Magneto-photoluminescence spectroscopy of bright and dark excitons in isolated semiconducting single-walled carbon nanotubes

Morgane Gandil[1*], Kazunari Matsuda[2], Philippe Tamarat[1] & Brahim Lounis[1]

[1] LP2N, Univ. Bordeaux - CNRS - Institut d'Optique Graduate School, F-33400 Talence, France
[2] Institute of Advanced Energy, Kyoto University, Uji, Kyoto 611-0011, Japan
* morgane.gandil@institutoptique.fr

Abstract

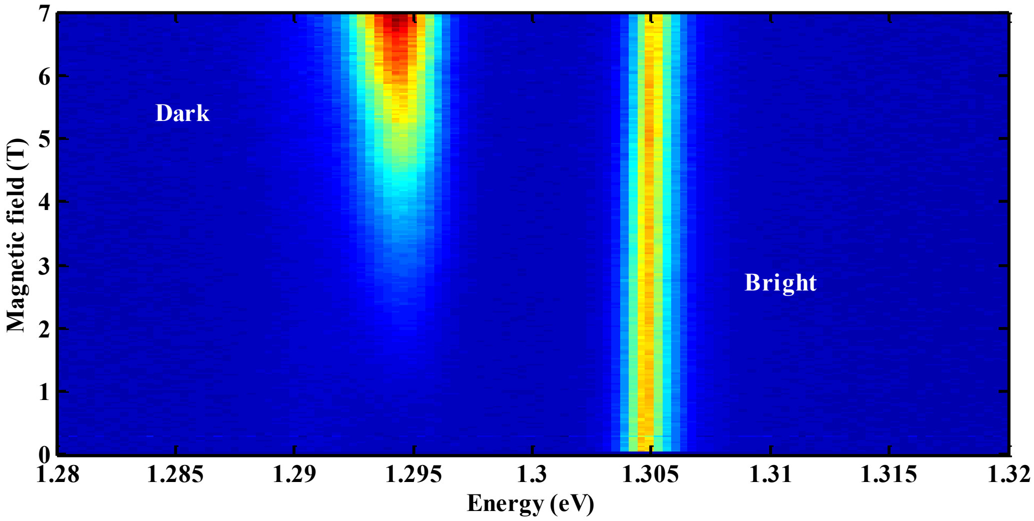
Since the first experimental evidence of photoluminescence of semiconducting single-walled carbon nanotubes (SWNTs) [1], studies have been conducted to investigate the optical properties of these nano-structures, motivated by possible applications in the fields of quantum information, biological labeling, opto-electronics or laser technology.

The unidimensional nature of SWNTs, through the combined effect of the strong spatial confinement and the low coulomb screening, leads to high electron-hole binding energies [1,2]. The photo-excitation of SWNTs results therefore in the formation of strongly correlated electron-hole pairs, so-called excitons, which dominate the photo-physical behavior of these nano-objects. Due to the configuration of the excitonic band structure, the luminescence of semiconducting SWNTs is mainly governed by the two lowest singlet states: the upper one is optically active (bright) whereas the lower one corresponds to a parity forbidden dipole transition (dark). A magnetic field applied along the SWNT axis induces the coupling of these two levels through the Aharonov-Bohm effect. The resultant magnetic brightening of the dark state opened up the field of magneto-photoluminescence spectroscopy [3,4] as a promising way to investigate the photo-physical properties of SWNTs.

Here, we report the study of isolated CVD-grown SWNTs suspended on lithographed trenches of a silicon substrate. Measurements were performed at the single-object level using a home-built confocal optical microscope with a large numerical aperture (NA = 0.95) operating at cryogenic temperatures (down to 2K) and high magnetic field (up to 7T). Photoluminescence spectra and decays of single SWNTs were acquired under various experimental conditions, including different magnetic fields, temperatures and optical excitation frequencies.

As displayed in the figure, the dark state spectacularly brightens with increasing magnetic fields, which reveals the dark/bright energy splitting. From the spectroscopic and time-resolved measurements, relaxation dynamics of the bright and dark excitons and their interactions with the phonons will be discussed.

Figure



Photoluminescence spectra of a single (6,5) SWNT resonantly excited on its S22 transition with a 561nm CW laser under various magnetic fields.

References

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