ULTRAHIGH MAGNETIC FIELD OPTICAL STUDY OF SINGLE-WALLED CARBON NANOTUBES FILM

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Abstract

Excitons in Single-Walled Carbon Nanotubes (SWNTs) have emerged as an ideal candidate for exploring one-dimensional (1-D) exciton physics. Exciton states which dominate optical properties of SWNTs even at room temperature, are not clarify yet. The optical absorption spectra of aligned SWNTs films under ultra high magnetic fields up to 190 T are examined to investigate this issue. Shifting and splitting of the absorption peaks due to Aharonov-Bohm effect was observed clearly above 80 T in the configuration where the magnetic fields were applied in parallel to the alignment of SWNTs. The lowest singlet exciton state has been determined through the analysis of energy splitting of excitons by the application of magnetic fields.

Keywords: blue shift, optically active, optically inactive, red shift, single-turn coil system,

Introduction

One-dimensional (1-D) excitons have been the subject of numerous theoretical studies over the last several decades [1]. Their properties are predicted to differ significantly from excitons in higher dimensions. Recently, excitons in Single-Walled Carbon Nanotubes (SWNTs) have emerged as an ideal candidate for exploring 1-D exciton physics due to their extreme quantum confinement with the resultant huge binding energies as well as their relatively low degrees of disorder.

In SWNTs, the short-range interactions will split the exciton states to be bright exciton or optically-active and dark exciton or optically-inactive. One of the important issues is which the lower singlet state to be assigned, optically active or optically inactive. However, the assignments of the lower exciton states are not clarify yet. Our research aims to determine the assignment experimentally by performing ultrahigh magnetic field optical absorption measurement in the visible spectral region for a stretch-aligned SWNTs film using a single-turn coil system.

Experimental

The stretch-aligned semiconducting SWNTs films in which the (6,5) and (7,5) chiral stuctures exist dominantly are used in this experiment. The films were prepared following the procedure developed by Y Kim, et al. [2]. CoMoCAT SWNT (SouthWest NanoTechnologies Inc.) was used as raw nanotube. Gelatine from alkali-processed bovine bone obtained from Wako Chemicals. Dispersion in D_2O (Deuterium Oxide, heavy water) solution of sodium dodecyl sulfate (SDS) by sonication and ultracentrifugation were performed following the procedure developed by M. J. O'Connel et al. [3].

The high magnetic fields up to 190 T in this study were generated by using the single-turn coil system of Megagauss Laboratory of the Institute for Solid State Physics (MGL

ISSP) of the University of Tokyo. Optical absorption spectra in the wavelength range between 520 nm and 700 nm (between 1.77 eV and 2.38 eV) under ultra-high magnetic fields up to 190 T were measured using a streak spectrometer with an image intensifier and a charge-coupled-device (CCD) camera. A xenon flash lamp was employed as a light source. Incident light and transmitted light were delivered using optical fiber system. The magneto-absorption measurements were performed in the Voigt configuration. A linear polarizer was inserted on the incident side of the SWNT/SDS/gelatin film. The polarization direction of the polarizer was chosen to be parallel to the alignment (*A*) of SWNTs in the film in order to exclude components of absorption by unaligned SWNTs from transmitted light. Magnetic fields (*B*) were applied in parallel or in perpendicular to the SWNT alignment.

Result and Discussion

Our single-turn coil system yield the pulse width around 7 μs (Fig. 1).

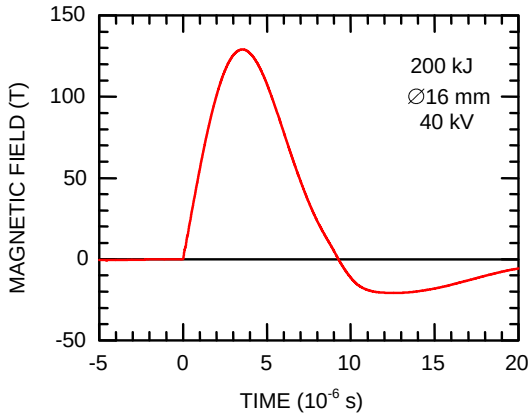
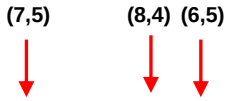


Fig. 1. Field pulse shape of a 200 kJ (40 kV) capacitor bank on to a 16 mm bore copper coil.

