

Magnetic-State Controlled Molecular Vibrational Dynamics at Buried Molecular–Metal Interfaces

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Abstract-

Self-assembled molecular (SAM) structures have been intensively used in molecular electronics and spintronics. However, detailed nature of the interfaces between molecular layers and extended metallic contacts used to bias the real devices remains unclear. Buried interfaces greatly restrict the application of standard techniques such as Raman or scanning electron microscopy. Here, we introduce low-frequency noise spectroscopy as a tool to characterize buried molecular–metal interfaces. We take advantage of vibrational heating of the molecules with incomplete contacts to the interface. Electrons, being the main spin and charge carriers propagating through the interfaces involving SAMs, interact inelastically with the nuclei and excite quantum molecular vibrations (phonons). Our detailed investigation of both conductance and conductance fluctuations in magnetic tunnel junctions with few nanometer perylenetetracarboxylic dianhydride (PTCDA) allows to map vibrational heating at specific biases taking place in hot spots such as where SAM layers make unstable contact to the metallic electrodes. We follow this effect as a function of PTCDA thickness and find the highest molecular–metal order for the lowest (three to five monolayers) barriers. Moreover, we show experimentally that the low-frequency noise depends on the relative alignment of electrodes well beyond expectations from fluctuation–dissipation theorem. In combination with modeling, we interpret this effect as due to a magnetic-state dependent molecular vibrational heating at the interfaces driven by the spin-polarized current.

Index Terms-

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