On the detection and classification of material defects in crystalline solids after energetic particle impact simulations

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Abstract

We present a fingerprint-like method to analyze material defects after energetic particle irradiation by computing a rotation invariant descriptor vector for each atom of a given sample. For ordered solids this new method is easy to use, does not require extreme computational resources, and is largely independent of the sample material and sample temperature. As illustration we applied the method to molecular dynamics simulations of deuterated and pristine tungsten lattices at 300 K using a primary knock-on atom (PKA) of 1 keV with different velocity directions to emulate a neutron bombardment process. The number of W atoms, that are affected after the collision cascade, have been quantified with the presented approach. At first atoms at regular lattice positions as well as common defect types like interstitials and vacancies have been identified using precomputed descriptor vectors. A principal component analysis (PCA) is used to identify previously overlooked defect types and to derive the corresponding local atomic structure. A comparison of the irradiation effects for deuterated and pristine tungsten samples revealed that deuterated samples exhibit consistently more defects than pristine ones.

Keywords: Tungsten, MD simulations, descriptor vectors, PCA, Deuterium

1. Introduction

Plasma facing materials (PFMs) in the next generation of nuclear reactors are subjected to extreme conditions due to the direct plasma interaction [1]. The induced material damage has to be analyzed by the identification, classification, and quantification of lattice defects. This is a complex analysis, complicated by potentially unforeseen atomic configurations after irradiation [2, 3].

Material defects on an atomic scale are often investigated using Voronoi tessellation and the analysis of Wigner-Seitz cell volumes to identify common defects like vacancies and interstitials [4, 5]. Also a description of the topology of individual grains, bubbles, and cells in three-dimensional polycrystals [6] is sometimes provided. However, non-standard and unexpected material defects provide a challenge to the existing methods. A robust method is needed to classify and quantify damage e.g. after the impact of high-energy particles from fusion reactions. For this reason, the International Atomic Energy Agency set out a competition [7] to detect material defects in different metals. In this paper, we present a novel method to identify, classify, and quantify material defects regardless of the sample composition and temperature, and impact energy. Our numerical procedure is able to identify new material defect types other than the common ones like interstitial sites and vacancies. It is based on the Smooth Overlap of Atomic positions method [8, 9] (SOAP) to compute the descriptor vector of individual atoms of a given sample, which describes their particular physical environment. SOAP is implemented in the QUantum mechanics and Interatomic Potentials package (QUIP) [10] that is a collection of software tools with a variety of interatomic potentials and tight binding packages. As an example, we analyze the defects in single-crystal tungsten after the interaction with a 1 keV of primary knock-on atom, due to tungsten is an important candidate as PFM for a fusion device [11]. It is also of interest to investigate the effect of deuterium decoration of W on the material defects formation [12, 13].

This paper is organized as follows: In section 2 we present the computational methods that are utilized in our research work, followed by a description of the preparation of the numerical cells used to perform MD simulations of a deuterated and pristine W lattice for 1 keV of primary knock-on atoms. We also briefly describe the computation of the descriptor vectors that define our crystal defects classifications, quantification and further results for the different systems are shown in the section 3. Finally, in section 4 we discuss our results and provide concluding remarks.

2. Theory

2.1. Descriptor vectors

An ideal tool for the identification of distortions in a crystalline sample should be sensitive to changes in a local environment, but at the same time insensitive to ”global” changes like rotation of the sample or permutations of its atoms. In addition, it should allow for some kind of calibration, e.g. not counting thermal fluctuations as defects. Furthermore, the possibility of a
probabilistic interpretation instead of a binary yes/no-response would be beneficial. Most of the suggested approaches so far fail in one or more of these aspects or can only be applied to specific kinds of defects. For example, just using the Cartesian coordinates of the neighboring atoms would result in a false positive identification of a changed environment under simple rotation. Similarly the use of a vector of nearest-neighbor distances (which are invariant under rotation) has deficiencies, where very different atomic configurations can result in the same vector [8].

