Exploring Seebeck-coefficient fluctuations in endohedral-fullerene, single-molecule junctions

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For the purpose of creating single-molecule junctions, which can convert a temperature difference ΔT into a voltage ΔV via the Seebeck effect, it is of interest to screen molecules for their potential to deliver high values of the Seebeck coefficient $S = -\Delta V/\Delta T$. Here we demonstrate that insight into molecular-scale thermoelectricity can be obtained by examining the widths and extreme values of Seebeck histograms. Using a combination of experimental scanning-tunnelling-microscopy-based transport measurements and density-functional-theory-based transport calculations, we study the electrical conductance and Seebeck coefficient of three endohedral metallofullerenes (EMFs) $Sc_3N@C_{80}$, $Sc_3C_2@C_{80}$, and $Er_3N@C_{80}$, which based on their structures, are selected to exhibit different degrees of charge inhomogeneity and geometrical disorder within a junction. We demonstrate that standard deviations in the Seebeck coefficient σ_S of EMF-based junctions are correlated with the geometric quantity σ and the charge inhomogeneity σ_q. We benchmark these molecules against C_{60} and demonstrate that both σ_q, σ_S are the largest for $Sc_3C_2@C_{80}$, both are the smallest for C_{60} and for the other EMFs, they follow the order $Sc_3C_2@C_{80}$ >$Sc_3N@C_{80}$ > $Er_3N@C_{80}$ > C_{60}. A large value of σ_S is a sign that a molecule can exhibit a wide range of Seebeck coefficients and if orientations corresponding to high values can be selected and controlled, then the molecule has the potential to exhibit high-performance thermoelectricity. For the EMFs studied here, large values of σ_S are associated with distributions of Seebeck coefficients containing both positive and negative signs, which reveals that all these EMFs are bi-thermoelectric materials.

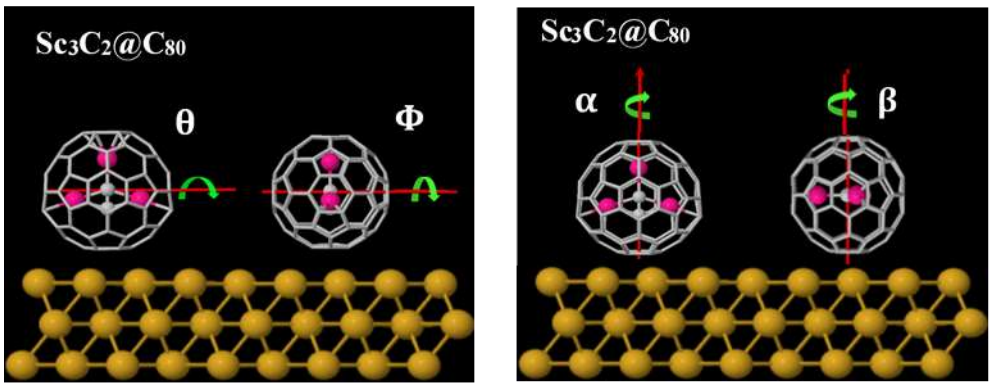

Figure 1. Illustration of the four rotation axes: θ and Φ are horizontal axes, α and β are vertical axes. This Figure shows how the axes pass through the I_h-C_{80} cage + metallic moiety.

[1] Rincón-García, Laura, Ali Ismael and *et al*. *Nature materials* **15**, no. 3 (2016).
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