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Letter to Russia

J. G. Tobin

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Disclaimer

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To: Alex Mirmelstein, VNIITF, Snezhinsk, Russia
Mikhail Ryzhkov, Russian Academy of Science, Yeketerinburg,Russia

From: Jim Tobin, LLNL

Re: Referee report on our manuscript

Dear Alex and Mikhail,

Below is the referee report. It is not as bad as it seems at first. The manuscript has not been rejected. Instead, the referee is "not recommending publication." On the APS website, the status is "with authors," instead of "not under consideration." Thus, this manuscript is still alive, but we will need to work on it.

Please take a look at what the referee says below and let me know how you would respond. I will do the same. Hopefully, we will be able to respond well and find a way for this manuscript to get into PRB.

The worst-case scenario is we try another journal, such as J. Condensed Matter Physics. However, let's not go to that option quite yet.

With best regards and many thanks,
Jim

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Re: BP11467
Probing actinide electronic structure through Pu cluster calculations
by M. V. Ryzhkov, A. Mirmelstein, S. W. Yu, et al.

Dear Dr. Tobin,

The above manuscript has been reviewed by one of our referees. Comments from the report appear below.

These comments suggest that the present manuscript is not suitable for publication in the Physical Review.

Yours sincerely,

Athanasios Chantis
Assistant Editor
Physical Review B
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See http://prb.aps.org/rapids for new information and statistics
on PRB Rapid Communications.

P.S. Another referee was consulted but we now assume that no
report will be received. If a useful report is received,
we will contact you.

Report of the Referee -- BP11467/Ryzhkov

According to the introduction of their manuscript, the authors intend to study the electronic structure of clusters of Pu atoms and, among other things, to illustrate how the properties of the cluster's central region approach those of the bulk Pu metal as the cluster size increases. It is then somewhat surprising to find out that all the "cluster" calculations discussed in the paper are in fact set up in such a way that they model the bulk properties - the clusters are embedded in a kind of mean field that is designed to approximate the rest of an infinite lattice (the authors call it the extended cluster scheme). Consequently, all the observed finite-size effects are essentially artificial since they represent the inaccuracies of the embedding procedure.

The results for the finite clusters themselves do not carry a direct physical meaning (which contradicts authors' statements from the introduction), only the extrapolation to the infinite cluster would, if done properly. The authors propose that the number of 5f electrons n_5f is a linear function of the cubic root of N, where N is the number of atoms in the cluster. This function fits the calculated data well (Fig. 8), but, as the authors indeed note, it cannot hold for very large N where n_5f must saturate at a finite value. The calculated data show no sign of such saturation (Fig. 8), which indicates that the considered clusters are too small to draw conclusions about the bulk properties. I find it puzzling that the authors nonetheless claim in their conclusions that "An evaluation of state occupations supports the proposal that the occupation of the 5f levels in bulk Pu must be near 5".

Apart from the aforementioned conceptual inconsistencies, there are a number of more technical aspects that are not discussed in sufficient detail. Among these are:

1/ The authors use LDA to approximate the electron correlations. A lively debate takes place in the literature whether this approximation can adequately describe the electronic structure of Pu metal or not, yet the authors do not discuss the choice of the approximation at all, which they should, in my opinion. They should also specify if their solutions are spin polarized or whether they use spin-restricted LDA.

2/ The quality of the employed basis set is not clear. Are the results converged with respect to the basis size? What is the estimated magnitude of the residual errors?

3/ There are statements in the manuscript indicating that the cluster calculations depend somehow on the calculations of the diatomic molecule. Namely: "Underpinning these calculations, there is a geometry optimization of diatomic molecules..." and "Underlying the Pu cluster simulations is the calculation of the electronic structure of a Pu2 dimer with the bond length 3.28 A[ngstrom] corresponding to the inter-atomic distances in delta-Pu." What does this underpinning/underlying mean in more technical terms? What role does the geometry optimization play when the cluster calculations seem to be performed at a fixed geometry corresponding to the delta-Pu?

Lastly, the manuscript contains a lot of material that was previously (and often multiple times) published elsewhere, including the Physical Review journals. For instance, the experimental part of Fig. 2 was shown already in Refs. 26, 27 and 28 in essentially the same graphical form; the top part of Fig. 9 appeared in Refs. 19, 4 and in PRL 90, 196404 (2003). I think that reprinting these results is not necessary and just referencing the earlier papers would be sufficient.

I do not recommend this paper for publication in Physical Review B.