In this section, we outline the approach to compute the reference descriptor vector based on the QUantum mechanics and Inter-atomic Potentials package (QUIP) [10], that can be considered as a footprint of the local environment of an atom [8,9].

2.2. Descriptor vector computation

The analysis of the defects of a given material structure is started by first computing the descriptor vectors (DVs) of all the atoms in the sample. The DV of the $i$-th atom of the sample, $\vec{\xi}^i$ (defined below), is a component vector and a representation of the atomic neighborhood within some cutoff-radius. The representation is based on an expansion of the local neighborhood of an atom in a product of spherical harmonic functions and radial basis functions. It has been shown in Ref. [8] that this representation exhibits the desired properties, i.e. that the descriptor vector introduced below is invariant to rotation, reflection, translation, and permutation of atoms of the same species, but sensitive to small changes in the local atomic environment [8]. In order to compute the DVs, let us define the atomic environment of an $i$-th atom by a sum of truncated Gaussian density functions as [8],

$$\rho^i(\vec{r}) = \sum_{i'j}^{\text{neigh}} \exp \left( -\frac{|\vec{r} - \vec{r}^{ij}|^2}{2\sigma^2_{\text{atom}}} \right) f_{\text{cut}} \left( \vec{r}^{ij} \right),$$

where $\vec{r}^{ij}$ denotes the difference vector between the atom positions $i$ and $j$. $\sigma^2_{\text{atom}}$ denotes the broadening of the atomic position, this parameter is set according to the lattice constant of the sample and takes into account the thermal motion. Finally, $f_{\text{cut}} \left( \vec{r}^{ij} \right)$ is a smooth cutoff function, which is required to limit the considered neighborhood of an atom. Then, $\rho^i(\vec{r})$ can then be expressed in a basis set, as obtained in [9], in terms of expansion coefficients, $c_{nlm}$:

$$\rho^i(\vec{r}) = \sum_{n,lm} c_{nlm} Y_{nlm}(\hat{\vec{r}}) \cdot$$

with $\hat{\vec{r}} = \vec{r}/|\vec{r}|$ as unit-vector in the direction of $\vec{r}$. The $c_{n0m} = (g_{n0m}|\rho^i)$ are the expansion coefficients that corresponds to the $i$-th atom in the lattice and $g_{n0m}(\vec{r})$ is a set of orthonormal radial basis functions $(g_{n0m}(\vec{r}) | g_{n0m}(\vec{r}) = \delta_{nm})$, and $Y_{nlm}(\hat{\vec{r}})$ are the spherical harmonics with the atom positions projected onto the unit-sphere. The inner product $\langle f | g \rangle$ used above is given by the integral over the surface of a unit-sphere

$$\langle f | g \rangle = \int f^\ast(\vec{r}) \cdot g(\vec{r}) \cdot d\Omega(\vec{r}).$$

To achieve invariance against rotations of the local environment of atom $i$, Eq. [3] needs to be averaged over all possible rotations. After some algebra [9], the desired rotation invariant result can be expressed by the multiplication of the $c_{n0m}$ with the complex conjugate coefficient $c_{n0m}^\ast$, summed over all $m$. This resembles some similarity to the power spectrum of Fourier coefficients. Thus the components of the DV of the $i$-th atom, $\vec{\xi}^i$, are given by [9]

$$\vec{\xi}^i = \left( \sum_{m} c_{n0m} Y_{nlm} \right)_{n,n',f},$$

where each component of the vector corresponds to one of the index triplets $\{n,n',f\}$. In the following we will use the normalized vector $\vec{q}^i$ as descriptor vector (DV) for the local environment of atom $i$: $\vec{q}^i = \vec{\xi}^i/|\vec{\xi}^i|$. We compute the DVs of the deuterated and pristine sample using the QUIP package with its python interface [10].

