Low-temperature ordering in a substitutional alloy with injecting nonequilibrium vacancies: The FePt case

N. I. Polushkin
Institute for Physics of Microstructures of RAS, 603807, Nizhny Novgorod, Russia

Achieving the compositionally ordered state in a substitutional alloy of two or more species can often be even critical for improving its functional properties. To produce a highly ordered alloy, a longtime high-temperature (up to $T \sim 1000$ K) treatment of the alloy is typically necessary because of insufficient vacancy concentration ($c_v$) and their mobility. However, such processing affects the morphology and complicates the technology of functional alloys. We show theoretically that the ordering in the practically important FePt system (FePt$_{1-x}$ with $x$ being close to 0.5) is already achievable at $T \sim 450$ K for reasonable times $t \sim 10^3$ s due to frozen nonequilibrium vacancies. Our simulation is based on the Dienes equation for relaxation of the long-range order parameter ($S$), with taking additionally into account that the ordering kinetics in the alloy is mediated by vacancies. Importantly, the results of such simulation are in good agreement with previous experimental data on the ordering kinetics. We also find that nanosecond laser pulses can be employed to achieve a sufficient level of $c_v \sim 10^{-5}$ for effective low-temperature ordering.

**Keywords:** long-range order parameter, order-disorder transformations, nonequilibrium vacancy, substitutional alloy, laser quenching

I. INTRODUCTION

Typically, a substitutional alloy prepared, for example, in its thin-film form by sputtering onto a cold substrate gets its disordered state, e.g., Refs. [1-8]. If the energy of mixing is negative, i.e., $|V_{ii}| > |V_{ij}|$, where $V_{ii}$, $V_{ij}$, and $V_{jj}$ are the interatomic potentials in a binary i-j alloy, it has a tendency to compositionally order via vacancy migration [9-16] at temperatures $T$ lower than the critical temperature $T_c$ for the order-disorder phase transition [17, 18]. Because of insufficient vacancy concentration, $c_v$, in the atomic lattice one should employ high enough annealing temperatures $T \sim 700-1000$ K $< T_c$, in order to activate the ordering process for long enough times [1-8]. However, a longtime high-temperature process would be undesirable in view of the compatibility for a practical device fabrication.

An example of functional alloys with order – disorder transformations is the Fe$_x$Pt$_{1-x}$ system, where $x \sim 0.5$ (FePt) [1-8, 11, 14, 19-26], which is crystallized in a face-centered cubic (fcc) lattice with the lattice constant of $a=0.381$ nm in the disordered alloy. Obtaining the ordered L1$_0$ state in the FePt lattice is of great practical importance [19-23]. As the melting temperature $T_m \approx 1800$ K is higher than $T_c \approx 1600$ K in FePt [27], the phase transition occurs in the solid state. To optimize the annealing temperature and timescale regimes for the ordering process, both experimental [1-8] and theoretical [25] efforts were previously taken with FePt thin films. For example, it was demonstrated that extensive ordering with formation of the ordered L1$_0$-FePt state occurs at $T \sim 800$ K for a time $t \sim 10^2$ s [1, 5]. The ordering timescale can be shortened even to milliseconds [6, 7] at expense of annealing temperature elevation to $T \sim 1000$ K [7]. To provide the ordering at lower $T$, different technological tricks were employed, e.g., fabrication of multilayers (alternate deposition of Fe and Pt monolayers) [1, 3, 5, 7], stress-promoted L1$_0$ ordering transformations [4], adding a third element into FePt [2, 21, 22].

Here, we show theoretically that the annealing temperature needed for long-range compositional ordering in FePt alloys occurring for reasonable times, $t \sim 10^4$ s, can be lowered even to $T \sim 450$ K. Such effective low-temperature ordering is conceivable under the two conditions as follows: (1) The initial concentration of vacancies, $c_v(0)$, in the alloy should be high enough, while (2) $T$ should be low enough, so that vacancies could not migrate and annihilate at boundaries of the alloy (crystallites). The simulation we have done is based on the Dienes equation for relaxation of the long-range order parameter $S$ [28]. In addition, we redefine the rate constants for order-disorder transformations to be proportional to the quantity of $c_v$ which is time-dependent. To derive $c_v(t)$, we use the linear differential equation of the Bloch type [29]. Qualitatively, it is obvious that the higher $c_v(t) \sim c_v(0)$ (when vacancies are frozen at low $T$ and not able to migrate and annihilate), the larger the ordering rate. However, there is a task of quantifying the effect of $c_v(0)$ on the ordering kinetics. Indeed, even if $c_v(t) \sim c_v(0)$ persists at high enough level, the ordering process can be not activated at too low $T$.

