Editors' Suggestion

Contributions to Diffusion in Complex Materials Quantified with Machine Learning

Soham Chattopadhyay and Dallas R. Trinkle*
Department of Materials Science and Engineering, University of Illinois, Urbana-Champaign, Illinois 61801, USA

(Received 11 January 2024; revised 17 March 2024; accepted 5 April 2024; published 30 April 2024)

Using machine learning with a variational formula for diffusivity, we recast diffusion as a sum of individual contributions to diffusion—called "kinosons"—and compute their statistical distribution to model a complex multicomponent alloy. Calculating kinosons is orders of magnitude more efficient than computing whole trajectories, and it elucidates kinetic mechanisms for diffusion. The density of kinosons with temperature leads to new accurate analytic models for macroscale diffusivity. This combination of machine learning with diffusion theory promises insight into other complex materials.

DOI: 10.1103/PhysRevLett.132.186301

The first verified laws for diffusion in fluids dates back to the nineteenth-century work of Fick [1], and the last century brought Einstein's major breakthrough connecting Brownian motion and diffusion [2], as well as Onsager's nonequilibrium thermodynamics [3]. In solids, systematic studies of diffusion in metals go back to Roberts and Austen's work on gold diffusing into lead [4]. As the fundamental kinetic process for atomic motion in a material, diffusion controls materials processing for metals, semiconductors, ceramics, and nanoparticles; the operation of batteries and fuel cells; and degredation from corrosion and irradiation [5]. The nano- or atomic-scale processes controlling diffusion are often thermally activated and driven by external forces; the understanding of thermally activated processes goes back to Arrhenius [6], Eyring [7], Polanyi [8], and Vineyard [9]. However, in complex materials with multiple competing processes, diffusivity can deviate from Arrhenius behavior, and is difficult to connect individual processes to macroscale behavior.

Theoretical approaches to diffusion abound, where a variety of approximations have been developed [10,11]. An incomplete list of approaches includes stochastic methods like kinetic Monte Carlo [12–17], master equation methods based on cluster expansions [18–20], kinetic mean-field approximations [21–23], path probability methods for irreversible thermodynamics [24–26], Green function methods [27–29], and Ritz variational methods [30–32]. Recent work on a variational method [33] connected many of these methods and provided a basis for comparing accuracy. The computational methods take different approaches to the underlying difficulty in diffusion: the long-time limit of trajectories complicates identifying important processes and obscures how individual states and transitions contribute to transport. Complex systems have a variety of rates that themselves may be Arrhenius, but together the diffusivity deviates from Arrhenius behavior. A state of the system may have fast transitions to other states, but without connected pathways, these fast transitions indicate trapping without contributing to diffusion, leading to emergent behavior such as percolation.

The variational principle for diffusion combined with modern machine learning techniques presents an opportunity: by rewriting the diffusivity of a system as a sum over individual contributions from every state and transition—what we call "kinosons" (corresponding to movement)—we analyze the transport processes in a new way and discover non-Arrhenius behavior in complex systems. Machine learning methods solve the optimization problem in the variational method to accelerate the computation of diffusivity and permit decomposition into kinosons. With the density of kinosons, we directly identify new analytic forms for the diffusivity and identify differences in the behavior of species within a system. We demonstrate these ideas with diffusion in a complex high-entropy alloy to find fingerprints of percolation and a new analytic form for diffusion.

Diffusivity can be alternatively computed from the mean squared displacement at infinite time, or as the minimum of average squared displacements. The Einstein-Smoluchowski form [2] of diffusion,

$$D = \lim_{t\to\infty} \frac{\langle (\mathbf{x}(t) - \mathbf{x}(0))^2 \rangle}{2dt}, \tag{1}$$

expresses diffusivity D in d dimensions as the average of the long-time limit of squared displacement $\mathbf{x}(t) - \mathbf{x}(0)$ divided by time. By contrast, for a Markovian system, a variational form [33] for Onsager transport coefficients,

$$D = \inf_{\mathbf{y}_\chi} \frac{1}{2d} \left\langle \sum_{\chi'} W(\chi \to \chi') \left(\delta\mathbf{x}(\chi \to \chi') + \mathbf{y}_{\chi'} - \mathbf{y}_\chi\right)^2 \right\rangle_\chi, \tag{2}$$

is expressed as an average over contributions from every transition between any pair of states χ and χ', with rate $W(\chi \to \chi')$ and displacement $\delta\mathbf{x}(\chi \to \chi')$. The diffusivity is

minimized by optimizing the "positions" of states $\mathbf{y}_\chi$. If every state is moved to its optimal position $\mathbf{y}_\chi$, then the mean displacement out of every state is zero, while the mean squared displacement grows linearly with time. The optimized displacements between states is $\widetilde{\delta\mathbf{x}}(\chi \to \chi') \coloneqq \delta\mathbf{x}(\chi \to \chi') + \mathbf{y}_{\chi'} - \mathbf{y}_\chi$, and the total diffusivity is a sum over contributions from every state χ with probability $P(\chi)$ to any other state χ':

$$\kappa(\chi, \chi') \coloneqq \frac{1}{2d} W(\chi \to \chi') \widetilde{\delta x}^2(\chi \to \chi'). \tag{3}$$

These contributions we call "kinosons" (for "little moves") [34]. With the optimal $\mathbf{y}_\chi$, the density of kinosons,

$$g(\kappa) \coloneqq \left\langle \sum_{\chi'} \int_0^\infty \delta(\kappa - \kappa(\chi, \chi')) \right\rangle_\chi, \tag{4}$$

defines the diffusivity for the system and can elucidate the diffusion process.