2.3. Calibration of the descriptor vectors

Using the sequence of Eq. [10] to [3] for each atom $i$ of the sample a DV, $\vec{q}^i$, can be computed. Depending on the choice of the expansion orders in Eq. [2] for the spherical harmonics and the radial basis functions the number of components of $\vec{q}^i$ varies. Here, we used $n = 4$, and $l = 4$ (with $-l \leq m \leq l$) which yields a DV with $k = 51 \times 50 \times 50$ components. The difference of two local environments of atom $i$ and atom $j$ can then be obtained by computing the distance $d$ between the two DVs, $d = d(\vec{q}^i, \vec{q}^j)$, which for the standard euclidean measure is defined as $d^2 = \sum_{k} (q^i_k - q^j_k)^2$, where $k$ is used as a component index. However, giving all components the same weight may not always be appropriate because some of them may be more fluctuating than others - although the standard euclidean distance works quite well in most cases that we have looked at. In order to select an appropriate measure two compare the DVs, we used a MD simulation to generate a thermalized tungsten bcc lattice at $T = 300$ K (see section 3) without defects, and computed the DV for all the atoms. This group of DVs for a defect free and thermalized environment has been used to compute a mean reference descriptor vector as: $\bar{v}(T) = \frac{1}{N} \sum_{i} \vec{q}^i(T)$, as well as the associated covariance matrix $\Sigma(T)$, where especially the covariance matrix depends on the sample temperature. Following this, the distance difference of the DVs of the thermalized environment from the mean bcc-lattice can be computed using the Mahalanobis distance [14]

$$d^M(T)(\vec{q}, \vec{v}(T) | \Sigma) = \sqrt{(\vec{q} - \vec{v}(T))^\ast \Sigma^{-1}(\vec{q} - \vec{v}(T))}$$

This yields to a distance distribution of the DVs for a thermalized and defect-free lattice, which sets the scale to judge
\[1\] Besides the zeroth-component there are for each $l \in \{0,4\}$ 10 ordered pairs $(n,n')$, i.e. $(1,1), (1,2), (1,3), (1,4), (2,2), (2,3), (2,4), (3,3), (3,4), (4,4)$, yielding a DV with $50 + 1$ components.
if an unexpected large distortion of the local environments is present. For the computation of the reference DVs for other common types of point defects a similar approach is chosen: A small numerical cell containing the defect of interest (e.g., an interstitial) was prepared and the computation of the DVs of the atoms defines the fingerprint for this specific atomic environment (see section 3).

2.4. Probability of being a lattice atom

The use of the Mahalanobis distance $d^{M}(T)$ allows a straightforward probabilistic interpretation of the distance. Given that the distance distribution $d^{M}(T)$ of a thermalized sample is close to a Gaussian distribution, then the probability, $P(q^{i} \mid \bar{v}(T))$, of an atom $i$ being in a locally undistorted lattice can be computed using

$$
P(q^{i} \mid \bar{v}(T)) = P_{0} \exp \left[-\frac{1}{2} d^{M}(T)^{2} \right],
$$

$$
d^{M}(T)^{2} = (q^{i} - \bar{v}(T))^{T} \Sigma^{-1}(T) (q^{i} - \bar{v}(T)),
$$

where $P_{0}$ is the normalization factor and $q^{i}$ is the DV of atom $i$. For our present analysis we only use the diagonal elements of the covariance matrix, i.e., set $\Sigma_{ij} = 0$ for $i \neq j$. This yields a quantitative measure for identifying/selecting defects, even for samples at different temperatures, where fixed criteria (e.g., maximum displacement) could easily fail.

2.5. MD simulations

The simulation box is initially prepared as a pristine single-crystalline W lattice sample with 48778 W atoms based on a bcc unit cell. The box has a dimension of $(28a, 28a, 28a)$ with $a = 0.316$ nm as the W lattice constant [15], for a sample temperature of 300K due to the majority of the experiments of tungsten damaging are done at room temperature [16, 17]. The deuterated W lattice is created by introducing 0.1 at% D (53 deuterium atoms) randomly distributed in the whole sample at tetrahedral interstitial positions, Fig. [1]. All the samples are first energy optimized and subsequently thermalized to 300 K using a Langevin thermostat with a time constant of 100 fs [18, 19].

