Nonequilibrium fluctuations, effective temperature, and effective interactions driven by irradiation of alloys

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We study alloy systems subjected to sustained particle irradiation, where short-ranged thermally assisted diffusion competes with the finite-ranged random atomic exchanges forced by irradiation. For the case of binary alloys undergoing phase separation, we introduce a kinetic continuum description of the evolution of the composition field under irradiation, with a self-consistent treatment of the composition fluctuations. We derive an analytical formula for the structure factor at steady state. In the limiting case of short-ranged ballistic jumps, this formula indicates that the effective temperature criterion originally derived by G. Martin [Phys. Rev. B **30**, 1424 (1984)] applies not only to the driving force but to the fluctuations as well. In the case of finite-ranged ballistic jumps, however, the formula indicates that the concept of an effective temperature breaks down, but that one can make use of a more general approach involving effective atomic interactions. In particular, under appropriate irradiation conditions, finite-ranged ballistic jumps lead to finite-ranged effective interactions, which translate into the dynamical stabilization of finite-scale composition patterns.

I. INTRODUCTION

The sustained irradiation of materials by energetic particles leads to continuous production of disorder.[1] This disorder can be structural, e.g., in the form of point defects and dislocation loops, or chemical, e.g., in the form of forced atomic mixing. The continuous production of such disorder, which may also result in nonzero net fluxes of defects and chemical species, tends to drive the irradiated material into nonequilibrium states.[2,3] At finite irradiation temperatures, however, thermally activated dynamics, driven by thermodynamic forces, promotes annealing of this nonequilibrium disorder. An important practical question is to determine the evolution of the irradiated material under the influence of these competing dynamics, since changes in the microstructure often translate into significant modifications of the properties of use. This question is also of fundamental interest since it pertains to the more general theme of the dynamical stability of dissipative systems.[4,5] We note that alloys under irradiation offer an ideal test bed for theoretical developments in this field. Indeed, the relevant atomistic mechanisms are well identified and their frequency and length scales can be varied experimentally in a controlled manner, thus allowing for direct confrontation of theoretical predictions with experiments.

Dynamical systems often reach some steady state or quasi-steady-state, as is the case for alloys under particle irradiation, and one appealing approach is to develop an effective thermodynamics framework for addressing the stability of these steady states. By analogy with equilibrium thermodynamics, such a framework should lead to the construction of nonequilibrium potentials with the following properties: the steady states that can be stabilized for a given set of irradiation conditions correspond to the extrema of these potentials, and, when several locally stable extrema coexist, the relative heights of these extrema determine the respective stabilities of the corresponding steady states. This second property is particularly demanding, since it requires that the description takes into account the fluctuations produced by the various dynamics, to predict correctly the transition rates from one steady state to another.[6]

In the simple case of an alloy fully characterized by its composition field, Martin introduced two decades ago a continuum mean-field kinetic model with two dynamics: the first one is thermally activated and assisted by point-defect migration, while the second one accounts for the forced atomic relocations resulting from nuclear collisions.[7] In the case of alloys that display two-phase fields at equilibrium, at temperatures and compositions within such two-phase fields, the two dynamics are competing since thermally activated diffusion promotes phase separation, while the forced, ballistic mixing promotes homogenization of the composition field. When the forced mixing is short range, in particular when nearly all atoms are relocated to first-nearest-neighbor sites, the forced mixing is equivalent to a random diffusion, with a ballistic diffusion coefficient proportional to the ballistic jump frequency. Within this model, Martin derived a rule of corresponding states: the steady state reached under irradiation at a temperature T corresponds to the equilibrium state of the alloy at a higher effective temperature $T_{eff} = T(1+\Delta)$, where $\Delta = D_b / \tilde{D}$ is the ratio of the ballistic diffusion coefficient to the chemical diffusion coefficient.