The paper is organized in the following way. First, in the methodological section we describe (A) the
approaches we use to simulate the ordering kinetics as well as the temporal evolution of the vacancy concentration. We also give (B) a description of material parameters, which are important for quantifying the effect of vacancy concentration on the ordering kinetics. Obtained results and their discussion are divided into the following sections: (C) Comparison of the simulation results to experimental data reported earlier by different authors; (D) Evaluation of the effect of initial vacancy concentration, $c_v(0)$, on the ordering rate at low temperatures and (E) the proposal how to increase $c_v(0)$ for achieving effective low-temperature ordering.

II. METHODOLOGY

A. Ordering kinetics

Figure 1 (a) illustrates a scenario for a diffusion act via the vacancy mechanism in the fcc lattice of a substitutional i-j alloy with negative energy of mixing, which increases its order degree. In the beginning, a jump of a “wrong” $i^a$ atom of the component j occurs into its correct position on the $\beta$-sublattice occupied initially by a vacancy (empty box), and then a “wrong” atom $i^b$ of the component i jumps into the released site on the $\alpha$-sublattice. The ordered L10 state can be formed in the FePt lattice due to such ordering events, whose number per unit of time exceeds the number of disordering events if $T<T_c$.

For previous decades there were proposed various approaches to modeling the ordering kinetics, e.g., Refs. [9, 10, 12, 16, 28]. A relatively simple formalism to describe the process has been developed by Dienes [28], who simulated compositional ordering/disordering upon the basis of the chemical balance equation for the change rate of “wrong” atoms, namely

$$\frac{d(W/N)}{dt} = -K_1\left(\frac{W}{N}\right)^2 + K_2\left(x - \frac{W}{N}\right)(1 - x - \frac{W}{N}), \quad (1)$$

where $W/N$ is the fraction of “wrong” atoms in the lattice and $K_{1(2)}$ is the rate constant for the ordering (disordering) process. Eq. (1) does not take explicitly into account the vacancy concentration in the lattice and can be used under the condition that $c_v < x < 0.5$. The equilibrium (thermal) vacancy concentration is given by

$$c_{eq}(T) = \exp\left[\frac{s_v}{k}\right] \times \exp\left[\frac{-E_v}{kT}\right] \quad (2)$$

where $s_v$ and $E_v$ are the vacancy formation entropy and energy, respectively, and $k$ is the Boltzmann constant.

According to Dienes presentation, the rate constants are as follows:

$$K_1 = v\exp\left[-\frac{U}{kT}\right], \quad K_2 = v\exp\left[-\frac{U + V_0S}{kT}\right], \quad (3)$$

where $V_0 = V_j - 1/2(V_{ij} + V_{jj}) = kT/\Gamma$ [17, 18], $\Gamma = \alpha(1 - \alpha)$, $S$ is the long-range order parameter which relates to the number of “wrong” atoms as $W/N = \Gamma(1 - S)$ [28], $v = D_0a^2$ is the vibration frequency, and $D_0$ is the prefactor in the Arrhenius law for atomic diffusion, i.e., $D = D_0\exp(-E/kT)$ with $E$ being the activation energy. The physical meaning of $U$ and $V_0S$ is illustrated in Fig. 1 (b), which schematically presents the barriers for ordering and disordering events at $T<T_c$.

As for the $K_{1(2)}$ coefficients, these quantities represent the atomic/vacancy diffusivity $D_m$. In a substitutional alloy, where diffusion occurs through vacancies, $K_i$ and $K_2$ have to be redefined to linearly depend on $c_v$. As a result, Eq. (1) can be rewritten as follows:

$$\frac{dS}{dt} = \frac{D_m}{a^2}c_v(t)\left[\Gamma(1 - S)^2\exp\left[\frac{V_0S}{kT}\right] - 1\right] - S \quad (4)$$

where $D_m = D_0\exp(-E_m/kT)$ and $E_m = U + V_0S$ is the migration barrier. It is also assumed that $c_v(t)$ is governed by a linear differential equation which describes the vacancy relaxation, namely [29]

$$\frac{dc_v}{dt} = \frac{c_{eq} - c_v}{\tau}, \quad (5)$$

![Fig.1. (a) Possible scenario for an ordering event via migration of a vacancy (empty box) in a substitutional alloy i-j. (b) The energy diagram for compositional order-disorder phase transformations at $T<T_c$.](image-url)
where \( \tau = L^2/D_0 \) is the characteristic time for vacancy annihilation at a boundary of the alloy or crystallite, and \( L \) is the linear size of the sample (film thickness) or crystallite diameter. The solution of Eq. (5) is

\[
c_v(t) = \frac{1}{M(t)}[c_v(0) + (1/\tau_0)\int_0^t M(t)\eta(t)dt], \tag{6}
\]

where \( M(t) = \exp[(1/\tau_0)\int_0^t \exp[-E_m/kT(t)]dt] \), \( \tau_0 = L^2/D_0 \), and \( \eta(t) = c_{eq}[T(t)]\exp[-E_v/kT(t)] \). In the isothermal regime, the solution (6) simplifies to be

\[
c_v(t) = c_v(0)e^{-\tau_0} + c_{eq}(1 - e^{-\tau_0}). \tag{7}
\]