MD simulations are performed by assigning an impact energy of 1 keV to a W atom, thus acting as a primary-knock on atom (PKA). Its initial position is located at the center of the sample. We consider ten velocity directions: $\langle 001 \rangle$, $\langle 101 \rangle$, $\langle 111 \rangle$, and 7 cases for $\langle r_{1}r_{2}r_{3} \rangle$ where $r_{i}$ are random numbers uniformly distributed in an interval of $[0, 1]$. A velocity-Verlet integration algorithm is applied to model the collision dynamics, which lasts for 10 ps with a time step of $\Delta t = 1 \times 10^{-3}$ ps. After that time the dynamics of the collision cascade has stopped and subsequent diffusive relaxation process are thus missed, but their proper consideration would in any case exceed the time scale accessible for MD method. The simulations were performed on a desktop computer using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS code) [20] with the reactive inter-atomic potential for the ternary system W-C-H of Juslin et al. [21], which is based on an analytical bond-order scheme. This potential has been used to study neutron damage in poly-crystalline tungsten [22], trapping and dissociation processes of H in tungsten vacancies [23], and cumulative bombardment of low energetic H atom of W samples for several crystal orientations [24].

3. Results

Initially, we computed reference DVs for the thermalized defect free bcc lattices for two temperatures, and two of the most common defects, interstitial atoms and W atoms next to single vacancy. For that, in order to compute the main reference vectors, we used a sample of 432 W atoms in a fully periodic box with lateral dimensions of $(5a, 5a, 5a)$, based on a bcc unit cell of dimension $a = 0.316$ nm. A tungsten atom is removed from this sample to calculate the reference vector for atoms next to a single vacancy. For the interstitial case, a W atom is introduced into the lattice at a tetrahedral interstitial site.

All the descriptor vectors are computed by QUIP with the SOAP descriptors package. We chose the following parameters: $l_{\text{max}} = 4$, $n_{\text{max}} = 4$, and a $r_{ij}$ cutoff of 0.31 nm, slightly smaller than the unit cell of W with its lattice constant of 0.316 nm at 0K. It is worth mentioning that with this choice all 8 nearest neighbours in the tungsten lattice are captured, as well as potential interstitials. A larger cut-off radius is possible but some preliminary tests indicated an increased tendency of the algorithm towards false positives. Reference DVs $\bar{v}(T)$ that take into account thermal displacement are calculated for thermalized bcc samples at 300 and 600 K. These vectors are subsequently used to compute the probability for each atom to be in an undistorted environment using Eq. [6]. Atoms which exhibit a very low probability are either potential defects (e.g. interstitial) or close to a defect (e.g. a vacancy). The computation
of the reference DVs takes \( \sim 7 \) sec. for this particular size of the W sample on a desktop computer. In figure 2 we show the reference DVs of three standard point defects, at 0 K, as a function of their components. A lattice W atom (without thermal fluctuations), a tungsten atom at an interstitial site and an atom next to a single vacancy. It can be seen that the structure of the non-zero components of the DVs for a tungsten atom in an ideal environment differs significantly from the structure of the DV of a W atom in a tetrahedral interstitial position. In contrast, the intensity pattern is close for atoms next to a vacancy compared to the DV of atoms in an undistorted lattice.