The forced mixing produced by energetic heavy ions, however, produces atomic relocations extending beyond first-nearest-neighbor sites, and molecular dynamics (MD) simulations indicate that the distribution of relocation distances can be well approximated by an exponential decay.[8] We have shown analytically[9] that this finite relocation range introduces a nonlocal term in the evolution equation of the composition field. As a result, when the decay length R exceeds a critical value R_c, and under appropriate irradiation temperature and flux, an irradiated alloy spontaneously

forms stable compositional patterns, with a maximum scale of $4\pi R$. This analytical prediction, which is supported by our atomistic kinetic Monte Carlo (KMC) simulations,[8,10] and by our preliminary experimental results on Ag-Cu multilayers irradiated with 1 MeV Kr$^+$ ions,[11] is not predicted by the effective temperature criterion. Indeed, within the mean-field approximation used to derive this criterion, a microstructure with compositional patterns of finite scale should be seen as a macroscopically homogeneous state with strong and long-lived composition fluctuations. Although fluctuations are not explicitly considered in Martin's model, if one assumes that the effective temperature possesses the properties of a thermodynamic temperature, the composition fluctuations observed under irradiation must be identical to those found at equilibrium, at the corresponding effective temperature. At equilibrium, however, for the class of alloys considered here, i.e., with short-range atomic interactions leading to phase separation, there are no composition fluctuations that would be equivalent to the compositional patterns stabilized by finite-range ballistic jumps. The main objective of this article is to address this limitation by constructing an analytical model that includes a self-consistent treatment of fluctuations. In particular, it would help us to understand whether, within Martin's approximation of short-range ballistic jumps, the effective temperature criterion applies to fluctuations as well, and how fluctuations are rescaled in the more complex case of a finite ballistic range.

A possible approach to include fluctuations in a kinetic model is to start from a probabilistic description. The most common one is the so-called master equation (ME), which can be approximated by a Fokker-Planck equation, or, even better, by Kubo's ansatz, in order to take properly into account the extensive nature of the effective potential that governs the steady-state probability distribution.[12] This approach has been successfully implemented for spatially homogeneous nonequilibrium systems,[6,12–15] leading to the analytical construction of nonequilibrium effective potentials, from which one can construct dynamical phase diagrams. The application to inhomogeneous nonequilibrium systems, however, has remained elusive because of the large dimensionality of the corresponding phase space and of the lack of detailed balance.[16] Nevertheless, Vaks and Kamyshenko have shown that mean-field approximations to their lattice model can be obtained in such cases.[17] These authors have derived an effective Krivoglaz-Clapp-Moss (KCM) formula,[18,19] which allowed them to generalize Martin's effective temperature rule. These results will be further discussed, and compared to our present results in Sec. III.

An alternative approach to incorporate fluctuations is to start from a deterministic description and transform it into a Langevin-type description by adding a noise term.[20] This noise term can be constructed by making use of a fluctuation-dissipation relationship, or by using the ME to derive kinetic equations for the relevant moments of the probability distribution. In the case of an alloy simply described by its composition field, as discussed in this paper, Cook[21] using a Langevin equation and Langer et al.[22] starting from the ME derived an identical expression for the thermal fluctuation term that has to be added to the classical deterministic Cahn-Hilliard diffusion equation. This addi-

tional term allows studying the effect of fluctuations on the early stages of spinodal decomposition.

In this article, we extend Cook's approach to the case of driven alloys with competing dynamics. We show that, for short-range ballistic jumps, Martin's effective temperature criterion applies both to the effective driving force and to the fluctuations, whereas in the presence of finite-range ballistic jumps, the concept of an effective temperature breaks down and should be replaced by the more generic concept of effective atomic interactions.

II. CONTINUUM MODELING

A. Deterministic model with competing dynamics

We recall the one-dimensional kinetic model that we introduced[9] to study the effect of the forced atomic relocation range on the stability of the composition field in a binary alloy $A_{1-\bar{c}}B_{\bar{c}}$ under irradiation. The temporal evolution of the local deviation from the nominal composition, $\psi(x) = c(x) - \bar{c}$, is comprised of two terms:

$$\frac{\partial \psi}{\partial t} = M\nabla^2\left(\frac{\delta\Omega F}{\delta\psi}\right) - \Gamma_b(\psi - \langle\psi\rangle_R). \tag{1}$$

The first term corresponds to the thermal dynamics for the conserved order parameter ψ. M is the thermal atomic mobility and is related to the chemical diffusion coefficient through $M = c(1-c)\tilde{D}/k_B T$, Ω is the atomic volume, and F is the free energy of the alloy, which we express using the Cahn-Hilliard form:

$$F = \frac{1}{\Omega}\int [f(\psi) + C|\nabla\psi|^2]dV, \tag{2}$$

where f is the free energy density of a homogeneous alloy and C the gradient energy term.

