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A machine-learning interatomic potential to understand primary radiation damage of silicon

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ABSTRACT

Harsh radiation environments cause displacement damages in semiconductor components, resulting in performance degradation. Molecular simulations provide a unique approach to study the dynamic processes of radiation-induced defect production, clustering, and evolution to design and reinforce novel semiconductor components. In this paper, we developed a more efficient machine learning (ML) potential with DFT accuracy to investigate the radiation damage in silicon material. The accuracy of the potential was verified by comparing the static properties, defect formation energy, and threshold displacement energy with experiments and DFT data. By simulating the excitation of a single PKA we found that with the ML potential PKA has an impact over a larger spatial area compared to the empirical potentials. Finally, by simulating multiple PKA excitations in sequence, we found that the first several excited PKAs produce an amorphous region. For the later excited PKAs, the energies are dissipated when they cross through the amorphous region, resulting in a 36% decrease in newly generated defects.

1. Introduction

In severe radiation environments like nuclear plants or deep space, neutrons, gamma rays, protons, α particles, heavy ions, etc. have radiation effects on semiconductor components and materials. Among them, the displacement damage effect can seriously affect the electrical properties of semiconductor components and lead to performance degradation [1–3]. Therefore, it is needed to study the displacement damage in semiconductor materials for designing and reinforcing novel semiconductor components.

To study the defects produced by the displacement damage, experimentalists have used advanced experimental facilities like transmission electron microscopy (TEM) and deep level transient spectroscopy (DLTS) [4]. Although the defects produced by the displacement damage can be observed and identified, the dynamic process of the defects production cannot be obtained since it is extremely fast and far from equilibrium. To obtain a deeper understanding of the production, clustering, and evolution of radiation-induced defects, atomic-scale simulations are needed to provide the necessary microscopic perspective [5].

The density functional theory (DFT) method offers a relatively accurate Born-Oppenheimer potential energy surface and can be used to obtain the structure of radiation-induced defects, defect formation energy, defect migration path, and threshold displacement energy [6–9]. However, due to the length and time scale limitations of this approach, it is difficult to investigate defect production, clustering, and evolution during radiation damage. Eventually, molecular dynamics (MD) simulations based on empirical potentials took the stage. MD simulations based on empirical potentials are much more computationally efficient than DFT [13]. The computational cost scale linearly with simulation time and the size of the system so that they can handle significantly larger time and length scales. MD simulations based on empirical potentials have been used to study the surface effect [10], the temperature effect [11], and the formation process [12] of the radiation damage. A more complete review is presented by Nordlund [5]. The accuracy of MD simulations depends entirely on the accuracy of the potential. However, the accuracy of the empirical potentials has always been a concern. The mathematical form of the empirical potential is derived from the physical understanding of the interatomic interactions, but this

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approximation tends to oversimplify the physical nature of interatomic interactions. Therefore, the potential energy surface obtained by the empirical potential is not accurate enough.

Recently, the ML potential has successfully implemented a potential energy surface with DFT accuracy while maintaining the efficiency of MD simulations [13–15]. The ML potential uses the DFT results as the data set to train the mapping between the local atomic environment and the atomic energy contribution. In this way, ML successfully builds a bridge between DFT and MD. The ML potential has been applied to different kinds of materials such as silicon [16], copper [17], water [13], zinc oxide [18], etc., which shows great generality. The ML potential has also been used to study radiation damage [19–21]. Silicon is one of the most important and widely used materials for the manufacture of semiconductor components. In the research on the radiation damage of silicon, experimentalists have investigated the production of vacancies [48] and divacancies [48,57] by electron radiation, threshold displacement energy [48–49] and sputtering yield [58–60]. Meanwhile, the simulation community has studied the threshold displacement energy [8], generation and long-time relaxation of damage structure [61] and PKA energy spectra [62]. In particular, Hamedani et al. [20] have used a modified GAP potential (one type of ML potential) to study radiation damage in silicon materials. However, the efficiency of this Gaussian process regression-based modified GAP potential is less than that of neural network-based ML potentials, especially when large DFT datasets are included [22]. Therefore, it is needed to develop a more efficient ML potential to perform radiation damage calculations of silicon.

In this paper, we developed a more efficient ML potential with DFT accuracy to describe radiation damage in silicon material. Based on the obtained model, we first calculated the static properties, defect formation energy, and threshold displacement energy to verify the accuracy of the ML potential. Then, we investigated the difference between the ML potential and the empirical potentials in terms of defect production, clustering, and evolution by studying the radiation damage with a single primary knock-on atom (PKA) excitation. Finally, based on the ML potential, we performed simulations of PKA excitations in sequence and investigated the effect of the first several excited PKAs on the later excited PKAs.

