Aharonov-Bohm Exciton Splittings in Carbon Nanotubes in Ultra-High Magnetic Fields

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Abstract. The Ajiki-Ando (A-A) splitting of single-walled carbon nanotubes was observed by the magneto-absorption measurements conducted up to a very high magnetic field, 78 T for PFO-samples. The well-resolved absorption spectra from the $E_{11}$ transitions in the PFO-samples showed a clear A-A splitting. The electro-magnetic flux compression method was used for generation of the field up to 360 T, where the absorption spectra of the $E_{22}$ transition of the HiPco samples were measured by the streak spectroscopy. Parameters for the A-A splitting were determined for different chirality of each case.

1. Introduction

A single-walled carbon nanotube (SWNT) is well known for showing up splitting of the energy bands upon applying an external magnetic field parallel to the tube axis, which is known as the Aharonov-Bohm effect, and the splitting is also called Ajiki-Ando (A-A) splitting [1]. Observation of the A-A splitting has been reported in various SWNTs by many authors either by means of the absorption [2] or by the photoluminescence (PL) spectra [3].

Recently, excitonic effects are recognized as very crucial in understanding the fundamental optical transition. Owing to interplay between the inter- and intra-K-K valley short range scattering, the exciton states become very complicated with 16 split states of the bright and dark excitons [4]. Application of a magnetic field causes a mixing of both states, and the complicated exciton states are expected to be clarified experimentally. So far, the dark exciton states are identified by magneto-PL in ensemble samples [5,6] or by micro-magneto-PL [7] in a single SWNT.

Aiming at observing the A-A splitting and the behaviors of these exciton states, we attempted the magneto-optical absorption measurements up to an ultra-high magnetic field. An oscillator strength of the optical transition is directly deduced from the absorption spectra, but not necessarily from those of PL.

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2. Experimental
The samples used are micelle-suspended HiPco or PFO-SWNTs dispersed in a liquid. We have performed measurements in ultra-high magnetic fields using the electro-magnetic flux compression (EMFC) techniques [8] up to 360 T, and also by a newly developed giant single-turn coil (GSTC) method up to 80 T [9].

2.1. PFO-SWNT up to 80 T
We attempted near-infrared magneto-optical absorption measurements to the PFO (poly(9,9-dioctylfluorenyl-2,7-diyl)) SWNTs [10] at a room temperature. The PFO-SWNTs show a very sharp and well-defined absorption spectra with respect to the chirality of SWNTs. We first focused on the first subband $E_{11}$ absorption spectra showing sharp peaks, observed in the near-infrared region. We have performed this work to study the exciton absorption spectral change in the region of a magnetic field where the exciton exchange interaction is manifested below 100 T.

2.2. HiPco-SWNT up to 360 T
We have conducted magneto-absorption measurements of HiPco SWNT samples in extremely high magnetic fields above 300 T by the EMFC system. In principle 700 T is now possible to generate in a bore of 6 mm. Due to restriction of the optical alignments around the sample cell, the large diameter of the sample holder limits the highest achievable magnetic field for the measurements. Fast rise time of the magnetic field (2 $\mu$s from 100 T to 360 T) also restricts the optical exposure time. The streak spectroscopy (Hamamatsu Photonics Co. Ltd.) is only the available methods of detecting the optical transmission signal during such a short time. Only the visible light region is detectable, which enable us to measure the interband $E_{22}$ transitions.

3. Results and Discussion
Well resolved absorption spectra are obtained for the PFO-SWNTs of the chirality (8,6), (7,6), and (7,5). The absorption peak is well resolved each other in the absence of a magnetic field. Upon applying magnetic field up to 53 T by a non-destructive pulse magnet, a new peak appeared at lower energy side of the main peak as seen in Fig.1. However, peak separation is not enough for the chirality (7,6) and (7,5) to undergo a process of reliable spectral deconvolution. The peak separation becomes more unambiguous by a further increase of the fields up to 78 T generated by the GSCT. Further increase of the field broadens the spectra due to a substantial contribution from those of the randomly oriented tubes with respect to the direction of magnetic field.

