Correlated electrons in optically-tunable quantum dots: Building an electron dimer molecule

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We observe the low-lying excitations of a molecular dimer formed by two electrons in a GaAs semiconductor quantum dot in which the number of confined electrons is tuned by optical illumination. By employing inelastic light scattering we identify the inter-shell excitations in the one-electron regime and the distinct spin and charge modes in the interacting few-body configuration. In the case of two electrons a comparison with configuration-interaction calculations allows us to link the observed excitations with the breathing mode of the molecular dimer and to determine the singlet-triplet energy splitting.

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The creation and control of electrons confined in semiconductor quantum dots (QDs) enables fundamental studies of Coulomb interaction at the nanoscale and allows to define optimal architectures for solid-state quantum information processing [1]. In the case of two electrons, singlet and triplet spin states with a well-defined energy splitting are under active investigation for the implementation of logic gates for quantum computing [1-3].

In the dilute limit of few electrons, a paradigm shift occurs as the liquid-like properties of the electrons – captured by a single-particle picture – cease to dominate and the impact of Coulomb correlation favors the formation of a molecular electron state with strong radial and angular order [4-9]. The molecular dimer formed by two electrons is especially relevant being the benchmark for the exploration of Coulomb correlation at the nanoscale. This state has been the subject of several theoretical investigations (e.g. [10-13]) albeit difficult to realize experimentally since it requires soft confinement potentials that are comparable to Coulomb interaction strengths. Two-electron correlated QDs were studied in potentials that are comparable to Coulomb interaction (CI) approach (aka exact diagonalization) that precisely reproduces the inelastic light scattering spectra. In the case of two electrons the CI analysis reveals links between observed excitations and the breathing and center-of-mass oscillations and rigid-rotor rotations of the molecular electron dimer.

In the CI interpretation the rotation energies are $\hbar^2 M_{rel}^2 / 2I$, with $I$ being the momentum of inertia and $M_{rel} = 0, \pm 1, \pm 2, \ldots$ the angular momentum in the relative frame. To comply with the antisymmetry of the total two-electron wave function, $M_{rel}$ must be even (odd) for a singlet (triplet) state. Therefore, the energy separation between the triplet and singlet ground states is the rotation quantum $\hbar^2 / 2I$, that as shown below is accessible by the inelastic light scattering experiments.

The sample is constructed as an array (100 $\mu$m × 100 $\mu$m) of identical QDs (10⁴ replica) to improve the
signal-to-noise ratio displaying the same optical properties \textsuperscript{[20]} and inter-dot spacing of 1 \( \mu \)m). It is fabricated by electron beam lithography and inductively coupled-plasma reactive ion etching on a 25 nm wide, one-side modulation-doped Al\(_{0.1}\)Ga\(_{0.9}\)As/GaAs quantum well. The measured low-temperature two-dimensional electron density and mobility are \( 1.1 \cdot 10^{11} \text{ cm}^{-2} \) and \( 2.7 \cdot 10^9 \text{ cm}^2/\text{Vs} \), respectively. SEM images of such QDs are shown in Figs. (a) and (b).

The inelastic light scattering experiments were performed in a backscattering configuration \( (q \leq 2 \cdot 10^4 \text{ cm}^{-1} \) where \( q \) is the wave-vector transferred into the lateral dimension) at \( T = 2 \text{ K} \). A tunable ring-etalon Ti:Sapphire laser with a wavelength of 789 nm in resonance with the QD absorption (whose onset is at 813 nm) was focused on the QD array with a 100 \( \mu \)m diameter area, and the scattered light was collected into a triple grating spectrometer with CCD detection.

Photo-depletion of the QD electrons was achieved by continuous-wave HeNe laser excitation at 633 nm that creates electron-hole pairs in the AlGaAs barrier. Whereas the photoexcited electrons in the AlGaAs barrier contribute to charge compensation of the ionized donors, the photoexcited holes are swept in the QDs flattening the parabolic in-plane potential (i.e., decreasing the confinement energy \( \hbar \omega_0 \) and contributing to the removal of the confined electrons by electron-hole recombination \textsuperscript{[21],[22]} [cf. inset to Fig. (d)]. As shown below the method allows to fully photo-deplete the QDs and tune their population with single-electron accuracy by changing the HeNe laser intensity \( I_{\text{HeNe}} \).

