Observation of low-lying excitations of electrons in coupled quantum dots

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Abstract

Tunneling excitations of electrons in dry-etched modulation-doped AlGaAs/GaAs coupled quantum dots (QDs) are probed by resonant inelastic light scattering. A sequence of intra- and inter-shell excitations are found at energies determined by the interplay between the QD confinement energy $\hbar \omega_0$ and the tunneling gap $\Delta_{SAS}$, the splitting between the symmetric and anti-symmetric delocalized single particle molecular states. The narrow line-widths displayed by electronic excitations in these nanostructures indicate promising venues for the spectroscopic investigation of entanglement of electron states in these artificial molecules.

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Semiconductor quantum dots (QD’s) are regarded as artificial atoms with electron properties that can be tailored on demand \[1\]. In this framework, artificial molecules can be realized with two interacting quantum dots \[2\]. It has been proposed that coupled quantum dots are of particular relevance for quantum computation schemes \[3, 4\]. In fact, the tunneling gap $\Delta_{SAS}$ that splits the QD levels into their symmetric and anti-symmetric combinations can modulate the exchange interaction and provides a route towards entanglement of two spins \[5\]. Electron states in coupled double QD are expected to be less sensitive to dephasing mechanisms linked to coupling of spin and charge states that occur in a single QD environment. Coupled QDs also offer ways to study novel spin and charge collective phases and quantum phase transitions at the nanoscale \[6\].

The remarkable physics and applications of coupled QDs is manifested in several experimental studies. Transport experiments based on tunneling have extensively investigated inter-dot coupling effects in the strong and weak coupling regimes \[7, 8, 9\]. These measurements have provided evidence for the impact of symmetric and anti-symmetric states in the Coulomb blockade spectra and have demonstrated slower relaxation rates for spin states in laterally coupled QDs and their coherent manipulation. Inter-band luminescence experiments also have been carried out in self-assembled InAs coupled QDs \[10\].

In this letter we report the observation of low-lying neutral excitations of electrons in vertically coupled GaAs/AlGaAs double QDs nanofabricated by e-beam lithography and dry etching. Resonant inelastic light scattering spectra access excitations linked to the tunneling and quantum confinement degrees of freedom of electrons. The spectra reveal sharp excitations, with FWHM below 200$\mu$eV, that demonstrates the high quality of these etched nanostructures. These QD systems could be suitable for exploration of entanglement of electron states that enter in quantum computation schemes based on the optical manipulation of spin and charge.

Eigenstates of a parabolic QD in the non-interacting scheme are described by the Fock-Darwin (FD) levels. In vertical QDs the coupling between the two dots leads to the splitting of the single dot levels in bonding (symmetric) and antibonding (antisymmetric) levels separated by the tunneling gap $\Delta_{SAS}$. The single particle eigen-energies of parabolic double QDs at zero magnetic field can thus be modeled by the following equation:

$$E_{N,P} = \hbar \omega_0 (2n + |m| + 1) + P \Delta_{SAS} = \hbar \omega_0 (N + 1) + P \Delta_{SAS},$$  \hspace{1cm} (1)
where $n = 0, 1, \ldots$, $m = 0 \pm 1, \ldots$ are the radial and azimuthal quantum numbers, respectively, $N = 2n + |m|$ identifies shells with well-defined atomic-like parities, $\hbar \omega_0$ is the confinement energy and $P$ is the extra degree of freedom labeled as a pseudospin. $P$ takes values -1/2 for symmetric levels and 1/2 for antisymmetric levels. This peculiar energy-level structure yields an excitation spectrum characterized by tunneling or pseudospin modes constructed from intra- or inter-shell excitations of electrons from the symmetric to the anti-symmetric QD levels. Figure 2a shows the FD sequence following Eq.1 in the case in which $\Delta_{SAS}$ is slightly larger than $\hbar \omega_0$.

Parity selection rules applied to parabolic double coupled QDs establish that monopole transitions with $\Delta m = 0$ ($\Delta N = 0, 2, 4, \ldots$) and $\Delta P = 0, \pm 1$ are the strongest intensity modes active in light scattering experiments in a backscattering geometry \[11, 12\]. The single-particle representations of intra- and inter-shell pseudospin excitations associated with changes of $N$ and $P$ are shown in Fig.2a as vertical arrows. In agreement with this scheme we found in our experiments two lowest-energy peaks in the low-temperature inelastic light scattering spectra at energies below 1 meV that were assigned to the intra-shell $\Delta P = 1$ excitation at $\Delta_{SAS}$ and to the inter-shell pseudospin mode at $2\hbar \omega_0 - \Delta_{SAS}$. These peaks are remarkably sharp with a full width half maximum (FWHM) of 200 $\mu$eV and allow direct measurement of $\Delta_{SAS} = 1 \pm 0.1 meV$ and $\hbar \omega_0 = 0.8 \pm 0.05 meV$ in our system. Consistent with this assignment we found a broader inter-shell monopole mode at $2\hbar \omega_0 = 1.8 \pm 0.1 meV$.