2. Machine learning potential

2.1. Database

To construct a ML model that can capture the evolution of defects in collision cascade in silicon material, a dataset related to the radiation conditions was built. The dataset contains four parts, corresponding to the static properties, points defects, liquid/amorphous phase, and radiation damage of silicon. All configurations come from either *ab initio* MD (AIMD) simulations or MD simulations with empirical force fields. The details are shown in Table 1 below. The type and number of configurations are based on physical intuition and past practice. The dataset contains a total of 66,000 configurations.

During the radiation damage evolution, the interatomic distances can be very short. In this case, the interatomic interactions are difficult to describe by the conventional DFT approaches [19]. In this article, we calculated the short-range interatomic interactions using the coupled-cluster singles and doubles with perturbative triples (CCSD(T)) [23–24] method. The calculations were performed with Python-based simulations of chemistry framework (PySCF) [25–27]. The cc-pVDZ basis set [28] was used to determine the repulsive interaction within the Si-Si bond (≤ 2.36 Å). The CCSD(T) results were then fitted in the form of a potential table and interpolated into the ML potential to describe the radiation damage evolution process. The DFT calculations were performed with the Vienna *Ab initio* simulation package (VASP) [29–31] using the projector augmented wave (PAW) method [31]. The Perdew-Burke-Ernzerhof (PBE) [32] exchange–correlation functional

Table 1

Summary of the database for the silicon model for accurate radiation damage simulations. The first column shows the corresponding properties of the configurations. The second column shows the type of configurations. The third column shows the number of atoms of the configurations. The fourth column shows the number of configurations. The last column shows the number of atomic environments, which is equal to the number of atoms times the number of configurations.

Corresponding properties	Structure type	No. atoms	No. configurations	No. atomic environments
Static properties	Diamond under strain ($\delta, 0, 0, 0, 0, 0$)	64	10,000	640,000
	Diamond under strain ($\delta, \delta, 0, 0, 0, 0$)	64	10,000	640,000
	Diamond under strain ($\delta, \delta, \delta, 0, 0, 0$)	64	10,000	640,000
Defects related properties	Diamond with vacancy under strain ($\delta, 0, 0, 0, 0, 0$)	215	2000	430,000
	Diamond with vacancy under strain ($\delta, \delta, 0, 0, 0, 0$)	215	2000	430,000
	Diamond with vacancy under strain ($\delta, \delta, \delta, 0, 0, 0$)	215	2000	430,000
	Diamond with dumbbell interstitial under strain ($\delta, 0, 0, 0, 0, 0$)	217	2000	434,000
	Diamond with dumbbell interstitial under strain ($\delta, \delta, 0, 0, 0, 0$)	217	2000	434,000
	Diamond with dumbbell interstitial under strain ($\delta, \delta, \delta, 0, 0, 0$)	217	2000	434,000
	Diamond heated to liquid and cooled down	216	12,000	2,592,000
	Diamond with PKA activation	144	12,000	1,728,000
	Liquid/amorphous phase			
Radiation damage				

was used and the cutoff energy for the plane wave basis set was set to 400 eV. A $2 \times 2 \times 2$ Monkhorst-Pack k-points mesh was used in all AIMD simulations. The energy of each configuration and the forces on each atom of the configuration were then calculated using both the short-range potential table and DFT. The difference between the short-range potential table and DFT was then used as the dataset for supervised ML training.

2.2. Model training

Based on the above data set, we trained a ML potential using DeePMD [13] artificial neural network architecture. Atoms within a cutoff radius of 6 Å were taken into account for the local atomic environment of the ML potential. The batch size was set to 4 and the training ran through 10^6 batches. The starting and ending learning rate was set to 5×10^{-3} and 5×10^{-8} , respectively and the learning rate was exponentially decayed during the training. Finally, a deep neural network of three hidden layers with 240, 240, and 240 nodes was employed to map the local atomic environment to the atomic energy contributions. The training took approximately 31 h on Intel Xeon E5-2692 v2 CPU. The

comparison of the computational efficiency of the trained ML potential and another ML potential [20,37] is shown in the [supporting information](#).

Comparisons between the database and ML results on energies and forces are shown in Fig. 1. The resulting root mean squared errors (RMSE) of the energies are 5.19 and 5.19 meV/atom for training and test sets, respectively. As for forces, the RMSE are 98.37 and 98.29 meV/Å for training and test sets, respectively. The RMSE values of the ML potential are consistent with the typical RMSE values for ML models [33].