Having a variational form in Eq. (2) permits a variety of methods to find the optimized displacements, including combining methods for new solutions [33]. For example, using kinetic Monte Carlo to generate finite-length trajectories, Eq. (1) is a variational solution, even if trajectories include only a single transition [33]. We can apply parametrized solutions that map our states χ into vectors $\mathbf{y}_\chi$, such as convolutional neural networks [35], cluster expansions [19,20], or a scaling of the average single-transition displacement out of a state (called the "velocity bias," $\mathbf{b}_\chi$, that contains the escape rates from a state). The neural networks contain the linear cluster expansion models as a subset and should outperform them, while the velocity bias contains explicit information about the rates. For all models, we use machine-learning training methods with Eq. (2) as our objective function; we construct two equal-sized sets of states with Monte Carlo: a "training set" and a distinct "validation set" of states. In this case, the ML algorithm *does not have access to the true diffusivity*, but instead optimizes to the lowest diffusivity, and our validation set verifies that we have not overtrained our models or used insufficient diversity in the training set. With a variational approach, the algorithm with the lowest diffusivity is closest to the true value, and we can combine multiple methods to increase the accuracy. Finally, once we have our optimized $\mathbf{y}_\chi$, we can find the density of kinosons to identify important diffusion processes.

We apply this new approach to the diffusion of one of the first and most studied high-entropy alloys, the Cantor alloy [36,37]; details are available in the Supplemental Material [38]. In this equiatomic Mn-Fe-Cr-Ni-Co alloy, diffusion occurs by the movement of a single vacant site (a vacancy) on a face-centered cubic lattice with a random arrangement of five different chemical species. Due to the range of different exchange rates for each species and the dependency on the local environment, complex behavior emerges: fast-exchanging species trap a vacancy when the vacancy cannot find a different atom to exchange with to escape. For a face-centered cubic lattice with twelve neighbors, this percolation-like behavior is expected when the concentration of fastest species drops below 20% [57,58]. Experimental and theoretical investigations of high-entropy alloys have identified improved mechanical properties [59–65] and high-temperature stability, which indicates slow kinetics for phase separation [66,67]. This slow kinetics is often surprising, due to the inclusion of "fast" exchanging species in the alloys, and it presents an intriguing test for the development of accurate theories of diffusion [68].

A complex material system lies along a spectrum from ordered to fully random; the two ends of the spectrum provide insight into the performance of different computational methods. We consider two model systems with five components to understand how these approaches behave on the ordered-to-random spectrum: one where the vacancy exchange rate is fixed for each chemical species, and another where the rate is sampled from log-normal distributions convolved with the local environment out to the third-nearest-neighbor distance of the vacancy. We select the mean rate (ordered) and the mean and variance (random) for the chemical species to match the distributions of our complex high-entropy alloy. Figure 1 shows that the ordered

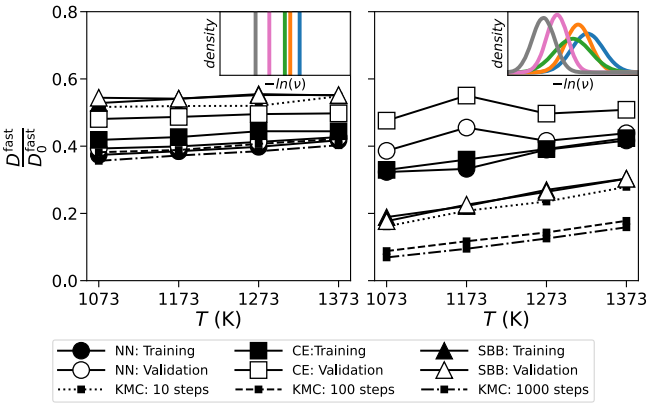

FIG. 1. Predicting diffusivity in an "ordered" (left) vs a "random" (right) five-component model system. Diffusivity is shown as a fraction of the one-step diffusivity prediction, with smaller values indicating increased trapping. The five atomic components have equal concentration, and they move via exchange with a single vacancy, either with a fixed rate (ordered) or a distributed rate (random) that depends on the chemistry. The fastest species dominates the exchanges with the vacancy, but it has difficulty moving over longer distances, as it needs to connect with other fast species to escape trapping. The kinetic Monte Carlo results converge to the true diffusivity as the number of steps increases (lower diffusivity is more accurate). In the ordered case, a neural network (NN) is most accurate, with only a single step; in the random case, the rate-informed scaled bias basis (SBB) method is the most accurate. A complex system lies in between ordered and random, and we expect to need a combination of methods to predict diffusivity.

system diffusivity can be accurately predicted by a nonlinear model (neural network, NN), which outperforms the linear model (cluster expansion, CE) and does better than a simple model relying only on the rates (scaled bias basis, SBB); the configuration around the vacancy provides sufficient information to build an optimized estimate of the mean displacement from a state. However, for a truly random system, the rates are much more diverse, and they flummox nonlinear and linear models based on the configuration of species around the vacancy; direct information about the rate is needed to approach what comes from ten steps in a trajectory. This type of random model has been previously used to estimate diffusivity in high-entropy alloys [68–70], but we note that a random model behaves differently than a *complex* system.

