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Time Resolved Photoelectron Spectroscopy of CdSe Quantum Dots in the Gas Phase¹ JENNIFER ELLIS, WEI XIONG, DANIEL HICK-STEIN, CHENGYUAN DING, MARGARET MURNANE, HENRY KAPTEYN, Department of Physics and JILA, University of Colorado and NIST, Boulder — We present the first photoelectron spectroscopy measurements of quantum dots (semiconductor nanocrystals) in the gas phase. By coupling a nanoparticle aerosol source to a femtosecond velocity map imaging photoelectron spectrometer, we apply robust gas-phase photoelectron spectroscopy techniques to colloidal quantum dots. Working with a flowing aerosol of quantum dots offers the advantages of providing fresh nanoparticles for each laser shot and removing perturbations from bonding with a surface or interactions with a solvent. In this work, we perform a two-photon photo to be to be to be a show that the photoelectron yield per exciton depends on the physical size of the quantum dot, increasing for smaller dots. Using effective mass modeling we show that the extent to which the electron wave function of the exciton extends from the quantum dot, the so-called "evanescent electron wavefunction", increases as the size of the quantum dot decreases and that the photoelectron yield is dominated by the evanescent electron density. Further, we measured the charge transfer rate from the quantum dots to attached dye molecules. This work shows that gas-phase photoelectron spectroscopy is a robust and general probe of the electronic structure and dynamics of quantum dots.

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> Jennifer Ellis Department of Physics and JILA, University of Colorado and NIST, Boulder

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