq. (17) reads

$$c_v(t)=(c_v(0)+M(t))/K(t), \tag{18}$$

where

$$K(t)=\exp\left[(1/\tau_0)\int_0^t \exp(-E_m/k_BT)dt\right],$$

$$M(t)=(1/\tau_0)\int_0^t \chi(t)K(t)dt,$$

$$\tau_0=L^2/D_0,\quad \text{and } \chi(t)=c_{eq}(T)\exp(-E_m/k_BT).$$

After multiplying by η^{-3}, Eq. (16) is convertible into a linear differential equation for a variable $\nu=\eta^{-2}$, so that

$$\frac{\mathrm{d}\nu}{\mathrm{d}t}-\alpha(t)\nu=\beta(t), \tag{19}$$

where 2α and 2β are replaced by α and β, respectively. As the solution of Eq. (19) is retrievable in the analytical form, one obtains finally that

$$\eta(t)=\left[R(t)\left(1/\eta^2(0)+\int_0^t\frac{\beta(t)}{R(t)}\mathrm{d}t\right)\right]^{-1/2}, \tag{20}$$

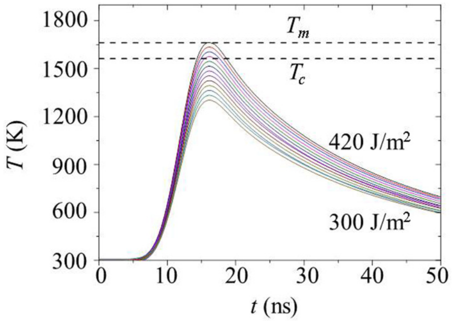

FIG. 3. Simulated $T(t)$ evolution in the 40-nm-thick alloy of $Fe_{0.6}Al_{0.4}$ onto SiO_2(150 nm)/Si under laser irradiation of 5-ns-duration at FWHM with different fluences Φ absorbed by the film within the range of 420–300 J/m^2. The melting point T_m and the CODT critical temperature T_c are indicated by the dashed lines. The transformations are expected to occur at T close enough to T_c, $|T-T_c|\ll T_c$.

where $R(t)=\exp(\int_0^t\alpha(t)\mathrm{d}t)$, so that $\eta(\infty)\rightarrow 0$ at $T=T_c$, which is in accordance to the Landau theory. Equation (20) is of central significance in this study to be valid at sufficiently small $\eta<0.5$ (Fig. 1).

III. RESULTS AND DISCUSSION

A. $T(t)$ evolution

To find out the laser-induced CODT kinetics, first we simulate the temporal evolution of temperature $T(t)$ inside the alloy. Figure 3 shows $T(t)$ curves calculated for different laser fluences Φ within the range of 300–420 J/m^2, which are absorbed by a thin ($L=40$ nm) film of the bcc $Fe_{0.6}Al_{0.4}$ alloy on (150 nm)SiO_2/Si substrate. The film is irradiated by a short laser pulse, whose full width at half maximum (FWHM) is 5.0 ns. The rest of details for the $T(t)$ simulations were given in Ref. [33]. In the context of our current study, it is important that $\Phi_c\approx 385$ J/m^2 heats the film up to the maximal temperature equal to $T_c=1563$ K [57] in $Fe_{0.6}Al_{0.4}$. We expect that a fluence, which is sufficiently high ($\Phi>\Phi_c$) to provide $T>T_c$ but still not sufficient for film melting occurring at $T>T_m=1662$ K [57], is able to provide extensive disordering ($\eta\rightarrow 0$). Another question is whether is it possible to recover the ordered state by short-pulse laser irradiation of lower intensity ($\Phi<\Phi_c$)?

