Electronic Structure and Magnetism of [(FePc)$_n$H$_2$Pc] Superlattices: A First-principles Study

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Recently Vargas et al. synthesized quasi-1D [(FePc)$_n$H$_2$Pc] superlattices and measured the magnetic properties. In this study we calculate electronic and magnetic properties of [(FePc)$_n$H$_2$Pc] with $n = 3$ and 4 using density functional theory (DFT). FePc systems are prone to local energy minima due to different *d*-orbital occupations. We identified a lower energy state compared to previous studies by applying *d*-orbital occupation matrix control. Based on DFT total energies with spin-orbit interaction, we estimate the exchange coupling constants and zero-field-splitting parameters for an effective spin Hamiltonian of each superlattice. We found that an FePc molecule adjacent to an H$_2$Pc molecule has smaller on-site magnetic anisotropy. The exchange interaction between two FePc molecules that are separated by an H$_2$Pc molecule is over 600 times smaller than that between two adjacent molecules. The antisymmetric exchange or the Dzyaloshinskii–Moriya interaction is two orders smaller than the isotropic exchange interaction. Finally, we present calculated magnetization and magnetic susceptibility for both [(FePc)$_3$H$_2$Pc] and [(FePc)$_4$H$_2$Pc] superlattices. We hope to shed some light on the application of MPc systems to quantum information science. Further studies of spin dynamics are underway.

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