The second term in Eq. (1) expresses the forced mixing induced by ballistic jumps, which occur at a frequency Γ_b, with relocation distances distributed according to a normalized weight function w_R. We showed that the ballistic rate of change of the order parameter is proportional to the difference between the local composition and nonlocal average, denoted by angular brackets and defined as

$$\langle\psi\rangle_R = \int w_R(x - x')\psi(x')dx'. \tag{3}$$

B. Langevin equation with thermal dynamics only

We briefly recall the procedure introduced by Cook.[21] In the absence of particle irradiation, the deterministic Eq. (1) is transformed into a Langevin equation by adding a random noise term $\xi_{th}(x, t)$, with suitably defined statistical properties (to be defined below)

$$\left(\frac{\partial\psi}{\partial t}\right)_{th} = M\nabla^2\left(\frac{\delta\Omega F}{\delta\psi}\right) + \nabla^2\xi_{th}. \tag{4}$$

For small fluctuations around $c(x) = \bar{c}$, i.e., $\psi(x) = 0$ everywhere, we can linearize Eq. (4). Now transforming to Fourier space, denoted by the carets, we obtain

$$\left(\frac{d\hat{\psi}}{dt}\right)_{th} = -\alpha_{th}(k)\hat{\psi} - k^2\hat{\xi}_{th}. \tag{5}$$

The amplification factor is given by $\alpha_{th}(k)=Mk^2[f''(\bar{c}) +2Ck^2]$, where f'' is the second derivative of the homogeneous free-energy density with respect to composition. Within a regular solution model, $f''=u''+k_BT/[\bar{c}(1-\bar{c})]$, where u is the internal energy term in f. The noise term is assumed to be uncorrelated in time, and has a spatial structure given by the quantity $Q_{th}(k)$:

$$\langle\hat{\xi}_{th}(k,t)\hat{\xi}_{th}^*(k,t')\rangle = Q_{th}(k)\delta(t-t'). \tag{6}$$

The statistical quantity $Q_{th}(k)$ can be obtained by calculating the equilibrium structure factor $S_{eq}(k)=\langle|\hat{\psi}(k,t\to\infty)|^2\rangle$. Following a standard procedure to solve Eq. (5), Cook obtains

$$S_{eq}(k) = \frac{k^4 Q_{th}(k)}{2\alpha_{th}(k)}. \tag{7}$$

This structure factor can also be evaluated through a fluctuation-dissipation relationship:[18,23]

$$S_{eq}(k) = \frac{k_BT}{f''(\bar{c}) + 2Ck^2}. \tag{8}$$

Therefore the functional dependence of $Q_{th}(k)$ that reproduces properly the structure factor is

$$Q_{th}(k) = \frac{2k_BTM}{k^2}. \tag{9}$$

C. Langevin equation with ballistic dynamics only

Following a similar strategy, we can write an equation including fluctuations for the irradiation-induced mixing dynamics:

$$\left(\frac{\partial\psi}{\partial t}\right)_b = -\Gamma_b\left(\psi - \int w_R(x-x')\psi(x')dx'\right) + \nabla^2\xi_b, \tag{10}$$

where the subscript b stands for ballistic. This is already a linear equation, so we can transform it into an ordinary differential equation in Fourier space:

$$\left(\frac{d\hat{\psi}}{dt}\right)_b = -\alpha_b(k)\hat{\psi} - k^2\hat{\xi}_b, \tag{11}$$

where the ballistic amplification factor is given by

$$\alpha_b(k) = \Gamma_b[1 - \hat{w}_R(k)]. \tag{12}$$

Now the steady-state structure factor $S_b(k)$ that we expect from the ballistic dynamics alone is the one of a random solid solution, i.e., $S_b(k)=\bar{c}(1-\bar{c})$. Applying Cook's solution to this case, we find that the spatial correlations of the ballistic fluctuations must be given by

$$Q_b(k) = \frac{2\bar{c}(1-\bar{c})\alpha_b}{k^4}. \tag{13}$$

D. Langevin equation with two dynamics in parallel

For the model with competing dynamics with fluctuations, we add the terms describing the deterministic evolution of the composition, as in Eq. (1), as well as the noise terms. The α terms are additive since we assume the two deterministic dynamics to be acting in parallel and independently. Within this assumption, the corresponding fluctuations are uncorrelated and therefore additive as well:

$$\frac{d\hat{\psi}}{dt} = -[\alpha_{th}(k) + \alpha_b(k)]\hat{\psi} - k^2(\hat{\xi}_{th} + \hat{\xi}_b). \tag{14}$$

