

We thank the reviewers for taking the time to review the manuscript and for their constructive comments. We have addressed each individual comment below. We believe that all comments have been appropriately addressed, and that the manuscript has improved due to these revisions.

Reviewer 1

My main concern is with the presentation of results. Since fluctuations are expected due to vibrations in the lattice, the authors should provide a perspective on how much are the fluctuations and how much do they affect the properties. I would like to see a plot on the supercell fluctuations of lattice parameters, along with the averaged one that they have provided. It would also be beneficial if the authors could provide numerical values of lattice constants in addition to normalized lattice constants. I could not understand why the actual lattice constants were not provided to begin with. On the defects front, it would be helpful to know how many calculations were performed to obtain the average defect formation energies for each type of defect. What is the effect of the fluctuations on the defect energies - clearly, the fluctuations of the nearest neighbor atoms due to temperature may affect the formation energy values. The inflection point at 400 K is interesting. At this point, the authors have not provided any physical reasoning behind it - it could either be true or could simply be an artifact of their calculation. I think, in order to prevent any wrong pursuit later by the community, they authors should provide some level of confidence to support their results that indeed some interesting mechanism operates at 400 K. Otherwise, at this point, it merely appears as an observational result.

-A new figure illustrating the variation in lattice parameter as a function of time is shown, and the data is quantified within the text.

-The room temperature lattice constants are included in the text and the full temperature-dependent lattice constants have been included in the appendix.

-The total number of simulations are included in the computational details section, and a further description of standard error in these calculations is included.

-For data at 400 K, it is currently an observational result, as no clear indication of a change in behavior was observed. A further discussion around the (un)certainty of this inflection has been included.

Reviewer 2

The authors present ab-initio molecular dynamic simulations of the temperature dependence of lattice parameters and point-defect formation in alpha-U. Overall the manuscript is clearly structured and well written with appropriate use of figures and tables. I recommend the manuscript for publication in Frontiers after the authors have addressed the issues below.

In line 140/141 the authors mention that 'significant scatter exists in the experimental datasets' that enter as averages in Fig.2. For the comparison of the AIMD results and experiment, it would be beneficial to include also the error bar of the experimental data in Fig.2.

The experimental data compilation from Touloukian summarized 48 individual experiments and provided a recommended fit for each a , b , c , and V thermal expansion. These experiments did not all necessarily quantify their uncertainty, however, Touloukian stated that the recommended values are considered accurate to within $\pm 5\%$. It was our intent to illustrate that while there is experimental literature available, and that literature is qualitatively consistent, there are quantitative discrepancies. Text revision and additional discussion along these lines has been included in the manuscript.

As far as I understand from Sec.2, the authors obtained the data in all figures from an average over 10 trajectories. As the error can increase substantially with temperature (Fig.5) I would suggest to include the error bar in all figures.

The standard error for equilibrium lattice constants, and thus for thermal expansion, is sufficiently miniscule as to be illegible on these graphs. Twice the standard error at 800 K, averaged over all three lattice constants, is 0.002 Angstroms. Text has been included to indicate such small deviations in the lattice constants. However, error bars have now been included in the heat capacity figure and in the defect lattice strain figure.

For the comparison with experiment (particularly for Fig.2) it would be helpful if the authors could mention if the experiments used single-crystalline or poly-crystalline samples. In the latter case, the anisotropy in the thermal expansion could be averaged out and explain the difference to the AIMD data. This would also explain why the volume-averaged thermal expansion is in better agreement than the thermal expansion of the individual lattice constants.

Further explanation of the experimental results has been included. The data includes both single crystal and polycrystalline samples.

Also regarding the anisotropy of the thermal expansion of the individual lattice constants, I would suggest to add DFT calculations of elastic constants at $T=0\text{K}$ to the manuscript. The extra calculations would be a moderate effort as only a primitive unit cell is required and the actual calculation can be done with one of the existing tools for computing elastic constants with DFT. If the DFT error as compared to experiment is larger for one of the elastic constants than for the bulk modulus, then this could explain the discrepancy to experiment in Fig.2 and the agreement in Fig.3.