Absorption spectra in coincide time with the appearance of the magnetic field were obtained by regulate the Xenon flash lamp, magnetic field generation, and streak camera by multi channel pulse generator. A representative of absorption streak image and pulse ultrahigh magnetic field graph is shown in Fig.2 below.



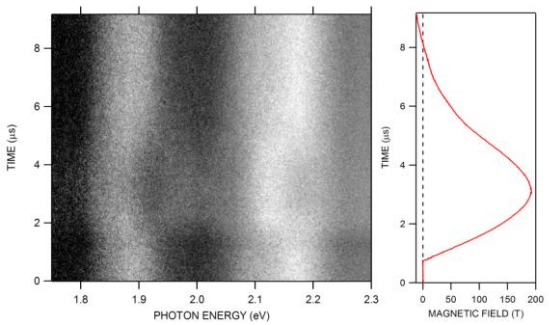


Fig. 2. Absorption streak image of CoMoCAT/SDBS/Gelatin Stretch-Aligned (x4) Film under 190 T pulse magnetic field.

From this obtained data, the optical absorption spectra might be taking from the each absorption image at the time of 3 μs. Magnetic field expansions of absorption spectra for stretch-aligned SWNT films in the arrangement of *B*//*A* (magnetic field parallel to the alignment) is presented in Fig. 3.

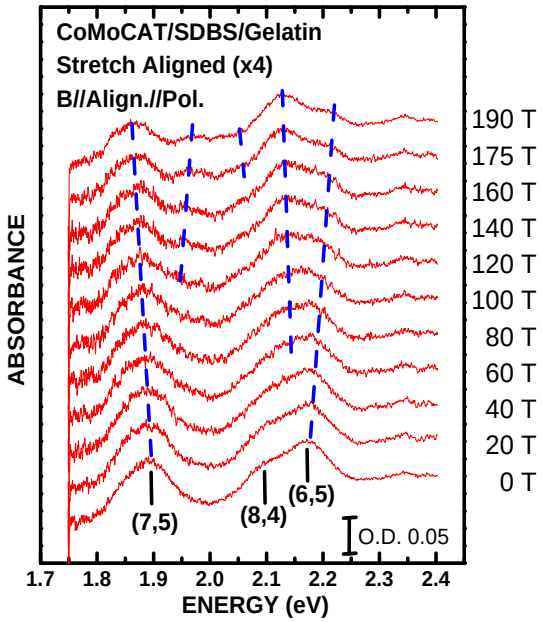


Fig. 3. Magneto-absorption spectra of SWNTs film up to 190 T.

In Fig.3 above, three dissociate absorption peaks are recognized. The three peaks observed around 1.9 eV, 2.1 eV and 2.18 eV can be assign to interband transitions in the second-subband gaps of semiconducting tubes with chirality (7,5), (8,4) and (6,5) respectively. At magnetic fields below 40 T, no significant change was recognized in these three peaks. However, broadening of the peaks was observed clearly above 80 T.

On the other hand, in the previous study with the HiPco sample as raw nanotube, there are no noticeable variation is observed in the magneto-absorption spectra in the arrangement $B \perp A$ up to 110 T (Fig. 4.) [4]. Therefore, the remarkable change of the absorption spectra is found only in the case that magnetic field passes through SWNTs aligned in the sample, which is consistent to the AB effect predicted theoretically.

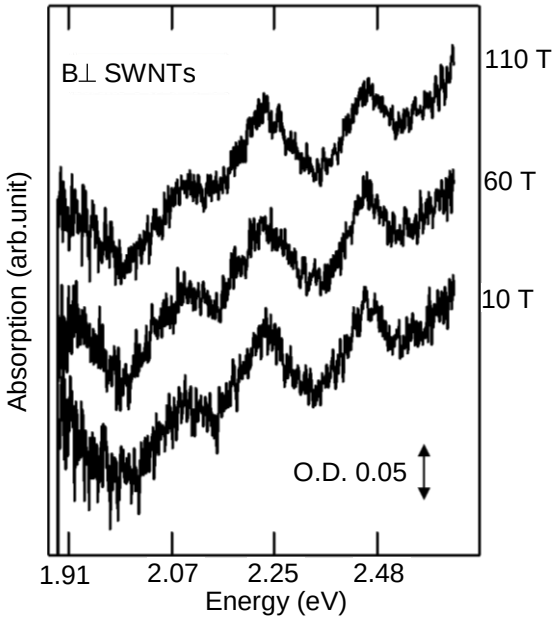


Fig. 4. Magnetic field expansions of absorption spectra for stretch-aligned SWNTs film in the arrangement $B \perp A$ [4].