3. Results and discussion

3.1. Static properties

To verify the validity of the ML potential, we first calculated several static properties using the trained ML potential and compared them with other empirical potentials, DFT results, and experiments. The static properties here include the lattice constant a , the cohesive energy E_{Coh} , and the diamond cubic structure elastic constants C_{11} , C_{12} and C_{44} . The detailed results are shown in Table 2.

From Table 2, our DFT results are consistent with the other DFT works. Meanwhile, the computed static properties based on our trained ML potential are very close to our DFT reference data. The results of another ML model [37] are also consistent with its DFT reference data. It should be noted that the ML models do not use the DFT calculated static properties as training data to fit the potential. The ML potentials only fit the potential energy surface of the reference data. Some of the empirical interatomic potentials use the experimental or DFT calculated static properties data to fit the potential, but the calculated static properties are still not as good as the ML models.

3.2. Defect formation energies and threshold displacement energies

Defect production, clustering, and its evolution in semiconductor materials is a crucial aspect to explore radiation degradation in electronic devices. Therefore, in addition to static properties, the ML

Table 2

Comparison of static properties based on a range of interatomic potentials, DFT references, and experiments.

Model	$a(\text{\AA})$	$E_{Coh}(\text{eV})$	$C_{11}(\text{GPa})$	$C_{12}(\text{GPa})$	$C_{44}(\text{GPa})$
Experiment	5.43 [34]	-4.63 [34]	165.7 [35]	63.9 [35]	79.6 [35]
DFT [36]	5.47	-4.55	144	53	75
DFT [37]			153.3	56.3	72.2
DFT (This work)	5.47	-4.55	153.45	57.03	74.51
DeePMD + short-range interaction (This work)	5.48	-4.55	151.71	58.97	73.27
GAP [37]	5.46	-4.55	149.4	58.58	69.92
EDIP [38]	5.43	-4.65	171.76	65.47	72.53
MEAM [39]	5.43	-4.61	135.23	65.15	59.47
Purja Pun [40]	5.43	-4.63	172.14	65.04	80.59
SW [41]	5.43	-4.34	151.4	76.77	56.44
Tersoff [42]	5.43	-4.63	121.65	86.28	10.33
TersoffScr [43]	5.43	-4.63	165.89	66.02	76.57

potential must accurately reproduce defect-related properties, including defect formation energies and threshold displacement energies. For the defect formation energy, the formation energies of vacancy and dumbbell interstitial position are considered as shown in Table 3. The AIMD trajectories of vacancy and dumbbell interstitial were included in the dataset in Table 1. Therefore, the ML potential should be able to reproduce the defect formation energy. From the results in Table 3, we can see that our DFT calculations are consistent with another DFT work [37]. Meanwhile, the results of our trained ML potential are very close to our DFT reference data, and the results of another ML model are also close to its DFT reference data. The two ML potentials yield better defect formation energies than other empirical potentials.

The threshold displacement energy E_d is the minimum energy applied to an atom to form a stable Frenkel pair [44]. It is the most fundamental physical quantity that describes the resistance of a material to radiation. To calculate E_d , we ran a series of MD simulations by applying kinetic energy to a single PKA along specific crystal directions. The simulation system contained 1,000 atoms ($8 \times 8 \times 8$ units cells) and

