Reply to "Comment on 'Accurate and fast numerical solution of Poisson's equation for arbitrary, space-filling Voronoi polyhedra: Near-field corrections revisited' "

Aftab Alam,[1,*] Brian G. Wilson,[2,†] and D. D. Johnson[1,3,‡]

[1]*Division of Materials Science and Engineering, Ames Laboratory, Ames, Iowa 50011, USA*

[2]*Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, California 94550, USA*

[3]*Department of Materials Science and Engineering, Iowa State University, Ames, Iowa 50011, USA*

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This is a Reply to the Comment by Gonis and Zhang[1] on our recent paper.[2] They discuss supposed issues with our "accurate and fast numerical solution of Poisson's equation for arbitrary Voronoi polyhedra" (VP).

The method of Gonis *et al*.[3] and Vitos and Kollár[4] correctly addresses the mathematical issues related to the solution of Poisson's equation [and near-field corrections (NFCs)] using a spherical harmonic basis (L's). However, as discussed in Ref. 2, their method is not numerically efficient and suffers from severe convergence issues from the multiply nested (conditionally convergent) multipole expansions to address $r_</r_>$ problems. Hence, the abstract in Ref. 2 clearly states that we provide a method to address a "*fast but accurate numerical solution of the Poisson equation*" for site-centered methods.

The method of Gonis *et al*. is exact, in principle, only for Bauer expansions with infinite L sums. By our own tests, none of the previous results have been converged with sufficient accuracy, including the fcc case where the potential at the far corner of the cube has a 10^{-2} error even for $L \sim 80$. Their method is correct for highly converged calculations but requires excessively large computational resources—more than required to solve the density functional theory equations! Nicholson and Shelton[5] had similar convergence issues using multipoles.

To emphasize this, consider a nonsymmetric VP, such as a sheared fcc lattice. For convergence, the Gonis *et al*. approach would require significantly higher internal L's and even more excessive computational resources—hour(s) of computing time on a single processor, compared to about 1 s for our method.[2] This estimate is obtained by a direct calculation of the van Morgan problem in Ref. 2 and includes the cost of calculating the shape function.[6] If one uses our new integration,[6] the cost drops dramatically, but the convergence issues and excessive cost for large L-sum integrations remain. Notably, this computational effort is required for each self-consistent field iteration during each molecular dynamics step.

Thus, from our abstract,[2] and a very practical point of view, our conclusion and summary are untarnished—we have indeed provided an accurate and fast numerical solution of Poisson's equation, using no large multipole sums or fast Fourier transforms (FFTs). We applied our method to two distinct problems, a van Morgan and a Madelung model of a solid, in two charge distribution limits, with exact agreement to analytic results using a single L sum up to 8. So, there can be no dispute that the method in Ref. 2 works and works much faster for any level of fixed accuracy.

We do concede that "arbitrary" VP is overstated, given the atypical cell (electrostatic) example in Fig. 1 of Ref. 1. In practical electronic-structure applications for such a cell, additional basis functions (i.e., empty sites) can be added, for example, and our method works.

Second, we do not dispute that we provided a solution based on the sum of two contributions for α_L for ease of numerical solution. This point is a "red herring." FFTs are used by plane-wave methods to avoid VP integrals and NFCs completely, but these FFTs limit cell sizes due to communication bottlenecks on parallel computers.

Lastly, the integration method that we used to solve the Poisson equation is generally quite useful, and it is very efficient for solving the integral representation of α_L to avoid the multipole approach and to yield an accurate and fast numerical solution of Poisson's equation in practical applications, which was all that was stated in Ref. 1.

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*aftab@ameslab.gov

†wilson9@llnl.gov

‡ddj@ameslab.gov

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