Anomalous Circular Polarization of Magneto-photoluminescence from Individual CdSe Nanocrystals

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A Zeeman splitting between otherwise degenerate spin eigenstates under applied magnetic fields is a fundamental quantum mechanical effect that has been studied in many different quantum systems ranging from atoms and molecules to electron-hole excitations (excitons) in both bulk and nanostructured semiconductors. Recently, a significant research effort has focused on field-dependent spin phenomena in semiconductor quantum dots (QDs) motivated by a desire to understand the fundamental spin structure of QD electronic states and by potential technological applications in solid-state spintronics and quantum information processing.

Zeeman effects in self-assembled QDs grown by epitaxial methods have been extensively studied in both QD ensembles and in single QDs using magneto-photoluminescence (PL). In cylindrically symmetric QDs, a magnetic field $B$ applied along the epitaxial growth direction ($\hat{z}$) splits the lowest-energy, spin-degenerate, optically-allowed (“bright”) exciton into two states with net spin projections $+1$ and $-1$. In the Faraday geometry ($\mathbf{k} \parallel \mathbf{B}$), these states emit right- or left-circularly polarized light ($\sigma^\pm$) as depicted in Fig. 1(a). However, many epitaxial QDs possess a significant asymmetry in the X-Y plane, normal to the growth direction $\hat{z}$. In these QDs, the long-range (anisotropic) part of the electron-hole exchange interaction mixes the $|\pm\rangle$ bright states to produce a “fine structure” of two linearly and orthogonally polarized dipoles, $|X,Y\rangle=\left(|+1\rangle \pm |-1\rangle\right)/\sqrt{2}$ with energy splitting $\Delta_{XY}$ at $B=0$. With increasing $B$, the PL polarization from these states evolves from linear elliptical to left/right circular in the limit where the Zeeman energy $E_z=g_e\mu_B B > \Delta_{XY}$ [Fig. 1(b)]. Here, $\mu_B$ is the Bohr magneton and $g_e$ is the bright exciton g-factor. This “traditional” picture has been experimentally confirmed in numerous studies of epitaxially-grown QDs.

Previous experiments have also addressed Zeeman effects in semiconductor nanocrystal quantum dots (NQDs) synthesized by colloidal chemistry. However, until now, magneto-PL methods have been applied exclusively to ensembles of NQDs, where the PL linewidth (>50 meV) is inhomogeneously broadened by the NQD size distribution. As a result, Zeeman splittings between right- and left-circularly polarized PL are not well resolved. In this work, we study polarized PL from individual CdSe NQDs at low temperatures and in magnetic fields to 5 T. The unusually strong anisotropic exchange splitting $\Delta_{XY}$ found in many NQDs dramatically modifies the magneto-PL polarization of the emitting “bright” exciton. We observe two distinctly different and systematic behaviors: In all NQDs exhibiting small $\Delta_{XY}$ (<0.5 meV) we observe a traditional $\sigma^+$-polarized Zeeman doublet, from which we measure, for the first time, the exciton g-factor of single nanocrystals. In contrast, all NQDs exhibiting large $\Delta_{XY}$ (>1 meV) show a highly anomalous PL polarization wherein the lower-energy PL peak becomes circularly polarized with increasing $B$ while the higher energy peak remains linearly polarized. We show that strong mixing between the emitting and absorbing bright-exciton states, which results from anisotropic exchange interactions in these NQD, accounts for this effect.

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