Contrary to the case of single QDs, this inter-shell excitation has a marked temperature dependence and disappears around 15K. The measured activation gap is consistent with the value of $2\hbar \omega_0 - \Delta_{SAS}$, offering further evidence of the impact of inter-dot coupling on the excitation spectrum. Finally an additional broad mode is observed around $2.4 \pm 0.4 meV$ corresponding to $4\hbar \omega_0 - \Delta_{SAS}$.

The sample was fabricated starting from an 18 nm wide, symmetrically modulation-doped Al$_{0.1}$Ga$_{0.9}$As/GaAs double quantum well with 6nm Al$_{0.1}$Ga$_{0.9}$As barrier separation (see Fig.1). The tunneling gap measured by light scattering in the double layers (data not shown) is 0.8 meV. The low-temperature electron density and mobility measured from Subnikov De Haas are $n_e = 3 \times 10^{11}$ cm$^{-2}$ and $2.7 \times 10^6$ cm$^2$/Vs, respectively. The lateral confinement was produced by inductive coupled plasma reactive ion etching (ICP-RIE). To this end a 30 nm thick nickel mask was first deposited on top of the sample. Coupled QD arrays (with sizes 100$\mu$m $\times$ 100$\mu$m containing $10^4$ single coupled QD replica) with lateral
dot diameters of about 400 nm were defined by electron beam lithography. This value of lateral diameter was chosen to yield a confinement energy of the order of the tunneling gap and a large electron occupation (above 100). In this many-electron case, in fact, the light scattering response at resonance is dominated by single-particle transitions linked to Eq.1 and not corrected by dynamical many-body effects [11]. Deep etching (below the doping layer) was then achieved by using a mixture of BCl₃/Cl₂/Ar in the ICP-RIE and applying low voltages. The nickel mask was removed before the optical studies. Top panels in Fig. 1 show scanning electron microscope (SEM) images of the coupled dots in the array. Light scattering experiments were performed in a backscattering configuration (q ≤ 2 × 10⁴ cm⁻¹ where q is the wave-vector transferred into the lateral dimension) with temperatures down to T = 1.9 K. A tunable ring-etalon Ti:sapphire laser was used and the scattered light was collected into a triple grating spectrometer equipped with a CCD detector.

Figure 2 shows the resonant inelastic light scattering spectra at different excitation energies and T=1.9K after conventional subtraction of the background due to interband lu-
FIG. 2: Left: schematic representation of energy levels and transitions in the coupled quantum dot. $N$ and $P$ are the shell and pseudospin quantum numbers, respectively. Black and red lines represent symmetric and antisymmetric levels, respectively. The dotted line marks the position of the Fermi level that accounts for the observed reduced intensities of the two lowest-energy modes. Right panel: Resonant inelastic light scattering spectra at 1.9K and at different laser energies (shown in the figure) in depolarized configuration. Intensities were scaled by factors indicated in the figure.

minescence. The laser with intensity of 0.1W/cm$^2$ was tuned between 1528.6meV (bottom spectrum) to 1534.5meV (top spectrum) to explore different resonances. The spectra were taken in a depolarized configuration with perpendicular polarizations of the incoming and outgoing light in order to reduce the stray laser light.

