Probing collective modes of correlated states of few electrons in quantum dots

Low-lying collective excitations above highly correlated ground states of few interacting electrons confined in GaAs quantum dots (QDs) are probed by resonant inelastic light scattering. We highlight that separate studies of the changes in the spin and charge degrees of freedom offer unique access to the fundamental interactions. The case of QDs with four electrons is found to be determined by a competition between triplet and singlet ground states that is uncovered in the rich light scattering spectra of spin excitations. These light scattering results are described within a configuration-interaction framework that captures the role of electron correlation with quantitative accuracy. In these studies, inelastic light scattering methods emerge as powerful probes of collective phenomena and spin configurations in QDs.

Vittorio Pellegrini

vp@sns.it

Collaborators
G. Goldoni
M. Gurioli
S. Kalliakos
E. Molinari
L. Pfeiffer
A. Pinczuk
M. Rontani
A. Singha
A. Vinattieri
K. West

Despite the large body of theoretical studies of the roles of correlation in QDs [1,2], probing experimentally the fundamental interactions underlying the emergence of a collective correlated state of few electrons in a QD represents a very demanding task. In our recent work we have shown that energies of low-lying spin and charge excitations of few electrons in nanofabricated GaAs QDs carry the signatures that identify electron correlation [3]. In the case of four electrons in the correlated regime the low-energy excitations manifest the formation of an electronic molecular state that has characteristic roto-vibrational modes [4]. These electron molecular excitations are revealed in the light scattering experiments, but are not directly accessible by transport methods nor in photoluminescence experiments. We recall that within time-dependent perturbation theory the resonant inelastic light scattering cross section incorporates two inter-band optical transitions close to the material band gap. Due to the conservation of energy, the energy difference between the incident and scattered photons is the energy of the QD excitation. Selection rules dictate that the active modes in light scattering experiments in QDs correspond to multipole inter-shell excitations (i.e., corresponding to even change in the angular momentum M) between Fock-Darwin (FD) shells with the same orbital parity.

As in conventional molecules where the excitation spectra have a roto-vibrational character, in the strong-correlation regime where the Coulomb interaction rigidly fixes the relative positions of the electrons in the QD, the electronic excitations have character linked to either a vibrational configuration (associated with changes in the relative-motion wave function) or rotational state (with changes in the angular momentum). This is in contrast with the weak-correlation regime where the Coriolis force tends to mix the rotational and vibrational motions. A direct evidence for the formation of the electron molecule would be the independence of the vibrational modes from the rotational state of the electron system as a whole. To this end tuning of the ground state angular momentum M is needed to scan the roto-vibrational spectrum. To probe the ground state angular momentum we performed inelastic light scattering experiments on a QD array in a relatively small magnetic field perpendicular to the QD plane [4]. This procedure enables tuning the total angular momentum of the ground state from $M = 0$ to $M = 2$ (and from total spin $S = 1$ to $S = 0$) (see upper-right part of Fig.1). 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 $n_e = 1.1 \bullet 10^{11}$ cm^{-2} and a mobility of $2.7 \bullet 10^6$ cm^2/Vs. QD arrays with sizes

100 X 100 mm containing 10^4 identical single QD replica [5], were defined by electron beam lithography and dry etching (see the SEM picture in Fig.1).

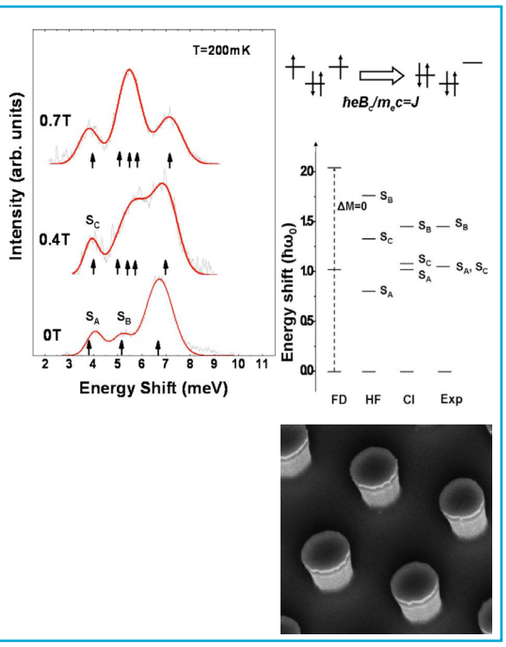

Fig. 1
Left panel: Light scattering spectra of spin excitations at different magnetic fields (the SEM picture of a quantum dot sample is also shown). Red lines are fits to the experimental data with Gaussians. At zero field the four-electron ground state is a spin triplet. As the magnetic field is turned on, when the cyclotron energy equals the exchange interaction contribution, the field induces a ground-state spin transition from the triplet (M=0,S=1) to a singlet state (M=2,S=0), (see top-right panel where the three lowest energy FD levels and electron occupation are shown). In our case, the transition occurs at ~ 0.27 T. Lower right panel: Calculations of spin excitation energies. Within CI approach, both S_A and S_C transitions occur at ~ $\hbar\omega_0$ (the energy quantum of the lateral harmonic potential, which is 3.75 meV in this particular case), and therefore strongly deviate from the Hartree-Fock calculation. The latter predicts a large exchange-energy splitting between S_A and S_C of $J \sim (\hbar\omega_0)^{1/2}$ (around 1.5 meV).

We plot in Fig.1 the spin excitations at different magnetic fields. The calculated energies of the spin modes using the Configuration-interaction (CI) model are indicated as black arrows. At zero field, the spin excitation at ~ 5.2 meV is identified as the Tripet-to-Singlet intershell monopole (ΔM=0) excitation (TS, here labeled S_B) confirming the four-electron population in the QDs [3]. The large intensity increase of the TS mode beyond 0.3 T is due to the emergence of three closely spaced spin excitations from the new ground state (with total spin S = 0 and M = 2) and provides the evidence of the ground state transition from (M = 0, S = 1) to (M = 2, S = 0). The key finding is that the lowest-energy spin excitation, that is, S_A for the triplet ground state and S_C for the singlet ground state, does not shift as we go through the ground-state transition. As pointed out above, this is precisely the molecular signature in the QD, where the rigid rotation of the electrons is decoupled from the relative-motion dynamics. This experimental result is in sharp contrast with that theoretically expected in the absence of correlation (see lower-right panel in Fig. 1).

These experimental results combined with refined theoretical analysis demonstrate the large sensitivity of inelastic light scattering methods in applications that uncover and monitor major impacts of electron-electron interactions in QDs with few-electrons. These are new venues that might provide much needed control of fundamental Coulomb interactions at the nanoscale.

References

[1] S. M. Reimann, M. Manninen, Rev. Mod. Phys. 74 (2002) 1283.
[2] A. Ghosal *et al.*, Nature Phys. 2 (2006) 336.
[3] C. P. García *et al.*, Phys. Rev. Lett. 95 (2005) 266806.
[4] S. Kalliakos *et al.*, Nature Phys. 4 (2008) 467.
[5] S. Kalliakos *et al.*, Appl. Phys. Lett. 90, 181902 2007.