

**Self-diffusion and solute diffusion in alloys under irradiation: Influence of ballistic jumps**

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We have studied the influence of ballistic jumps on thermal and total diffusion of solvent and solute atoms in dilute fcc alloys under irradiation. For the diffusion components that result from vacancy migration, we introduce generalized five-frequency models, and show that ballistic jumps produce decorrelation effects that have a moderate impact on self-diffusion but that can enhance or suppress solute diffusion by several orders of magnitude. These could lead to new irradiation-induced transformations, especially in the case of subthreshold irradiation conditions. We also show that the mutual influence of thermal and ballistic jumps results in a nonadditivity of partial diffusion coefficients: the total diffusion coefficient under irradiation may be less than the sum of the thermal and ballistic diffusion coefficients. These predictions are confirmed by kinetic Monte Carlo simulations. Finally, it is shown that the method introduced here can be extended to take into account the effect of ballistic jumps on the diffusion of dumbbell interstitials in dilute alloys.

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**I. INTRODUCTION**

It is well documented that irradiation by energetic particles can modify the kinetic evolution of an alloy.<sup>1,2</sup> The physical origins of these modifications are known for metallic alloys. On one hand, nuclear collisions between energetic particles and atoms of the target lead to a forced atomic mixing. For recoil energies higher than a displacement threshold energy (typically 25 eV in metals) but less than a few hundreds of eV, this forced mixing takes place in linear replacement collision sequences, while for higher recoil energies (typically above 1 keV in metals) most of this mixing takes place in displacement cascades.<sup>3</sup> For low to moderate recoil energies, this forced mixing can be well modeled by a ballistic, i.e., random, diffusion process.<sup>4</sup> On the other hand, nuclear collisions initiated by recoils above the threshold energy lead to the formation of stable Frenkel pairs. This results in a supersaturation of point defects, and hence in an acceleration of thermal diffusion. This supersaturation can reach several orders of magnitude, particularly at low temperature as rates for point defect recombination and annihilation at sinks are suppressed.<sup>5</sup> Note that for recoil energies below the displacement threshold energy, while no new point defect can be created, existing point defects can be forced to jump from one lattice site to a neighboring one. Such subthreshold events can also contribute to noticeable evolutions of alloy microstructures under mild irradiation conditions.<sup>6-8</sup> Again, this forced migration can be approximated by a ballistic process.

In many situations, ballistic mixing and thermally activated diffusion compete since they tend to drive the alloy microstructure in different directions. Indeed, on one hand ballistic mixing tends to randomize atomic configurations, e.g., to promote the dissolution of precipitates and the disordering of chemically ordered phases, while thermally activated diffusion, on the other hand, is driven by chemical potential gradient and often results in the formation of precipitates and ordered phases. Irradiation parameters such as the irradiation temperature, the flux, the mass, and the energy

of the projectile, as well as material parameters determine whether thermal or ballistic processes are predominant. In the latter case, irradiation-induced phase transformations may be observed, while in the former case irradiation-enhanced transformations may take place. These transformations often lead to significant changes in the properties of the irradiated material. Both from a practical and a theoretical viewpoint, it is therefore important to develop the modeling of these alloys driven by competing dynamics.

To study quantitatively the competition between thermal and ballistic diffusion, Martin introduced a kinetic model with two dynamics in parallel.<sup>4</sup> In this model the composition field of an alloy under irradiation evolves in time due to thermal diffusion, enhanced by point defect supersaturation, and due to ballistic diffusion, which results from nuclear collisions. This model has been successfully used and developed to rationalize order-disorder transformations<sup>9</sup> and dissolution-precipitation reactions<sup>10</sup> or to analyze ion-beam mixing experiments.<sup>11</sup> This approach was also recently extended to include the finite range of atom relocations in energetic displacement cascade.<sup>12</sup> Another extension of Martin's approach has been developed to study dynamical phase transitions in alloys subjected to intense shearing.<sup>13</sup> In that context, the tools initially derived by Martin to address phase stability in alloys under irradiation have been shown to be useful to rationalize phase transformations forced by ball milling.<sup>14-16</sup> More generally, atomic systems with competing dynamics have been widely used as prototypes to study the general behavior of nonequilibrium dynamical systems.<sup>17-19</sup>

The presence of several independent dynamics can easily be incorporated in kinetic modeling at the microscopic level: the configurational transition rates appearing in a microscopic master equation are written as the sum of the rates due to each dynamics.<sup>2</sup> While this approach lends itself very well to atomistic kinetic Monte Carlo simulations, no general analytical results can be obtained at this microscopic level to predict the dynamical stability of microstates.<sup>18</sup> Approximate solutions, however, can be derived through continuum modeling, as for instance shown by Martin.<sup>4</sup> An important ingre-

dient of such continuum modeling is how the coexistence of several dynamics is taken into account. In fact, while the coupling between chemical and point defect fluxes in alloys under irradiation has received a lot of attention (see Ref. 2 and references therein), no specific attention has been paid to the possible effect of ballistic jumps on thermal diffusion. To illustrate this point, let us concentrate on how diffusion in a dilute alloy would be written following Martin's model. For the sake of simplicity, we concentrate here on the part of point defect-assisted diffusion that results from vacancy migration, but we will indicate how the proposed approach can be extended to diffusion by dumbbell interstitials. As thermally activated and ballistic migration events are assumed to take place in parallel, the approach currently in use is to write the total tracer diffusion coefficient in an irradiated alloy as

$$D_X^{\text{irr}} = a^2 f^{\text{th}} c_v^{\text{irr}} \omega_v^{\text{th}} + D_X^{\text{bal}}, \quad (1)$$

where  $X$  is the tracer atom,  $a$  the lattice parameter of the crystal, which is assumed to possess cubic symmetry here,  $f^{\text{th}}$  a thermal correlation factor,  $c_v^{\text{irr}}$  the vacancy concentration in the presence of irradiation,  $\omega_v^{\text{th}}$  the  $X$  atom-vacancy exchange frequency, and  $D_X^{\text{bal}}$  is the ballistic diffusion coefficient. As we will later discuss irradiation situations where only subthreshold collision events take place, let us notice that, by analogy with Eq. (1), the total diffusion coefficient in that case would read

$$D_X^{\text{irr,sub}} = a^2 f^{\text{th}} c_v \omega_v^{\text{th}} + D_X^{\text{bal,sub}}, \quad (2)$$

where  $c_v$  is the vacancy concentration at thermal equilibrium, and  $D^{\text{bal,sub}}$  is the diffusion coefficient corresponding to the atomic jumps forced by subthreshold collisions.