B. Nonequilibrium vacancy concentration

As the relaxation process we consider occurs via vacancies in the atomic lattice, the next step is to clarify how many vacancies can be produced in the alloy by its preliminary treatments. Figure 4(a) shows the equilibrium vacancy concentration c_{eq} in $Fe_{0.6}Al_{0.4}$ as a function of T under isothermal annealing. For this plotting, the factor of s_v/k_B and the energy for vacancy formation in $Fe_{0.6}Al_{04}$ were taken to be equal to 5.7 [58] and $E_v=1.0$ eV [58–60]. As seen, the quantity of c_{eq} can be high enough, up to ≈25 at.%, at $T\rightarrow T_m$. Using Eq. (17), which empirically gives the nonequilibrium vacancy concentration when T depends on t, we find that the

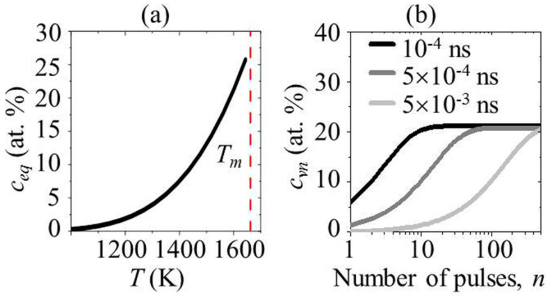

FIG. 4. (a) Equilibrium vacancy concentration c_{eq} versus T in $Fe_{0.6}Al_{0.4}$ up to $T_m = 1662$ K [57]. (b) Vacancy concentration c_{vn} in the thin-film ($L = 40$ nm) $Fe_{0.6}Al_{0.4}$ alloy as a function of number of pulses n with $\Phi = 420\,\mathrm{J/m^2}$ at different τ_0–10^{-4} ns, 5.0×10^{-4} ns, and 5.0×10^{-3} ns.

dependence of this quantity on number of laser pulses (n) is

$$c_{vn} = c_v(0)K^{-n}(\infty) + M(\infty)\frac{1-K^{-n}(\infty)}{K(\infty)-1}.$$

Figure 4(b) shows the $c_{vn}(n)$ dependence in the thin-film ($L = 40$ nm) $Fe_{0.6}Al_{0.4}$ alloy at different values of τ_0–10^{-4} ns, 5.0×10^{-4} ns, and 5.0×10^{-3} ns. Under the assumption that vacancies annihilate at boundaries of the film, we estimate that $\tau_0 = 6.0\times10^{-4}$ ns provided that $D_0 = 2.6\times10^{-3}\,\mathrm{m^2/s}$ [59,60]. Thus, a train of $n \sim 10^2$ 5-ns-duration (FWHM) pulses with $\Phi = 420\,\mathrm{J/m^2}$ provides a value of c_{vn} close to $c_{eq}(T_m) \approx 25$ at.% even at a small initial vacancy concentration, $c_{eq}(0) \sim 0.01$ at.%, which can be obtained by isothermal annealing at $T \sim 800$ K [61].

C. $\eta(t)$ evolution

The central result of our study is illustrated in Figs. 5(a)–5(d) where we show the temporal evolution of η in the alloy at different values of Φ within the range of 300–420 J/m². The plotted $\eta(t)$ curves are calculated at $E_m = 1.7$ eV [58–60], $a = 0.29$ nm, e.g., Ref. [36], and $c_v(0) = 21.0$ at.%. Such a high level of the vacancy concentration can be achieved with a train of laser pulses, as illustrated in Fig. 4(b).