![Figure 1. Magneto-absorption spectra showing well-defined AB splitting, measured up to 78 T for the chirality (8,6), (7,6), and (7,5), respectively, in PFO-SWNTs.](image-url)

In order to estimate the peak splitting against an effective magnetic field $B_{||}$, we need to consider the randomness of the tube orientation against a magnetic field as well as the field induced dynamical
orientation of the ensemble nano-tubes in an aqueous solution subjected to a pulse magnetic field [11].

Details of evaluation for $B_r$ from the observed peak separation will be discussed elsewhere. Figure 2 is a plot of the results of the peak splitting against $B_r$ after the corrections. Due to short time nature of the GSCT in comparison to that of the long pulse magnet, $B_r$ becomes much less. The peak splitting against $B_r$ was fitted by the relation employed by other authors [6,7], and the results are shown by dotted lines in Fig. 2. The values of the dark and bright exciton splitting $\Delta_{bd}$ and of the coefficient representing the A-A splitting $\mu_{ex}$ (defined by $\Delta_{A-A}(B) = \mu_{ex} B_r$) were listed in Table 1 together with the positions of the dark and bright exciton in energy known from the relative intensity of the split peaks.

Figure 2. The A-A splitting observed for the chirality (7,5) of PFO-SWNTs. Circles are from the long pulse magnet, and triangles are from the GSTC. The dotted lines are from calculation. Values of magnetic field generated by the GSTC is denoted each by numbers for data of triangles.

Figure 3 is a plot of the time evolution of a magnetic field produced by the EMFC and an obtained streak image of the absorption spectra of the HiPco SWNTs for the chirality (10,2) (9,5) and (7,5). Owing to the broadness of the absorption spectra of the $E_{22}$ transition in comparison to that of $E_{11}$ and also to the peak overlaps of each chirality, spectral resolution of the peak splitting in magnetic fields is less qualified than that in Fig.1. However, the substantial peak splitting is observed, which amounts to about 160 meV at $B_r = 260 \text{T}$. The results of the splitting against $B_r$ is summarized in Fig. 4, where the characteristic parameters are evaluated from the fitting, and results are tabulated in Table 1.

Figure 3. The signal of a magnetic field produced by the EMFC, and the magnet coil (inset). The streak image of the absorption spectra of the HiPco SWNTs with the chirality (10,2),(9,5), and (7,5).
4. Summary

We have shown the results of the magneto-absorption spectra of PFO- and HiPco-SWNTs. The absorption spectra of the $E_{11}$ transition for PFO samples up to 78 T and those of the $E_{22}$ for HiPco samples up to 360 T were measured. The important parameters associated with the A-A splitting were determined for several different chirality samples. The values of $\mu_{ex}$ determined from the experiments are always almost 70% of those of the theory, $\mu_{th}$. The values of the bright-dark exciton splitting as well as the position of each state were determined from the spectral fitting owing to the A-A splitting.

Table 1. Results of the parameters from the A-A splitting. $d$ denotes the diameter of the nanotube.

| Sample | Transition | Chirality | $d$ [nm] | $\mu_{ex}$ | $\mu_{th}$ | $\mu_{ex}/\mu_{th}$ | $\Delta_{bd}$ [meV] | Lower State |
|--------|------------|-----------|----------|-----------|-----------|-----------------|-----------------|-------------|
| PFO    | $E_{11}$   | (7,5)     | 0.83     | 0.66      | 0.93      | 0.71            | 10.4            | Dark        |
|        |            | (7,6)     | 0.89     | 0.71      | 0.98      | 0.72            | 9.8             | Dark        |
| HiPco  | $E_{22}$   | (7,5)     | 0.83     | 0.56      | 0.75      | 0.75            | 32              | Bright      |
|        |            | (10,2)    | 0.88     | 0.57      | 0.77      | 0.74            | 55              | Bright      |
|        |            | (9,5)     | 0.97     | 0.68      | 0.98      | 0.69            | 51              | Dark        |

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