The central point addressed in this paper is to question the validity of Eqs. (1),(2). For the rare cases where each dynamics results in a random walk of tracer atoms, e.g., for interstitial atoms migrating via a direct interstitial mechanism, it is correct to add the thermal and ballistic diffusion coefficients determined *in the absence* of the second dynamics, as done in Eqs. (1),(2). This procedure is also correct in the limiting case where both dynamics proceed by the same mechanism and lead to identical correlation factors. In nearly all practical situations, however, thermal and ballistic dynamics proceed by different mechanisms [as in Eq. (1)], or result in correlated walks with different correlation factors. This is particularly true when thermally activated diffusion of solvent and solute atoms proceeds by vacancy migration, as illustrated later in this work. In all these important practical cases, we show here that *the presence* of a second dynamics can significantly alter correlations during tracer atom migration by a first dynamics, leading to suppression or enhancement of the corresponding diffusion coefficients by several orders of magnitude. For these situations, a second point that we address is to question whether the total diffusion coefficient can even be written as the sum of the thermal and the ballistic diffusion coefficients. Indeed, as the migration path of a given atom is comprised of alternating thermal and ballistic segments, it is not *a priori* guaranteed that the total mean-square displacement of this atom is the sum of the

mean-square displacements due to thermal and ballistic events taken separately. In this paper, for the sake of simplicity, we study these two points when thermal diffusion proceeds by vacancy migration, and we restrict our study to pure and dilute fcc alloys, for which elaborate diffusion models are available.<sup>20,21</sup>

The paper is organized as follows. First in Sec. II, we introduce generalized five-frequency models and propose expressions for thermal, ballistic, and total diffusion coefficients in a dilute fcc binary alloy in the presence of thermally activated and ballistic dynamics. Then, in Sec. III we compare some of these diffusion coefficients with kinetic Monte Carlo results. A few model alloys are selected to illustrate how thermal diffusion is modified by the presence of ballistic jumps. Finally, we discuss some practical consequences for the decomposition kinetics of an alloy during subthreshold irradiation, and we indicate how to extend the present approach, derived for vacancy migration, to the case of dumbbell interstitial migration.

## II. MODEL

The classical theory of atomic diffusion provides us with analytical expressions for self-diffusion and solute diffusion coefficients in an alloy at thermal equilibrium.<sup>20,21</sup> After briefly recalling these expressions for an fcc dilute  $A_{1-c}B_c$  alloy, we show how they can be extended to the case of an alloy where both thermal and ballistic jumps take place.

*Thermal diffusion.* In addition to  $A$  solvent atom-vacancy exchange frequency  $\omega_0$ , there are four exchange frequencies to be considered:<sup>20,21</sup> the frequency of  $B$  solute-vacancy exchanges  $\omega_2$ , the frequency of  $A$  atom-vacancy exchanges that do not dissociate the solute-vacancy pair  $\omega_1$ , the frequency of an  $A$  atom-vacancy exchange that dissociates the solute-vacancy pair  $\omega_3$ , and the frequency of an  $A$  atom-vacancy exchange that associates the solute and the vacancy  $\omega_4$ . In this model, the thermal diffusion coefficient of an isolated solute atom  $B$  is calculated to be

$$D_B^{\text{th}} = D_B^{\text{th}}(\omega_0, \omega_1, \omega_2, \omega_3, \omega_4) = a^2 \omega_2 c_v \frac{\omega_4}{\omega_3} f_2, \quad (3)$$

where  $c_v \omega_4 / \omega_3$  is the vacancy concentration at a neighboring site, and  $f_2$  is the correlation factor between successive solute-vacancy exchanges:

$$f_2 = \frac{\omega_1 + \omega_3 3.5F(\omega_4/\omega_0)}{\omega_1 + \omega_2 + \omega_3 3.5F(\omega_4/\omega_0)}, \quad (4)$$

where

$$F(x) = 1 - \frac{1}{7} \frac{10x^4 + 180.5x^3 + 927x^2 + 1341}{2x^4 + 40.2x^3 + 254x^2 + 597x + 435} \quad (5)$$

for  $x = \omega_4 / \omega_0$ .

Formula (3) gives also the thermal self-diffusion coefficient of an  $A$  tracer atom when setting all  $\omega$ 's equal to  $\omega_0$ :

$$D_A^{\text{th}} = D_B^{\text{th}}(\omega_0, \omega_0, \omega_0, \omega_0, \omega_0) = a^2 \omega_0 c_v f_0, \quad (6)$$

where the geometric correlation factor  $f_0 = [1 + 3.5F(1)] / [2 + 3.5F(1)] = 0.7815$ .

*Ballistic diffusion above displacement threshold.* Following Martin's idea,<sup>4</sup> the forced atomic jumps produced by recoil energies above the displacement threshold energy, but below recoil energies producing displacement cascades, are modeled by performing isolated random exchanges between solvent atom, solute atom, and vacancy.<sup>2</sup> At these moderate recoil energies, it is correct to assume that such exchanges take place between nearest neighbor sites only. The frequency of these jumps can be simply calculated by multiplying the displacement rate per atom by the number of replacements produced by one displacement.<sup>4</sup> Then, defining  $2\omega_b$  as the ballistic atom exchange frequency per pair, the ballistic diffusion coefficient of any tracer element  $X$  ( $X = A$  or  $B$ ) reads

$$D_X^{\text{bal}} = a^2 \omega_b. \quad (7)$$

*Ballistic diffusion below displacement threshold.* An extension of the previous procedure is proposed for the forced atomic jumps produced by subthreshold collisions. In that case only atoms that are nearest neighbors of a vacancy can jump. Furthermore, since the energy required to initiate such a jump is of the order of 1 to a few eV, we will here assume that the frequency of such an event is independent of the local atomic configuration. This simplification is justified for alloys with moderate heats of mixing because, in that case, the configuration-dependent part of site energies are well below 1 eV. Defining  $\omega_b^{\text{sub}}$  as the vacancy ballistic jump frequency, the subthreshold ballistic diffusion coefficient of any tracer element  $X$  ( $X = A$  or  $B$ ) now reads

$$D_X^{\text{bal,sub}} = a^2 \omega_b^{\text{sub}} c_v f_0. \quad (8)$$

*Thermal and above threshold ballistic diffusion.* We now want to derive an expression for the total diffusion coefficient of an  $X$  tracer atom,  $D_X^{\text{tot}}$ , when both thermal and ballistic dynamics are active. The direct addition of the thermal coefficients from Eq. (3) or Eq. (6) to the ballistic coefficient given by Eq. (7) does not provide accurate results. Indeed, both the vacancy concentration around the tracer atom ( $c_v \omega_4 / \omega_3$ ) and the correlation factor ( $f_2$ ) are modified by the ballistic jumps performed by the vacancy. To account for these effects, we propose here to write the total diffusion coefficient as a sum of a modified thermal diffusion coefficient and the ballistic diffusion coefficient:

$$D_X^{\text{tot}} = \hat{D}_X^{\text{th}} + D_X^{\text{bal}}. \quad (9)$$

The modified thermal diffusion coefficient,  $\hat{D}_X^{\text{th}}$ , is simply obtained by adding the atom ballistic frequency, i.e.,  $2\omega_b$  to the thermal frequencies  $\omega_0, \omega_1, \omega_3, \omega_4$  in Eq. (3), so as to take into account the effects of the ballistic jumps on the correlations of the tracer atom thermal migration. No ballistic term should be added to  $\omega_2$  since ballistic jumps between the solute and the vacancy are already accounted for in the solute ballistic diffusion coefficient. This thermal diffusion coefficient modified by the presence of ballistic jumps is thus given by

$$\hat{D}_X^{\text{th}} = \frac{c_v^{\text{irr}}}{c_v} D_X^{\text{th}}(\hat{\omega}_0, \hat{\omega}_1, \hat{\omega}_2, \hat{\omega}_3, \hat{\omega}_4),$$

with

$$\begin{aligned} \hat{\omega}_0 &= \omega_0 + 2\omega_b, & \hat{\omega}_1 &= \omega_1 + 2\omega_b, & \hat{\omega}_2 &= \omega_2, \\ \hat{\omega}_3 &= \omega_3 + 2\omega_b, & \hat{\omega}_4 &= \omega_4 + 2\omega_b. \end{aligned} \quad (10)$$

