Peculiar many-body effects revealed in the spectroscopy of highly charged quantum dots

M. EDIGER[1], G. BESTER[2]*, A. BADOLATO[3], P. M. PETROFF[3], K. KARRAI[4], A. ZUNGER[2] AND R. J. WARBURTON[1]*

[1]School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh EH14 4AS, UK
[2]National Renewable Energy Laboratory, Golden, Colorado 80401, USA
[3]Materials Department, University of California, Santa Barbara, California 93106, USA
[4]Center for NanoScience and Department für Physik, Ludwig-Maximilians-Universität, 80539 München, Germany
*e-mail: G.Bester@fkf.mpg.de; R.J.Warburton@hw.ac.uk

Published online: 14 October 2007; doi:

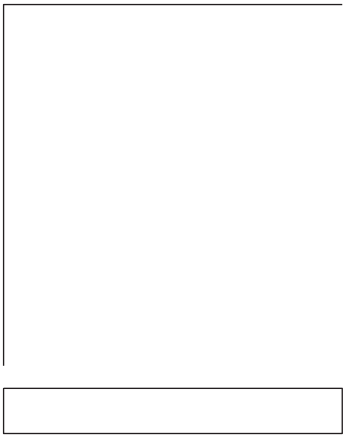

Figure 3 Calculated leading configurations for charged excitons. Left: one hole and N electrons; right: one electron and N holes. For the positively (negatively) charged states, the single electron (hole) in the initial state is in the lowest electron (hole) state. The last line indicates whether fine-structure effects are predicted (Y) or not (N).

captures the multiband, intervalley and spin–orbit interactions

Figure 4 Measured photoluminescence spectra from dot A for different exciton charges. Photoluminescence intensity (840 s integration time) is plotted against energy for V_g corresponding to the centre of each charging plateau. Individual peaks are labelled: red circle denotes a photoluminescence peak from the particular exciton with charge n; blue circle emission from exciton with charge n − 1; black circle biexciton-related emission; green circle emission from an excited initial-state configuration.

predict that the hole charging sequence is perturbed by the presence of the electron: without the electron, the second *p* state is not occupied at all[15]. Curiously, the predicted initial configuration is open shell, yet the photoluminescence is almost unpolarized, both in the experiment and in the theory, signifying a zero-spin-state coupling of the unpaired holes. Small fine-structure e□ects

are still present in the theoretical results, originating from the admixed (27%) configurations in the initial state; these effects are beyond the experimental resolution. Theoretically, the signature of the open-shell X^{5+} is the presence of a multitude of peaks with comparable intensity, whereas the closed-shell X^{5+} configuration has one strong peak accompanied by many very weak transitions. Experimentally, there are several strong photoluminescence lines, strongly supporting the open-shell configuration. The non-Aufbau filling of hole states continues for X^{6+} where p_2 is left half empty. A polarized experimental spectrum (dots A and B) with a few peaks agrees with the theoretical prediction but the X^{6+} photoluminescence is very weak.

(2) *Non-perturbative Coulomb interactions.* A perturbation treatment of the Coulomb interactions predicts a blue-shifted X^{1+} on the basis of a red-shifted X^{1-} (ref. 18). Indeed, our calculated Coulomb energies[17] $J_{hh} = 25.9\,\text{meV} > |J_{eh}| = 25.3\,\text{meV} > J_{ee} = 24.9\,\text{meV}$ lead to a $J_{hh} - J_{eh} = 0.6\,\text{meV}$ blue-shift of X^{1+} with respect to X^0. However, this effect is countered by the non-perturbative mixing of the $h_S^2 e_S^1$ configuration with other configurations, a mixing that produces an overall red-shift of X^{1+}, a clear feature in both experiment (Fig. 4) and theory (Fig. 2). This simple fact pro

our theory, we extrapolate in Fig. 4 the photoluminescence spectra to zero electric field. This is crucial as it reveals a red-shifted X^{1+}, not a blue-shifted X^{1+} as a cursory inspection of Fig. 1 might suggest. None of the splittings in the experimental photoluminescence depend on electric field demonstrating the validity of this method.