Distinct spin and charge intershell monopole modes (with \( \Delta M = 0, M \) is the total angular momentum) were detected as a function of \( I_{\text{HeNe}} \) by varying the linear polarization of the scattered photons (parallel (perpendicular) to that of the incoming laser for charge (spin)). The condition of photo-depleted QDs (number of confined electrons \( N = 0 \)) is defined by the absence of any inter-shell excitations. This is achieved at \( I_{\text{HeNe}} = 1.82 \text{ W/cm}^2 \) as displayed in Fig. (d) \textsuperscript{[24]}. The \( N = 1 \) regime is characterized by a single excitation at \( 2\hbar \omega_0 \) (degenerate in spin and charge) between FD levels with the same angular momentum. Therefore its energy position measures directly \( \hbar \omega_0 = 1.1 \text{ meV} \), as shown in Fig. (c). This condition is reached at \( I_{\text{HeNe}} = 1.46 \text{ W/cm}^2 \). Peculiar to this regime is the absence of multiplets at higher energy, as confirmed by calculation of the light scattering cross section within a perturbative approach under the resonant conditions employed here \textsuperscript{[24]}. Additional spin and charge modes appear at higher energy [see Figs. (a) and (d)] as we further reduce \( I_{\text{HeNe}} \). The emergence of multiplets in the spectra experimentally defines the occurrence of two electrons in the QDs at \( I_{\text{HeNe}} = 0.68 \text{ W/cm}^2 \). In this regime of shallow confinement, \( \hbar \omega_0 \approx 1-2 \text{ meV} \), the electrons are highly correlated \textsuperscript{[6],[7],[9]}.

The spectra predicted by CI calculations for \( N = 2 \) [Fig. (b)] are remarkably similar to the experimental ones. In the evaluations the only free parameter, \( \hbar \omega_0 \), is adjusted to fit the energy of the four peaks shown in Fig. (a). The spectra appear as two doublets — each formed by two peaks in the charge (blue lines) and spin (red lines) channels, labeled respectively as \( C_1, S_1 \) and \( C_2, S_2 \) in Fig. (b). Both spin excitations of Fig. (a) are slightly red shifted (\( \approx 0.2 \text{ meV} \)) from their charge counterparts. These splittings are also seen in the theoretical calculations in Fig. (b), which also reproduce the relative intensity of the peaks in each channel. The value of \( \hbar \omega_0 = 1.6 \text{ meV} \) used in Fig. (b) is larger than that measured at \( N = 1 \), as expected for a lower \( I_{\text{HeNe}} \) \textsuperscript{[22]}. The down triangles in Fig. (b) summarize instead the results of a self-consistent Hartree-Fock (HF) calculation.
of spin and charge transition energies that neglects correlation effects. This approach was extensively used to interpret previous light scattering spectra in the many-electron regime \[20-28\]. By fitting the position of \( S_1 \) [lowest down triangle in Fig. 2(b)] to its measured value [Fig. 2(a)] by properly tuning \( \hbar \omega_0 \), then the remaining three HF excitations are blue shifted more than 1 meV from their measured values. On the other hand, by fitting the position of the higher HF doublet (not shown) then \( S_1 \) is red shifted by \( \approx 1 \) meV. This analysis therefore rules out an interpretation in terms of single-particle transitions between FD shells and establishes the onset of electron correlation as we increase the electron population from \( N = 1 \) to \( N = 2 \).