In Fig. 3a) and b) we present the DVs of all atom of the defect-free and thermalized BCC samples at 300 and 600 K, respectively. To provide a better visualization of the thermal descriptor vectors, we have used a bee-swarm plot: for component indices with intensities (> 0), the position is given by \( x + 0.5 \cdot x' \), where \( x' \) is a random number uniformly distributed in the interval \([-1, 1]\). It is observed that the distance in vector space at 300 K still allows a good identification, because regular lattice atoms which are close in one component may differ in another component, Fig. 3a). The situation is different for a sample at 600 K, as it is shown in Fig. 3b). Here the overlap is too strong to allow for a reliable identification of atoms next to a single vacancy because of DV method is centered around atoms. Thus, the DVs for the W sample at 600 K have - as to be expected - a bigger variance than those at 300 K. Nevertheless, the mean of the components of the DVs at 300 and 600 K is very close to the components of the lattice DV at 0 K. It is therefore less suited for the identification of larger voids (because there are no atoms). In practice these kinds of defects are more easily detected using Voronoi-based methods, e.g. by computing the largest empty sphere around a given location. In our work, we calculate the number of vacancies in the damaged W sample by identifying their spatial location by a k-d-tree algorithm, explained in Appendix A. Thus, a manual inspection of the atoms next to an identified single vacancy is recommended to visualize all W atoms around these kinds of defects, which helps to avoid double-counting of defects (e.g. 8 tungsten atoms next to a single vacancy are in a distorted lattice position - but this should be counted as 1 defect site only - not as 8). The lattice W atom reference descriptor vector, the interstitial and the ‘atom next to a vacancy’-reference descriptor vectors as well as their geometry are provided in the supplementary material (Appendix B).

In Fig. 4a), we show the probability of an atom to be considered at a distorted site, i.e. \( 1 - P(q) \), as a function of the distance difference by using the DVs for a thermalized W sample to 300 (x-symbol) and 600 K (+-symbol) as a reference. Already the histogram of the distance from the DV for an atom of a bcc-lattice position of a thermalized tungsten lattice provides a good hint for a suitable distance value for the defect threshold. Here, a manual inspection of the local atom environment for different threshold values provides a visualization of the local disorder around the atom with the highest probability to be a point defect, which can be done by using an open visualization tool like OVITO [25] for example. Therefore, we noticed that a threshold set at 0.24 gives the number of atoms with the highest probability, which are frequently associated with the total number of Frenkel pairs. Here, a threshold set at 0.15 shows us the distorted regions around the point defects.

In Fig. 4b) we present the results for the euclidean distance between the mean reference descriptor vector \( \bar{v}(300 \text{ K}) \) for the thermalized sample at 300 K with the DVs of each atom in the damaged pristine W sample after interaction with the 1 keV PKA. W atoms that have a distance larger than the threshold set to 0.15 are classified as atoms being in a distorted environment. The subsequent identification of these atoms (e.g. as being close to a vacancy site or being an interstitial atom) is done by calculating the distance difference between the atomic descriptor vector and the corresponding reference DVs of the different types of defects.
3.1. Defect classification in a pristine W sample

For the classification of the type of the distorted environment the descriptor vectors of the previously identified atoms are compared with reference descriptor vectors of a number of common defect structures (like an interstitial atom). If the distance of the atom descriptor vector and the reference vector of the defect is below some threshold, the atom is correspondingly labelled.

In Fig. 5 we show the point defects of the pristine W sample, for a PKA velocity direction of ⟨001⟩. W atoms that are in a defect-free lattice environment have been removed. Interstitial atoms in the W sample are represented by blue spheres, and the atoms in their distorted local region are shown in light-blue sphere. The formation of a dumbbell defect, where two atoms share a lattice site [26], is observed at the right hand side of the picture and can be easily identified by the DV for an interstitial site. W atoms next to a vacancy and type-a defects atoms are presented as red and green spheres, respectively. All these atoms are identified by our DV based method. In addition vacancy sites, black spheres, are formed along the trajectory path of the primary projectile, and are identified by our method. A black arrow is added to indicate the ⟨001⟩ velocity direction. This figure was created by using the Visual Molecular Dynamics (VMD) tools [27].

