High-Field Magnetoluminescence of Colloidal Mn^{2+}-doped CdSe Quantum Dots

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Introduction

Semiconductors doped with magnetic impurities, often referred to as diluted magnetic semiconductors (DMSs), possess unique magnetic, optical, magneto-electronic, and magneto-optical properties and have generated interest for applications in spin-based electronics.[1] Mn^{2+} is a common dopant for DMSs because it can be incorporated into various II-VI semiconductor lattices and possesses a relatively large magnetic moment (S = 5/2). In small- to intermediate-band-gap materials like CdSe, the Mn^{2+} ligand field excited states lie outside the semiconductor optical gap and the photoluminescence (PL) spectrum is dominated by excitonic emission. The recent availability of colloidal Mn^{2+}:CdSe quantum dots has allowed the strong dopant-exciton magnetic exchange interactions to be examined via zero-field and magnetic circularly polarized luminescence (MCPL). Notably, antiferromagnetic superexchange interactions of nearest neighbor Mn^{2+}-Mn^{2+} dimer pairs influence the magnetic properties of DMSs. Because of antiferromagnetic nearest-neighbor superexchange, the zero-field ground states of these dimers have S = 0 spin, while nearby excited states possess greater net spin (S = 1–5). At large magnetic fields, components of the Zeeman split excited spin states may cross below the S = 0 state, establishing a new ground state with net magnetization. This occurrence is characterized by an abrupt step in the magnitude of the excitonic Zeeman splitting, and five magnetization steps are expected as the antiferromagnetically bound Mn^{2+} pairs "unlock."[2] Additionally, in DMSs like Mn^{2+}:CdSe, the *sp-d* exchange splittings oppose the intrinsic Zeeman splittings, but far exceed the intrinsic splittings at moderate magnetic fields. However, the *sp-d* exchange splittings saturate when the dopant spins are fully magnetized in an applied magnetic field, whereas the intrinsic contributions to the excitonic Zeeman splittings continue to increase linearly. At sufficiently high fields, it is expected that the intrinsic contributions will rival the saturated *sp-d* contributions, and the Zeeman splitting will begin to decrease with increasing field strength. We sought to observe these phenomena at NHMFL.

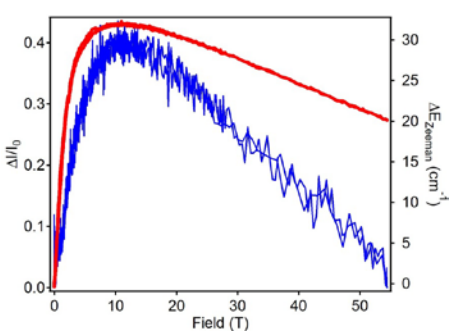

Figure 1. MCPL polarization ratios and excitonic Zeeman splittings for colloidal 0.4% Mn^{2+}:CdSe quantum dots, measured as a function of magnetic field at 1.6 K.

Experimental

Colloidal Mn^{2+}:CdSe quantum dots were synthesized as previously reported.[3] High-magnetic-field MCPL measurements were made with the 60 T long-pulse magnet supported by a 1400 MVA motor generator. Excitation was provided by a diode laser (λ = 405 nm). PL was dispersed using a single monochromator and detected with a liquid nitrogen cooled CCD camera. Circularly polarized PL was detected through a quarter-wave plate and a linear polarizer.

Results

MCPL data were measured for doped CdSe quantum dots with low Mn^{2+} concentrations. We successfully observed decreasing excitonic Zeeman splittings at large magnetic fields (**Figure 1**), allowing the rivalry between intrinsic and *sp-d* Zeeman splittings in colloidal nanocrystals to be analyzed quantitatively. The fields required to observe this effect (>~10T) exceed those available outside the NHMFL. Magnetization steps were sought in a variety of colloidal Mn^{2+}:CdSe quantum dots via MCPL but were not observed.

Acknowledgments

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References

[1] Furdyna, J. K. Ed. Diluted Magnetic Semiconductors, Academic Press; New York, **25** (1988).
[2] Crooker, S. A., *et al.*, Phys. Rev. B, **60**, 2173-2176 (1999).
[3] Beaulac, R., *et al.*, Science, **325**, 973-976 (2009).