A complex high-entropy alloy—with energies and barriers from a modified-embedded atom model [67]—includes aspects of ordered and fully random systems, and so we combine a convolutional neural network with a scaled residual bias correction in Fig. 2. The diffusivity is converged after 100 transitions in each trajectory using Eq. (1), or an ML approach gets similar results with *single* transitions and Eq. (2). The ML approach starts with a neural network to transform local environments around the vacancy into an estimate for $\mathbf{y}_\chi$ by minimizing the diffusivity in Eq. (2); we can further correct that estimate with the residual velocity bias by directly incorporating information about the escape rates. We note that the neural network can be optimized using high-temperature kinetics and applied out of domain at lower temperatures. This combined method outperforms other computational approaches to the diffusivity, while also requiring orders of magnitude less computational effort than computing long-time trajectories. As we have a good approximation for our optimized displacements, we also evaluate the kinosons for this material system.

Figure 3 shows how the density of kinosons differs from the distribution of rates in the problem, with a fingerprint of percolation of the fast species in the optimized displacements. The complex high-entropy alloy has five different normal distributions for the energy barriers to exchange with a vacancy, depending on the chemical species; the distribution of rates is then approximately log-normal and dominated by the fastest species (Mn). However, the density of kinosons follows a different distribution: a log-skewed normal, which we identify with an exponentially modified Gaussian form [71–73]. The mean of $\ln \kappa$ moves to lower values, while the skewness also favors smaller values. The change in form is due to the relaxed displacements $\widetilde{\delta x}$, which also show their own interesting behavior. The vacancy exchanges with all five species in the alloy, albeit with the highest probability for Mn. However, the distribution of $\widetilde{\delta x}$ looks remarkably different for Mn—where it collapses to highest probability at *zero displacement*—than for the other four species. While Mn is

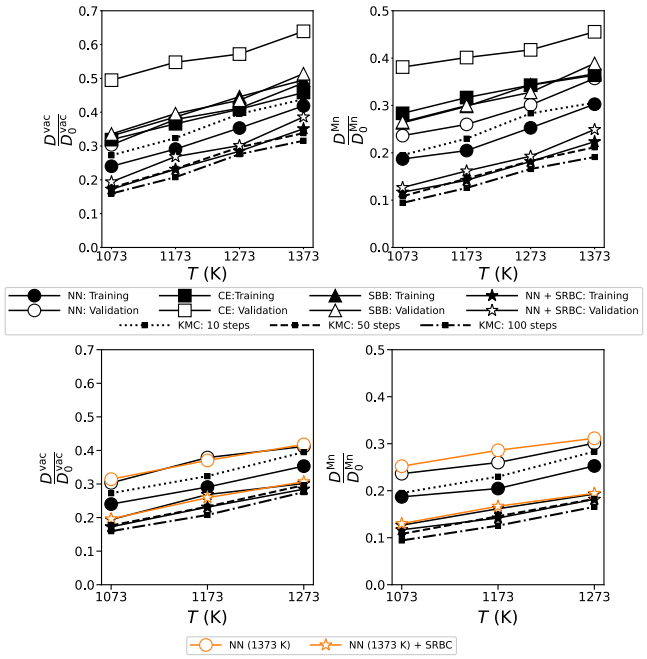

FIG. 2. Predicting vacancy (left) and Mn (right) diffusivity in a five-component complex high-entropy alloy. Diffusivity is shown as a fraction of the one-step diffusivity prediction, with smaller values indicating increased trapping. The real alloy has atomic exchanges with a vacancy that depend on the local atomic environment, behaving as a combination of an ordered and a random system. The kinetic Monte Carlo results converge to the true diffusivity as the number of steps increases (lower diffusivity is more accurate). By combining a neural network with a rate-informed scaled residual bias correction (NN + SRBC, stars), it is possible to predict diffusivity very close to the true value, while only taking a single transition from one state to another. The neural network trained on higher-temperature states can work at lower temperatures in the same phase, with similar accuracy (orange stars). This accuracy allows the computation of the density of kinosons for this complex alloy, and an understanding of the diffusion process.

the fastest exchanger, at 20% concentration most exchanges are later undone, and hence there is the maximum probability for zero displacement. The collapse of the distribution indicates the percolation limit, without connected paths for Mn to transit.

