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Mechanical Control of Spin States in Spin-1 Molecules and the Underscreened Kondo Effect[1]

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The ability to make electrical contact to single molecules creates opportunities to examine fundamental processes governing electron flow on the smallest possible length scales. We report experiments in which we controllably stretched individual molecules with spin S = 1/2 and S = 1 while simultaneously measuring Kondo-assisted current flow through the molecule as a probe of its spin states. For molecules with S = 1/2, the temperature dependence of the Kondo signal is in excellent agreement with the predicted universal scaling curves for the S = 1/2 Kondo effect and the molecular spin states exhibit no energy splitting with stretching, consistent with Kramers' theorem. However, for cobalt complexes we observe temperature scaling curves that are very different from the S = 1/2 Kondo predictions, and instead are in quantitative agreement with the predictions of the underscreened Kondo model for S = 1, in which conduction electrons only partially compensate the molecular spin. This allows us to identify the spin state as S = 1. As a function of stretching, the S = 1 molecules exhibit energy splittings of the spin states in the absence of magnetic field due to magnetic anisotropy arising from modification of the molecular symmetry. These findings demonstrate a mechanism of spin control in single-molecule devices and establish that they can serve as model systems for making precision tests of correlated-electron theories.

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