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Polarization-resolved fine structure and magneto-optics of single CdSe nanocrystal quantum dots[1]

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Low-temperature photoluminescence (PL) microscopy of single colloidal quantum dots has proven a very effective tool for probing the emission properties of the band-edge excitons in isolated CdSe nanocrystals (NCs). Past studies employing high spectral resolution have resolved the narrow 'atomic-like' emission lines from single NCs, while separately, polarization-resolved measurements have shown that the $|+1>$ and $|-1>$ bright exciton states are nominally degenerate with transition dipoles oriented isotropically in the plane normal to the crystallographic c-axis of the NC. To date, however, these two powerful techniques have not been simultaneously employed. To this end we constructed a low-temperature (4 K) microscope to measure both polarization- and spectrally- resolved PL of individual nanocrystals. Both orthogonal polarizations (horizontal/vertical linear or right/left circular) are simultaneously recorded to minimize the effects of spectral diffusion and blinking. The data clearly show [1] that many NCs possess a clear bright exciton "fine structure" consisting of two linearly-(and orthogonally-) polarized peaks split in energy by $\delta \sim 1-2$ meV. This splitting is attributed to a breaking of the nanocrystal's cylindrical symmetry, leading to an anisotropic electron-hole exchange that mixes the $|\pm 1>$ bright excitons. Inferred orientation of the NCs will be discussed. Finally, we study the interplay between the anisotropic exchange and magnetic Zeeman energy in single NCs by incorporating a 5 T magnet into the microscope. With increasing magnetic field, the fine structure states become elliptically polarized and eventually approach pure circular polarization in the limit where the Zeeman energy $1/2g\mu_B B > \delta$. We extract the exciton g-factor of individual NCs from the variation of the observed energy splitting with field in this regime.

[1] M. Furis, H. Htoon, T. Barrick, M. Petruska, V. I. Klimov, S. A. Crooker, Phys. Rev. B Rapid Comm. 73, 241313 (2006).

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