This new formulation can also be used for the self-diffusion coefficient by setting all thermal  $\omega$ 's to  $\omega_0$ . For high ballistic frequency,  $\omega_b$  suppresses both the vacancy segregation around the solute atom  $X$  [ $(\omega_4 + 2\omega_b) / (\omega_3 + 2\omega_b) \rightarrow 1$ ] and the correlation between consecutive solute jumps [ $\hat{f}_2 = f_2(\hat{\omega}_0, \hat{\omega}_1, \hat{\omega}_2, \hat{\omega}_3, \hat{\omega}_4) \rightarrow 1$ ]. These effects are indeed expected due to the random nature of the ballistic jumps. At low ballistic jump frequency we expect the new formula Eq. (10) to provide a rather accurate description of thermal diffusion coefficients since it includes the effect of the ballistic events on the path that the vacancy performs between two consecutive exchanges with the tracer atom. A first limitation of Eq. (10), however, is that it ignores the fact that the solute can migrate through ballistic jumps between two thermal jumps. The second effect that is ignored is that ballistic exchanges between the tracer and a solvent atom provide additional ways for dissociation or reassociation of the tracer-vacancy pair. It is therefore not guaranteed that this new expression will retain its accuracy for intermediate ballistic jump frequency. Note that the influence of the ballistic jumps on thermal diffusion is expected to be much more pronounced for solute diffusion than for self-diffusion. Indeed, in the latter case, the correlation factor will only increase from  $f_0 = 0.7815$  to 1 as the ballistic frequency goes from zero to infinity, whereas the solute correlation factor will increase from  $f_2$ , which can be very small in a trapping situation, to 1. The probability to find the vacancy next to the solute atom, however, may be reduced because of the ballistic jumps. Depending upon the specifics of the alloy considered, the ballistic jumps could therefore enhance or reduce thermally activated diffusion. These effects will be illustrated on a few model alloys in Sec. III. It should be stressed that, for comparison with experimental data, one should evaluate the steady-state vacancy concentration under irradiation, as  $\hat{D}_X^{\text{th}}$  is directly proportional to  $c_v^{\text{irr}}$ . This can be done by using rate equation models.<sup>5</sup>

*Thermal and subthreshold ballistic diffusion.* Here again ballistic jumps will modify both the correlation factors and the probability to find the vacancy next to the tracer atom  $X$ . Following the approach used in the preceding paragraph, an improved expression for the thermal diffusion coefficient in the presence of subthreshold ballistic diffusion is proposed:

$$\hat{D}_X^{\text{th,sub}} = D_X^{\text{th}}(\hat{\omega}_0, \hat{\omega}_1, \hat{\omega}_2, \hat{\omega}_3, \hat{\omega}_4),$$

with

$$\begin{aligned} \hat{\omega}_0 &= \omega_0 + \omega_b^{\text{sub}}, & \hat{\omega}_1 &= \omega_1 + \omega_b^{\text{sub}}, & \hat{\omega}_2 &= \omega_2, \\ \hat{\omega}_3 &= \omega_3 + \omega_b^{\text{sub}}, & \hat{\omega}_4 &= \omega_4 + \omega_b^{\text{sub}}. \end{aligned} \quad (11)$$

For the ballistic diffusion term, since all ballistic jumps proceed by migration of the vacancy, the correlation factor for this migration, as well as the probability to find the vacancy next to a tracer atom, are in fact perturbed by the thermal jumps. In order to propose an expression consistent with Eq. (11), we simply apply the same rules used in the derivation of Eq. (11), except that now the roles of the two dynamics are permuted. This improved expression for the ballistic diffusion coefficient can be rewritten as

$$\hat{D}_X^{\text{bal,sub}} = D_X^{\text{th}}(\hat{\omega}_0, \hat{\omega}_1, \hat{\omega}_2, \hat{\omega}_3, \hat{\omega}_4),$$

with

$$\begin{aligned} \hat{\omega}_0 &= \omega_b^{\text{sub}} + \omega_0, & \hat{\omega}_1 &= \omega_b^{\text{sub}} + \omega_1, & \hat{\omega}_2 &= \omega_b^{\text{sub}}, \\ \hat{\omega}_3 &= \omega_b^{\text{sub}} + \omega_3, & \hat{\omega}_4 &= \omega_b^{\text{sub}} + \omega_4. \end{aligned} \quad (12)$$

For this case of mixed thermal and subthreshold ballistic diffusion, since all jumps take place by an atom-vacancy exchange mechanism, we can also directly use the classical five-frequency formula to write the total diffusion coefficient of a tracer atom  $X$ . All five jump frequencies are simply the sum of a thermal frequency and of the ballistic frequency. This exact expression for the total diffusion coefficient can be rewritten as

$$D_X^{\text{tot,sub}} = D_X^{\text{th}}(\hat{\omega}_0, \hat{\omega}_1, \hat{\omega}_2, \hat{\omega}_3, \hat{\omega}_4),$$

with

$$\begin{aligned} \hat{\omega}_0 &= \omega_0 + \omega_b^{\text{sub}}, & \hat{\omega}_1 &= \omega_1 + \omega_b^{\text{sub}}, & \hat{\omega}_2 &= \omega_2 + \omega_b^{\text{sub}}, \\ \hat{\omega}_3 &= \omega_3 + \omega_b^{\text{sub}}, & \hat{\omega}_4 &= \omega_4 + \omega_b^{\text{sub}}. \end{aligned} \quad (13)$$

