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Enhanced Magnetoresistance in Molecular Junctions by Geometrical Optimization of Spin-Selective Orbital Hybridization SOUMYAJIT SARKAR, DAVID RAKHMILEVITCH, ORA BITTON, LEEOR KRONIK, OREN TAL, Weizmann Institute of Science — Molecular junctions based on ferromagnetic electrodes allow the study of electronic spin transport near the limit of spintronics miniaturization. However, these junctions reveal moderate magnetoresistance that is sensitive to the orbital structure at their magnetic tip - molecule interfaces. The key structural parameters that should be controlled in order to gain high magnetoresistance have not been established, despite their importance for efficient manipulation of spin transport at the nanoscale. Here, we show that single-molecule junctions based on nickel electrodes and benzene molecules can yield a significant anisotropic magnetoresistance of up to $\sim$200% near the conductance quantum G_0. The measured magnetoresistance is mechanically tuned by changing the distance between the electrodes, revealing a nonmonotonic response to junction elongation. These findings are ascribed with the aid of first-principles calculations to variations in the metalmolecule orientation that can be adjusted to obtain highly spin-selective orbital hybridization. Our results demonstrate the important role of geometrical considerations in determining the spin transport properties of metalmolecule interfaces.

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