Thus, the solution of Eq. (5) reflects the fact that the factor of \( c_v(0) \) contributes to \( c_v(t) \) only if \( \tau_0 \ll \tau \) for migration and annihilation at boundaries of the alloy or crystallites. It is also obvious that \( c_v(t) \rightarrow c_{eq} \) in the other limit \( (\tau_0 \gg \tau) \).

**B. Determination of material parameters**

To quantify the effect of \( c_v(t) \), there is a necessity to determine the material parameters in Eqs. (1-7), i.e., \( D_0, E_m, E_v \), and \( c_{eq}(T) \). Figure 2 (a-b) shows the experimental \( D(T) \) dependence in FePt bulk, which was obtained in Ref. [26]. It is important to note that diffusion processes were studied by authors of Ref. [30] at such high temperatures \( T \gg T_c \) that the behavior of the component concentration was obeyed the Fick’s law under the approximation of ideal solution. Thus, it was possible to measure atomic diffusivities \( D \) with the help of the Boltzmann-Matano method [30] which is typically used to investigate diffusion processes in metallurgical systems.

The \( D(T) \) dependence shown in Fig. 2 (a) enables us to determine the prefactor \( D_0 \) in the Arrhenius law as well as the diffusion activation energy \( E = E_m + E_v \) from the following relation:

\[
\ln D = \ln D_0 - (E/k)T^{-1}, \tag{8}
\]

which is illustrated in Fig. 2 (b). As a result, one gets that \( E = 2.5 \pm 0.2 \) eV and \( D_0 = (2.5 \pm 0.5) \times 10^{-6} \) m\(^2\)/s. The quantity of \( E_v \) can be taken from the simulations of this parameter in Ref. [14], where it was shown that \( E_v \) depends on \( T \) over a broad temperature range (600–1800 K) and can be approximated as

\[
E_v = 0.000295 \times T (\text{eV K}^{-1}) + 1.245. \tag{9}
\]

Then, the quantity of \( E_m \) can be found by subtraction of \( E \) from \( E_v \) which, in turn, can be determined from Eq. (8). Finally, by taking into account Eq. (9) for \( E_v \) and that \( s/Jk = 2 \) [15], one can find \( c_{eq}(T) \) in FePt, which is plotted in Fig. 2 (c).

**III. RESULTS AND DISCUSSION**

**C. Comparison to the experiments**

Firstly, we compared the results of our simulations with experimental data reported earlier on ordering kinetics in FePt films. Figure 3 (a) shows the S-versus-\( t \) (number of pulses) dependence (circles) retrieved in Ref. [7] by detecting the increase X-ray diffraction from the formed L1\(_0\) superstructure. Although the signal from the superstructure was observed after irradiation only, the extrapolation of the experimental curve into the time moment \( t = 0 \) indicates the partial ordering \( S(0) = 0.47 \) in an as-prepared Fe/Pt multilayer. This conclusion is consistent with other studies, where the formation of the partially ordered structure with \( S \) up to \( S = 0.5 \) was observed in course of the multilayer film growth at elevated temperature of the substrate, e.g., \( T = 160^\circ \text{C} \) [3].

It is interesting to check if the experimental data \( S-t \) presented in Fig. 3 (a) can be compatible to the results of solving numerically the Dienes equation [28] with taking into account the factor of \( c_v(t) \) what we do in the paper. In the same plot we show theoretical dependences \( S\)-versus-\( t \) derived at \( T = 700 \) °C and different values of \( E = 2.7 \) eV, 2.56 eV and 2.45 eV, whereas the rest of parameters were retained to be fixed: \( c_v(0) = c_{eq}(T=700^\circ \text{C}), S(0) = 0.47, D_0 = 2.5 \times 10^{-6} \) m\(^2\)/s, \( L = 10 \) nm, and \( E_v = 1.53 \) eV. As we see, the best
agreement between the modeling results and experiment occurs at $E=2.56$ eV. We note here that the most important parameter is the barrier $E_v=E-E_c$ for vacancy migration, which affects the ordering rate via the exponential factor in the Arrhenius law for the vacancy mobility $D_m$.

