Probing the triplet lifetimes of a single molecule by atomic force microscopy

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The nonequilibrium triplet state of molecules plays an important role in photocatalysis, organic photovoltaics, and photodynamic therapy. Recently, the single-electron sensitivity of AFM [1] in detecting electrostatic forces has been exploited to sense individual electron tunneling events between tip and investigated structures[2-4]. Here we report the direct measurement of the triplet lifetime of an individual pentacene molecule on an insulating surface with atomic resolution by introducing an electronic pump-probe method in atomic force microscopy (AFM)[5]. Strong quenching of the triplet lifetime is observed if oxygen molecules are coadsorbed in close proximity. By means of single-molecule manipulation techniques, different arrangements with oxygen molecules were created and characterized with atomic precision, allowing for the direct correlation of molecular arrangements with the lifetime of the quenched triplet. Such electrical addressing of long-lived triplets of single molecules, combined with atomic-scale manipulation, offers previously unexplored routes to control and study local spin-spin interactions.

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