Spectra shown in Fig.2 are remarkably different from those found in single QDs with the same lateral diameter [13]. In single QDs the resonant inelastic light scattering spectra display a sequence of peaks equally spaced in energy by $2\hbar\omega_0$ with a FWHM of 1 meV in agreement with data obtained by other groups [11]. The spectra of double QDs are instead characterized by two sharp (FWHM = 0.2 meV) low-energy peaks at $2\hbar\omega_0 - \Delta_{SAS}$ and $\Delta_{SAS}$. The peak at 1.8meV corresponding to $2\hbar\omega_0$ is thus assigned to the conventional inter-shell $\Delta N=2$ mode also observed in the single QDs. The peculiar energy level structure of the coupled QDs is additionally revealed by the fourth highest-energy peak shown in Fig.2 and observed at an energy corresponding to $4\hbar\omega_0 - \Delta_{SAS} = 2.4$ meV. The energy of this peak shifts from 2.2 to 2.6 meV depending on the laser excitation wavelength. This behavior can be linked to nonparabolicity effects whose impact increase with the energy of the mode and
FIG. 3: Temperature dependence of the monopole inter-shell transition at $2\hbar\omega_0$. Laser intensity and energy are 0.1 W/cm$^2$ and 1567 meV. Spectra are presented after conventional subtraction of background due to luminescence. The inset shows an Arrhenius plot of the integrated intensity with an activation energy of 0.8 meV.

to partial overlap with the $2\hbar\omega_0 + \Delta_{SAS}$ inter-shell pseudospin mode expected at $\sim$2.8 meV. More excitations were detected at higher energies (data not shown) with decreasing intensity and with broader signal corresponding to higher inter-shell excitations combined with the symmetric and antisymmetric states. It can be noted that the energy of the intra-shell pseudospin mode ($\Delta_{SAS} = 1$ meV) is higher than the the single-particle excitation energy at the bare tunneling gap measured in the double quantum well prior to nanofabrication ($\sim$0.8 meV). This is probably due to partial depletion of electrons caused by the etching processes.

The difference in the intensities between the modes below 1 meV and those at higher energies is remarkable. It suggests partial population of the two highest-energy occupied levels as indicated by the position of the Fermi level shown by the dotted line in the left part of Fig.2. These two levels are the excited states associated with the two sharp low-lying transitions. Their partial population explains the reduced intensities of the two modes due to phase space filling effects. The results in Fig.2 therefore suggests that light scattering can be applied to determine both energies and population of molecular states in coupled QDs.

Further evidence of the impact of inter-dot coupling arises from the temperature behavior of the conventional intra-shell monopole excitation at $2\hbar\omega_0$. Contrary to the single quantum dot case, in fact, where this excitation remains unchanged up to temperatures above 30K, here a significant change of the signal intensity occurs at much lower temperatures and the
inter-shell mode disappears around 15K with an activation energy of 0.8 meV as displayed in Fig.3. The activation gap is consistent with the value of \(2\hbar\omega_0 - \Delta_{SAS}\), the gap separating the highest-energy occupied level (an antisymmetric state with shell number \(N\)) from the lowest-energy unoccupied level (a symmetric state with shell number \(N+2\)). This behavior thus offers further evidence of the impact of inter-dot coupling in the excitation spectrum.

In conclusion, we reported the first measurements of excitations of electrons in nanofabricated vertically-coupled quantum dots. The spectra reveal a low-lying intrashell pseudospin mode across the tunneling gap as well as inter-shell excitations resulting from the interplay between the confinement energy and the tunneling gap. The results presented here suggest that, by offering access to molecular-like excited states in the coupled QDs, the light scattering methods can provide a wealth of quantitative information on the energy level sequence, level occupation and tunneling gap in double QDs. Further work in the few-electron occupation regime should address the interplay between spin and pseudospin excitations in the coupled dots.

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[1] S.M. Reimann and M. Manninen, Rev. Mod. Phys. 74, 1283 (2002).
[2] V.G. Van Der Wiel et al., Rev. Mod. Phys. 75, 1 (2003)
[3] D.P. DiVincenzo et al., Nature 408, 339 (2000)
[4] D. Loss and D. P. DiVincenzo, Phys Rev. A 57, 120 (1998).
[5] G. Burkard et al., Phys. Rev. B 59, 2070 (1999)
[6] M. Rontani et al., Phys. Rev. B 69, 085327 (2004)
[7] T. Hatano et al., Science 309 268 (2005)
[8] J.R. Petta et al., Science 309 2180 (2005)
[9] T.H. Wang, S. Tarucha, Appl. Phys. Lett. 71 2499 (1997)
[10] H.J. Krenner et al., Phys. Rev. Lett. 94, 057402 (2005)

[11] R. Strenz et al., Phys. Rev. Lett. 73 3022 (1995); D.J. Lockwood et al., Phys. Rev. Lett. 77, 354 (1996); C. Schuller et al. Phys. Rev. Lett. 80, 2673 (1999)

[12] C. Pascual García et al. to appear in Physical Review Letters (2005).

[13] C. Pascual García et al. to appear in Physica E (2005).