Since we now have an exact expression for the total diffusion coefficient and two improved expressions for the thermal and ballistic components, we can test whether the total diffusion coefficient is indeed given by the sum of the partial diffusion coefficients. By inspection of Eqs. (11),(12),(13) it is seen that the equality is in general not fulfilled. The difference comes from the fact that in the expression of the modified correlation factors  $\hat{f}_2$  the ‘‘second’’ jump frequency reads  $(\omega_2 + \omega_b^{\text{sub}})$  in the total diffusion coefficient, whereas it reads  $\omega_2$  and  $\omega_b^{\text{sub}}$  in the thermal and ballistic diffusion coefficients, respectively. As all frequencies are positive quantities, it is therefore predicted that the total ballistic diffusion will be less than or equal to the sum of the thermal and ballistic diffusion measured separately. Note that in the limiting cases where one diffusion mechanism is strongly predominant, i.e., when  $\omega_b^{\text{sub}}$  is much smaller or much larger than  $\omega_2$ , this difference disappears and the total diffusion coefficient is indeed recovered as the sum of the partial diffusion coefficients. It is expected that the deviation from additivity is maximum when the dynamics are operating at the same frequency. In the case of self-diffusion, since all thermal jump frequencies are equal to  $\omega_0$ , the maximum relative deviation can be obtained analytically by setting  $\omega_i = \omega_0 = \omega_b^{\text{sub}}$  in Eqs. (11),(12),(13). Simple algebra yields

$$\frac{\hat{D}_A^{\text{th,sub}} + \hat{D}_A^{\text{bal,sub}} - D_A^{\text{tot,sub}}}{D_A^{\text{tot,sub}}} = \frac{1 - f_0}{1 + f_0} = 0.1226. \quad (14)$$

While this difference of about 12% is small, larger effects are again possible in the case of solute diffusion. The physical origin of this reduction of the total diffusion coefficient is quite clear: when two consecutive jumps of the tracer atom occur by different dynamics in opposite directions the total migration distance of the tracer atom is zero, while each jump taken separately can contribute to the partial migration distances, and therefore to the partial diffusion coefficients. It should be remembered that the predicted deviation from additivity of partial diffusion coefficients relies on the use of two approximated expressions, one for the thermal and the other for the ballistic diffusion coefficients, see Eqs. (11),(12). It is therefore necessary to test these predictions, as well as the other results we derived in this section, by Monte Carlo simulations.

### III. SIMULATION

The Monte Carlo simulations are performed on a rigid face-centered cubic crystal using periodic boundary conditions. Only one vacancy is introduced in the crystal. Other sites are occupied by  $A$  atoms. For the solute diffusion case, one  $A$  atom is replaced by a  $B$  one. A crystal size of 512 sites is enough to calculate the quantities of interest. The vacancy concentration is thus fixed constant at  $1.9 \times 10^{-3}$ . For comparison with the analytical predictions, however, a correction is sometimes necessary since  $c_v$  and  $c_v^{\text{irr}}$  in all previous equations refer to concentrations in the matrix, whereas in the simulations it is the total vacancy concentration that is fixed. We will explicitly discuss this correction when it becomes significant. The simulations are performed following the procedure we used to study phase evolution under irradiation.<sup>12,22</sup> Let us simply recall that activation energies for vacancy-atom exchanges are described in the framework of a kinetic Ising-like model where atom interactions are restricted to pairwise energies between nearest-neighbor atoms. An important consequence of this simplification is that there are only three independent frequencies in this model,  $\omega_0$ ,  $\omega_1$ , and  $\omega_2$ , since  $\omega_3 = \omega_0$  and  $\omega_4 = \omega_1$ . A residence-time algorithm is used to construct a kinetic evolution of the system through vacancy jumps and ballistic exchanges (see Refs. 22,23 and references therein for a discussion on this algorithm). To calculate the tracer diffusion coefficients, we label every  $A$  or  $B$  atom and first compute the time-dependent diffusion coefficient  $D_i(t) = \delta \langle R^2(t) \rangle_i / 6\delta t$ , where  $R$  denotes the displacement of the atom and  $\langle \rangle_i$  stands for an average over the  $A$  tagged atoms (no such average is performed for the solute diffusion since there is only one  $B$  atom in the simulation cell). The tracer diffusion coefficients  $D_A^{\text{tot}}$  or  $D_B^{\text{tot}}$  are then recovered at large time. A tracer trajectory obtained in the simulation, in general, contains segments corresponding to thermal and ballistic jumps. The thermal and ballistic diffusion coefficients are calculated by putting together all thermal segments and all ballistic segments, respectively. The corresponding diffusion coefficients  $D_X^{\text{th,MC}}$ ,

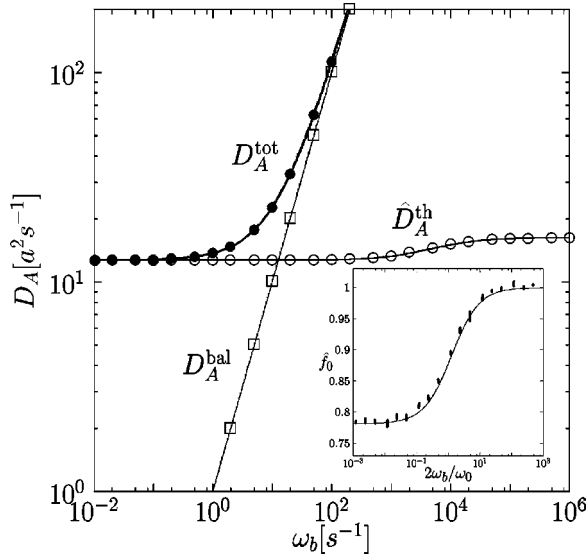


FIG. 1. Total  $D_A^{\text{tot}}$  (●), thermal  $\hat{D}_A^{\text{th}}$  (○), and ballistic  $D_A^{\text{bal}}$  (□) self-diffusion coefficients as a function of the above-threshold ballistic frequency  $\omega_b$  from MC simulations and analytical expressions (lines) [see text]. Data are obtained for  $\omega_0 = 8.33 \times 10^3 \text{ s}^{-1}$  at  $T = 400 \text{ K}$  and  $c_v = 1.9 \times 10^{-3}$ . The inset shows the correlation factor  $\hat{f}_0$  as a function of  $2\omega_b/\omega_0$ .

$D_X^{\text{bal,MC}}$ , and  $D_X^{\text{tot,MC}}$  are then calculated and compared to the analytical expressions proposed in Sec. II.