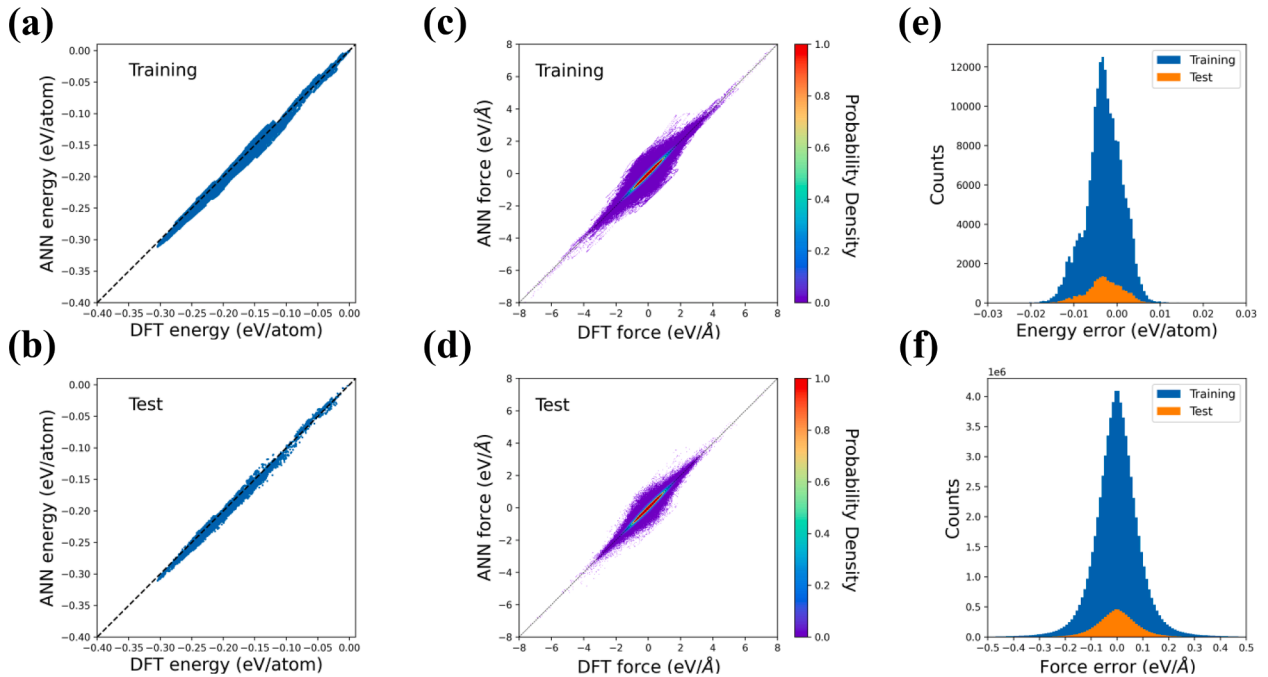


Fig. 1. Comparisons of the ML results with respect to the references DFT data. (a) and (b) show the comparisons of energies per atom for the training and test set, respectively. (c) and (d) exhibit force comparisons for the training and test set. 2D histogram plots are used to show the density distribution of errors. (e) and (f) display the error distributions for energies and forces for training and test set.

Table 3

Comparison of defect formation energies based on a range of interatomic potentials and DFT references.

Model	Defect formation energy (eV)	
	vacancy	dumbbell interstitial position
DFT [37]	3.67	3.66
DFT (This work)	3.59	3.59
DeePMD + short-range interaction (This work)	3.49	3.56
GAP [37]	3.61	3.58
EDIP [38]	3.66	3.92
MEAM [39]	3.26	3.00
Purja Pun [40]	3.86	3.50
SW [41]	2.64	4.87
Tersoff [42]	1.29	3.49
TersoffScr [43]	4.00	3.26

the length of the cubic simulation box is about 27 Å. Periodic boundary conditions were used and the time step of the simulations is set to 0.5 fs. The system was first relaxed by energy minimization and the simulation box is allowed to fluctuate to reach zero system pressure. Then, kinetic energy was applied to a single PKA at the center of the system and the system was evolved with NVE ensemble for 4 ps. The initial PKA kinetic energy was 5.0 eV and this energy was increased in steps of 0.5 eV in subsequent simulations until a stable Frenkel pair was formed. The minimum PKA kinetic energy to form a stable Frenkel pair is identified as threshold displacement energy. The Wigner-Seitz defect analysis [45] method was used to detect the Frenkel pairs. This method has proven to be effective in identifying crystalline defects in materials [46–47]. The results of the E_d calculations are shown in Table 4.

From Table 4, with our trained ML model, our calculated E_d is within the error bars of the DFT results [8]. It is also consistent with the experimental results [48–49]. The results of another ML model [37] are also consistent with the DFT results and the experiments. The two ML potentials yield better E_d than other empirical potentials.

3.3. Defect production and clustering with a single primary knock-on atom

The defect production and clustering of radiation-induced defects were studied by carrying out cascade simulations of a single 1.0 keV PKA

Table 4

Comparison of threshold displacement energy E_d based on a range of interatomic potentials, DFT references, and experiments.

