Evidence of Correlation in Spin Excitations of Few-Electron Quantum Dots

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We report inelastic light scattering measurements of spin and charge excitations in nanofabricated AlGaAs/GaAs quantum dots with few electrons. A narrow spin excitation peak is observed and assigned to the intershell triplet-to-singlet monopole mode of dots with four electrons. Configuration-interaction theory provides precise quantitative interpretations that uncover large correlation effects that are comparable to exchange Coulomb interactions.

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Electrons confined to semiconductor quantum dots (QDs) have novel ground and excited states that manifest Coulomb interactions at the nanoscale [1]. States of very few electrons are prime candidates for spintronics applications and for the implementation of quantum bits in nanoscale devices [2]. Great attention is, therefore, devoted to the study of spin physics in the regime of few-electron occupation and to experimental methods capable of reading the state of spin in the quantum dot (QD) [3].

The interpretation of experiments on few-electron QDs often requires descriptions beyond mean-field Hartree-Fock (HF) theory [1,4]. In addition to their relevance for quantum information encoding, these correlated states have significant interest for the investigation of fundamental effects [5,6]. Transport in few-electron QDs coupled to leads and excitonic optical recombination measurements have explored exchange energies and spin relaxation times. These remarkable experiments offer evidence for roles played by interactions that emerge as the number of electrons in the QD is changed [3,4,7–15].

In this Letter, we report resonant inelastic light scattering experiments in low electron density GaAs/AlGaAs QDs that probe low-neutral excitations. These are intershell monopole excitations [with change in the Fock-Darwin (FD) shell and without change in angular momentum, as required by light scattering selection rules [16,17]]. We detected two broad intershell modes that we interpret as excitations of electrons from the two populated lowest shells. Each of these two modes is split by exchange and depolarization effects into a \( \Delta S = 1 \) (spin) and a \( \Delta S = 0 \) (charge) excitation [18,19], where \( \Delta S \) represents the change of the total spin of the QD associated to the electronic mode.

A prominent feature of the spectra is an additional mode peculiar to the regime of few-electron occupation that emerges at low temperature and low excitation power. It occurs as a very narrow peak with light scattering polarization selection rules for spin excitations and is interpreted as a \( \Delta S = -1 \) intershell spin mode characteristic of a \( S = 1 \) triplet ground state with four electrons. We argue that the observed splitting between the \( \Delta S = 1 \) and \( \Delta S = -1 \) spin modes represents a direct manifestation of the role of interactions in the excitation spectra of few-electron QDs.

Numerical evaluations within a configuration-interaction (CI) theory [20] support the interpretation that links the new spin mode to the triplet-to-singlet (TS) intershell excitation of a QD with four electrons and offer quantitative insights on the role of interactions in this regime. The CI evaluations reproduce the experimental light scattering spectra with a great precision that is not achieved by HF theory. Comparisons of mean-field and CI calculations uncover large exchange and correlation terms of electron interactions that, in the case of the four-electron triplet state, are found to be comparable to quantum confinement energies.

Samples were fabricated from a 25 nm wide, one-side modulation-doped \( Al_{0.1}Ga_{0.9}As/GaAs \) quantum well with measured low-temperature electron density \( n_e = 1.1 \times 10^{11} \text{ cm}^{-2} \) and a mobility of \( 2.7 \times 10^6 \text{ cm}^2/\text{Vs} \). QDs were produced by inductive coupled plasma reactive ion etching. QD arrays (with sizes \( 210 \text{ nm} \) (shown in Fig. 1 side panels) that we expect to be comparable to quantum confinement energies.

A convenient description of single-particle QD levels is provided by FD orbitals [1] with energies given by \( e_{nm} = \hbar \omega_0 (2n + |m| + 1) \), where \( n = 0, 1, \ldots \), \( m = 0, \pm 1, \ldots \)
are the radial and azimuthal quantum numbers, respectively, and $\hbar \omega_0$ is the harmonic confinement energy. The FD shells are defined by an integer value of $N_{\text{shell}} = 2n + |m|$ with well defined atomiclike parity. QD states can be classified in terms of the $z$-component total angular momentum $M$, total spin $S$, and its $z$ component $S_z$. Studies of selection rules in QDs indicate that the monopole transitions with $\Delta M = 0$ ($\Delta N_{\text{shell}} = 2, 4, \ldots$) are the intershell modes active in light scattering experiments in backscattering geometry [16,17,22]. This noninteracting picture of intershell transitions is shown in the left part of Fig. 4, where the lowest-energy dipole ($\Delta N_{\text{shell}} = 1$) and monopole ($\Delta N_{\text{shell}} = 2$) modes are represented.