![Graph showing $S$-versus-$t$ dependence at $T=700 \, ^\circ\text{C}$ for different $E = 2.56$ eV, 2.70 eV, and 2.45 eV. The simulation results were compared to the experimental $S$-versus-$t$ curve obtained (circles) in Ref. [7].](image1)

![Graph showing theoretical and experimental data for $S$-versus-$t$ at $T=450$ K.](image2)

![Graph showing $S$-versus-$t$ at $T=450$ K, $c_v(0)=10^{-5}$ and $10^{-6}$, $S(0)=0.3$ and 0.47.](image3)

We also compared the modeling results in terms of the temperature dependence of the characteristic timescale $t^*$, on which the ordering ($S\rightarrow1$) occurs, with experimental data obtained in different studies [1, 5, 7] of the ordering kinetics in FePt multilayers. As illustrated in Fig. 3 (b), despite different experimental conditions and technological peculiarities of sample preparation, we find that the experimental data, that is, $t^*\sim0.05$ s at $T=700 \, ^\circ\text{C}$ [7] and $t^*\sim10$ s at $T=550 \, ^\circ\text{C}$ [1, 5], are compatible with the theoretical $t^*$-versus-$T$ dependence derived at $E=2.56$ eV and $S(0)=0.47$. Note here that, at such time and temperature regimes, the ordering rate depends on $c_{eq}(T)$ but not $c_v(0)$.

**D. Low-temperature ordering**

Obviously, nonequilibrium vacancies, that can be injected into the alloy by quenching, should quickly migrate and annihilate at the boundaries of the alloy or crystallites at high $T$, so that the vacancy concentration $c_v$ will tend to the equilibrium $c_{eq}(T)$ if $t\gg\tau=L/D_m$. Formally, such a behavior is reflected in Eq. (5) [29], whose solution describes the temporal evolution of $c_v$. Therefore, if $T$ is high enough, it is hard to expect the effect of the initial vacancy concentration $c_v(0)$ on the ordering kinetics. However, the factor of $c_v$ becomes effective in the other limit, $t<\tau$. The relaxation time $\tau$ of nonequilibrium vacancies (and hence the ordering rate) can be increased by enlarging the sample or crystallites, which is actually not desirable in view of possible practical applications of such alloys [20, 23].

Another possibility for increasing $\tau$ is to lower $T$. By contrast, the usage of low-temperature standard annealing would be useful for improving the functional properties of the ordered alloys [3]. However, the lower $T$, the lower vacancy migration mobility $D_m$ and thus the lower the ordering rate. Therefore, there is a competition between these two opposing factors in Eq. (4) which describes the relaxation of $S$ by taking into account the factor of $c_v$.

Our simulations show that there is a region of low $T$, in which there can be the complete ordering ($S\rightarrow1$) of the initially partially ordered [$S(0)>0.3$] alloy [3] for $t<10^4$ s provided that the initial vacancy concentration is not so small, $c_v(0)\geq10^{-5}$. This is illustrated graphically in Fig. 4, where we show $S$-versus-$t$ dependences simulated (a) at $T=450$ K, $c_v(0)=10^{-5}$ and $10^{-6}$, and $S(0)=0.47$ and 0.3 as well as (b) at $c_v(0)=10^{-5}$ and $T$ varied from 370 K to 430 K. We see that the factor of $c_v(0)$ is critical for effective
low-temperature ordering. For example, at \(c_v(0)=10^{-6}\) and \(T=450\) K the ordering does not practically occur for \(t<10^4\) s (Fig. 4, a). However, an increase of \(c_v(0)\) to \(10^{-5}\) at the same \(T\) provides \(S\to1\) for \(t\approx10^2\) s. As seen from plotting \(c_v\) as a function of \(T\) (Fig. 2, c), the level of \(c_v(0)=10^{-5}\) in FePt is achievable at \(T=1500\) K. Such temperatures are still lower than \(T_c=1600\) K at which the alloy becomes completely disordered \((S_v=0)\) in the equilibrium state.

![Graph of order parameter vs. temperature](image)

**Fig. 5.** The order parameter \(S_v\) at the equilibrium \((t\to\infty)\) versus \(T\). The \(S_v\) level sufficient for effective low-temperature ordering (Fig. 4) is indicated.