Model	E_d (eV)		
	[100]	[111]	$\bar{[111]}$
Experiment	21.0 ± 2.0 [48]		12.9 ± 0.6 [49]
DFT [8]	19.5 ± 1.5	14.5 ± 1.5	12.5 ± 1.5
DeePMD + short-range interaction (This work)	18.5 ± 0.5	14.0 ± 0.5	12.0 ± 0.5
GAP + short-range interaction [20]	20.5 ± 0.5	14.5 ± 0.5	12.5 ± 0.5
EDIP [38]	11.0 ± 0.5	17.5 ± 0.5	10.0 ± 0.5
MEAM [39]	13.5 ± 0.5	9.5 ± 0.5	8.0 ± 0.5
Purja Pun [40]	9.0 ± 0.5	9.5 ± 0.5	9.5 ± 0.5
SW [41]	23.5 ± 0.5	20.5 ± 0.5	17.5 ± 0.5
Tersoff [42]	9.0 ± 0.5	9.0 ± 0.5	7.5 ± 0.5
TersoffScr [43]	16.5 ± 0.5	9.5 ± 0.5	8.0 ± 0.5

at 300 K by Large-scale Atomic/Molecular Massively Parallel Simulation (LAMMPS) Package [50]. A $22 \times 22 \times 22$ unit cell containing 85,184 atoms was constructed. Periodic boundary conditions were applied in all directions. The system was first equilibrated at zero external pressure and at a temperature of 300 K (NPT ensemble) for 2 ps. After that kinetic energy was added to one single PKA in the center of the structure to model primary damage. The directions of the velocities of the PKA were randomly-selected and 10 simulations were performed to average the fluctuations due to randomly-selected directions. The NVT ensemble was applied to the borders (5.4 Å in length) of the simulation cell to dissipate the excess heat by PKA to the interior part of the material. Electronic stopping [51–52] was applied to atoms with kinetic energies above 10 eV within the interior region as a non-local friction force. The simulations can be divided into two stages. During the initial stage, the number of defects rapidly increased up to ~ 0.2 ps. While in the following stage (~ 0.2 –2 ps), the number of defects decreased due to the recombination of vacancies and interstitials, which is often called a heat spike stage.

The number of defects was used to quantify the radiation effect of a single PKA. The PKA simulations were performed with ML potential and the Wigner-Seitz defect analysis [45] method was used to detect the defects. Two empirical potentials, namely, SW [41], and Tersoff [42] were also used to perform the same collision simulations for comparison purposes. The results are shown in Fig. 2.

From Fig. 2, the variation of interstitials with time shows the same trend based on the three different models. In the initial stage (~ 0.2 ps), the number of interstitials grows rapidly. Then, in the heat spike stage (~ 0.2 –2 ps), the number of interstitials starts to decrease, implying the “healing” of the defects. The maximum value of the number of interstitials appears in this phase. In the final stage (~ 2 ps-), the number of interstitials starts to stabilize, suggesting the formation of stable defects. Our calculations based on the SW potential agree with another paper [20], while the final number of interstitials based on the ML model in this work is between the SW and Tersoff potentials. Our ML potential predicts the resulting final number of interstitials to be 2–3 times higher than that of SW.

At the final stage of the collision simulation, stable defects are formed. Among these defects, there are not only isolated defects but also clusters of defects, which play an important role in the mechanical and electrical properties of the material [11]. Neighboring interstitials located within two times of lattice constant (about 10.8 Å) [53] are defined as interstitial clusters and the number of interstitials in each cluster refers to the size of clusters. The interstitial cluster size

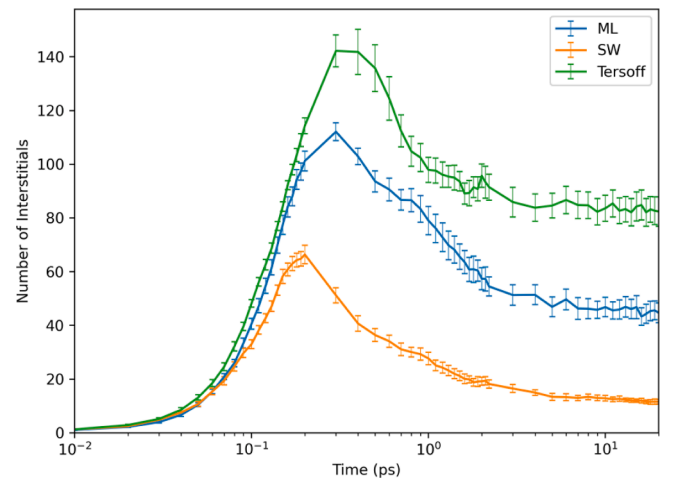


Fig. 2. Comparison of the number of interstitials vs time during collision simulations initiated by a 1 keV PKA. The number of interstitials was obtained by Wigner-Seitz analysis. The error bars referred to the standard errors of 10 simulations with randomly-selected PKA directions.

distributions based on the ML model in this work, SW, and Tersoff potentials are shown in Fig. 3.