Figure 1(a) shows representative low-temperature spectra of intershell spin and charge excitations that are detected with crossed and parallel polarization between incident and scattered light, respectively [23]. Pairs of peaks are seen at energies close to 4 and 7–8 meV and interpreted as monopole excitations with $\Delta S = 1$ (spin) or $\Delta S = 0$ (charge). In the noninteracting FD picture, these two excitations are degenerate but they split in the presence of exchange and depolarization contributions. A characteristic feature of these doublets is their significant linewidth that is attributed largely to inhomogeneous broadening due to the distribution of electron occupations of the dots as described below.

The spectra in Fig. 1(a) reveal a much sharper excitation in the spin channel at 5.5 meV. To interpret the origin of this sharp spin mode, we note that an additional TS intershell spin mode with $\Delta S = -1$ can occur if the ground state is a triplet with $S = 1$ and the excited state is a $S = 0$ singlet state (see the calculated levels shown in Fig. 4). Such TS excitation is split from the $\Delta S = +1$ mode seen at lower energy by the difference in exchange and correlation contributions. On this basis, we identify the sharp peak at 5.5 meV with the TS ($\Delta S = -1$) intershell spin excitation. According to Hund’s rules, a triplet ground state occurs only when two electrons are in a partially populated shell, as is the case of QDs with four electrons. This is confirmed by the calculations described below. In this interpretation, the narrow width is simply explained as due to the absence of inhomogeneous broadening from the distribution of the electron population of the QDs.

We used the full CI approach [20,24,25] for the numerical evaluation of the energy and intensity of low-lying spin and charge excitations of the interacting system with $N$ electrons. The correlated wave functions of ground and excited states are written as superpositions of Slater determinants (SDs), $|\Phi_{(\alpha_j)}\rangle = \prod_{j=1}^{\ell} c_{\alpha_j} |0\rangle$, obtained by filling in the single-particle spin orbitals $\alpha$ with the $N$ electrons in all possible ways [$c_{\alpha}^\dagger$ creates an electron in level $\alpha = (n, m, \uparrow, \downarrow)$]. The resulting Hamiltonian is first block diagonalized, fully exploiting symmetries [25]. Finally, the Hamiltonian is diagonalized via Lanczos method in each $(M, S, S_z)$ sector, giving the low-energy excited states. The resonant Raman transition matrix elements $M_{\text{FI}}$ between the fully interacting ground and excited states $|I\rangle$ and $|F\rangle$, respectively, are obtained, after the CI calculation, from $M_{\text{FI}} = \sum_{\alpha} \gamma_{\alpha} |F| c_{\alpha}^\dagger c_{\alpha} |I\rangle$, where $\gamma_{\alpha}$ is the two-photon process matrix element between $\alpha$ and $\beta$ spin orbitals, as defined in Ref. [26] within second order perturbation theory in the radiation field and containing resonant denominators. $\gamma_{\alpha}$ causes the enhancement of the spectrum intensity when the laser energy resonates with the optical gap [27].

Figure 1(b) displays the calculated spectra for $N = 4$ and $\hbar \omega_0 = 4$ meV. The latter value was determined after an overall best-fitting procedure of the energy positions of the experimental peaks shown in Fig. 1(a). An independent check for this value of $\omega_0$ and $N$ comes from the empirical relation given in Ref. [1] [Eq. (11)] linking $N$, $\omega_0$, and the electron density, $n_e$, which gives $n_e = 1.2 \times 10^{11}$ cm$^{-2}$, in good agreement with the experimental value. It can be seen that, among all calculated excitations, only a few of them with $\Delta M = 0$ turn out to have significant intensities, generating discrete spectrum lines (with a phenomenological broadening chosen to reproduce the measured TS linewidth) in very good agreement with the experimental ones. It can also be noted that more than one excitation

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**FIG. 1 (color).** Side panels: Scanning electron microscope images of the studied QDs. (a) Experimental low-temperature ($T = 1.8$ K) polarized (parallel incident and scattered photon polarizations, in red) and depolarized (perpendicular incident and scattered light polarizations, in black) resonant inelastic light scattering spectra after conventional subtraction of the background due to laser light. Incident laser energy is 1567 meV, intensity is $I = 0.08$ W/cm$^2$, and integration time is 30 minutes. Fits of the data with three Gaussians are shown. (b) Theoretical spectra for electron number $N = 4$. Gaussian distributions with a phenomenological standard deviation $\sigma = 0.18$ meV were used.
gives a significant contribution to the spectra at energies above the TS mode. This is consistent with the observed larger linewidths for the higher-energy excitation pairs. Figure 2 reports the evolution of the calculated spectra as a function of \(N\). As expected, the TS (\(\Delta S = -1\)) mode is peculiar to \(N = 4\) and it is not observed at any other explored electron occupation configurations.