First, our interest was to clarify whether the disordering can be induced at $T > T_c$ but still without melting, i.e., at $T < T_m$. Figure 5(a) shows the $\eta(t)$ evolution simulated at different fluences $\Phi > \Phi_c$, which provide such heating (Fig. 3). As the approach used to obtain the $\eta(t)$ is not valid at $\eta > 0.5$, the $\eta(t)$ curves are shown by the dashed lines at $\eta > 0.5$. We find that a fluence of $\Phi = 420\,\mathrm{J/m^2}$, which induces the film overheating by T–$T_c \approx 100$ K, provides a highly disordered state $\eta(\infty) \to 10^{-3}$ if $\eta(0) = 0.48$. However, such disordering is not achievable even with a slightly lower fluence, e.g., $\Phi = 410\,\mathrm{J/m^2}$. As shown in Fig. 5(b), the minimum of $\eta(t)$ achieved at $\Phi = 420\,\mathrm{J/m^2}$ is essentially deeper (down to $\eta \sim 10^{-7}$) than that achieved at $\Phi = 410\,\mathrm{J/m^2}$. This difference in the depth of the minimum in $\eta(t)$ provides strongly different final states, $\eta(\infty)$, produced by even slightly different fluences [Fig. 5(a)].

Second, it is interesting that a fluence close to Φ_c, within the range of 350–390 J/m², provides the ordering in the alloy from a disordered state $\eta(0) = 10^{-3}$, at least, to a partially ordered state η~0.5, as shown in Fig. 5(c). We find that even a fluence that slightly exceeds Φ_c, e.g., $\Phi = 390\,\mathrm{J/m^2}$, is sufficient for extensive ordering in the nanosecond regime. However, the minimum in $\eta(t)$ achievable with a higher fluence, e.g. $\Phi = 410\,\mathrm{J/m^2}$, is so deep [Fig. 5(d)] that the final state remains highly disordered, $\eta(\infty) \to 0.3\times10^{-2}$. Obviously, the laser irradiation of too low intensity ($\Phi < 330\,\mathrm{J/m^2}$) is not able to provide temperature elevation that would be sufficient ($T < 1300$ K) to induce significant changes in η.

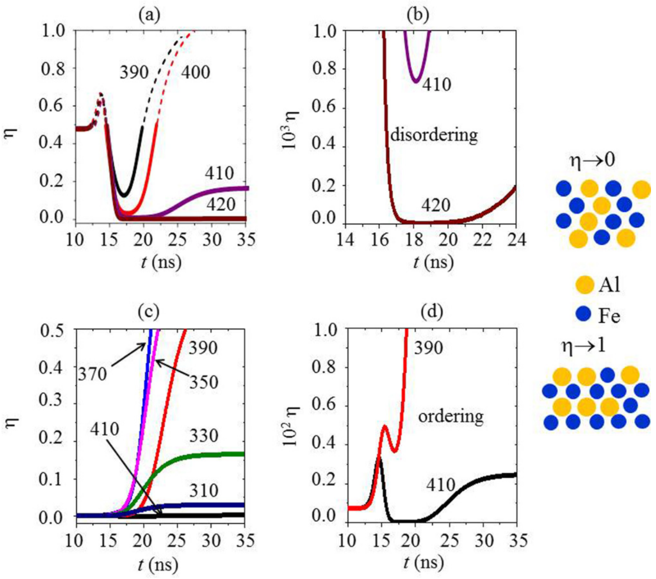

FIG. 5. η versus t in the thin-film ($L = 40$ nm) $Fe_{0.6}Al_{0.4}$: (a) The $\eta(t)$ evolution at different values of Φ, starting from a partially ordered state, $\eta(0) = 0.48$. The final state obtained with a high enough fluence $\Phi = 420\,\mathrm{J/m^2}$ is strongly disordered, $\eta(\infty) \to 10^{-3}$. As the approach used to obtain $\eta(t)$ is not valid at $\eta > 0.5$, the $\eta(t)$ curves are shown by the dashed lines at $\eta > 0.5$. (b) The achieved minima of $\eta(t)$ under disordering at $\Phi = 410\,\mathrm{J/m^2}$ and $\Phi = 420\,\mathrm{J/m^2}$. (c) The $\eta(t)$ evolution at different values of Φ, starting from a highly disordered state, $\eta(0) = 10^{-3}$. Fluences close to Φ_c provide the ordering, at least, to a partially ordered state, $\eta\to0.5$. (d) The achieved minima of $\eta(t)$ under the ordering at $\Phi = 390\,\mathrm{J/m^2}$ and $\Phi = 410\,\mathrm{J/m^2}$. The numbers in the plots are values of Φ given in units of J/m².