From Fig. 3, for all three models, isolated interstitials are dominant. The ML potential predicts the largest proportion of isolated interstitials, followed by the SW potential, and finally the Tersoff potential. Meanwhile, the Tersoff potential predicts the largest proportion of large interstitial clusters (>30 interstitials within the cluster), followed by the ML potential, and almost none by the SW potential. Finally, the SW potential predicts the largest proportion of interstitial clusters containing 2–10 interstitials. Then comes the Tersoff and ML potential. Our calculation of the interstitial cluster size distribution based on the SW potential is consistent with another paper [20]. The three models predict very different clustering behavior.

To investigate the different clustering behavior of the three models, the evolution of the interstitials is studied. The interstitials in the heat spike and final stage are shown in Fig. 4. We can see that the ML potential produces interstitials in a larger area during the heat spike stage, eventually producing most isolated interstitials in a larger area and a few medium-sized and large interstitial clusters, which is consistent with the size distribution of interstitial clusters with ML potential in Fig. 3. The SW and Tersoff potentials produce interstitials only in a relatively small area during the heat spike stage, while only isolated and medium-sized interstitial clusters are eventually retained based on the SW potential, and Tersoff potential produced large interstitial clusters and a few medium and small interstitial clusters, which is also consistent with the size distribution of interstitial clusters with SW and Tersoff potentials in Fig. 3. By looking at the evolution of interstitials, we can see that the major difference between the ML potential and the empirical potentials is that with ML potential PKA has an impact over a larger spatial area. In contrast, based on the empirical potentials, the effect of PKA is more localized. This is the essential reason for the different clustering behavior of different potential models.

3.4. Effects of multiple primary knock-on atoms excited in sequence

Chapter 3.3 analyzes the production, clustering, and evolution of defects based on ML potential using a single PKA. However, more than one PKA may exist at the same time. Previous study has shown that within the preexisting amorphous region, both interstitials and vacancies move towards the center of the region when it resolidifies and recombine or cluster there [63]. The cumulative effect of multiple PKAs in a material is significantly dependent on the degree of overlap between the influence regions of PKAs. In this paper, we limited our investigation

to the scenario that the material is first damaged by a portion of PKAs and then followed by other PKAs. We studied the effect of defect production and evolution when multiple PKAs are excited sequentially. A larger system containing 117,600 atoms was used to study the effect of multiple PKAs sequentially excited. Periodic boundary conditions were applied in all directions. One atom was selected at a time in a small region in the upper part of the model as a PKA and a random but downward directed kinetic energy is applied. The successive PKA was excited after the number of defects generated by the previous PKA remained unchanged (after 20 ps). Twelve 1 keV PKAs were sequentially excited in twelve successive simulations. Again, the NVT ensemble was applied to the borders (5.4 Å in length) of the simulation cell to dissipate the excess heat by PKA to the interior part of the material. Electronic stopping [51–52] was applied to atoms with kinetic energies above 10 eV within the interior region as a non-local friction force.

The number of defects produced by the sequential PKAs is shown in Fig. 5. The PKA simulations were performed with ML potential and the Ovito software [54–55] was used to detect atoms that are not arranged in a diamond lattice (coordination defects). From Fig. 5, it can be seen that the increase in the number of coordination defects produced by the first four PKAs is close to the multiples of defects produced by a single PKA. However, starting from the fifth PKA, the increase in the number of coordination defects per PKA is 36 % less than that produced by a single PKA. This indicates that when PKA excitation is sequential, the defects produced by the previous PKA affect the microstructure so that the later PKA produces fewer defects. PKAs excited in a sequence should be considered in the radiation damage research.

To investigate why the first several PKAs produced more defects and the following PKAs produced fewer defects. We first checked the defects produced by the first 4 PKAs and that produced by all 12 PKAs. We found that the defects produced by the first 4 PKAs constituted a spherical region containing many amorphous local structures, as shown in Fig. 6 (a). For the following PKAs, the PKA first passes through the amorphous region. Then defects are formed at a deeper place in the structure. Fig. 6 (b) shows the migration paths of the energetic atoms during the fifth PKA activation. The PKA first crosses through the amorphous region and then generated new defects outside the amorphous region. Fig. 6(c) shows the final configuration after all 12 PKA activations. We further investigated the evolution of defects during the sequential PKA simulations. First, we calculated the average maximum atomic kinetic energy with time during the simulation, as shown in Fig. 6(d). We found that the average maximum atomic kinetic energy decreases more rapidly for the first four PKA excitations. For the next eight PKA excitations, the average maximum kinetic energy decreases more slowly at the initial stage. This is attributed to the fact that for the first four PKA excitations, the PKAs hit the diamond structure. However, for the next eight PKA excitations, the PKA first hit the amorphous structure. The diamond structure has a higher modulus than the amorphous structure [56]. For the simulation of the next 8 PKA excitations, part of the kinetic energy of the PKAs is dissipated when they pass through the amorphous structure. When the PKA passes through the amorphous structure and starts to generate defects in the diamond structure, the kinetic energy of the last eight PKAs is smaller than that of the first four PKAs, thus the number of newly generated defects is smaller, as shown in Fig. 6(e).

