

Kiran *et al.* Reply: Our original paper [1] made two important observations regarding Al_nH_m clusters. First, hydrogen can alter the total electron count of a given Al_n cluster by adopting different orientations. In bridging and capping mode it adds an electron to the total count, while in the radial mode it takes away an electron from the system. Secondly, in Al_nH_m clusters, hydrogen adopts a position in such a way that the total electron count corresponds to electronic shell closure of an otherwise Jellium cluster. Such clusters then have large HOMO-LUMO gaps and gain enhanced stability. A rule was provided to count these electrons and to demonstrate the stability of certain Al-H clusters by computing the energies of low-lying isomers and verifying the computed electronic properties of the magic clusters with experiments. Explicit calculations were provided for all clusters containing 20 electrons and two clusters containing 40 electrons. None of these calculations have been found to be wrong by Jung and Han in the preceding Comment [2]. As a matter of fact, our lowest energy isomers, for most of which we also presented experimental evidence, were validated by Jung and Han.

However, the authors have found low-energy structures in the case of $Al_{13}H_3$, $Al_{13}H_5$, $Al_{14}H_4$, and $Al_{14}H_6$, which do not obey the rule we have prescribed. For two of the clusters ($Al_{14}H_4$ and $Al_{14}H_6$) the authors have found isomers that lie, respectively, 0.1 and 0.38 eV lower in energy than the structures prescribed by our rule. These energies are within the numerical uncertainty such as the approximations used for exchange-correlation energy functional in density-functional theory and the choice of basis sets. Besides, finding global minima for larger structures is a very difficult task. For example, an icosahedric cluster containing 13 Al atoms has 20 triangle and 40 bridge positions and 12 radial sites for hydrogen atoms to adopt. With 3 and 5 H atoms, there are an enormous number of isomers. It is not clear from their work if there are other 40 electron systems with H occupying different on-top and bridge and capping sites that may have lower energy than what the authors have given.

It should be stated that our rule is meant to guide in the selection of possible structures. Even for those clusters,

which do obey the rule, the structures are not unique as several arrangements of hydrogen atoms are possible and accurate computations are required to determine the best arrangement. In addition, the rule developed in our paper is explicitly based on the Jellium model (the total number of electrons, N , are equated to 20, 40, etc.; Eq. 1). Obviously, the rule is as good as the Jellium model itself. If the Jellium model fails so does our rule. There are several instances where a non-Jellium cluster, preferably a lower electron count system, was shown to be more stable. For example, Kumar [3] had shown that an $Al_{10}Li_{18}$, 38 electron aluminum cluster, with 1.62 eV HOMO-LUMO gaps, shows classic behavior of a magic cluster. In fact, even though there is an apparent failure of the rule, it should be noted that none of the low-energy structures predicted by Jung and Han have “higher electron” count than predicted by the rule; i.e., for a 40 electron system, no 42 or 44 electron structures were found to be lower in energy.

B. Kiran,¹ P. Jena,¹ X. Li,² A. Grubisic,² S. T. Stokes,²
G. F. Ganteför,² K. H. Bowen,² R. Burgert,³ and
H. Schnockel³

¹Physics Department
Virginia Commonwealth University
Richmond, Virginia 23284, USA

²Department of Chemistry and Materials Science
Johns Hopkins University
Baltimore, Maryland 21218, USA

³Institute of Inorganic Chemistry
University of Karlsruhe (TH)
78128 Karlsruhe, Germany

Received 26 March 2008; published 12 May 2008

DOI: [10.1103/PhysRevLett.100.199702](https://doi.org/10.1103/PhysRevLett.100.199702)

PACS numbers: 73.22.-f, 71.15.Mb, 79.60.-i

- [1] B. Kiran *et al.*, Phys. Rev. Lett. **98**, 256802 (2007).
- [2] J. Jung and Y.-K. Han, preceding Comment, Phys. Rev. Lett. **100**, 199701 (2008).
- [3] V. Kumar, Phys. Rev. B **60**, 2916 (1999).