Equation (14) yields the following expression for the steady-state structure factor:

$$S(k) = k^4\frac{Q_{th}(k) + Q_b(k)}{2[\alpha_{th}(k) + \alpha_b(k)]}. \tag{15}$$

After simple algebraic manipulations we obtain

$$S(k) = \frac{k_BT\left(1 + \dfrac{\alpha_b}{\tilde{D}k^2}\right)}{2Ck^2 + u''(\bar{c}) + \dfrac{k_BT}{\bar{c}(1-\bar{c})}\left(1 + \dfrac{\alpha_b}{\tilde{D}k^2}\right)}. \tag{16}$$

Equation (16) is the central result of Sec. II. We now consider specifically two cases for the ballistic dynamics.

E. Case 1: Short-range ballistic jumps

In this case, the dynamics can be approximated by a diffusional process with a diffusion coefficient D_b,[7] and thus $\alpha_b(k)=D_bk^2$. The equation for the structure factor now becomes

$$S_1(k) = \frac{k_BT\left(1 + \dfrac{D_b}{\tilde{D}}\right)}{2Ck^2 + u''(\bar{c}) + \dfrac{k_BT}{\bar{c}(1-\bar{c})}\left(1 + \dfrac{D_b}{\tilde{D}}\right)}. \tag{17}$$

This equation admits a straightforward interpretation in terms of Martin's effective temperature criterion: the effective temperature $T_{eff}=T(1+D_b/\tilde{D})$ describes *both* the effective driving force *and* the effective fluctuations of the alloy under irradiation at steady state. We should also notice that in Martin's derivation of the effective temperature criterion, the ratio $D_b/\tilde{D}$ is assumed to be composition independent. It thus implies that the fluctuations are also composition independent. Therefore, for short-range ballistic jumps, Martin's nonequilibrium potential has the two desired properties of a nonequilibrium potential discussed in the second paragraph of the Introduction, and the common tangent rule, as used in Ref. 7, is a valid construction to determine the globally stable steady states.

F. Case 2: Finite-range ballistic jumps

Following the MD results,[8] we choose w_R to be an exponential decay with decay length R. Its Fourier transform is

then given by $\hat{w}_R(k) = 1/(1+k^2R^2)$, and thus $\alpha_b(k) = \Gamma_b k^2 R^2/(1+k^2R^2)$.[24] The equation for the structure factor now becomes

$$S_2(k) = \frac{k_B T \left(1 + \dfrac{R^2\Gamma_b}{\tilde{D}}\dfrac{1}{1+k^2R^2}\right)}{2Ck^2 + u''(\bar{c}) + \dfrac{k_B T}{\bar{c}(1-\bar{c})}\left(1 + \dfrac{R^2\Gamma_b}{\tilde{D}}\dfrac{1}{1+k^2R^2}\right)}.$$

(18)

Again, one notes that the same rescaling applies to the temperature in the numerator, which corresponds to the amplitude of the fluctuations, and in the denominator, which corresponds to the second derivative of the nonequilibrium potential. The concept of effective temperature, however, is no longer appropriate since the effective temperature would be a function of the wave vector k. A better approach is to divide both the numerator and the denominator in Eq. (18) by the rescaling factor of the temperature. Before doing so, we note that C and u'' in Eq. (18) are both related to the physical interactions between A and B species. This point becomes very clear if we use a Bragg-Williams approximation with a pairwise interaction energy between nearest neighbors, v, so that the internal energy term becomes $u(\bar{c}) = \bar{c}(1-\bar{c})v/2$. Note that $v < 0$ for alloy systems with a tendency toward phase separation. Simple algebra shows that in Eq. (18) $u'' = \hat{v}(k=0)$ and $2C = \hat{v}''(k=0)/2$.[23] Since $\hat{v}(k)$ is an even function of k by symmetry, the first two terms in the denominator in Eq. (18) correspond to the first two terms of the expansion of the interaction energy around $k=0$. We can therefore reinterpret Eq. (18) as the structure factor, evaluated at the physical temperature T, of an alloy system with effective pairwise interactions defined as

$$\hat{v}_{eff}(k) = \frac{\hat{v}(k)}{1 + \dfrac{R^2\Gamma_b}{\tilde{D}}\dfrac{1}{1+k^2R^2}}.$$

(19)

A similar equation was originally derived by Vaks and Kamyshenko [see Eq. (15) in Ref. 17], albeit from a discrete description. The implications of such equations for effective interactions are discussed in the next section.

