High-Performance Single-Molecule Memory Based on Redox-Active N-Heterocyclic Carbenes

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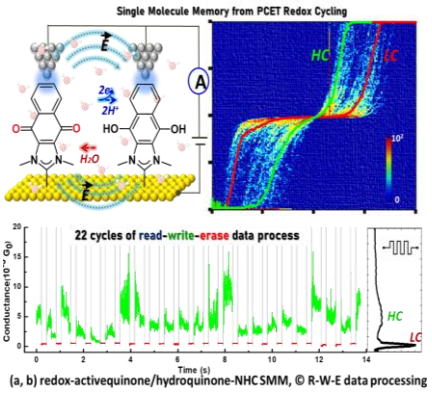
Alternative memoristive elements, such as molecular memory, are extensively searched as valuable complimentary approaches to the conventional CMOS architectures. "Single molecular memory" (SMM) emerges when conductance switching can be reversibly controlled in single-molecule junctions (SMJs). Stable switching, especially at room temperature, remains challenging. Here we report a stable SMJ based on a redox-active quinone-functionalized N-heterocyclic carbenes (Q-NHCs) covalently anchored to a gold electrode. By combining the redox-switching of the quinone/hydroquinone pair and the stable carbene-Au interface, we obtained robust, non-volatile single molecule memory effect at room temperature. Scanning-tunneling-microscopy break-junction (STM-BJ) and static conductance–time (G(t)) techniques demonstrate an initial high conductance state (HC, ~$4\times10^{-3}G_0$) corresponding to the oxidized quinone form which reversible switches to low conductance state (LC, ~$10^{-5}G_0$) evidenced by highly reproducible hysteretic I–V loops under electric field stimulation. This works records the current best molecular memory performance, with an ON/OFF ratio of 60 ± 20 and a switching yield larger than 80%. Density functional theory combined with nonequilibrium Green's function transport calculations confirm that the electrically driven redox quinone/hydroquinone interconversion is the mechanism behind the memory effect. Taking advantage of the stable switching, we demonstrate non-volatile memory switching over 20 cycles of read–write–erase-read operations between the HC and LC states. Our findings establish that carbene chemistry combined with redox-switching is a promising route to develop high-performance solid state single-molecule memory with potential for non-volatile molecular-scale storage application.



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