Negative Differential Resistance in Single-Molecule Junctions Based on Heteroepitaxial Spherical Au/Pt Nanogap Electrodes

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Single-molecule junctions have established a variety of functional quantum devices working based on the molecular orbitals of individual molecules[1]. The single-molecule junctions exhibiting negative differential resistance (NDR) behaviors, characterized by a decrease in current with increasing voltage, have attracted considerable attention due to their potential application as ultra-fast resonant tunneling diodes. Thus, several pioneering studies have investigated the mechanisms behind this nonlinear NDR behavior through theoretical modeling and experimental evaluation[2]. However, the peak-to-valley (PV) ratios of NDR observed in most single-molecule junctions are relatively small (< 10). The large-scale fabrication and integration of electronic devices also highlight a demand to create a platform for constructing solid-state single-molecule junctions.

Recently, we have developed heteroepitaxial spherical (HS)-Au/Pt nanogap electrodes (Figure 1a) prepared by electron-beam lithography (EBL) and self-termination electroless gold plating (ELGP)[3]. Their molecular length gap separation, small radii, and robust thermal stability enable large-scale multiple fabrication of single-molecule junctions on a Si substrate using interested molecules.

Here, we report a pronounced NDR effect with a PV ratio of 30.1 (Figure 1b) on a single-molecule junction consisting of a π-conjugated quinoidal-fused oligosilole derivative, Si2×2[4], embedded between HS-Au/Pt nanogap electrodes. This NDR effect persists over a consecutive 180 current traces and showed stable temperature dependence between 9 K and 300 K[5]. Density functional theory calculations under electric fields suggest that the NDR effect arises from bias-dependent resonant tunneling transport via the polarized highest occupied molecular orbital (HOMO). Our findings demonstrate a promising electrical platform for constructing functional quantum devices at the single-molecule level.

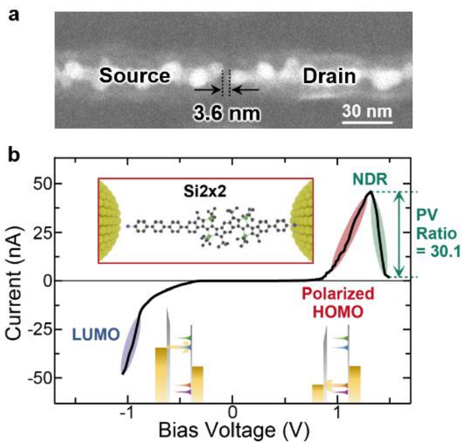

Figure 1. (a) SEM image of top view of HS-Au/Pt nanogap electrodes. (b) I_d−V_d characteristics of Si2×2 single-molecule junctions with a pronounced NDR effect.

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