Finally, we can examine the behavior of the density of kinosons with temperature, and we derive a new functional form for the diffusivity of high-entropy alloys in Fig. 4. An exponentially modified Gaussian distribution has three parameters—mean μ, variance σ^2, and decay parameter λ^{-1}—that control the skewness. The parameters follow simple temperature behavior that allows us to easily fit the macroscopic diffusion behavior and extrapolate to lower temperatures. The change in the density of kinosons with temperature can be modeled with a few parameters, producing a new functional form for this diffusivity in

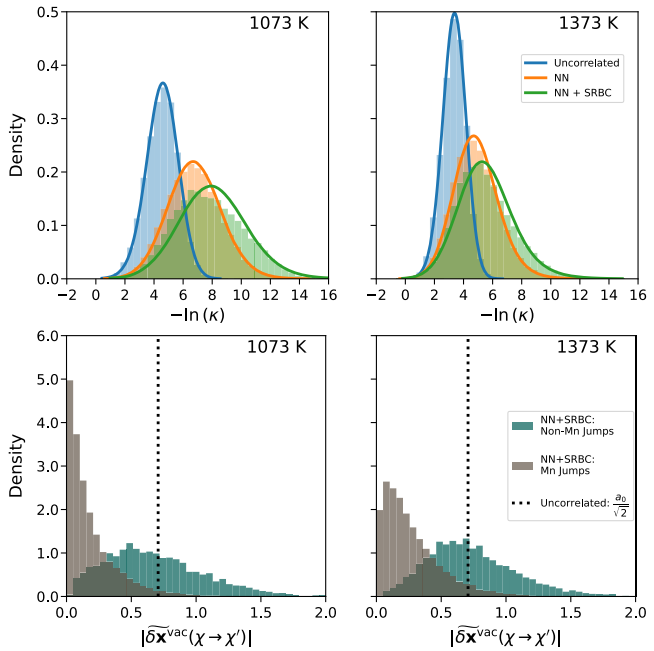

FIG. 3. Density of kinosons κ (top) and optimized displacements $\widetilde{\delta \mathbf{x}}$ (bottom) for the vacancy in a high-entropy alloy. Without the optimized displacements, the distribution of exchange rates provides an "uncorrelated" estimate of the density of kinosons that contribute to diffusivity; for this alloy, this is a nearly log-normal distribution. Once the displacements are known, the kinoson distribution skews to lower values and follows a log–exponentially modified Gaussian. The optimized displacements show a distinct distribution depending on whether they are with a fast Mn or one of the slower elements; the Mn displacements collapse to a peak at zero, while the other elements are distributed around the jump distance in the face-centered cubic lattice. This change is a signature of percolation-like behavior for Mn.

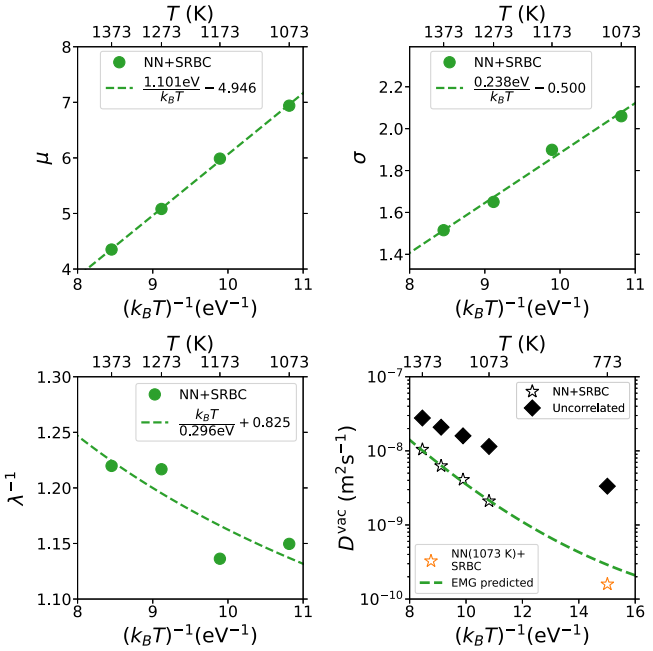

FIG. 4. The density of kinosons' mean, standard deviation, and decay parameters with temperature for the complex high-entropy alloy, and extrapolation to lower temperatures. The log–exponentially modified Gaussian form in Fig. 3 is quantified with three parameters, and their temperature dependence is empirically fit. This reveals a new non-Arrhenius analytic form for the vacancy diffusivity [Eq. (5)] that can be extrapolated accurately to lower temperatures.

this class of alloys. In terms of our parameters, the diffusivity is

$$D^{(\mathrm{EMG})} = \frac{\lambda}{\lambda + 1} \exp\left(-\mu + \frac{1}{2}\sigma^2\right). \tag{5}$$

If σ and λ^{-1} were both zero, the diffusivity would follow an Arrhenius form, as μ is linear in inverse temperature; however, σ is also linear in inverse temperature and λ^{-1} is finite with a weak temperature dependence, producing *non-Arrhenius behavior* in this complex high-entropy alloy. We note that the vacancy and Mn have nonzero λ^{-1}, while Fe and Cr are better represented by log-normal distributions ($\lambda^{-1} = 0$). The result of this high-temperature fit also extrapolates well to even lower temperatures. The functional form of Eq. (5) comes from the density of kinosons, revealed by our machine-learning analysis, and we expect it to be applicable to other high-entropy alloys.