Acquirement the original peak component of the magneto-absorption spectra (Fig. 3) was performed by deconvolution analysis. Curve plotting of (7,5), (8,4) and (6,5) peak components of SWNTs was depicted in Fig. 5.

Fig. 5. shows that both of absorption peaks, (7,5) and (6,5) semiconducting SWNTs, exhibited broadening and shift. Moreover splitting was observed to emerge with increasing magnetic field. Changing of absorption peaks originated from second sub-band exciton in semiconducting SWNTs was perceived very clearly with increasing magnetic field in parallel to the alignment of SWNTs (Fig. 3.).

For the (7,5) SWNTs, the absorption peak observed at 0 tesla shifts toward the *lower* energy side (red-shift) and a new one appears on the *higher* energy side. Whereas for the (6,5) SWNTs, the absorption peak observed at 0 tesla shifts toward the *higher* energy side (blue-shift) and a new one appears on the *lower* energy side (guided by the blue dashed line in Fig. 3.). On the other hand, similar to the previous experimental result (Fig. 4.), there are no modification spectra has been detected by the application of magnetic fields perpendicular to the alignment of SWNTs up to 200 T, which suggests that the spectral changes mentioned above are attributed to the AB effect.

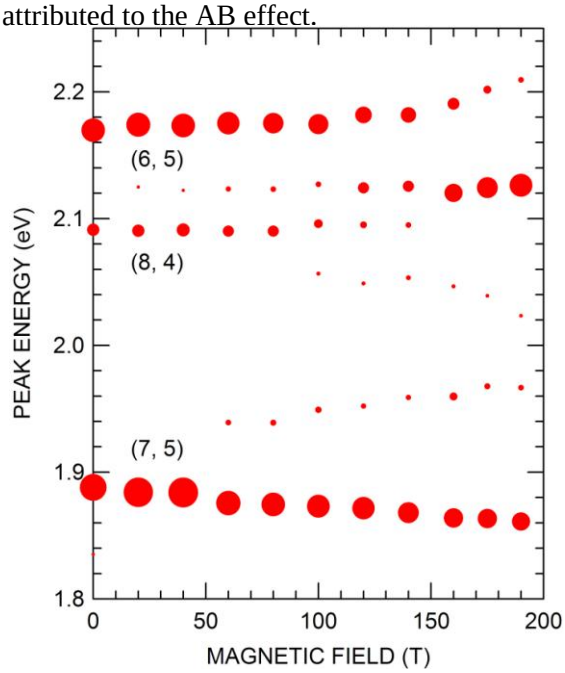


Fig. 5. Magneto-absorption spectra of SWNTs film up to 190 T in the view of peak components.

These experimental results prove that the lower energy side in the case of the singlet splitting of the second sub-band excitonic state is assigned as the "bright" excitonic state in (7,5) SWNTs and the "dark" one in (6,5) SWNTs. This configuration is different from that reported in the other magneto-luminescence studies where the "dark" exciton situates at lower energies in all of the investigated SWNTs [1, 5-7].

Conclusion

Study of the exciton state splitting in SWNTs can be carried out by measurement the magneto-absorption spectra of SWNTs sample under ultra high magnetic field. The (7,5) semiconducting SWNTs exhibited the red-shift and a new peak appears on the *higher* energy side. Whereas the (6,5) SWNTs exhibited the

blue-shift and a new peak appears on the *lower* energy side.

These experimental results prove that the lower energy side in the case of the singlet splitting of the second sub-band excitonic state in (7,5) and (6,5) SWNTs are assigned as the "bright" and "dark" excitonic state, respectively. This configuration is different from the investigation results in the other magneto-luminescence studies where the "dark" exciton situates at lower energies in all of the SWNTs sample [1, 5-7].

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