The approach presented so far does only account for known types of defects, unexpected/unforeseen structures can be also found. A principle component analysis of the obtained DVs in the various bombardment simulations (see sec. 3.2 for more details) revealed that besides interstitials and single vacancies a third type of a local defect structure was consistently present, which was subsequently labelled as type-a defect. In the following, the structure of this type-a defect was elucidated by manual inspection of the involved atoms and verified by computing again the DV of the idealized structure. In Fig. 6, eight bcc unit cells are used to illustrate the atomic arrangement of the type-a W defect (green spheres), as a W atom between two vacancies (vacancies are represented by transparent pink spheres).

We note that the W atom can be found either between two consecutive vacancies or in a split vacancy. It is worth mentioning that the DV of a type-a W atom can be stored as a new defect reference vector and used in further analysis. In addition, further formation energy calculations can be used to investigate whether or not this kind of defect is indeed stable or perhaps an unexpected artifact of the MD potential used.

In order to relate the identified atoms which are in a distorted environment (in our case on average 35 W atoms at a threshold of 0.15) to specific classes of defects like Frenkel pairs, a subse-
Table 1: Defect quantification of pristine W samples, after collision cascade, at different velocity directions. \( r_i \) with \( i = 1, 2, 3 \) are uniform random numbers in the interval \([0,1]\). The number of atoms at the highest probability are reported in parentheses.

| Vel. dir. | Inter. | Next to vac. (Vac.) | Type-a | Total |
|-----------|--------|---------------------|--------|-------|
| (001)     | 24 (3) | 7 (3)               | 2      | 33    |
| (011)     | 28 (2) | 6 (2)               | 2      | 36    |
| (111)     | 24 (2) | 7 (2)               | 4      | 35    |
| (\(r_1r_2r_3\)) | 25 (3) | 7 (3)               | 2      | 34    |
| Average   | 25 (3) | 7 (3)               | 3      | 35    |

sequent analysis step is necessary. In the ideal case (sample temperature at 0K) the 8 W atoms which surround a single vacancy identified by our method are all counted as being in a single empty lattice site environment. However, our method identifies \(2-5\) W atoms around a vacancy due to thermal motion. Thus, by applying this post-processing to the case of the pristine W sample, we obtain an average number of 3 Frenkel pairs and the correct visualization of the W atoms around the vacancies. In Table 1 we list the total number of W atoms that are identified as being located at imperfect sites; the percentages of interstitial sites, W atoms next to a vacancy, and W atoms between two vacancies (type-a) for different velocity directions at a threshold of 0.15, while the number of atoms at the highest probability are reported in parentheses at a threshold of 0.24. In the same table, we report the average of the obtained results for the seven MD simulations, that are performed for random velocity directions. The total number of defects for each velocity direction are: \(36 (3), 34 (3), 36 (3), 33 (3), 35 (3), 31 (2), 33 (2)\). The majority of the sample defects, found by our method, are interstitial sites with several atoms in their local atomic environment. A minority of the defects is consistently appearing as type-a. A comparison with the results of A. E. Sand et al. [28] and K. Nordlund et al. [29] who obtained 3 Frenkel pairs shows good agreement at 1 keV of PKA. The result can be also compared to the expected number of Frenkel pairs (FP) as given by Setyawan et al. [13]: \(\text{FP} = 0.49 \left( E_{\text{PKA}}/E_d \right)^{0.74} = 2.24 \) at a sample temperature of 300 K with \(E_d = 128\) eV. In the supplementary material, we provide a video to visualize the sample defects under different angles.