The new prediction of diffusivity agrees much better with experimental measurements [74] in Fig. 5. The ratio of diffusivity for the second- and third-fastest species (Fe and Cr) to the fastest (Mn) removes the unknown vacancy concentration, tracer correlation factor, and thermodynamic factors in our random alloy—assumed to be equal for our species—to robustly compare with the measured

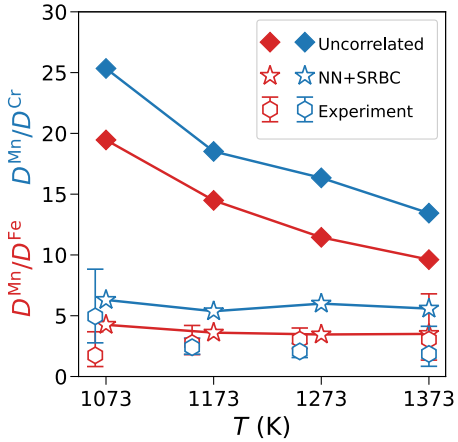

FIG. 5. Ratio of the diffusivity of Mn to those of Fe and Cr in the cantor alloy. Using the average rates ("uncorrelated") disagrees with experimental tracer diffusion measurements [75], while the neural network calculations of the optimized kinosons are within experimental uncertainty.

tracer diffusivity from experiments. The unrelaxed kinoson prediction of "uncorrelated" diffusivity—what you would expect using the distribution of transition rates—shows a significant deviation from the experimental values. When the optimized kinosons are used instead, the agreement is within the experimental error estimates.

Working with a variational approach for diffusivity, a machine-learning approach for optimization correctly predicted diffusion in a complex high-entropy alloy with a fraction of the effort needed for trajectories. We take advantage of a physics-informed neural network, leveraging underlying crystal symmetries including translation and the locality provided by a convolutional neural network to gain insight into the underlying physical processes. With the optimized displacements, we express diffusivity as a sum of discrete jumps, called kinosons, which follow a different statistical distribution than the transition rates in the material. The distribution of optimized displacements indicates the approach of a percolation transition for the fastest species in alloy, helping to illustrate the complexity of this material that lies between ordered and random. The density of kinosons provides a new analytic form for the diffusivity of high-entropy alloys, and the parameters can be easily fit from the density of kinosons. This new analysis technique highlights the power of physics-based machine learning to model and even *understand* diffusion processes in complex materials, transforming the high-dimensional kinetic problem of diffusion into a one-dimensional density of kinosons. This should prove a powerful tool for understanding diffusion in other solid materials, including glasses, and the analysis of long-time kinetic processes may be applied to other nonequilibrium problems in materials or chemistry.

The authors thank Dr. Danny Perez and Professor Lee DeVille for helpful conversations, and Professor Sergiy Divinski for helpful conversations and the experimental data in Fig. 5. This work is sponsored by the NSF under Program No. MPS-1940303. This work made use of the Illinois Campus Cluster, a computing resource that is operated by the Illinois Campus Cluster Program (ICCP) in conjunction with the National Center for Supercomputing Applications (NCSA), and which is supported by funds from the University of Illinois at Urbana-Champaign. The code is available at [76], and the data are available at [77].

