Optical spectroscopy of single quantum dots at tunable positive, neutral, and negative charge states

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We report on the observation of photoluminescence from positive, neutral, and negative charge states of single semiconductor quantum dots. For this purpose we designed a structure enabling optical injection of a controlled unequal number of negative electrons and positive holes into an isolated InGaAs quantum dot embedded in a GaAs matrix. Thereby, we optically produced the charge states -3, -2, -1, 0, $+1$, and $+2$. The injected carriers form confined collective ''artificial atoms and molecules'' states in the quantum dot. We resolve spectrally and temporally the photoluminescence from an optically excited quantum dot and use it to identify collective states, which contain charge of one type, coupled to few charges of the other type. These states can be viewed as the artificial analog of charged atoms such as H^-, H^{-2}, H^{-3}, and charged molecules such as H_2^+ and H_3^{+2}. Unlike higher dimensionality systems, where negative or positive charging always results in reduction of the emission energy due to electron-hole pair recombination, in our dots, negative charging reduces the emission energy, relative to the charge-neutral case, while positive charging increases it. Pseudopotential model calculations reveal that the enhanced spatial localization of the hole wave function, relative to that of the electron in these dots, is the reason for this effect.

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I. INTRODUCTION

Real atoms and molecules often appear in nature as charged ions. The magnitude and sign of their ionic charge

the wider of two coupled GaAs QW's, separated by a thin AlAs barrier layer.[5]

We spatially, spectrally and temporally resolved the PL emission from single SAQD's in both samples using a variable temperature confocal microscope setup.[15] Figure 2 compares the pulse excited PL spectra of the neutral [Fig. 2(a)] and mixed type [Fig. 2(b)] samples. The excitation in both pulse and cw (not shown here) excitations was at photon energy of 1.75 eV and the repetition rate of the picosecond pulses was 78 MHz ($\simeq$13 ns separation between pulses)

and the *e*-*h* pairs recombine one by one. Therefore, all the pair numbers that are smaller than N_x

ratio between the maximum intensity of the X^0 PL line under cw excitation and that from the negatively charge states [10:1, see Fig. 4(c)], since the emission intensity at maximum is inversely proportional to the state lifetime.[22,27] From the measured decay time of the X^0 line ($\simeq 1.3$ ns, not shown) we deduce the hopping times and find them to be slightly longer than the pulse repetition time ($\simeq 13$ ns). Therefore, under pulsed excitation, once the QD is optically depleted from its initial charge, it remains so for times longer than the time difference between sequential pulses. Thus, PL lines that result from exciton recombination in the presence of negative charge evolve similar to neutral lines under cw-mode excitation [Fig. 3(a)], and their intensity weakens with the increase in excitation power. We use this behavior to sort out the neutral states PL emission from that of negatively charged ones, in general, and for identifying the X^0 line in particular.

The larger the number of deionized donors which participate in the depletion process, the shorter is the lifetime of the charge depleted state that they generate. This can be straightforwardly deduced from Fig. 3(d) and more quantitatively by simple rate equation model simulations.[27] Since four and five deionized donors are involved in generating the $q=+1$ and $q=+2$ charge states, respectively, the lifetime of these states is shorter than that of the charge-neutral state and shorter from the pulse repetition time. Hence, the evolution of the PL intensity of the X^{+1} and X^{+2} lines with increasing pulse excitation power, is similar to that of the X^0

calculations[19] in order to realistically estimate the various terms in Eq. (1). The first three terms were obtained via first-order perturbation theory whereas the last term (the correlation energy) was obtained via configuration-interaction calculations.[14,29] The QD calculated here,[22] has a slightly elongated lens shape, with major and minor axis 45 and 38 nm (in the 110 and $\overline{1}10$ directions, respectively), and a height of 2.8 nm. Both the QD's and the two monolayer wetting layer have a uniform composition of $In_{0.5}Ga_{0.5}As$, and they are embedded in a GaAs matrix. The shape, size, and composition are based on experimental estimations, and they are somewhat uncertain.

The pseudopotential treats the alloy atomistically, and it includes spin-orbit interaction and strain. We include the first six bound electron and hole states in our configuration-interaction expansion. The calculated *S*-*P* shells splitting ($\varepsilon_{e_2}-\varepsilon_{e_1}+\varepsilon_{h_2}-\varepsilon_{h_1}\simeq 37$ meV), well agrees with the measured one and so does the energy splitting of the *P* shell ($\varepsilon_{e_3}-\varepsilon_{e_2}+\varepsilon_{h_3}-\varepsilon_{h_2}\simeq 6$ meV).

Figure 4 shows isosurface plots of the calculated density of probability for electrons and holes in their three lowest energy states. The electric charge that these isosurfaces contain amount to 75%. The figure clearly demonstrates that the

tons, unlike their analog free positively charged molecules,[17] have larger recombination energies. This novel observation may prove to be very useful in future applications of semiconductor quantum dots, where their optical emission can be discretely varied by controlled carrier injection.

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