The impact of Coulomb correlations is represented by the dimensionless radius \( r_s \) of the circle whose area is equal to the area per electron (in units of the Bohr radius \[16\]). For the CI calculation of Fig. 2(b) \( r_s = 3.39 \). In this dilute regime we expect the two electrons to form a molecule. This is confirmed in Fig. 2(c) by comparing the CI excitations of Fig. 2(b) [dashed lines in Fig. 2(c)] with the analytical predictions for a perfectly formed “Wigner” molecule \[11\] (WM) [solid lines in Fig. 2(c)]. The WM excitations have a composite nature: \( C_1 \) (\( C_2 \)) is a single (double) excitation of the breathing mode, with energy \( \hbar \omega_B \) (\( 2 \hbar \omega_B \)). \( S_1 \) is a dipolar center-of-mass excitation \( \hbar \omega_0 \) plus a rotational quantum \( \hbar^2/2I \) (the system as a whole keeps \( M = 0 \)), and \( S_2 \) adds a breathing mode quantum. Remarkably, spin and charge excitations scale differently with \( \hbar \omega_0 \), since \( \omega_B = \sqrt{3} \omega_0 \) whereas

\[
I \propto \omega_0^{-4/3}.
\]

The WM estimate for \( \hbar^2/2I \), and hence the singlet-triplet splitting, is 0.517 meV.

The agreement between CI and WM predictions in Fig. 2(c) is very good —with small deviations only for the higher excitations— implying that electrons in the ground state freeze their relative distance (cf. Fig. 3 and Supplementary Discussion). The formation of this Wigner molecule, which develops after the emergence of rotational bands \[9\] as \( r_s \) increases, occurs relatively soon for the dimer since it involves only nearest-neighbor particles (cf. Supplementary Discussion).

In Fig. 3(a) we plot the conditional probability \( P(x, y; x_0, y_0) \) of finding an electron at position \((x, y)\) provided the other one is fixed at \((x_0, y_0)\) (black dot).

In the ground state (GS, left panel) the well developed correlation hole around the fixed electron suggests the freezing of the relative distance \( r \). This is best seen from the pair correlation function \( g(r) \) [black line in Fig. 3(b)], which for \( N = 2 \) is the square modulus of the relative-motion wave function. Since \( g(r) \) is the probability that two electrons are at distance \( r \), its well developed maximum around \( r \approx 1.5 \) [in units of the characteristic QD length \( \ell_{QD} = (\hbar/m^* \omega_0)^{1/2} \)] identifies the inter-electron equilibrium distance of the molecule. Besides, the plots of \( C_1 \) and \( C_2 \) in Fig. 3(a) [Fig. 3(b)] show respectively one
and two additional nodal surfaces in the $xy$ plane (nodes along $r$), pointing to the excitation of respectively one and two quanta of the breathing mode. Note that the molecule is not perfectly formed due to residual electron delocalization $[g(r = 0) \neq 0$ in Fig. 3(b)]. Spin excitations [red lines in Fig. 3(c)] have a different nature: $g(r = 0) = 0$ for symmetry $[g(r) \propto r^{M_{\text{rel}}}$ as $r \rightarrow 0$ and $M_{\text{rel}} = 1]$ and $S_1$ ($S_2$) has no node (one node) at finite $r$, consistently with the the excitation of no (one) breathing mode quantum. A similar analysis in the center-of-mass coordinates confirms the assignments of Fig. 3(c).

Figure 4 summarizes the evolution of the measured excitations as a function of $I_{\text{HeNe}}$. The data show that: (i) $\hbar \omega_0$ increases at lower $I_{\text{HeNe}}$ since the modes are blue shifted $[22]$; (ii) $N$ increases for $I_{\text{HeNe}} < 0.68 \text{ W/cm}^2$ since the two low-lying spin and charge modes do not split significantly, contrary to the expected behavior for $N = 2$ $[29]$. Indeed the spectra at $I_{\text{HeNe}} = 0.16 \text{ W/cm}^2$ agree well with the CI excitation energies for $N = 3$, shown as up triangles in Fig. 4. The best agreement with the experimental data is obtained with $r_s = 2.18$ ($\hbar \omega_0 = 2.8 \text{ meV}$), a value significantly smaller than the one for $N = 2$. This decrease of $r_s$ weakens electron correlations and it stops at $I_{\text{HeNe}} = 0$. Then the spectra resemble those obtained in similar QDs studied previously by us $[9, 16]$ and interpreted as due to the low-lying spin and charge excitations of $N > 3$ electrons with $r_s \approx 2$.

In conclusion, we have reported the characteristic excitations of two correlated electrons (a molecular dimer) in nanofabricated GaAs quantum dots. Charge tunability with single electron accuracy is obtained by photodepletion. These studies open new opportunities for understanding the ground states of few electrons in semiconductor quantum dots in the regime of strong Coulomb interaction and weak confinement potential.