*dtrinkle@illinois.edu

[1] Adolf Fick, Ueber diffusion, Ann. Phys. (Berlin) **170**, 59 (1855).
[2] Albert Einstein, Über die von der molekularkinetischen Theorie der Wärme geforderte Bewegung von in ruhenden Flüssigkeiten suspendierten Teilchen, Ann. Phys. (Berlin) **322**, 549 (1905).
[3] Lars Onsager, Reciprocal relations in irreversible processes: I, Phys. Rev. **37**, 405 (1931).
[4] William Chandler Roberts-Austen, Bakerian lecture: On the diffusion of metals, Phil. Trans. R. Soc. A **187**, 383 (1896).
[5] Robert W. Balluffi, Samuel M. Allen, and W. Craig Carter, *Kinetics of Materials* (John Wiley & Sons, Inc., New York, 2005).
[6] Svante Arrhenius, Über die reaktionsgeschwindigkeit bei der inversion von rohrzucker durch säuren, Z. Phys. Chem. **4U**, 226 (1889).
[7] Henry Eyring, The activated complex in chemical reactions, J. Chem. Phys. **3**, 107 (1935).
[8] M. G. Evans and M. Polanyi, Some applications of the transition state method to the calculation of reaction velocities, especially in solution, Trans. Faraday Soc. **31**, 875 (1935).
[9] George H. Vineyard, Frequency factors and isotope effects in solid state rate processes, J. Phys. Chem. Solids **3**, 121 (1957).
[10] Paul Heitjans and Jörg Kärger, *Diffusion in Solids: Fundamentals, Methods, Materials, Diffusion-Controlled Processes*, Springer Series in Solid-State Sciences (Springer-Verlag, Berlin Heidelberg, 2005).
[11] Helmut Mehrer, *Diffusion in Solids: Fundamentals, Methods, Materials, Diffusion-Controlled Processes*, Springer Series in Solid-State Sciences (Springer-Verlag, Berlin Heidelberg, 2007).
[12] G. E. Murch, Simulation of diffusion kinetics with the Monte Carlo method, in *Diffusion in Crystalline Solids*, edited by G. E. Murch and A. S. Nowick (Academic Press, Orlando, Florida, 1984), Chap. 7, pp. 379–427.
[13] I. V. Belova and G. E. Murch, Collective diffusion in the binary random alloy, Philos. Mag. A **80**, 599 (2000).
[14] I. V. Belova and G. E. Murch, Behaviour of the diffusion vacancy-wind factors in the concentrated random alloy, Philos. Mag. A **81**, 1749 (2001).
[15] I. V. Belova and G. E. Murch, Computer simulation of solute-enhanced diffusion kinetics in dilute fcc alloys, Philos. Mag. **83**, 377 (2003).
[16] I. V. Belova and G. E. Murch, Solvent diffusion kinetics in the dilute random alloy, Philos. Mag. **83**, 393 (2003).
[17] Manuel Athénes, Gilles Adjanor, and Jérôme Creuze, Estimating linear mass transport coefficients in solid solutions via correlation splitting and a law of total diffusion, Phys. Rev. Mater. **6**, 013805 (2022).
[18] M. Nastar, V. Yu. Dobretsov, and G. Martin, Self-consistent formulation of configurational kinetics close to equilibrium: The phenomenological coefficients for diffusion in crystalline solids, Philos. Mag. A **80**, 155 (2000).
[19] M. Nastar, A mean field theory for diffusion in a dilute multi-component alloy: A new model for the effect of solutes on self-diffusion, Philos. Mag. **85**, 3767 (2005).
[20] Thomas Schuler, Luca Messina, and Maylise Nastar, KineCluE: A kinetic cluster expansion code to compute transport coefficients beyond the dilute limit, Comput. Mater. Sci. **172**, 109191 (2020).
[21] K. D. Belashchenko and V. G. Vaks, The master equation approach to configurational kinetics of alloys via the vacancy exchange mechanism: General relations and fea-

