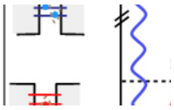

A new spectroscopy to unravel spin states in GaAs/AlAs quantum dots

The on-demand generation of entangled photons is an essential achievement leading to important applications for quantum technologies. In semiconductor quantum dots (QDs), the radiative biexciton-exciton cascade is successfully employed. Resonant two-photon excitation (TPE), coherently preparing the biexcitonic state, significantly improves the entangled photon-pairs emission rate.
Members of INSP have found a novel use of this technique (TPE) to resonantly probe the spin properties of excited biexcitonic states - impossible to be detected by more conventional techniques - where the two electron-hole (e-h) pairs occupy distinct orbital levels.

TPE is a nonlinear optical absorption process providing the simultaneous creation of two e-h pairs in the QD (fig.(a)). While this technique was restricted to the lower energy valence and conduction orbitals, its extension to excited levels allows to investigate the spin properties of composite biexcitons constructed with one e-h pair on the s_c and s_v and a second on the p_c and p_v conduction and valence levels- where s and p denote the confined electronic states of the dot having the same symmetry as their atomic S and P- counterparts-. The radiative recombination of the pc-pv pair is detected at higher energy and leaves an e-h pair on s_c and s_v (fig.(a)). A doublet structure is revealed on fig.(b), and it is related to a fine structure depending on the spin configurations.

The determination of the fine structure requires to consider all the spin configurations resulting from these 4 interacting e & h particles, i.e. a total of 2^4=16 states, 9 of which are constructed with triplet states, and the other 7 with singlet spin states, either electron or hole.
The spectral doublet results from the recombination of triplet states identified by eliminating the so-called «dark» configurations (i.e. not coupled to light) on the one hand, and by discarding the singlet states that relax efficiently via non-radiative processes on the other hand. In particular, the lowest energy emission line is attributed to electron and hole triplet states with zero spin projections. This latter signal serves as a probe to detect the higher energy states whose emission is almost undetectable.

Fig.(c) shows the evolution of the luminescence intensity when the laser energy finely scans the two-photon resonance. The main peak associated with the TPE is accompanied by a series of smaller amplitude resonances. The first two resonances were attributed successively to the singlet states of holes and electrons. The TPE provides the way to address resonantly these singlet states, which thermalize efficiently to the triplet states and explains the succession of resonances observed in Fig. (c) . It results that the exchange integrals $J_{t\text{-}t}$ for the hole and e-e $J_{e\text{-}e}$ for the electron can be evaluated without measuring the weak luminescence of the singlet states.

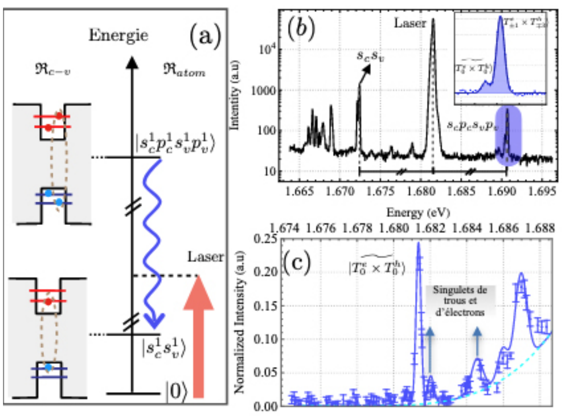

Figure
(a) Level diagram representing the resonant 2-photon excitation between the exciton and the biexciton. Both «conduction-valence» $\mathfrak{R}_{c-v}$ and «atomic» notation $\mathfrak{R}_{atom}$. are used to represent these quasi-particles to improve clarity
(b) Luminescence spectrum showing the experimental implementation of the resonant 2-photon excitation. The insert magnifies the luminescence from the p_c-p_v excited levels, where the doublet appears
(c) Luminescence intensity of one of the triplet states as a function of the laser energy.

This new spectroscopy probes the singlet states very efficiently, and has already highlighted many-body effects between carriers, which are important in GaAs/AlAs dots since the strength of the quantum confinement remains relatively weak. In addition, it makes possible the coherent driving of localised carriers on higher energy orbital states, providing new degrees of freedom for all-optical spin control protocols in quantum dots.

Reference

"Unveiling the spin-singlet states of two electron-hole pair complexes using two-photon excitation in a GaAs/AlAs quantum dot"
S. Germanis, P. Atkinson, A. Bach, R. Hostein, R. Braive, M. Vabre, F. Margaillan, M. Bernard, V. Voliotis , and B. Eble
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