3.2. Defect classification of deuterated W samples

The defect analysis of multi-component samples (here with W and D) can in principle follow two different approaches. In the first approach one tries to get along with the DVs for tungsten only, indirectly accounting for the displacement due to presence of interstitial D atoms. The displacement has to be taken into account because it happens also in defect-free systems. Initially, the reference DVs for the tungsten atoms surrounding an interstitial deuterium atom are computed by keeping the W atoms positions fixed after (virtually) removing the interstitial D atoms. This allows to remain the computation of the DV identical to the mono-species case. However, this approach will fail for larger interstitial atoms because the atom displacement becomes too large, and will be cumbersome for systems with more than two species. We identify the DVs for a W lattice atom and a W atom next to a D using a principal component analysis (PCA). As shown in Fig. 7a) all the projected DV (referring to defect-free tungsten with interstitial D atom) show a considerable scatter. The reason for this is simple: the interstitial deuterium displaces the W atoms from their equilibrium bcc-lattice positions, which complicates the subsequent identification of defects and can result in false classifications. The quantification and classification of the point defects, found in the damaged sample by this first approach, are reported in the table. Please note that a number of W atoms next to a vacancy turned out to be incorrectly labelled by this first approach, which we therefore do not recommend - despite its simplicity.

Since the first approach does not provide viable results, the information about the D atoms needs to be included in the computation of new multi-component DVs for W atoms next to a
D atom. This second approach is straightforward but increases the number of basis functions and the nature of the reference DVs. In Fig. 7b, we show the PCA results of the DVs of the damaged-free and deuterated W sample, where the first and second principal components capture 49.03% and 31.90% of the variance of the data. We notice that the cluster around the origin is related to the DVs of a lattice W atom in a defect-free and pristine environment. There are two clearly separated clusters with centers at the points (0.021, 0.08) and (0.055, 0.13), that are associated to W atoms next to the D atom as a first and second neighbors, respectively. Closer inspection reveals that W atoms that are next to an octahedral interstitial D atom can be assigned to a first-neighbor DV, and W atoms next to a tetrahedral D atom are identified by a second-neighbor DV.

In Fig. 7b), we show the DVs for a lattice W atom (× symbol); for a W atom next to a D atom, where (a) are first neighbors (□) and (b) are considered as second neighbors (○); and for a D atom (●). Due to the inclusion of the D atom in the DVs computation, more components are needed to describe the D atom in the local neighborhood of the W atoms resulting in DVs with \( k = 181 \) (0…180) components. To be consistent with the number of components, the reference DVs for the pristine W case are extended to 181 components by adding additional vector components with their value set to zero. We then used these new DVs to identify W atoms next to D atoms in deuterated W samples. An example for that is given in Fig. 1.

To analyze the deuterated and damaged W sample, we first identified the W atoms that are in interstitial sites. Following the approach described in the previous section, a histogram is computed for the distances of the atoms DVs to the reference DV for an interstitial atom \( \vec{v} (0 \text{ K}) \), and shown in Fig. 8b). A comparison of the same quantity for the pristine case is given in Fig. 8a). The number of interstitial atoms or atoms next to an interstitial is larger for the pristine case. However, the picture changes if all atoms in a distorted environment are considered. As is detailed below, the additional deuterium in the sample changes the relative occurrence of the observed defect-types.

We tabulated the number of defects in the deuterated W sample as a function of the PKA velocity direction in the Tab. 2 where total number of Frenkel pair is reported in parentheses and the total number of defects for the random velocity directions are \{35(1), 36(1), 36(1), 35(1), 31(1), 35(1)\}. Comparing the results of Sec. 3.1 (pristine W) with the present data for a sample with D, the results reveal a small but persistent difference in the total number of atoms in a distorted environment between the two samples.

We provide a movie of the classification of defects for a deuterated and damaged W sample at a (001) velocity direction in the supplementary material. A new type of defect can be identified for the deuterated case after the collision cascade: Almost all (> 80%) of the displaced tungsten atoms are accompanied by a deuterium in the near vicinity (c.f. table 2 approach 2). In addition, for all simulations except one the collision cascades yield a larger number of defects in the deuterated sample compared to the pristine one. Although the statistics is not very good, such a results should happen by chance in less than 1% of all cases if the defect probability is the same for both cases.

We conclude that the effect of the presence of deuterium in the W sample is modest but that on average the number of point defects is larger than in the pristine one. This could point towards a stabilization of defects by hydrogen - but here many more simulations are needed to substantiate this hypothesis.

