

Abstract Submitted
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Rotational Excitation Spectroscopy with the Scanning Tunneling Microscope – Distinction of Nuclear Spin States¹ FABIAN DONAT NATTERER, FRANÇOIS PATTHEY, HARALD BRUNE, École Polytechnique Fédérale de Lausanne — The appeal of inelastic electron tunneling spectroscopy with the scanning tunneling microscope (STM) stems from its unmatched spatial resolution and the ability to measure the magnetic, electronic and vibrational properties of individual atoms and molecules. Rotational excitations of molecules could provide additional information of surface processes but have hitherto remained elusive. Here we demonstrate rotational excitation spectroscopy (RES) with the STM for hydrogen and its isotopes on graphene and hexagonal boron nitride. Since the Pauli principle imposes restrictions on the allowed rotational levels J for molecules with identical nuclei, a certain alignment of the nuclear spins entails a specific set of rotational levels. Conversely, measuring the rotational levels allows characterizing the molecular nuclear spin state. We measured excitation energies at 44 meV and 21 meV, corresponding to rotational transitions $J = 0 \rightarrow 2$ for hydrogen and deuterium. We thereby identify the nuclear spin isomers para- H_2 and ortho- D_2 . For HD, we observe $J = 0 \rightarrow 1$ and $J = 0 \rightarrow 2$ transitions, as expected for heteronuclear diatomics. Our measurements demonstrate the potential of STM-RES in the study of nuclear spin states with unprecedented spatial resolution.

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