Density Functional Studies of Magic Metal-(C$_{60}$)$_2$ Clusters

EVA ZUREK, Max Planck Institute for Solid State Research, JOCHEN AUTSCHBACH, Department of Chemistry, University at Buffalo — Previous experimental studies of C$_{60}$-metal clusters revealed that clusters with composition C$_{60}$Ba$_{32}$ and (C$_{60}$K$_6$)$_n$K$^+$ appeared as magic peaks in the Time-of-Flight mass spectra due to geometric and electronic stability, respectively. Recent experiments using a new heating technique have revealed a different set of magic peaks, which cannot be explained by either one of the aforementioned mechanisms. We present theoretical studies addressing the stability and bonding of these newly observed magic clusters. Molecular density functional calculations have been performed to determine the most energetically stable geometrical configurations for $M_n$(C$_{60}$)$_2$ clusters, with $M$ = K, Ba and $1 \leq n \leq 6$. The bonding mechanisms have been analyzed in some detail. The results indicate that for barium containing clusters, ionic bonding (transfer of the valence 6$s$ electrons to the C$_{60}$ $- \pi^*$ orbitals) and covalent bonding (between the barium 5$d$ and C$_{60}$ $- \pi^*$ orbitals) are the dominant mechanisms. Moreover, a metal cluster is formed between the two fullerenes (Ba-Ba bonding). For the potassium containing clusters, only ionic bonding was found. Calculations of the Gibbs free energies indicate that Ba$_3$(C$_{60}$)$_2$ and K$_4$(C$_{60}$)$_2$ are the most stable structures, in agreement with experimental results. The role of the entropy is found to be very important in determining which clusters are magic.

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