We thank the reviewers for their comments, and we feel the paper has been strengthened and has gained in clarity due to their efforts. Comments to their revision requests are below in blue.

Reviewer #1: In the presented article, Beeler, et al. study gamma-U using AIMD simulations at temperatures from 900 to 1400 K. Using this approach equilibrium volumes, the bulk modulus, the radial distribution function, the interstitial and vacancy formation energies, and the diffusion coefficients are calculated. In my opinion authors conducted a thorough and reliable study of the mentioned above properties of gamma-U. Yet, I have some comments:
1) "Uranium (U) is an actinide exhibiting delocalized f-electrons that exists in three solid phases: … " - I think that considered P-T conditions should be mentioned, because at higher pressures bct-U is predicted to be stable.

We have revised the sentence to indicated that this is at atmospheric pressures.

2) "There have been no investigations of the free energy via the temperature dependent effective potential (TDEP) technique" - there have been investigations with TDEP technique. Please add these two references in the text: Ladygin, et al. "Lattice dynamics simulation using machine learning interatomic potentials." Computational Materials Science 172 (2020), Kruglov, et al. "Phase diagram of uranium from ab initio calculations and machine learning." Physical Review B 100.17 (2019).

We thank the reviewer for pointing this out and we apologize for missing these references. They have been added.

3) I think that averaged over the final 1000 timesteps P and E values can be shown on Fig. 1.

The average over the final 1000 timesteps has been included in Fig 1.

4) "The melting point of uranium is 1408 K" - please put a reference to this value.

A reference has been added.

5) Is there an explanation why experimental values of thermal expansion coefficients are different in two experiments by Touloukian and Basak (Fig. 3)? I'm asking because later in the text authors say that DFT leads to the underestimation of the lattice constant, so this may lead to a different from experiment values of thermal expansion coefficients (yet of Fig. 3 there is almost a perfect match between calculated points and experimental values from Basak, et al.)

The thermal expansion is adjusted such that a value of zero is obtained at 1100 K, regardless of the magnitude of the lattice parameter. In this sense, we tried to only look at lattice variation, and remove differences in the estimated magnitudes of lattice constants. As far as discrepancies between the two experiments, the metal fuels handbook, which is effectively the combined reference on this material system, provides no insight. This is simply a challenging system to investigate experimentally.

6) There is a typo in the axis label on Fig. 2.

Thank you for catching this error. This has been fixed.

7) There is no reference to Table 1 in the text.

Thank you, we have corrected out labeling and now point to both Table 1 and Table A2 in the appendix.

8) About rdf: authors explain broadening of the peaks at T = 900 K, yet at T close to the melting point (1400 K) I would also expect broadening. Why there is no broadening at this temperature?

The rdfs from 1200K to 1400K are effectively identical. In such a small supercell, one would likely need to dramatically superheat in order to get a phase change into a liquid structure. Provided additional heating beyond the melting point, we would expect to see broadening of the rdfs. However, it was not observed at 1400K.

9) Vacancies formation energies are almost constant at T < 1300 K, and then there is a big growth (yet for interstitials formation energies are almost constant in the whole temperature region). Is there an explanation for such behavior?

Thank you for the comment, and this is an excellent question. First, we would argue that vacancy formation energy is slightly increasing over the temperature range, and that the rate of increase accelerates above 1300K, but not dramatically so. The error bars in the figure represent one standard deviation, and if two standard deviations are included, then a linear fit to the entire dataset is included within the error bars. As such, the statistics are not sufficiently refined to establish the veracity accelerated increase above 1300K. However, quadratically increasing vacancy formation energies have been observed in bcc U by Smirnov and Stegailov, in the paper you referenced. A quadratic fit is the best-fit to the dataset. Thus, we believe the behavior of the vacancy formation is reasonable. But we do not provide an unique reason for the behavior.

I also would suggest citing the following work, where formation energies were calculated using empirical potential (yet show similar behavior): Smirnov, G. S., and V. V. Stegailov. "Formation free energies of point defects and thermal expansion of bcc U and Mo." Journal of Physics: Condensed Matter 31.23 (2019).

This paper has been referenced, citing the entropic behavior observed, as well as the defect formation energy behavior.

Overall, mentioned above comments do not change the importance of the presented work, so I would recommend minor revisions.

Reviewer #2: In this manuscript, Beeler, Andersson, Jiang and Zhang perform ab initio molecular dynamic (AIMD) simulations in order to compute the bulk (thermal expansion and bulk modulus) and defect (formation energies and diffusion coefficients) thermal properties of gamma-U between 900 K and 1400 K. If the bulk properties and the vacancy formation energy

are in very good agreement with experiments, the self-diffusion coefficient is one or two orders of magnitude above the experiments.

The calculation of defect properties in gamma-U as a function of temperature is very important in order to properly describe the thermodynamic and transport properties in this material but also in other related bcc uranium compounds (UMo, UZr…). Moreover, a comprehensive understanding of the physics behind the creation of point defects in a strongly anharmonic system, only stable at high temperature, would be very useful for the community. I thus believe that this manuscript can be of interest to the readership of Journal Nuclear Materials.

However, I consider that at the present stage the manuscript cannot be accepted for publication and needs some rewriting. I list below my comments or questions:
1. Page 2, 2nd paragraph: "There have been no investigations of the free energy via temperature dependent effective potential (TDEP) technique [...]". At least three recent works use TDEP for gamma-U and compute its free energy as a function of P, T, Mo content…: Bouchet et al. [1], Kruglov et al. [2] and Castellano et al. [3]. I suggest to the authors to amend this bibliographical part.

