Contribution ID: **245** Type: **Talk**

【176】Surface single-molecule dynamics controlled by entropy at low temperatures

Thursday 24 August 2017 18:00 (15 minutes)

Transitions of individual molecules on surfaces are traditionally described with energy barriers and attempt rates using an Arrhenius law. This yields consistent energy barrier values, but the attempt rates are orders of magnitude below expected oscillation frequencies of particles in potential wells. Using LTSTM to measure an individual dibutylsulfide molecule on Au(111), we find that the transition's barrier and attempt rate depend in a highly correlated fashion on the tip-molecule position. We establish a single-molecule entropy-enthalpy compensation, and discuss an interpretation in terms of multi-excitation entropy. Motivated by these findings, we study the conservative as well as the dissipative interactions between the molecule and the tip in combined SFM/STM measurements.

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Session Classification: Surfaces, Interfaces and Thin Films

Track Classification: Surfaces, Interfaces and Thin Films