

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

M. EDIGER<sup>1</sup>, G. BESTER<sup>2\*</sup>, A. BADOLATO<sup>3</sup>, P. M. PETROFF<sup>3</sup>, K. KARRAI<sup>4</sup>, A. ZUNGER<sup>2</sup>  
AND R. J. WARBURTON<sup>1\*</sup>

<sup>1</sup>School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh EH14 4AS, UK

<sup>2</sup>National Renewable Energy Laboratory, Golden, Colorado 80401, USA

<sup>3</sup>Materials Department, University of California, Santa Barbara, California 93106, USA

<sup>4</sup>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<sup>15</sup>. 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 effects

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<sup>17</sup>  $J_{hh} = 25.9$  meV  $> |J_{eh}| = 25.3$  meV  $> J_{ee} = 24.9$  meV lead to a  $J_{hh} - J_{eh} = 0.6$  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

simple fact pro

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.