### Table 2: Defect quantification of a deuterated damaged W samples for different PKA velocity directions. Total number of Frenkel pairs are reported in parentheses.

| Vel. dir. | Inter. | Vac. | New type | Total |
|----------|--------|------|----------|-------|
|          |        |      |          |       |
| (001)    | 3 (1)  | 5 (1)| 35       | 43    |
| (011)    | 3 (1)  | 5 (1)| 36       | 44    |
| (111)    | 3 (1)  | 5 (1)| 33       | 41    |
| (r_1/r_2) | 3 (1) | 5 (1)| 37       | 45    |
| Average  | 3 (1)  | 5 (1)| 35       | 43    |

|          |        |      |          |       |
|----------|--------|------|----------|-------|
| (001)    | 3 (1)  | 1 (1)| 31       | 35    |
| (011)    | 3 (1)  | 1 (1)| 33       | 37    |
| (111)    | 3 (1)  | 1 (1)| 30       | 34    |
| (r_1/r_2) | 3 (1) | 1 (1)| 31       | 35    |
| Average  | 3 (1)  | 1 (1)| 31       | 35    |

Figure 8: (Color on-line) Histogram of the distance between the atom DVs and the reference DV \( \vec{v} (0 \text{ K}) \) for the deuterated case, in a), and for the pristine case in b). The velocity direction for the considered sample is (001) in both cases. Please note that in both panels only interstitial defects are identified - although the pristine sample has eventually fewer atoms in a distorted environment, the number of Frenkel pairs is larger.

### 4. Concluding Remarks

MD simulations are commonly used to study radiation damage in crystalline materials. The analysis of the results are sometimes misleading by the relative small volume fraction of the modified parts of the sample and by the formation of unforeseen defect types after irradiation. In this work, we present a fingerprint method capable to identify defects in crystalline
samples. It is based on a descriptor vector of the local environment of an atom. The DV is sensitive to local modifications of the atomic configuration but is insensitive to global changes (like rotations) at the same time. The proposed approach is suited to identify and classify defects of a given sample in a semi-automated manner. This opens the door for a semi-automated continuous monitoring of the defect evolution during collision cascades - something which has rarely been done so far because of the large manual effort involved.

It provides also a probabilistic quantification for each atom of the sample to be in a distorted local environment. We applied our method to irradiated pristine and deuterated W samples. For this, we used QUIP to compute the descriptor vectors of all atoms of the samples to describe the neighborhood of each atom. The MD simulations have been performed with LAMMPS using a primary tungsten knock-on atom of 1 keV and sample temperature of 300 K. For the deuterated case, we used a principal component analysis to identify suitable descriptor vectors for W atom as first and second nearest neighbors of D atoms. These DVs are subsequently used to identify and quantify defects in the D-W samples after irradiation. It turns out that the deuterated samples exhibit, with high probability, more defects than the pristine samples - although improved statistics is needed to substantiate this claim further. Future work is on the one hand side focused on the effect of the impact energy and the deuterium concentration on the formation of defects in tungsten. The other line of research addresses the identification of new types of defects and the application of the DV based approach to other systems.

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Appendix A. Vacancies identification

The number of vacancies is calculated by computing the nearest neighbor distance to the closest atom of a thermalized defect-free W sample and the damaged sample, by using the k-d-tree algorithm [30]. A comparison between atom-atom distances indicates the vacancy location in the damaged sample, where distance values exceed a lattice dependent threshold. In Fig. 9 we present a histogram of the results of identification of vacancies for a velocity direction of \langle 001 \rangle. We notice that the distance difference values beyond a threshold set at 4 are associated to the location of vacancies. In order to obtain a visualization of all the defects classification, the W atoms that are next to these vacancies are identified and shown in the final analysis, Fig. 5.

Appendix B. Supplementary material

DV s and visualization videos

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