III. DISCUSSION

We first discuss the case of short-range ballistic dynamics. The present approach and the effective KCM formula derived by Vaks and Kamyshenko[17] provide self-consistent results establishing the existence of an effective temperature for both the driving force and the fluctuations under irradiation. It is interesting to note, however, that, if one assumes the existence of an effective temperature, its expression can be very simply deduced by making use of Einstein's relation. This relation links the atomic mobility to the chemical diffusion coefficient through $k_B T = c(1-c)\tilde{D}/M$. As we have assumed that the ballistic and thermal dynamics are uncorrelated, the total diffusion coefficient under irradiation is the sum of the thermal and ballistic diffusion coefficients.[25]

Similarly, the thermal and ballistic mobilities are additive. The ballistic dynamics, however, has a zero mobility since it corresponds to an "infinite-temperature" dynamics[7] with a finite ballistic diffusion coefficient. Now, if an effective temperature exists, it must obey Einstein's relation, and it should therefore satisfy $k_B T_{eff} = c(1-c)(\tilde{D}+D_b)/M$. After eliminating the mobility through the use of the equilibrium Einstein relation, we find that $T_{eff} = T(1+D_b/\tilde{D})$, i.e., we recover Martin's effective temperature.

The concept of an effective temperature is identical to a uniform rescaling of the physical interactions that exist between chemical species, $\hat{v}_{eff}(k) = \hat{v}(k)/(1+D_b/\tilde{D})$, as seen from taking the limit $R \to 0$ in Eq. (19). Effective interactions can be directly measured in kinetic Monte Carlo simulations by inverting the steady-state probability of selected configurations:[26] for the case of short-range ballistic, the effective interactions indeed follow the rescaling predicted from the effective temperature.[27]

It is interesting to note that the concept of an effective temperature has recently received much attention in the context of jammed granular systems,[28] sheared fluids,[29] and glassy systems.[30] Ono *et al.*[29] consider a jammed foam at zero temperature driven by shearing. They demonstrate that five independent definitions of an effective temperature, relying on linear response theory and generalized fluctuation-dissipation (GFD) relationships, yield consistent values and consistent dependence on the shear rate. Similarly, Berthier and Barrat[28] showed that an effective temperature can be used to account for several GFD relationships for a sheared two-component fluid at finite temperature.

One remarkable feature of the effective temperature derived by Martin is that it becomes infinite in the limit $T \to 0$. This is simply a consequence of the randomness of the mixing forced by irradiation. Recently, however, Lund and Schuh have reported the stabilization of nonrandom structures in Lennard-Jones binary alloys forced at 0 K by irradiation or shearing:[31] such a behavior appears to be specific to systems that do not exist on a lattice, fluids or amorphous solids, or possibly to alloy systems on a lattice but with very large size mismatch. In such systems, even though attempts at ballistic mixing are randomly chosen, the net effect may not be equivalent to a randomizing force, in the sense that large local stresses may prevent a ballistic switch of atoms from yielding a locally stable configuration in the potential energy landscape. As a result, athermal relaxation can undo the forced mixing attempted by the ballistic switch of two neighboring atoms. More work is needed to elucidate this point fully. We note, however, that our results are not in contradiction with Lund and Schuh's results, since the present model is restricted to an alloy on a lattice in the absence of stress effects.

