Answers to the Reviewer (2) comments.

Comment: The comments by the reviewer were very insightful and useful, and the manuscript has improved significantly by incorporating the suggestions. In addition to the responses given below, the section of the manuscripts has been improved by making changes on the figures and slight changes in the abstract and conclusion. The results section has been improved significantly to clarify on UNCLE cluster expansion, addition of reference for UNCLE code, analysis of PHONON dispersion curves and the slab depth of the surfaces and their relaxation.

Minor Corrections Answers

1). Question: In the second last sentence of the abstract the word "surface" is used four times. The last two uses are redundant and should be removed.

Answer: The two words "surface" in the abstract line were removed and the sentence reads as "The calculated thermodynamically equilibrium morphologies of the relaxed surface structures indicated that the (100) surface was the most dominant for all the studied structures."

2). Question: The last sentence of the Abstract is identical to the last sentence of the Conclusion. These sentences must be modified to be sufficiently different from each other.

Answer: Last sentence in the abstract was modified to be different from the last sentence in the conclusion. Abstract reads: The findings offer more insight on the stability of these minerals and their surface stability. And the conclusion reads: These outcomes gave more insights on the bulk and surface stability of these minerals which demonstrated the preferred plane cleavage of these minerals during mineral grinding.

3). Question: A reference is needed for why knowing the preferred cleavage plane could benefit the recovery of these minerals.

Answer: The statement in the abstract and conclusion "which demonstrated the preferred plane cleavage of these minerals and may be applicable in their recovery" was removed.

4). Question: Top of page 2: the sentence beginning "The Brillouin zone k-points..." is too long and complex and the use of the word "respectively" at the end of the sentence is not clear. The sentence should be split into two and rewritten.

5). Question: The use of "x" in cases such as "4x4x4" is lazy. Word (and latex) both have easily usable multiplication signs. All such instances must be corrected.

Answer (4) and (5): The statement was amended and the "x" replaced with "*" in the test and the sentence reads, "The Brillouin zone k-points sampling of 4*4*4 for $PtAs_2$ and PtAsS and 7*7*7 for Pd_2As were used. In the case of surfaces the 4*4*1 for $PtAs_2$ and PtAsS and 5*3*1 for Pd_2As were employed. These were chosen according to the scheme proposed by Monkhorst-Pack [8]."

6). Question: The acronym "UNCLE" is defined the second time that it is used. It should be defined the first time. Also, it is not clear why in one case it is referred to as "MedeA-UNCLE" and elsewhere as "UNCLE".

Answer: The word UNCLE was only defined in its first appearance in the abstract "The phase stability of PtAsS was obtained using UNiversal CLuster Expansion (UNCLE) code.", and the "MedeA-UNCLE" word was modified to "UNCLE". In addition the sentence "In addition, the

UNCLE code within Materials design (MedeA) was performed to generate new stable phase of PtAsS model [10].” was modified to show that the UNLCE code is within the materials design software.

7). Question: Page 2, first sentence: change “infinite extension” to “infinite extension, i.e., using periodic boundary conditions,”.

Answer: The statement was amended to “To model the surface of the periodic boundary conditions, a slab of finite thickness perpendicular to the surface but infinite extension, i.e. using periodic boundary conditions was used.” as suggested.

8). Question: Reference 11 is a reference to the theory behind the cluster expansion. A reference should be added for the code that was used. I am not sure if it is the 2009 paper of Lerch et al. or not. If the programme was written by the authors themselves then this should be clearly stated.

Answer: A correct reference was added “[12] Lerch D, Wieckhorst O, Hart G L W, Forcade R W and Muller S 2009 UNCLE: a code for constructing cluster expansions for arbitrary lattices with minimal user-input Modelling Simul. Mater. Sci. Eng. 17 1–19.” as required.

9). Question: Section 3.2: The supercell size or sizes should be given.

Answer: The bulk structures were used in their unit cell not supercells.

10). Question: Section 3.3: It is stated that positive frequencies are observed in all directions. However in Figures 3(b) and (c) the scale of the graph goes to -1, which suggests that the phonon frequencies do actually go negative. A careful zoom on (b) shows that at gamma the phonon frequencies are actually slightly negative. It should not change the overall result, but should be mentioned.

Answer: A statement was added to address the negative scale “However, the PtAsS and Pd_2As system were found to give a scale up to –1 THz, which may suggest a slight overlap of the vibrations to negative. These were ignored and considered as imaginary modes.”

11). Question: Figure 4(e): The slabs are not identical. I assume that this is due to the stacking configuration. Whatever the reason, it should be mentioned.

Answer: The slabs in Figure 4(e) was modified to have three slabs that display identical slabs.

12). Question: Section 3.4: The reason for using 15 layers should be given. Did the calculation converge at this number?

Answer: The reason for using 15-layers has been provided by inserting Figure 4(g) and 4(h) and for Pd_2As the number of layers was recounted as shown in the Figure 4(e). And a statement “The slab depths were varied as shown in Figure 4(g) for $PtAs_2$ and PtAsS (100) surface and 4(h) for Pd_2As (100) surface. These showed a linear decreasing trend of total energy with an increase in slab thickness. The principal concern is that the slab depth should be sufficient such that both surfaces act as an effectively infinite amount of the bulk solid (i.e. the surfaces do not interact with one another through the solid). Ideally to achieve this, a large slab depth would be required but the computational cost of modelling very deep slabs was avoided. As such the 15-layer slab depth for $PtAs_2$ and PtAsS (100) surface and 21-layers for Pd_2As (100) surface were chosen and considered as thick enough for adsorption.” was added to support the inserted Figures.