*Thermal and above threshold ballistic diffusion.* We first focus on self-diffusion under irradiation. The following parameters are used for that purpose. The vacancy migration energy is set to 0.8 eV and the vacancy attempt jump frequency  $\nu$  is set to  $10^{14} \text{ s}^{-1}$ , giving an  $A-V$  exchange frequency equal to  $\omega_0 = 8.33 \times 10^3 \text{ s}^{-1}$  at a temperature  $T = 400 \text{ K}$ . According to our model in Sec. II, the corrected thermal self-diffusion coefficient under irradiation  $\hat{D}_X^{\text{th}}$  is obtained by replacing in Eq. (10) all  $\omega$ 's by  $\omega_0$ , i.e.,  $\hat{D}_X^{\text{th}} = a^2 \omega_0 c_v \hat{f}_0$  where the correlation factor now writes  $\hat{f}_0 = (\omega_0 + 2\omega_b) \alpha / [\omega_0 + (\omega_0 + 2\omega_b) \alpha]$  with  $\alpha = 1 + 3.5F(1)$ . In Fig. 1, we plot  $\hat{D}_X^{\text{th}}$ ,  $D_X^{\text{bal}}$ , and  $D_X^{\text{tot}}$  as function of the ballistic frequency  $\omega_b$ , and  $\hat{f}_0$  as a function of  $2\omega_b/\omega_0$ , as well as the Monte Carlo results. Clearly the agreement is very good. In particular the corrected expression for the thermal diffusion coefficient predicts remarkably well the decorrelation effect due to the ballistic jumps, even in the region  $\omega_b \approx \omega_0$ . Note that, with the current parameters, the moderate increase in the thermal diffusion coefficient will be masked by the large increase of the ballistic diffusion coefficient. Another important conclusion drawn from Fig. 1 is that the total diffusion coefficient is accurately predicted by adding the thermal and the ballistic diffusion coefficients. The same conclusion holds also for solute diffusion for all alloys and irradiation conditions that we have tested in the case of above-threshold ballistic jumps.

We now investigate the case of solute diffusion. In comparison to self-diffusion, we expect the influence of ballistic jumps to produce more significant modifications of  $\hat{D}_B^{\text{th}}$ . In-

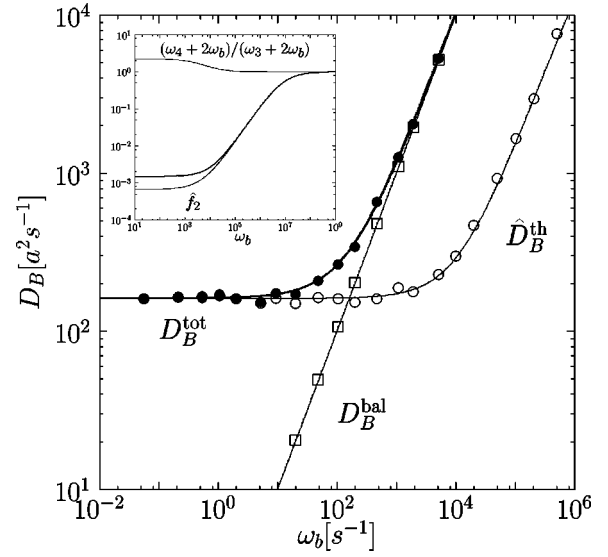


FIG. 2. Total  $D_B^{\text{tot}}$  (●), thermal  $\hat{D}_B^{\text{th}}$  (○), and ballistic  $D_B^{\text{bal}}$  (□) solute diffusion coefficients as a function of the above-threshold ballistic frequency  $\omega_b$  from MC simulations and analytical expressions (lines) [see text]. Data are obtained for the model 1 alloy at  $T = 400 \text{ K}$  and  $c_v = 1.9 \times 10^{-3}$ . The correlation factor  $\hat{f}_2$ , the factor  $(\omega_4 + 2\omega_b)/(\omega_3 + 2\omega_b)$ , and the product of these two quantities are shown in the inset.

deed, the correlation factor  $\hat{f}_2$  may now vary between 0 and 1, and the situation is complicated by the fact that vacancy segregation around the tracer  $B$  atom may also change due to the presence of ballistic jumps. We first want to illustrate the possible annihilation of correlations between successive solute-vacancy jumps. For this purpose we consider a model alloy (model 1) presenting initially strong correlations, i.e., a small value of  $f_2$ . This condition can be fulfilled by choosing a large solute-vacancy frequency  $\omega_2$  with respect to all  $\omega$ 's. In this example, activation energies are set to 0.50 eV for  $\omega_2$ , 0.80 eV for  $\omega_0$  and  $\omega_3$ , and 0.77 eV for  $\omega_1$  and  $\omega_4$  (these parameters correspond to the model alloy studied in Ref. 22, and are typical of a Co impurity in a Cu matrix). In absence of irradiation the correlation factor  $f_2$  is very low and equal to  $\approx 7 \times 10^{-4}$  while the rate  $\omega_4/\omega_3$  that gives the vacancy segregation tendency around the solute atom is equal to 2.2. In Fig. 2 the resulting diffusion coefficients are shown as a function of the ballistic frequency  $\omega_b$ . Here again the agreement between the Monte Carlo simulations and our analytical expressions is very good. In this case, as expected, the decorrelation effects are quite pronounced, resulting in an acceleration of thermal diffusion by almost three orders of magnitude. It is worth noticing that this large acceleration results in fact from two opposite effects: the vacancy segregation around the solute atom decreases slightly as  $\omega_b$  increases [the factor  $(\omega_4 + 2\omega_b)/(\omega_3 + 2\omega_b)$  goes from 2.2 to 1], but the detrapping of the vacancy is so large ( $\hat{f}_2$  increases from  $7 \times 10^{-4}$  to 1) that it determines the overall evolution of the solute thermal diffusion coefficient.

Let us now consider a second model alloy (model 2) to illustrate that the opposite situation can take place as well. We choose this model 2 alloy so that it presents a large

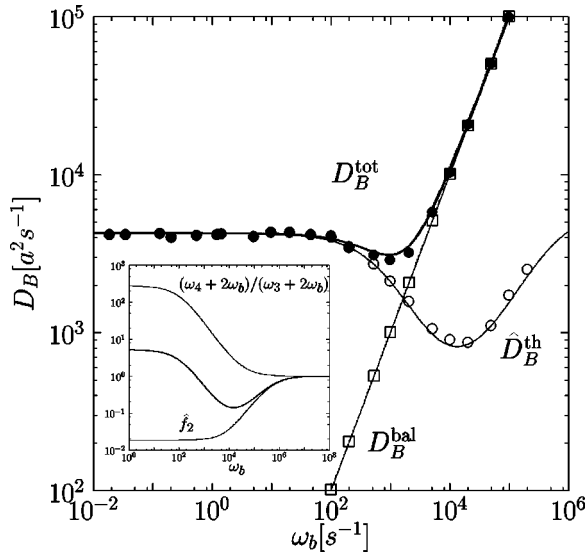


FIG. 3. Total  $D_B^{\text{tot}}$  (●), thermal  $\hat{D}_B^{\text{th}}$  (○), and ballistic  $D_B^{\text{bal}}$  (□) solute diffusion coefficients as a function of the above-threshold ballistic frequency  $\omega_b$  from MC simulations and analytical expressions (lines) [see text]. Data are obtained for the model 2 alloy at  $T=400$  K and  $c_v = 1.9 \times 10^{-3} / \{1 + c_b [Z(\omega_4 - \omega_3) / (\omega_3 + 2\omega_b) - 1]\}$ . The correlation factor  $\hat{f}_2$ , the factor  $(\omega_4 + 2\omega_b) / (\omega_3 + 2\omega_b)$ , and the product of these two quantities are shown in the inset.

vacancy segregation around the solute  $B$  atom. Activation energies are set to 0.60 eV for  $\omega_2$ , 0.93 eV for  $\omega_0$ ,  $\omega_3$ , and 0.73 eV for  $\omega_1$ ,  $\omega_4$ , so that  $\omega_4/\omega_3 \approx 275$  and  $f_2 \approx 0.02$  at  $T=400$  K. In Fig. 3 the corresponding diffusion coefficients are plotted as a function of  $\omega_b$ . With these parameters, the difference of bond strength between  $A$  and  $B$  atoms results in a large increase of the local vacancy concentration  $c_v^B$  around a solute  $B$  atom with respect to the bulk vacancy concentration  $c_v$ . As mentioned above, ballistic jumps tend to reduce this difference of vacancy concentration since under irradiation  $c_v^B = (\omega_4 + 2\omega_b) / (\omega_3 + 2\omega_b) c_v$ .