D. Relationship between the chemical order and magnetism

In view of our considerations of Fe_xAl_{1-x} alloys as phase-change materials, [32–35] it is important that a change in η mediates the change of Fe magnetic moments [32–53]. Figure 6 shows the expected temporal change in the Fe magnetic moment $\langle\mu\rangle$ averaged on lattice sites and given in units of Bohr magneton (μ_B) under the irradiation of the alloy with a fluence of $\Phi = 380\,\mathrm{J/m^2}$. This dependence was extracted by combining the $\eta(t)$ (Fig. 5) and $\langle\mu\rangle(\eta)$ curves. The latter dependence was taken from Ref. [51] where it had been calculated from first principles. It is seen that the magnetic moment $\langle\mu\rangle \to 2.4\,\mu_B$ at $\eta \to \eta(0) = 0.01$, while $\langle\mu\rangle \to 1.8\,\mu_B$ at $\eta \to 0.5$. Qualitatively, the enhancement of magnetization upon chemical disordering can be explained by strengthening Fe-Fe bonds. The changes in $\langle\mu\rangle$ under

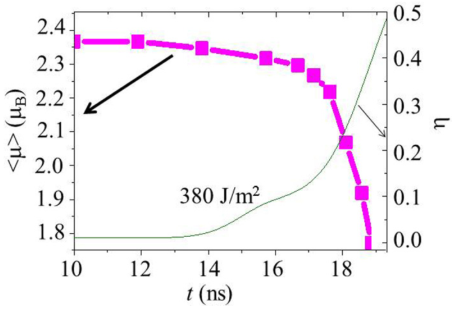

FIG. 6. The expected change of the averaged Fe magnetic moment $\langle\mu\rangle$ in an $Fe_{0.6}Al_{0.4}$ alloy under nanosecond laser irradiation. The dependence has been extracted by combining the $\eta(t)$ (Fig. 5) and $\langle\mu\rangle(\eta)$ [51] curves.

the CODT relaxation were observed experimentally in thin films of Fe_xAl_{1-x} [32–34,47,48,52], and so, CODTs in such specimens are controllable by measuring the magnetization with magnetometry techniques [33].

IV. SUMMARY

We have theoretically studied the temporal evolution of the chemical order parameter η in the thin-film (40-nm thickness) $Fe_{0.6}Al_{0.4}$ alloy under nanosecond laser irradiation (5-ns duration at FWHM). For our simulations, we employed the MKR equation [Eq. (1)] combined with the Landau theory for second-order phase transitions. The theoretical approach we used has allowed us to obtain the analytical expression for the $\eta(t)$ evolution [Eq. (20)], which is assumed to be valid for $\eta < 0.5$. Firstly, we have explored whether this approach predicts chemical disordering in the *solid* alloy at the nanosecond time scale. To check this, the absorbed fluence was chosen to be sufficient for heating the alloy above the CODT critical temperature $T_c = 1563\,\mathrm{K}$ but to be still too low to induce its melting occurring at $T > T_m = 1662\,\mathrm{K}$. It has been found that such a fluence provides the formation of a highly disordered state ($\eta \rightarrow 0$), starting from, at least, a partially ordered state $\eta \sim 0.5$. The disordered state produced persists upon cooling the alloy down to room temperatures. It is also interesting that a lower fluence of the same pulse duration is able to recover the ordered state. Temperatures, at which the CODT relaxation occurs, satisfy the condition that $|T - T_c| \ll T_c$, which is necessary for employing the Landau theory to account for the relaxation rate under CODTs. We note, finally, that the relaxation process can be controlled experimentally by measuring the magnetization of the alloy.

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