Fan *et al.* Reply: The development of atomistic methods capable of following microstructural evolution over time scales beyond the reach of traditional molecular dynamics simulations is a continuing challenge. We offer two observations concerning the preceding Comment [1] regarding the accuracy of the autonomous basin climbing (ABC) method and the benchmarking of new methods such as ABC and the kinetic activation-relaxation technique (k-ART). First, our experience with ABC suggests that it is a simple and robust algorithm for escaping from the potential energy surface (PES) minima in providing the dominant transition pathway [2–6]. In the present published form of ABC, however, it is likely to give an overestimation of effective transition time because of the 1D nature of sampling and implementation. We note, on the other hand, that the ABC method is not inherently limited to producing only a 1D trajectory for the system evolution. To give an illustration we consider a synthetic comparison between ABC and kinetic Monte Carlo (KMC) methods using a preconstructed two-dimensional PES [Fig. 1(a)]. The ABC trajectory was generated for the same prescribed initial state, and used to estimate the temperature-dependent system evolution time and the effective activation energy. Six different sets of activation penalty function parameters [2] were employed, and all the runs gave the same governing transition pathway. To simulate a nonequilibrium driving force that resembles the vacancy clustering problem (see Fig. 2(a) in [7]), we randomly introduced 100 intermediate minima with varying depths and widths [seen in Fig. 1(a)] to form a rough energy landscape, as well as a forward bias connecting the initial and final states. As shown in Fig. 1(b), ABC gives the same effective barrier (1.28 eV) as KMC simulations, while overestimating the evolution times by about 1–2 orders of magnitude. We believe this is a result of the 1D nature of the sampled trajectories in the published version of ABC rather than missing the dominant transition pathways. That is, the residence time at each state is governed by $1/(k_{\text{forward}} + k_{\text{backward}})$, whereas in KMC simulations it is governed by $1/\sum k_i$, considering all possible pathways i. This suggests that the evolution time in the vacancy clustering problem [7] could have been overestimated to some extent; however, it does not seem reasonable that the difference in sampling effectiveness alone can account for evolution times differing by 8 orders of magnitude [1,7]. Moreover, we are currently extending ABC to sample multiple transition pathways, akin to on-the-fly KMC simulations, by blocking the explored transitions. The ABC method, inherently due to its algorithm, favors finding the lowest-energy activation paths. Therefore, this new procedure can efficiently construct the event catalog from highly likely events to low-probability events. As expected, the results from this extended version of the method (ABC-E) show a higher numerical accuracy, as demonstrated for the same 2D PES in Fig. 1.

Second, the usefulness of an atomistic method in producing a minimum-energy path for the system evolution lies in determining not only the activation barrier, but more importantly, the reaction coordinates in terms of the atomic configurations along the path. It is the latter that reveals the associated governing mechanism. In [7], we showed that, in the vacancy supersaturated bcc Fe, the transition to rapid vacancy cluster growth is governed mainly by the onset of vacancy cluster mobility. This is a high-energy-barrier process often ignored at dilute defect concentrations [8]. We suggested this mechanism could provide an explanation of the critical temperature for the rapid growth upon annealing observed in PAS experiments [9]. For assessing the relative sampling effectiveness of the two methods, such as k-ART and ABC, one should ensure that equivalent transition state pathways are being examined, that is, the pathways which play the same dominant role in giving rise to a particular system-level behavior of interest. In doing so, the effects of the cutoff radius in topology discretization and the use of the mean rate method [10] on the connectivity of the sampled trajectories in k-ART could be an issue because of the high defect concentrations and the associated long-range interactions between defect clusters (see Supplemental Material in [7]). This step of benchmarking is important for gaining insights into the physical basis of each method, the understanding needed by the community in developing more robust methods for linking specific transitions to macroscale behavior.

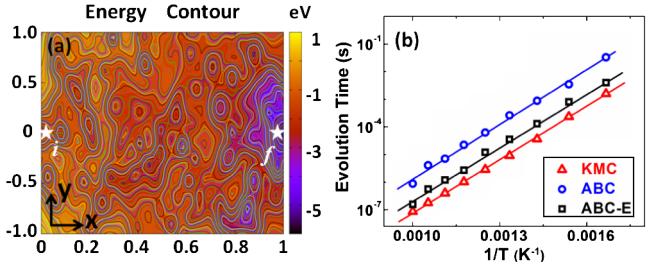

FIG. 1 (color online). (a) A preconstructed PES with bias from initial, i, to final state, f. (b) Variation of the evolution time with temperature sampled by KMC, ABC (average of 6 runs), and ABC-E.

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