We now turn to the case of finite-range ballistic mixing. As seen from Eq. (18), the concept of an effective temperature loses its relevance since T_{eff} would be a function of the wave vector k. A better approach is to use the concept of effective interactions:[16,17] the alloy reaches under irradiation a steady state that is identical to the equilibrium state reached at temperature T in a modified alloy, where atoms interact through effective interactions. Starting from a discrete micro-

scopic master equation, and using a mean-field approximation of its steady-state solution, Vaks and Kamyshenko derived an analytical expression for effective pair interactions. Their result [see Eq. (15) in Ref. 17], is in fact the discrete counterpart of the continuum expression derived here in Eq. (19). As discussed and exemplified in a previous paper,[26] effective interactions characteristic of a steady state can be systematically evaluated numerically through appropriate KMC simulations. One remarkable feature of Eq. (19) is that the range of the effective interactions introduced by the ballistic mixing scales with the range of that ballistic dynamics. It is also interesting to compare Eq. (19) to the analytical expression obtained if one ignores the fluctuations, i.e., using Eq. (1) instead of Eq. (14): the effective interactions introduced by the ballistic dynamics then appear as additive corrections to the physical interactions [see Eqs. (6) and (7) in Ref. 9 or Eq. (3) in Ref. 10]. This additivity of the effective interactions is however problematic since, as discussed in Ref. 10, it gives rise to inconsistencies, in particular in the limits $\Gamma_b \to 0$ and $R \to 0$. These inconsistencies are completely absent from Eq. (19), and it is therefore concluded that a correct derivation of effective interactions require a model that includes a self-consistent treatment of the fluctuations, as is done in this article.

The derivation of a nonequilibrium steady-state structure factor points to an experimental route to explore the phenomenology we study in this work. Small-angle scattering experiments can be used to measure structure factors of thin films subjected to ion-beam mixing, in particular by taking advantage of the high brightness of synchrotron x-ray sources. This article provides a theoretical background that can be confronted with experimental results. Equation (18) suggests that under heavy-ion irradiation we must expect the structure factor to evidence finite-range correlations in the form of a peak at a finite wave vector k, as opposed to the Lorentzian tail of the classic KCM formula [Eq. (17)]. Another approach for testing experimentally the present theoretical results is to take advantage of the recent advances in three-dimensional atom probe microscopy.[32–35] Three-dimensional atomic maps can be constructed with a subnanometer resolution. The presence of patterning could therefore be directly assessed from these maps. Furthermore, structure factors can be calculated as Fourier transforms of atomic maps,[10] thus allowing for a quantitative comparison with the present predictions. If confirmed by experiments, the current modeling suggests that one can take advantage of the spontaneous formation of nanoscale compositional patterns during ion-beam processing to synthesize nanocomposites with characteristic length scales that can be tuned by varying the irradiation parameters, so as to optimize their optical, magnetic, or electric properties.

As a final comment, we note that the present approach to derive effective temperature and effective interactions should be applicable to alloy systems with an ordering tendency as well, for instance by extending the kinetic model introduced by Cook *et al.* to describe ordering kinetics.[36] The study of this different class of alloy systems would be important to assess the general validity of the concepts of effective temperature and effective interactions, in particular since the analytical results obtained by Vaks and Beiden[37] for this class of alloys suggest that irradiation may stabilize chemically ordered but spatially frustrated states. These peculiar states could correspond to the patterns identified in KMC simulations, where both decomposition and chemical ordering take place at a finite scale.[38]

IV. CONCLUSION

We have introduced a kinetic model for the evolution of the composition field in an irradiated alloy, where short-ranged thermally assisted diffusion competes with the finite-ranged random atomic exchanges forced by irradiation. For the case of binary alloys undergoing phase separation, we have extended Cook's approach[21] to build a self-consistent expression for the nonequilibrium fluctuations of composition in such alloys. This model made it possible to derive an analytical expression for the steady-state structure factor. This expression is the continuum counterpart of the discrete one derived by Vaks and Kamyshenko.[17] In the limiting case of short-ranged ballistic jumps, this structure factor yields a simple interpretation: both the driving force and the fluctuations under irradiation follow their equilibrium expression, albeit evaluated at a higher, effective temperature. This is a direct generalization of the effective temperature criterion originally derived by Martin[7] for the driving force. In the more general case of finite-ranged ballistic jumps, the concept of an effective temperature is no longer correct, since this effective temperature would be a function of the wave vector of the composition modulations. It is nevertheless possible to interpret the structure factor equation as that of an equilibrium system at the true temperature T, but with effective, rescaled atomic interactions. These effective interactions are finite ranged, and they offer a direct explanation for the dynamical stabilization of compositional patterns under irradiation predicted analytically[9] and observed in atomistic KMC simulations.[8,10]

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