We thank the reviewer for pointing this out and we apologize for missing these references. They have been added.

2. Page 3, lines 6-7: Authors say that they didn't perform any spin-polarized calculations. What about the spin-orbit? Is it important in gamma-U at high temperature?

This has not been demonstrated to be important for gamma U at 0 K, but at high temperature, it is unknown. As this would dramatically increase the computational expense, SOC is potentially beyond the scope for a wide-ranging defect study such as this. A note has been added in the computational details indicating that SOC is not included.

3. Page 3, lines 8-9: What are the units ("10-4" and "-10-4")? Are these relative quantities?

Units have been added to these quantities.

4. Page 4, line 4: "[…] where the gamma phase is stable […]": I suggest to say that, in this range of temperature, the gamma-U phase is DYNAMICALLY stable (the phonon frequencies are real - omega2 positive), but not THERMODYNAMICALLY (the free energy is not the lowest).

We thank the reviewer for the clarification and have made the appropriate revisions.

5. Page 8, Table 1 (but also Appendix, Table A2): The noise is strong. It is almost chaotic. Do the authors have any explanations?

For the bulk modulus, fitting is to a series of volumes, and any errors in individual volume-pressure determinations will have an impact on the bulk modulus. These errors are not correlated with one another; therefore, it is possible to have errors compounding or negating one another, leading to more chaotic trends. The GENERAL behavior though is one of softening, which is as

expected. The authors would suggest to perform a fitting procedure to establish a more refined function of bulk modulus with temperature. Such a fitting was not included here because we felt it would distract from the emphasis on point defects. Data is included such that other researchers may utilize the softening for their own purposes.

6. Page 10, line 2: "The formation energy of both vacancies and interstitials increase with increasing temperature.". I don't see any clear increasing of the interstitial formation energy in Fig. 5. This assertion is evident for vacancies but seems slightly exaggerated for interstitials. Same comment in the conclusion: "Point defect formation energies increase with temperature […]"

We thank the reviewer for this comment, we agree and have modified the text accordingly.

7. Page 10, lines 4-5: "[…] due to the hexagonal-type ground state structures for both elements […]". There is definitely no hexagonal stable structure in the whole phase diagram of U. The only appearance of an hcp structure is metastable, when an uranium film grows above a substrate [4].

The alpha phase can be described as a distorted HCP lattice. An increase in the internal y parameter of the alpha phase from 0.1 (as in experiments), to 0.1667, would lead to an HCP structure, albeit with non-ideal c/a.

8. Page 13, below Eq. 4: The authors say that the entropic contribution is set equal to one. I'm not convinced by this strong approximation and I encourage the authors to argue about that. On one hand, Smirnova and coworkers [5] (I don't see any citation/comparisons with the results of this work in the present manuscript. It would be interesting to do that.) show that almost one order of magnitude of variation is obtained when one accounts for the explicit temperature effects on the entropic contribution (for U and Zr). On the other hand, at high temperature, significant modifications of the entropy (due to the huge variations of the phonon frequencies [1]) are expected when the gamma-U phase is stabilized by anharmonic effects. One can refer to the work of Grabowski et al. [6] in order to take into account such kind of contributions. Is it possible for the authors to go beyond their approximation and evaluate this term (DeltaSvib)? By computing the velocity auto-correlation function (vacf) of the pristine and defective systems, for each temperature, then performing a fast Fourier transform, it seems possible to access to the vibrational density of states (vDOS) as a function of temperature and evalutate DeltaSvib(T), using the harmonic approximation.

The results from Smirnova et al. have been included in Fig 9 for comparison. We agree that it would be valuable to explicitly determine the entropies in order to include a more accurate description of the activation energy for diffusion. However, we feel such a study would dramatically extend the scope of this work (in order to do it properly), and provide minimal additional benefit. A study led by a co-author into beta U migration energies (unpublished, to be submitted) has shown interstitial entropies on the order of 0.5kB, which actually points towards this approximation of exp(S/kB) = 1 as reasonable. Smirnov and Stegailov, JPCM 31 (2019) 235704, also showed very small entropic effects for interstitials in bcc U, and this reference has been added. We believe this validates the reasonableness of the assumptions on entropic effects.

9. Page 14, second paragraph: "[…] this is considered reasonable agreement and provides confidence […]". This point is related to the previous one. I'm not convinced that a difference of one or two orders of magnitude is a "reasonable agreement". Is this strong discrepancy also seen in other materials? Same comment in the conclusion: "[…] however this is considered reasonable agreement."

We have compared to the empirical potential results from Smirnova, showing similar levels of accuracy as this work. Both computational models err on the order of 1-2 magnitudes, with this work overestimating and the MD studies underestimating the self-diffusion. This discussion has been included to provide context to the results from AIMD. The conclusion has also been modified.

References:
1. J. Bouchet and F. Bottin, PRB 95, 154113 (2017)
2. Ivan A. Kruglov, Alexey Yanilkin, Artem R. Oganov and Pavel Korotaev, PRB 100, 174104 (2019)
3. A. Castellano, F. Bottin, B. Dorado and J. Bouchet, PRB 101, 184111 (2020)
4. R. Springell, B. Detlefs, G. H. Lander, R. C. C. Ward, R. A. Cowley, N. Ling, W. Goetze, R. Ahuja, W. Luo, and B. Johansson, PRB 78, 193403 (2008)
5. D.E.Smirnova, A.Yu.Kuksin and S.V.Starikov, JNM 458, 304 (2015)
6. B. Grabowski, T. Hickel, and J. Neugebauer, PSS B 248, 1295 (2011)