Nevertheless, there are at least two questions at this point. One of them is how rapidly should the sample be quenched after heating up to the maximum temperature \(T_{\text{max}}\) close to \(T_c\), in order to prevent thermalizing vacancies at a lower level of their concentration, as illustrated in Fig. 2 (c)? Another question is whether can the partial ordering \(S(0)\approx0.3\), which is sufficient for effective low-temperature ordering up to \(S\to1\) (Fig. 4, a), be conserved after quenching? Indeed, quenching from \(T_{\text{max}}\) close to \(T_c\) can reduce \(S(0)\) to \(S_v\), which approaches zero at \(T_{\text{max}}\toT_c\). The \(S_v(T)\) dependence, obtained by numerically solving Eq. (4), is shown in Fig. 5.

### E. Injection of nonequilibrium vacancies

Both of the above issues can be solved by treating preliminarily the alloy with short-pulse laser irradiation. Figure 6 (a) shows the simulated temporal evolution of the temperature averaged across a film of FePt with thickness of \(L=10\) nm on a glass substrate irradiated with a Gaussian laser pulse of duration of \(\sigma=25\) ns. The \(T(t)\) dependence was calculated according to the formula as follows [31]:

\[
T(t) = T(0) + I \int_0^t \frac{q(u)g(t-u)du}{\delta/L/(pc)}
\]

where \(I=F(1-\exp(-\delta L)/\delta/L/(pc))=409\) K/s, \(F=1.5\times10^{17}\) W/m\(^3\) is the power density absorbed by a sample, \(L=10\) nm, \(\delta=0.05\) nm\(^{-1}\) is the coefficient of light attenuation in a sample, which depends on the light wavelength, \(\rho=1500\) kg/m\(^3\) is the density of an FePt film [8], \(c_v=200\) J/kg/K is the specific heat capacity of an FePt film [8], \(q(t)=\exp(-4ln2(t-t_0)^2/\sigma^2)\) is the temporal evolution of pulse intensity, \(g(t)=\exp(\gamma t)Er\cd(\gamma T^{1/2})\), \(\gamma=(pc)\lambda_0^{1/2}/L/(pc)\) is the factor that reflects the substrate effect on film heating, \(\rho_c=2300\) kg/m\(^3\) is the substrate density, \(c_v=840\) J/kg/K is the specific heat capacity of the substrate, \(\lambda_c=0.8\) W/m/K is the heat conductivity of the substrate (glass). The heating regime for an FePt sample on a glass substrate, illustrated in Fig. 6 (a), can be provided with a pulsed nanosecond laser with fluence of ~100 mJ/cm\(^2\).

![Graph of temperature vs. time](image)

**Fig. 6.** (a) Temporal profile of \(T\) in an FePt film provided by a laser pulse of the Gaussian shape of duration of \(\sigma=25\) ns. (b) \(c_v\)-versus-\(t\) during the pulse of \(\sigma=25\) ns duration, which heats the film up to \(T_{\text{max}}=T_c\). The inset shows the \(S\)-versus-\(t\) evolution at \(S(0)=0.47\) during the pulse.

Having simulated the \(T(t)\) profile (Fig. 6, a), one may derive \(c_v(t)\) under nonisothermal conditions by using Eq. (6). Figure 6 (b) shows changes in \(c_v\) during the pulse that heats the film up to \(T_c=1600\) K. We find that it produces the vacancy concentration \(c_v\approx1.1\times10^5\) which would be sufficient for effective low-temperature ordering (Fig. 4). Importantly, there is no a decrease in \(S\) during such short-pulse irradiation which heats the film even up to \(T_{\text{max}}=T_c\), as shown in inset of Fig. 6 (b).

### IV. CONCLUSIONS

Using the example of a practically important Fe-Pt alloy, it is theoretically shown that, due to
nonequilibrium vacancies artificially injected into the alloy, compositional ordering in this alloy becomes achievable at temperatures of ~450 K, which are significantly lower than the temperatures usually used in the technology of such functional alloys. The ordering kinetics was simulated on the basis of the chemical balance equation for order-disorder transformations proposed by Dienes [28]. Being rather phenomenological and not sensitive to neither the type of atomic lattice nor the kind of phase transformation (spinodal ordering [24] or nucleus growth [25]), the Dienes approach is relatively simple and universal to be potentially attractive for analyzing obtained experimental data. However, in its original form this approach does not take into account such a critical parameter as the vacancy concentration \( c_v \) in the alloy. To treat the effect of \( c_v \), we have redefined the coefficients \( K_1 \) and \( K_2 \) in the Dienes equation [Eq. (1)], which characterize rates of the transformations. An experimental verification of the predictions reported here on low-temperature ordering after injecting nonequilibrium vacancies would be helpful for producing ordered alloys with good functional properties.

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