Visualizing exciton wavefunctions confined in a quantum dot

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Nano-resolution optical microscopy maps shrinkage and distortion of the wavefunctions of excited states caused by local perturbations to a quantum system.

Producing, characterizing, and manipulating single quantum states are basic techniques critical for the functioning of nanodevices and the implementation of quantum information processing. Semiconductor quantum dots (QDs) represent ideal quantum systems because their electrons are three-dimensionally confined, resulting in a discrete density of states. An important characteristic of this atomlike energy-level structure is its ability to suppress the interaction of an electron with a lattice vibration. Consequently, electrons in QDs can store quantum phase coherence for very long periods of time and show ultranarrow absorption and emission spectra. Such properties are advantageous in applications that include low-energy-consumption optoelectronic devices and quantum information processing. Moreover, the dots can be engineered to have desired optical and electronic properties by controlling their size and shape, as well as appropriately selecting their material.

Our research efforts focus on experimentally determining the physical shape and potential profile of QDs to provide vital information on the spatial distribution of strain, fluctuations of alloy components, and interface roughness. In particular, visualizing weak perturbations of confinement will enable us to detect disorder and impurities at the single-atom level. Photoluminescence (PL) spectroscopy using a near-field scanning optical microscope (NSOM) is promising tool for measuring the physical shape and potential profile of QDs. It combines the nondestructive nature of optical microscopy with high spatial resolution that far exceeds the diffraction limit of light. So far, we have realized real-space mapping at resolutions as high as 10–30nm of the wavefunctions (wavelike behavior) of excited states known as excitons and biexcitons confined in a gallium arsenide (GaAs) interface fluctuation QD (IFQD), a monolayer-high island formed in a narrow quantum well: see Figure 1.1,2

For an exciton confined in a shallow potential, the degree of extension of the wavefunction is sensitive to the exciton mass. As the mass decreases, the energy barrier for the confined exciton also decreases, and the wavefunction penetrates more deeply into the barrier. Thus, the profile of the wavefunction of an exciton confined in an IFQD with a shallow potential fluctuation should differ greatly from that of a biexciton, which has a mass twice that of the exciton: see Figure 2(a) and (b). Conversely, some characteristic differences in the wavefunctions of excitons and biexcitons can be used to measure the size and depth of the shallow potential.

We found a significant difference in spatial PL maps between exciton and biexciton emissions for several IFQDs: see Figure 2(c) and (d).3 To gain a quantitative understanding of the difference, we numerically calculated the wavefunctions of an exciton and a biexciton confined in an IFQD with an irregular potential profile. The calculation revealed that a shallow potential dip, which probably originates from atomic-scale interface fluctuations, is responsible for the difference in the PL images.

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Figure 2. (a, b) Isotropic shrinkage of a biexciton wavefunction (BX) with respect to that of an exciton (X). (c, d) Anisotropic shrinkage of the biexciton wavefunction due to a shallow potential formed in a QD.

Figure 3. Distortion of an exciton wavefunction (a) compared with that of a charged-exciton wavefunction (b) induced by a local electric field.

Charged excitons in IFQDs are good candidates for an optically driven spin-based qubit (quantum computing) system. However, due to the weak lateral confinement of charge carriers in IFQDs, frequent switching between neutral and charged exciton states may occur by thermal activation or quantum tunneling of the excess electron to a nearby trap. To understand and control this switching, it is important to directly observe the excess electron path in real space. We experimentally observed distortion of the exciton wavefunction due to the electric field produced by an excess electron at a nearby trap state (see Figure 3). The findings show switching between neutral and charged exciton emission that accompanies excess carrier tunneling.

Confinement of both electrons and light at the nanoscale gives rise to unique interactions that are completely different from those described in conventional textbooks. To be more specific, when the spatial resolution of a NSOM is less than the size of a QD, it enables direct mapping of the distribution of its wavefunction. More interestingly, it makes it possible to break the ‘optical selection’ rule: a dark state—an exciton state that cannot absorb or emit photons in the far-field regime and therefore appears dark—can be excited, opening new radiative decay channels and thus controlling energy storage and transport in exciton systems. The light-matter interaction at the nanoscale offers guiding principles for future nanophotonic devices. We will extend our research to include nano-optical and nanomechanical active control of quantum states using ultrafast phase-change materials.

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Toshiharu Saiki received his PhD from the University of Tokyo (1993). He joined the Kanagawa Academy of Science and Technology, Japan, where he later became a project leader. He became an associate professor in the Department of Electronics and Electrical Engineering at Keio University in 2002 and professor in 2009.

References