To compare the Monte Carlo results to our analytical expressions, a rescaling of the vacancy concentration is now required. Indeed, since the number of vacancy is fixed in the simulations (and equal to 1), and since the vacancy is strongly segregating on the solute atom, the true matrix vacancy concentration is reduced. Using a conservation equation for the vacancy in the simulation cell, it is calculated that the value of  $c_v$  in Eq. (10) is reduced by a ratio  $1 + c_b [Z(\omega_4 - \omega_3) / (\omega_3 + 2\omega_b) - 1]$ , where  $c_b$  is the  $B$  solute concentration and  $Z$  is the coordination number ( $=12$ ). Note that this correction is added only to address in a simple way the limitations of the canonical ensemble used for the Monte Carlo simulations. In Fig. 3, one can observe that for this model 2 the agreement between analytical expressions and Monte Carlo results is again very good. For this model 2 alloy, at low  $\omega_b$  the decrease of  $(\omega_4 + 2\omega_b) / (\omega_3 + 2\omega_b)$  occurs before the increase of  $\hat{f}_2$ . This leads to a marked decrease of  $\hat{D}_B^{\text{th}}$ , and produces even a small decrease of  $D_B^{\text{tot}}$ . In this example we arrive at the surprising conclusion that ad-

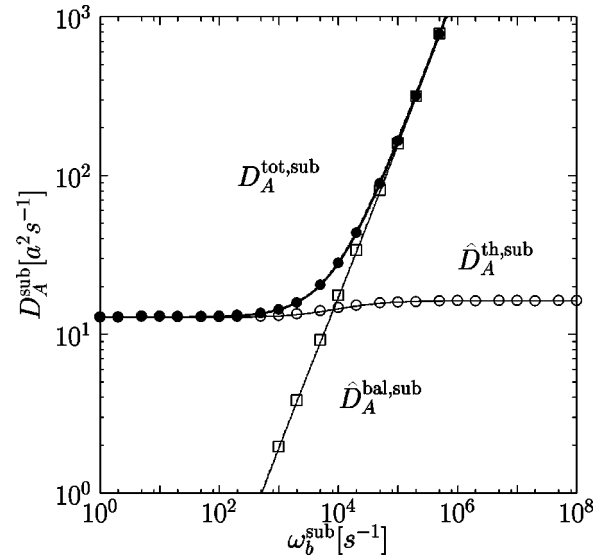


FIG. 4. Total  $D_A^{\text{tot,sub}}$  (●), thermal  $\hat{D}_A^{\text{th,sub}}$  (○), and ballistic  $\hat{D}_A^{\text{bal,sub}}$  (□) self-diffusion coefficients as a function of the sub-threshold ballistic frequency  $\omega_b^{\text{sub}}$  from MC simulations and analytical expressions (lines) [see text]. Data are obtained for the model 1 alloy at  $T=400$  K and  $c_v = 1.9 \times 10^{-3}$ .

ditional ballistic jumps may reduce the solute thermal and total diffusion coefficients.

*Thermal and subthreshold ballistic diffusion.* In the case of above-threshold ballistic jumps, the relative importance of thermal to ballistic diffusion coefficient is directly proportional to the vacancy concentration [see Eqs. (7),(10)]. In the case of subthreshold ballistic diffusion, however, both diffusion coefficients are proportional to the vacancy concentration [see Eqs. (11),(12)]. Furthermore, as both processes proceed by vacancy jumps, one may expect the mutual influence of the two processes on the partial and total diffusion coefficients to be more pronounced. The model 1 alloy introduced in the previous subsection will be used to study these points.

We first consider self-diffusion and solute diffusion in this model 1 alloy. Figures 4 and 5 provide the results obtained from the Monte Carlo (MC) simulations and from our analytical expressions for self-diffusion and solute diffusion coefficients, respectively. In both cases the overall agreement is very good. For self-diffusion, the small decorrelation effects predicted by the formulas are indeed reproduced in the MC simulations. As  $\omega_b^{\text{sub}}$  increases the thermal diffusion increases by a factor  $1/0.7815$ , whereas the ballistic diffusion divided by  $\omega_b^{\text{sub}}$  decreases by a factor 0.7815. Furthermore, even in the region where  $\omega_0 \approx \omega_b^{\text{sub}}$ , the approximated analytical expressions for the partial diffusion coefficients are validated by the MC results. As for the total diffusion coefficient, the MC results are in excellent agreement with the formula [Eq. (13)], but this is not unexpected because the formula is exact for that coefficient. Now for solute diffusion, while the decorrelation effect on the ballistic diffusion coefficient is similar to that for self-diffusion, the increase in the thermal diffusion coefficient is more pronounced; it is in fact quite similar to what was observed in the case of above-threshold

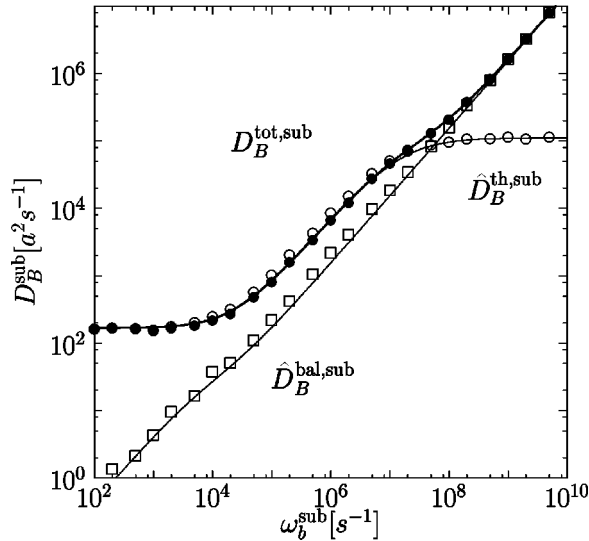


FIG. 5. Total  $D_B^{\text{tot,sub}}$  (●), thermal  $\hat{D}_B^{\text{th,sub}}$  (○), and ballistic  $\hat{D}_B^{\text{bal,sub}}$  (□) solute diffusion coefficients as a function of the sub-threshold ballistic frequency  $\omega_b^{\text{sub}}$  from MC simulations and analytical expressions (lines) [see text]. Data are obtained for the model 1 alloy at  $T=400$  K and  $c_v=1.9\times 10^{-3}$ .

ballistic diffusion (Fig. 2). Furthermore, this increase results in a similar increase of the total diffusion coefficient. As a consequence, these decorrelations could now be easily detected experimentally by measuring the solute diffusion coefficient.