The excitations of Fig. 2 show a redshift of the lowest-energy features in both channels as \(N\) (and \(n_e\)) increases [28] due to screening effect. Because of the exchange energy gain of excited states, the spin channel energy is systematically lower than the charge excitation energy. This large sensitivity of the light scattering spectra on particle occupation is at the origin of the difference between the observed linewidths of our intershell excitations. Comparing the evolution of peak energies shown in Fig. 2 with measured linewidths, we conclude that a distribution of electron occupation between 4 and 6 characterizes our QD arrays. It also indicates that the light scattering method allows one to probe excitation spectra of few-electron QDs with single-electron accuracy despite the relatively large number of QDs illuminated. Consistent with the assignment that links the \(\Delta S = -1\) mode to those selected QDs that have four electrons is the observed sharp linewidth of 0.4 meV, which is much lower than the linewidths of the other spin and charge transitions.

The evolution of the spin transitions at different incident laser intensities shown in Fig. 3(a) confirms that the QDs are in the few-electron regime. As the intensity increases, we expect additional electrons to be photogenerated. Consistent with Fig. 2, we found that the peaks display a redshift and that the TS transition disappears at around \(I = 1\) W/cm\(^2\), suggesting that at this intensity all the QDs have more than 4 electrons, and, therefore, the number of those photogenerated is at least one. In addition, contrary to the other intershell modes, the intensity of the TS spin excitation decreases significantly as temperature increases [Fig. 3(b)] with an estimated activation gap of 0.7 ± 0.3 meV. At such low energy, a possible thermally populated excited level is the singlet state without any change in orbital occupation. This energy thus provides an estimate of the low-lying intrashell singlet-triplet transition of the four-electron QDs and it compares well with the CI estimate of 0.8 meV.

A specific feature of the low-\(N\) regime studied here is that states are represented by superpositions of many different SDs to incorporate both radial and angular spatial correlations [29]. The side diagrams in Fig. 4 represent the weighted SDs in the CI expansion of the \(N = 4\) ground and excited states involved in the three transitions indicated by arrows in Fig. 1(b). We depicted the states corresponding to the maximum allowed \(S_z\), while in the actual calculation we considered only the degenerate states with \(S_z = 0\) [25].

Figure 4 also shows ground- and excited-state energies calculated with different approximations that provide evidence for correlation effects in the excitation spectra. In the FD picture, the energy difference between consecutive levels is given by \(\hbar \omega_0 = 4\) meV. In the HF approach, spin orbitals are computed self-consistently [30]. The energy difference between the three spin configurations is given by bookkeeping the exchange energy \(K_{ab}\) gained each time two electron spins, occupying any orbitals \(a\) and \(b\),
and \( b \), are parallel to each other. This gain is accounted for by the Coulomb exchange integral between orbitals \( a \) and \( b \) described by FD wave functions. This approach neglects spatial correlation among electrons.

Correlation effects are included in the CI approach, leading to the theoretical spectra in Figs. 1 and 2 and to the quantitative agreement with experiments shown in Fig. 4. The comparison between HF and CI (Fig. 4) suggests that correlation affects the relative splittings between excited states, even reversing their relative amplitudes: The \( S = 1 \) state is nearer to \( S = 0 \) than to \( S = 2 \) in HF, while the opposite is true in CI, in agreement with the experiment. As suggested by the decreasing contribution of the most-weighted SD configurations indicated on the right in Fig. 4, correlation effects are small for the ground and the \( S = 2 \) excited states but become increasingly important for excited states with smaller \( S \). As \( S \) decreases, exchange interaction is less effective in keeping electrons far apart and excited states become more correlated. Note that the relative amplitudes of the calculated HF and CI gaps are quite insensitive to the specific value of \( \hbar \omega_0 \), and we found that the measured splittings among the spin modes can be reproduced only by CI calculations, no matter the value of \( \hbar \omega_0 \).

In conclusion, we reported inelastic light scattering measurements of spin transitions in nanofabricated quantum dots. The characteristic excitations of the triplet configuration with four electrons have been identified and theoretically evaluated. We have shown that light scattering methods offer a wealth of information on the physics of spin states in QDs with few electrons.

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[25] Details of the method can be found in M. Rontani et al., cond-mat/0508111.
[27] At difference with Ref. [26], we neglected Coulomb interaction between electrons and heavy holes and we considered a finite-width quantum well for both electrons and holes. Spectra were computed under resonance conditions, with laser energy larger than the optical gap by an amount of 8 meV, and \( q = 1 \times 10^4 \, \text{cm}^{-1} \).