Calculations of elastic constants in $\alpha\text{-U}$ have been previously performed by the authors. This work is summarized and included in the manuscript. However, it is difficult to draw conclusions from the discrepancies between the 0 K elastic constant data and room temperature experimental data. DFT overestimates all primary (C_{11} , C_{22} , C_{33}) elastic constants, and subsequently overestimates the bulk modulus. Some degree of overestimation is to be expected when comparing data at room temperature to 0 K data. The percent overestimated is: C_{11} 39%; C_{22} 16%; C_{33} 36%; B 32%. Given this information, it is not readily obvious that DFT should predict more dramatic thermal expansion/contraction in the x/y directions, while matching both the z thermal expansion and volumetric expansion. Discussion indicating that such comparisons were made but no clear conclusions could be drawn, has been included.

In lines 173-175 the authors mention DFT calculations of the vacancy at $T=0\text{K}$. I would suggest to add this data also to Fig.5 for a direct visual comparison.

The 0 K vacancy and interstitial formation energies from previous work at 0 K have been included in Fig. 5 (now Fig. 6).

In line 176 the authors state that their lower value of the interstitial defect formation energy is due to additional reaction pathways or defect reorientation due to temperature in the AIMD simulations. I don't agree with this statement as it stands in the manuscript as the $T=0\text{K}$ calculations can in principle identify all relaxation pathways. If the AIMD detects a new configuration then this may have to do with a break of symmetry that is not accessible to $T=0\text{K}$ relaxation algorithms. With their statement the authors claim implicitly that they found a new configuration of the interstitial that is lower in energy than the configuration assumed in Huang and Wirth 2011. Therefore, in order to make this statement, the authors need to analyse their MD trajectories to identify the new configuration and confirm the lower energy by additional $T=0\text{K}$ DFT calculations similar to Huang and Wirth 2011. If there is no new low-energy configuration, then the lower value in this manuscript has a different origin. In this case the authors could not make this statement and would need to discuss the discrepancy differently.

In principle, DFT can access any prescriptible state, however the identification of a potential low energy state is often complex, or incredibly difficult to identify utilizing strictly 0 K simulations. Additionally, the identified low-energy defect location could in fact be metastable, whereas the local minima can be overcome by the means of added temperature fluctuations. Additionally, atomic vibrations may preferentially stabilize alternate defect orientations compared to the equilibrium configuration at 0 K. We only state that the defect configuration presented has a lower formation energy than the calculated defect energy from Wirth at 0 K. We have modified the text to better represent this information. If the reviewer continues to believe we should perform 0 K DFT calculations on a snapshot of our observed defect configuration, then we will do so, given additional time to perform such simulations.

In Fig.2 and Fig.6, the authors compare the temperature dependence of lattice strain without and with point defects, respectively. The plots have the same y-axis $\Delta L/L_0$ but as far as I understand the plots, the choice of the reference length L_0 is different. In Fig.2, L_0 is taken as the lattice constant at $T=300\text{K}$. In Fig.6, however, I think that the authors have not chosen a constant value of L_0 but rather the values of the lattice constant at the particular temperatures, i.e. $L_0(T)$ in contrast to $L_0(300\text{K})$. This effectively separates the thermal expansion of the ideal lattice from the influence of the defect. This is the correct procedure but it needs to be made clear in the discussion which value of L_0 is used.

We thank the reviewer for identifying this. The distinction and clarification have been included.

The discussion of RDF and DOS is a bit vague in my opinion and not very helpful. The authors could consider to extend it, e.g. by including figures, or to leave it out.

An additional figure and increased discussion have been included to illustrate the changes in the rdf as a function of temperature. The DOS analysis has not been included as the data does not

present any distinguishable information, and we feel should only serve as a point to describe how we looked for differences pointing towards inflection at 400 K, but found no obvious indicators. We hope the additional discussion increases the utility of this section.

Some minor wording issues:

- line 12: delete extra 'expansion' in 'thermal expansion expansion/contraction'
- line 150: 'Data point_s_ are shown ..'

We thank the reviewers for identifying these grammatical issues. They have been resolved.

Some of the names in the references are missing special characters, e.g. Blöchl, Furthmüller, Söderlind. In line 368 it should be 'Söderlin_d_'

The bibliography file has been updated to include umlauts, as well as the correction to Söderlind.