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Supplementary Discussion: Theoretical analysis of Wigner localisation in a broad range of $r_s$

Since the electron dimer is a finite system, there is no clear-cut transition to the WM state as $r_s$ increases. A good indicator of the crossover to the WM exploits the irrelevance of the spin degree of freedom in this classical limit $[4] [6]$. Specifically, the pair correlation functions $g(r)$ for the ground and $S_1$ states shown in Fig. 3(c) must overlap in the WM limit. In fact, these represent the square moduli of the relative-motion wave functions. Since in the WM limit $S_1$ and the ground state differ only for the excitation of one rotational rigid-body quantum plus a center-of-mass oscillation (cf. main text), the relative motion —and hence $g(r)$— is left unchanged. However, the two states have different spin symmetries: $S_1$ and the ground state are a spin triplet and singlet with one node and no node for $g(r)$ at $r = 0$, respectively. Therefore the $g(r)$’s are generically different and overlap only in the WM limit, when all the wave function weight is removed from the origin. Consistently, electrons fully localise in space.

This crossover to the WM is analyzed quantitatively by evaluating the functional distance $d(r_s)$ between the pair correlation functions $g(r)$ of the ground state $[g_{GS}(r)]$ and $S_1$ $[g_{S_1}(r)]$:

$$d(r_s) = 2\pi \int_0^\infty \! dr \sqrt{|g_{GS}(r) - g_{S_1}(r)|}.$$ (1)

$d(r_s)$ is plotted as a function of $r_s$ in Fig. 5. Note that the evolution is smooth $[8] [9]$, with $d(r_s) \rightarrow 0$ only for $r_s \rightarrow \infty$. Nevertheless, one may locate the crossover to the WM state at $r_s \approx 4$ where the slope of $d(r_s)$ significantly changes. This estimate is consistent with the theoretical analysis of Ref. 6. Note the proximity of the experimental value (filled black circle in Fig. 5) to
FIG. 5: Functional distance $d(r_s)$ vs $r_s$ as obtained by CI calculations. The unit length is $\ell_{\text{QD}} = (\hbar/m^*\omega_0)^{1/2}$. The continuous line is a guide to the eye.

FIG. 6: CI lowest-state energy of the electron dimer vs relative angular momentum $M_{\text{rel}}$ for $r_s = 3.39$. The continuous red line is the lowest rotational band of a perfectly formed WM (with an arbitrary offset).

the crossover region, which confirms our interpretation in terms of the WM dimer.

In a previous work [9] we have demonstrated the emergence of rotational bands for $N = 4$ at relatively high density ($r_s = 1.71$). This transition to a correlated molecular state is distinct from the formation of the WM, which implies electron localisation and occurs at larger values of $r_s$ [30]. This may be seen in Fig. 5 where the value of $d(r_s)$ for $N = 2$ corresponding to the same density as in Ref. 9 (red filled circle in Fig. 5) is significantly distant from the crossover region.

The assignment of rotational bands in Ref. 9 identifies states with different values of $M_{\text{rel}}$ and same profiles of $g(r)$. A feature peculiar of the WM is the parabolic dispersion of the rotational band, $\sim \hbar^2 M_{\text{rel}}^2/2I$, with the momentum of inertia $I$ univocally fixed by $r_s$. This may be seen in Fig. 6 where we compare the CI lowest-energies of the dimer for each $M_{\text{rel}}$ sector (black circles) with the parameter-free WM rotational band (continuous red line). The agreement is satisfactory up to $M_{\text{rel}} = 2$ and then deteriorates as $M_{\text{rel}}$ increases. The rationale is that the WM melts as the energy ($E_{\text{CI}}$) increases, the larger $r_s$ the higher the energy. According to Ref. 10 the results of Fig. 6 show that the WM is "floppy" for this value of $r_s$.

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The criterion we have used in Ref. [9] to investigate the formation of the WM (cf. Fig. 4c in [9]) relies on the spin polarization of the four-electron ground state and therefore it is not applicable to the unpolarized dimer ground state.