Let us now investigate the possible deviation from additivity of the partial diffusion coefficients, using the data shown in Figs. 4 and 5. For that purpose, we calculate the difference between the sum of the partial diffusion coefficients and the total diffusion coefficient, and we normalize this difference by the total diffusion coefficient [as in Eq. (14)]. The results obtained with the analytical expressions and the MC simulations are plotted in Fig. 6. As discussed in Sec. II, the analytical expressions predict that the sum of the partial diffusion coefficients will overestimate the true total diffusion coefficient. For both self-diffusion and solute diffusion, the MC results confirm this prediction. In the self-diffusion case, the measured excess is in very good quantitative agreement with the analytical curve, whereas in the solute case, the measured excess is about twice as large as the one predicted, and reaches about 60%. This quantitative difference between the analytical expressions and the MC results is not surprising since, as stressed in Sec. II, the proposed formulas for the partial diffusion coefficients ignore the solute-vacancy exchanges due to one dynamics in the calculation of the solute diffusion coefficient for the second dynamics.

#### IV. DISCUSSION

The two main questions addressed in the introduction were first to evaluate the effects of ballistic jumps on thermal diffusion coefficients in an alloy under irradiation, and second to determine whether it is correct to calculate the total diffusion coefficient of a tracer atom by adding its thermal

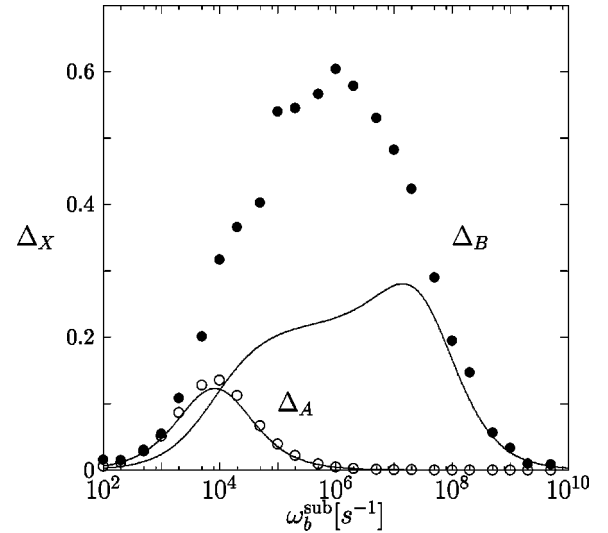


FIG. 6. Deviations from additivity of the partial diffusion coefficients  $\Delta_X = (\hat{D}_X^{\text{th,sub}} + \hat{D}_X^{\text{bal,sub}} - D_X^{\text{tot,sub}}) / D_X^{\text{tot,sub}}$  for  $X=A$  (○) and  $X=B$  (●) using MC data and analytical expressions (lines) shown in Figs. 4 and 5.

and ballistic contributions. As for the first point, Monte Carlo simulations clearly establish that ballistic jumps can have a large impact on thermal diffusion coefficients, especially for solute atoms that are strongly interacting with vacancies. By generalizing the classical five-frequency formula for solute diffusion in an fcc alloy, we obtain analytical expressions for these modified thermal diffusion coefficients that are in very good agreement with Monte Carlo simulations for all the parameters used in this study. This can be understood by recognizing that the modified formulas Eqs. (10),(11) take into account the effects of the ballistic jumps on the path that a vacancy performs between two consecutive exchanges with a tracer atom. These formulas, however, do not take into account that the tracer atom may exchange with the vacancy through ballistic jumps between two thermal exchanges. This limitation should become more apparent when the two dynamics operate at similar rates, because then the tracer migration path consists of mixed thermal and ballistic jumps. It is however observed that this limitation results in minimal deviations. The proposed modified analytical expressions for thermal diffusion in the presence of ballistic jumps are thus found to be useful for the whole range of ballistic jump frequency. In the case of subthreshold ballistic jumps, a similar formula even provides an accurate description of the decorrelation effects produced by thermal jumps on the ballistic diffusion coefficient. In view of this ensemble of results, it is also reasonable to propose that similar modified expressions can be used for bcc crystals, starting from the classical four-frequency formula that has been established for thermal diffusion.<sup>21</sup>

As an example of application of our analytical expressions, we now consider other alloy systems. For that purpose, we use the results published by Adams *et al.*,<sup>24</sup> which give numerical values for all five thermal frequencies for several alloy systems. Consider for instance the case of the diffusion of a Au impurity in Ni. As seen in Fig. 7, the Au

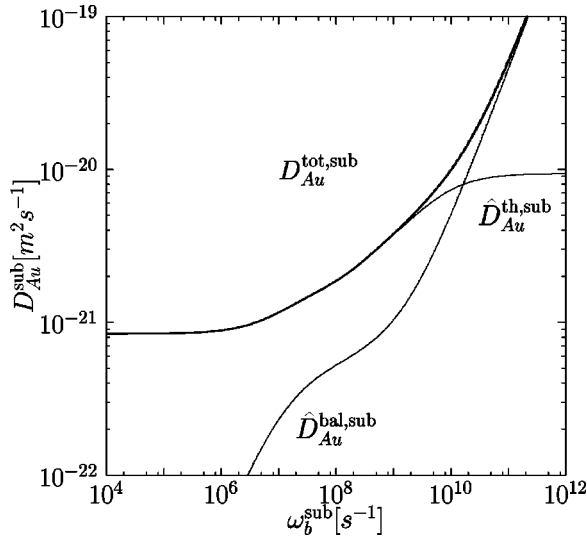


FIG. 7. Total and partial diffusion coefficients of a Au impurity in Ni as a function of the subthreshold ballistic frequency  $\omega_b^{\text{sub}}$ . The curves are plotted using Eqs. (11), (12), and (13) for  $T=800$  K and  $c_v=4.57 \times 10^{-12}$ . Values of the thermal activation energies used for Ni(Au) are equal to 1.1 eV, 1.41 eV, 0.6 eV, 1.07 eV, and 0.77 eV, respectively for  $\omega_0$ ,  $\omega_1$ ,  $\omega_2$ ,  $\omega_3$ , and  $\omega_4$ .<sup>24</sup> The attempt frequency  $\nu$  is set to  $10^{14} \text{ s}^{-1}$ .

thermal and total diffusion coefficients are enhanced by more than one order of magnitude at  $T=800$  K. The lower the temperature, the larger this enhancement gets. A possible experiment to observe this increase would be to irradiate with 300 keV electrons a supersaturated solution of Au in Ni, obtained by quenching the alloy from a temperature above the solvus temperature. The maximum energy that 300 keV electrons can transfer to Au and Ni atoms during elastic collisions is 4.3 eV and 14.4 eV, respectively. This energy is well below the displacement threshold energy and one should thus be in the case where thermal and below-threshold jumps will take place in parallel. Such an irradiation could even be performed *in situ* in a transmission electron microscope. It is anticipated that at moderate irradiation intensities the acceleration of thermal diffusion will lead to an acceleration of the decomposition of the supersaturated solid solution. However, as the irradiation flux increases, ballistic diffusion will become predominant leading to a stabilization of the solid solution. In order to address the possible decomposition of such a solid solution, one would first need to know the cross sections for subthreshold migration of vacancy, so as to convert the electron flux into a subthreshold ballistic jump frequency. Second, one should also take into account the effect of solute concentration on correlation factors. While these tasks are beyond the scope of the present work, a possible approach to address the second point would be to extend to the case of alloys under irradiation the approach recently introduced by Nastar and co-workers<sup>25</sup> to derive analytical expressions of correlation factors in concentrated alloys.