tures of microstructural evolution, J. Phys. Condens. Matter **10**, 1965 (1998).
[22] V. G. Vaks, A. Yu. Stroev, I. R. Pankratov, and A. D. Zabolotskiy, Statistical theory of diffusion in concentrated alloys, J. Exp. Theor. Phys. **119**, 272 (2014).
[23] V. G. Vaks, K. Yu. Khromov, I. R. Pankratov, and V. V. Popov, Statistical theory of diffusion in concentrated BCC and FCC alloys and concentration dependences of diffusion coefficients in BCC alloys FeCu, FeMn, FeNi, and FeCr, J. Exp. Theor. Phys. **123**, 59 (2016).
[24] Ryoichi Kikuchi, The path probability method, Prog. Theor. Phys. Suppl. **35**, 1 (1966).
[25] Hiroshi Sato and Ryoichi Kikuchi, Theory of many-body diffusion by the path-probability method: Conversion from ensemble averaging to time averaging, Phys. Rev. B **28**, 648 (1983).
[26] Hiroshi Sato, Takuma Ishikawa, and Ryoichi Kikuchi, Correlation factor in tracer diffusion for high tracer concentrations, J. Phys. Chem. Solids **46**, 1361 (1985).
[27] Elliott W. Montroll and George H. Weiss, Random walks on lattices: II, J. Math. Phys. (N.Y.) **6**, 167 (1965).
[28] Masahiro Koiwa and Shunya Ishioka, Integral methods in the calculation of correlation factors for impurity diffusion, Philos. Mag. A **47**, 927 (1983).
[29] Dallas R. Trinkle, Automatic numerical evaluation of vacancy-mediated transport for arbitrary crystals: Onsager coefficients in the dilute limit using a Green function approach, Philos. Mag. **97**, 2514 (2017).
[30] Zbigniew W. Gortel and Magdalena A. Załuska-Kotur, Chemical diffusion in an interacting lattice gas: Analytic theory and simple applications, Phys. Rev. B **70**, 125431 (2004).
[31] Magdalena A. Załuska-Kotur and Zbigniew W. Gortel, Ritz variational principle for collective diffusion in an adsorbate on a non-homogeneous substrate, Phys. Rev. B **76**, 245401 (2007).
[32] Magdalena A. Załuska-Kotur, Variational approach to the diffusion on inhomogeneous lattices, Appl. Surf. Sci. **304**, 122 (2014).
[33] Dallas R. Trinkle, Variational principle for mass transport, Phys. Rev. Lett. **121**, 235901 (2018).
[34] Note also that $dD/d\ln W(\chi \to \chi') = P(\chi)\kappa(\chi,\chi')$, and $P(\chi)\kappa(\chi,\chi') = P(\chi')\kappa(\chi',\chi)$.
[35] Taco Cohen and Max Welling, Group equivariant convolutional networks, in *Proceedings of the 33rd International Conference on Machine Learning*, Proceedings of Machine Learning Research Vol. 48, edited by Maria Florina Balcan and Kilian Q. Weinberger (PMLR, New York, New York, USA, 2016), pp. 2990–2999.
[36] B. Cantor, I. T. H. Chang, P. Knight, and A. J. B. Vincent, Microstructural development in equiatomic multicomponent alloys, Mater. Sci. Eng. A **375–377**, 213 (2004).
[37] Zhouran Zhang, David E. J. Armstrong, and Patrick S. Grant, The effects of irradiation on CrMnFeCoNi high-entropy alloy and its derivatives, Prog. Mater. Sci. **123**, 100807 (2022).
[38] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.132.186301 for detailed description of parametrics models and objective function, algorithms and dataset construction, and results for lattice gas and high-entropy alloys, which includes Refs. [39–56].
[39] Xavier Glorot, Antoine Bordes, and Yoshua Bengio, Deep sparse rectifier neural networks, in *Proceedings of the Fourteenth International Conference on Artificial Intelligence and Statistics*, Proceedings of Machine Learning Research Vol. 15, edited by Geoffrey Gordon, David Dunson, and Miroslav Dudík (PMLR, Fort Lauderdale, FL, USA, 2011), pp. 315–323.
[40] Adam Paszke *et al.*, Pytorch: An imperative style, high-performance deep learning library, in *Advances in Neural Information Processing Systems 32* (Curran Associates, Inc., Red Hook, 2019), pp. 8024–8035.
[41] Diederik P. Kingma and Jimmy Ba, Adam: A method for stochastic optimization, arXiv:1412:6980.
[42] Ken-ichi Ebihara, Tomoaki Suzudo, and Masatake Yamaguchi, Modeling of phosphorus transport by interstitial dumbbell in α-iron using first-principles-based kinetic Monte Carlo, Mater. Trans., JIM **58**, 26 (2017).
[43] A. R. Allnatt, T. R. Paul, I. V. Belova, and G. E. Murch, A high accuracy diffusion kinetics formalism for random multicomponent alloys: Application to high entropy alloys, Philos. Mag. **96**, 2969 (2016).
[44] John R. Manning, Correlation factors for diffusion in nondilute alloys, Phys. Rev. B **4**, 1111 (1971).
[45] L. K. Moleko, A. R. Allnatt, and E. L. Allnatt, A self-consistent theory of matter transport in a random lattice gas and some simulation results, Philos. Mag. A **59**, 141 (1989).
[46] Daan Frenkel and Berend Smit, *Understanding Molecular Simulation: From Algorithms to Applications* (Elsevier, New York, 2023).
[47] Aidan P. Thompson, H. Metin Aktulga, Richard Berger, Dan S. Bolintineanu, W. Michael Brown, Paul S. Crozier, Pieter J. in't Veld, Axel Kohlmeyer, Stan G. Moore, Trung Dac Nguyen, Ray Shan, Mark J. Stevens, Julien Tranchida, Christian Trott, and Steven J. Plimpton, LAMMPS: A flexible simulation tool for particle-based materials modeling at the atomic, meso, and continuum scales, Comput. Phys. Commun. **271**, 108171 (2022).
[48] Mohamed N. Nounou and Bhavik R. Bakshi, Multiscale methods for denoising and compression, in *Data Handling in Science and Technology* (Elsevier, New York, 2000), pp. 119–150.
[49] Graeme Henkelman and Hannes Jónsson, Improved tangent estimate in the nudged elastic band method for finding minimum energy paths and saddle points, J. Chem. Phys. **113**, 9978 (2000).
[50] Weinan E, Weiqing Ren, and E. Vanden Eijnden, String method for the study of rare events, Phys. Rev. B **66**, 052301 (2002).
[51] E. Maras, O. Trushin, A. Stukowski, T. Ala-Nissila, and H. Jónsson, Global transition path search for dislocation formation in Ge on Si(001), Comput. Phys. Commun. **205**, 13 (2016).
[52] Sebastián Echeverri Restrepo and Predrag Andric, ABC-FIRE: Accelerated bias-corrected fast inertial relaxation engine, Comput. Mater. Sci. **218**, 111978 (2023).
[53] Julien Guénolé, Wolfram G. Nöhring, Aviral Vaid, Frédéric Houllé, Zhuocheng Xie, Aruna Prakash, and Erik Bitzek,

Assessment and optimization of the fast inertial relaxation engine (fire) for energy minimization in atomistic simulations and its implementation in LAMMPS, Comput. Mater. Sci. **175**, 109584 (2020).

[54] Thomas Garnier, Venkateswara R. Manga, Dallas R. Trinkle, Maylise Nastar, and Pascal Bellon, Stress-induced anisotropic diffusion in alloys: Complex Si solute flow near a dislocation core in Ni, Phys. Rev. B **88**, 134108 (2013),

[55] David A. Porter, Kenneth E. Easterling, and Mohamed Y. Sherif, Thermodynamics and phase diagrams, in *Phase Transformations in Metals and Alloys* (CRC Press, Boca Raton 2021).

