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)_nK^+ 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 6s electrons to the C_{60} − π^* orbitals) and covalent bonding (between the barium 5d and C_{60} − π^* 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.