As for the second point addressed in this paper, the additivity of partial diffusion coefficients, Monte Carlo simulations indicate that this additivity is well followed in the case

where ballistic jumps take place everywhere in the crystal (above-threshold ballistic jumps). However, in the case where these forced jumps only involve vacancy-atom pairs (below-threshold ballistic jumps), it is predicted analytically and confirmed by MC simulations that the sum of the partial diffusion coefficients overestimates the true total diffusion coefficient. This deviation from additivity primarily originates from the back and forth jumps that the tracer performs using different dynamics for the forward and backward jumps. Because of the fundamental nature of this phenomenon, it appears that there is no simple way to correct the partial diffusion coefficients if one were to use one continuum kinetic equation with additive dynamics. It is believed that, in that case, kinetic descriptions that include the evolution of pair and multiple-body correlation functions could provide a more accurate description of these alloys with competing dynamics.<sup>26,10,25</sup>

We now turn to a discussion of the effects of ballistic jumps on interstitial migration. Indeed, in most cases, interstitial defects migrate through correlated walks, and by extension of our present results for vacancy migration, it is anticipated that ballistic jumps will perturb these correlations, and therefore will affect interstitial diffusion. In metallic alloys, interstitial defects often adopt a dumbbell configuration. In fcc alloys, these dumbbell interstitials are dissociated along  $\langle 100 \rangle$  directions and they migrate to one of the eight available neighbor sites by jumps that combine a  $90^\circ$  rotation and a translation.<sup>21</sup> For dilute alloys, Bocquet<sup>27</sup> and Chaturvedi and Allnatt<sup>28</sup> have derived expressions for solute diffusion via dumbbell interstitial migration. These expressions have a structure analogous to the expression obtained for vacancy diffusion [Eqs. (3),(4)]. For instance, in the absence of dumbbell rotation, the solute diffusion coefficient can be written as<sup>27</sup>

$$D_{B,i}^{\text{th}} = \frac{4}{3} s^2 \omega_2^i c_i \frac{\omega_4^i}{\omega_3^i} \frac{\omega_{BSI}}{\omega_{BIS}} f_2^i, \quad (15)$$

where  $s$  is the jump distance,  $c_i$  is the total interstitial concentration,  $\omega_4^i$  and  $\omega_3^i$  are the rates of association and dissociation of a solute atom with a self-interstitial dumbbell, respectively;  $\omega_{BSI}$  is the frequency at which a solute atom  $B$  initially on a substitutional site ( $S$ ) is transformed into a mixed dumbbell ( $I$ ), and  $\omega_{BIS}$  is the frequency for the reverse process. The product of  $c_i$  by the two fractions of jump frequencies in Eq. (15) corresponds in fact to the equilibrium concentration of mixed dumbbells.  $\omega_2^i$  is an effective rate of solute interstitial transport given by  $(\omega_2^i)^{-1} = (\omega_{BI})^{-1} + (\omega_{BIS})^{-1}$ , where  $\omega_{BI}$  is the jump frequency of mixed dumbbell from one site to a neighboring site. Finally,  $f_2^i$  is a correlation factor given by

$$f_2^i = \frac{\omega_3^i 5 F_i (\omega_4^i / \omega_0^i)}{\omega_2^i \omega_{BSI} / \omega_{BIS} + \omega_3^i 5 F_i (\omega_4^i / \omega_0^i)}, \quad (16)$$

where  $F_i$  is a function of the sole ratio  $\omega_4^i / \omega_0^i$ . Note that  $\omega_1^i = 0$  due to geometrical constraints on the migration path of the dumbbell (see Ref. 27 for details on the above expres-



sions). Despite the more complex nature of Eq. (15) compared to Eq. (3), the analogy between the two equations allows us to propose an expression for the interstitial solute diffusion coefficient *in the presence* of ballistic jumps by simply adding the interstitial ballistic jump frequency,  $\omega_b^i$ , to  $\omega_0^i$ ,  $\omega_3^i$ ,  $\omega_4^i$ ,  $\omega_{BSI}$ , and  $\omega_{BIS}$ . Similarly to the vacancy migration case, it is expected that ballistic jumps can strongly enhance or suppress solute interstitial migration depending upon alloy parameters. More work is required to test the expression proposed for interstitial migration and to investigate these decorrelation effects.

Finally, for a realistic description of solute and solvent diffusion in alloys undergoing above-threshold irradiation, modified thermal and ballistic diffusion coefficients for vacancy and interstitial migration should be combined. As the defect concentrations enter these expressions, it appears to the present authors that a proper self-consistent approach requires that these modified diffusion coefficients should also be used in the calculations of point defect concentration. These points are left for future work.

## V. CONCLUSION

It is shown that, in an alloy under irradiation, self-diffusion and solute diffusion via vacancy migration can be strongly modified by the presence of ballistic jumps. Indeed, these ballistic jumps can change the correlations that take place during the thermally activated migration of solvent and solute atoms. As a result, thermal self-diffusion increases

slightly, while thermal solute tracer diffusion can be suppressed or enhanced by several orders of magnitude. By generalizing the classical five-frequency model for diffusion in dilute fcc alloys, we proposed formulas that take into account these decorrelations. Monte Carlo simulations are performed and used to validate these analytical expressions. From a practical point of view, enhancements of thermal diffusivity by several orders of magnitude could lead to sizeable accelerations of the kinetics of phase transformations, in particular at low irradiation temperatures. In the case where the ballistic jumps take place only between atoms and vacancies (subthreshold ballistic jumps), it is furthermore established analytically and confirmed with Monte Carlo simulations that the sum of the thermal and ballistic diffusion coefficients exceeds the true total diffusion coefficient. This deviation from diffusion coefficient additivity, which is quite pronounced for solute diffusion, points to a fundamental limitation of continuum kinetic models with additive diffusional contributions. Finally, it is shown that the present approach to take into account the effect of ballistic jumps on solvent and solute diffusion can be extended to the case of dumbbell interstitial migration.

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