[56] Kazuki Sugita, Naoki Matsuoka, Masataka Mizuno, and Hideki Araki, Vacancy formation enthalpy in CoCrFeMnNi high-entropy alloy, Scr. Mater. **176**, 32 (2020).

[57] D. S. Gaunt and M. F. Sykes, Series study of random percolation in three dimensions, J. Phys. A **16**, 783 (1983).

[58] H. Ouyang and B. Fultz, Percolation in alloys with thermally activated diffusion, J. Appl. Phys. **66**, 4752 (1989).

[59] A. Gali and E. P. George, Tensile properties of high- and medium-entropy alloys, Intermetallics **39**, 74 (2013).

[60] Y. D. Wu, Y. H. Cai, T. Wang, J. J. Si, J. Zhu, Y. D. Wang, and X. D. Hui, A refractory $Hf_{25}Nb_{25}Ti_{25}Zr_{25}$ high-entropy alloy with excellent structural stability and tensile properties, Mater. Lett. **130**, 277 (2014).

[61] J. Y. He, H. Wang, H. L. Huang, X. D. Xu, M. W. Chen, Y. Wu, X. J. Liu, T. G. Nieh, K. An, and Z. P. Lu, A precipitation-hardened high-entropy alloy with outstanding tensile properties, Acta Mater. **102**, 187 (2016).

[62] Y. F. Ye, Q. Wang, J. Lu, C. T. Liu, and Y. Yang, High-entropy alloy: Challenges and prospects, Mater. Today **19**, 349 (2016).

[63] Daixiu Wei, Xiaoqing Li, Weicheng Heng, Yuichiro Koizumi, Feng He, Won-Mi Choi, Byeong-Joo Lee, Hyoung Seop Kim, Hidemi Kato, and Akihiko Chiba, Novel co-rich high entropy alloys with superior tensile properties, Mater. Res. Lett. **7**, 82 (2018).

[64] Weidong Li, Peter K. Liaw, and Yanfei Gao, Fracture resistance of high entropy alloys: A review, Intermetallics **99**, 69 (2018).

[65] X. J. Fan, R. T. Qu, and Z. F. Zhang, Remarkably high fracture toughness of HfNbTaTiZr refractory high-entropy alloy, J. Mater. Sci. Technol. **123**, 70 (2022).

[66] K.-Y. Tsai, M.-H. Tsai, and J.-W. Yeh, Sluggish diffusion in Co-Cr-Fe-Mn-Ni high-entropy alloys, Acta Mater. **61**, 4887 (2013).

[67] Won-Mi Choi, Yong Hee Jo, Seok Su Sohn, Sunghak Lee, and Byeong-Joo Lee, Understanding the physical metallurgy of the CoCrFeMnNi high-entropy alloy: An atomistic simulation study, npj Comput. Mater. **4**, 1 (2018).

[68] Spencer L. Thomas and Srikanth Patala, Vacancy diffusion in multi-principal element alloys: The role of chemical disorder in the ordered lattice, Acta Mater. **196**, 144 (2020).

[69] Josua Kottke, Daniel Utt, Mathilde Laurent-Brocq, Adnan Fareed, Daniel Gaertner, Loïc Perrière, Łukasz Rogal, Alexander Stukowski, Karsten Albe, Sergiy V. Divinski, and Gerhard Wilde, Experimental and theoretical study of tracer diffusion in a series of $CoCrFeMn_{100-x}Ni_x$ alloys, Acta Mater. **194**, 236 (2020).

[70] Biao Xu, Jun Zhang, Shihua Ma, Yaoxu Xiong, Shasha Huang, J. J. Kai, and Shijun Zhao, Revealing the crucial role of rough energy landscape on self-diffusion in high-entropy alloys based on machine learning and kinetic Monte Carlo, Acta Mater. **234**, 118051 (2022).

[71] Eli Grushka, Characterization of exponentially modified Gaussian peaks in chromatography, Anal. Chem. **44**, 1733 (1972).

[72] Jake Olivier and Melissa Norberg, Positively skewed data: Revisiting the box-cox power transformation, Int. J. Psychol. Res. **3**, 68 (2010).

[73] Sajid Ali, Jehan Ara, and Ismail Shah, A comparison of different parameter estimation methods for exponentially modified Gaussian distribution, Afr. Mat. **33**, 58 (2022).

[74] M. Vaidya, K. G. Pradeep, B. S. Murty, G. Wilde, and S. V. Divinski, Bulk tracer diffusion in CoCrFeNi and CoCrFeMnNi high entropy alloys, Acta Mater. **146**, 211 (2018).

[75] Daniel Gaertner, Josua Kottke, Yury Chumlyakov, Fabian Hergemöller, Gerhard Wilde, and Sergiy V. Divinski, Tracer diffusion in single crystalline CoCrFeNi and CoCrFeMnNi high-entropy alloys: Kinetic hints towards a low-temperature phase instability of the solid-solution?, Scr. Mater. **187**, 57 (2020).

[76] https://github.com/TrinkleGroup/VKMC.

[77] Dallas R. Trinkle and Soham Chattopadhyay, Kinoson data for FCC random and high entropy alloys (2023),https://zenodo.org/doi/10.5281/zenodo.10214333.