4. Conclusion

In this paper, we constructed a more efficient ML potential with DFT accuracy that can be used to describe radiation damage in silicon. We constructed a large DFT dataset containing conformations related to static properties, point defects, fluid and amorphous phases, and radiation damage. Then, to describe the short-range interactions which are critical in radiation damage, we calculated the short-range interactions between silicon atoms using the CCSD(T) method. Finally, we use ML methods to learn the difference between the DFT and the short-range interaction. The final model is made up of a ML potential and a short-

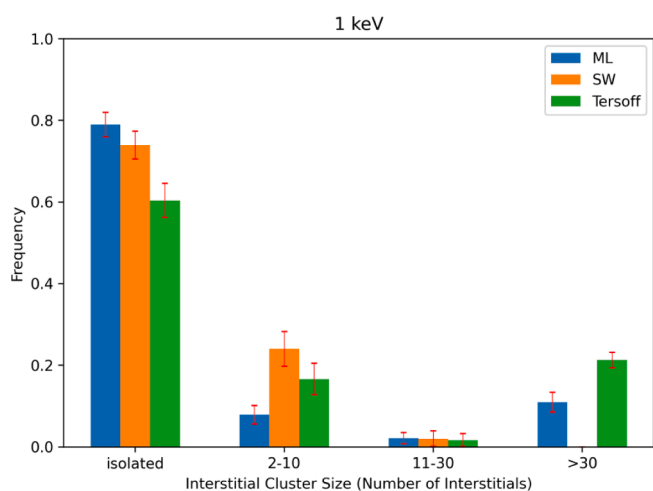


Fig. 3. Size distribution of interstitial clusters produced in one cascade initiated by a 1 keV PKA. The frequency has been normalized so the sum of the size distribution is one. The error bars referred to the standard errors of 10 simulations with randomly-selected PKA directions.



Fig. 4. Snapshots of interstitials in 1 keV PKA collision simulations at the heat spike and final stage. The ML potential produces interstitials in a larger area during the heat spike stage, eventually producing most isolated interstitials in a larger area and a few medium-sized and large interstitial clusters, which is consistent with the size distribution of interstitial clusters with ML potential in Fig. 3. The SW and Tersoff potentials produce interstitials only in a relatively small area during the heat spike stage, while only isolated and medium-sized interstitial clusters are eventually retained based on the SW potential, and Tersoff potential produced large interstitial clusters and a few medium and small interstitial clusters, which is also consistent with the size distribution of interstitial clusters with SW and Tersoff potentials in Fig. 3. Interstitials were obtained by Wigner-Seitz analysis.

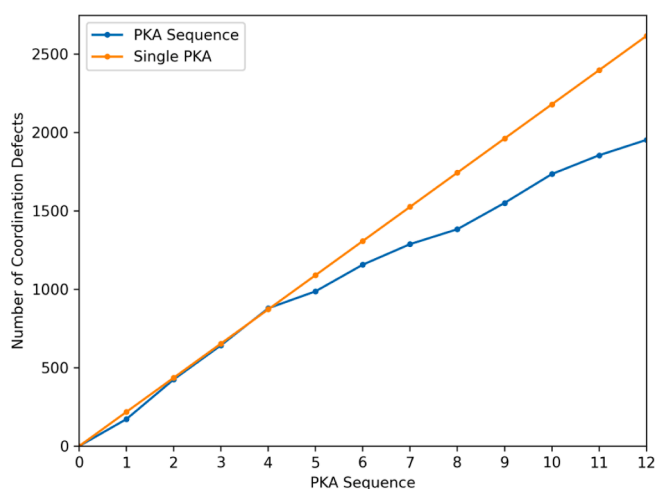


Fig. 5. Total coordination defects produced by sequential PKAs (blue line). The orange line represents the coordination defects produced by single PKA times the total number of sequential PKAs. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

range interaction component.

After obtaining the ML model, we calculate the static properties, defect formation energy, and threshold displacement energy of

crystalline silicon, and the results agree well with the DFT reference data and are better than the empirical potentials. This is a verification of the accuracy of the ML potential. Then, based on the ML potential, we investigated defect production, clustering, and evolution using a single PKA excitation. We found that based on the empirical potentials, the effect of PKA is more localized. In contrast, based on the ML potential, it has an impact over a larger spatial area. The defects generated by the ML potential are still dominated by isolated defects, and there are also some medium and large defect clusters. Finally, we studied the sequential excitation of the PKAs. We found that the first several PKAs produce an amorphous region, while the later PKAs first pass through the amorphous regions and then produce defects at greater depth. Due to the dissipation of kinetic energy while going through the amorphous region, the later excited PKAs produces fewer defects. Therefore, it is necessary to consider the case of sequentially PKA excitations in atomic simulations for radiation damage.

CRediT authorship contribution statement

Hongwei Niu: Investigation, Writing – original draft. **Junqing Zhao:** Supervision. **Huyang Li:** Investigation. **Yi Sun:** Conceptualization, Methodology, Supervision. **Jae Hyun Park:** Supervision. **Yuhang Jing:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Weiqi Li:** Supervision. **Jianqun Yang:** Supervision. **Xingji Li:** Conceptualization, Methodology, Supervision.

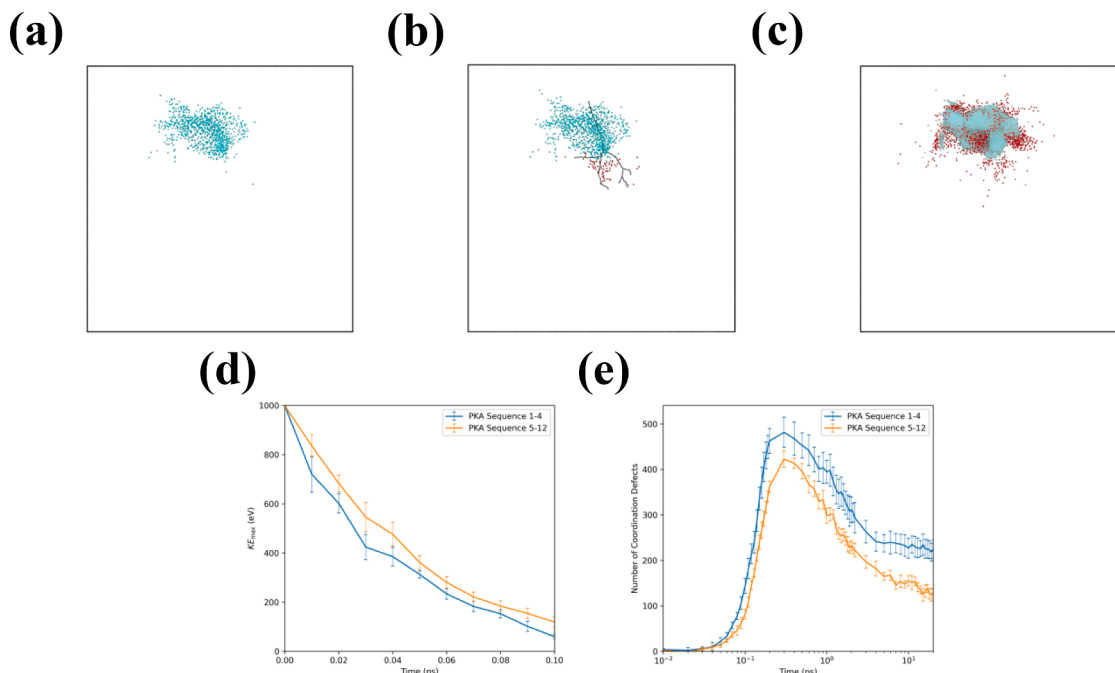


Fig. 6. (a) The spherical region of defects (in blue) produced by the first 4 PKAs. (b) The fifth PKA passes through the spherical region of defects (in blue) and generated new defects at greater depth (in red). The black lines are the migration paths of the energetic atoms during the fifth PKA activation (c) PKAs passing through the spherical region (in blue) of defects by the following 8 PKAs. Newly generated defects are in red. (d) The average maximum atomic kinetic energy of the first 4 PKAs and the following 8 PKAs. The error bars refer to the standard errors of the first 4 PKA simulations and the following 8 PKA simulations. (e) Number of newly generated coordination defects produced by the first 4 PKAs and the following 8 PKAs. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Data availability

The raw/processed data required to reproduce these findings are available to download from https://github.com/hongweiniu/primary_radiation_damage_of_silicon.

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.commatsci.2022.111970>.

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