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Magneto-Absorption Spectra from Selected Chirality of Single-Walled Carbon Nanotubes

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Abstract Magneto-absorption spectra of single-walled carbon nanotubes (SWNTs) in the region of near infrared were measured under high magnetic fields up to 54T. We succeeded to observe clear peak shifts and splittings for four different chiralities of SWNTs, (7,5), (7,6), (8,6), and (8,7). We employed PFO (poly(9,9-dioctylfluorenyl-2,7-diyl))-SWNTs. Absorption peaks are very sharp and we could observe well-defined peaks of each chirality with less mixture of the other peaks of different chirality. Energy positions of band-edge bright and dark excitons were determined distinctly in each chirality.

Keywords single walled carbon nanotube, PFO, pulse magnetic field, magneto-absorption

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1 Introduction

A single-walled carbon nanotube (SWNT) is well known as showing the Aharanov-Bohm (AB) splittings of the energy bands upon applying an external magnetic field parallel to the tube axis[1]. Observation of the AB effect has been reported in SWNTs by many group either by the absorption[2] or by the photoluminescence (PL) spectra[3,4,5]. Recently, excitonic effects are recognized as very

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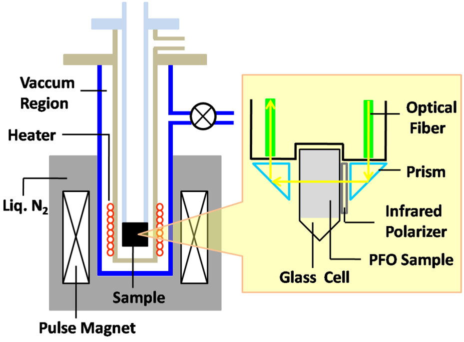

Fig. 1 (Color online) Schematics of the experimental setup: a measurement probe (left-hand) and a sample holder (right-hand)

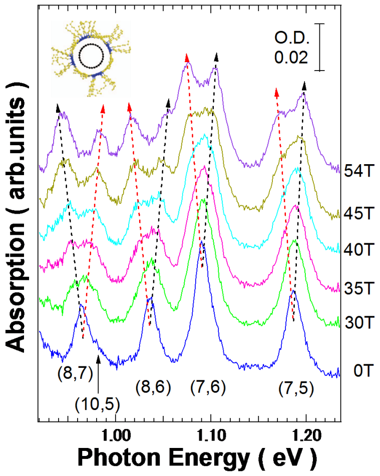

Fig. 2 (Color online) Magneto-absorption spectra of four chiralities (7,5), (7,6), (8,6), and (8,7). Inset shows a SWNT surrounded by PFO polymers.

crucial in understanding the fundamental optical transition. Owing to interplay between the inter and intra K-K valley Coulomb scattering, the exciton states become very complicated with many split states of the bright and dark excitons[6]. 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[2] or by micro magneto-PL[4,5] in a single SWNT. However, PL in general is sensitive to unknown impurities or localized states, and is not necessarily a good method to determine and to discuss the intrinsic and coherent energy states.

We attempted near infrared magneto-optical absorption measurements to the PFO (poly(9,9-dioctylfluorenyl-2,7-diyl)) SWNTs[7] at a room temperature which allow us to discuss directly an oscillator strength from the absorption spectra. The PFO-SWNTs show a very sharp and well-defined absorption spectra. The full-width-at-half-maximum (FWHM) of the spectrum is so narrow as about 20 meV. 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 spectral change in the region of a magnetic field where the exciton exchange interaction is manifested up to about 50 T.

2 Experimental details

A long pulse magnet with a bore of 20 mm was used. The pulse duration is 40 msec. The transmission infrared light was detected by an InGaAs charge-coupled device detector. The gate of the detector was synchronously opened at the top of the pulse field (the exposure time : 1.5 ms), and the absorption spectra were taken at each magnetic field. The sample used in this study is the PFO-SWNT whose

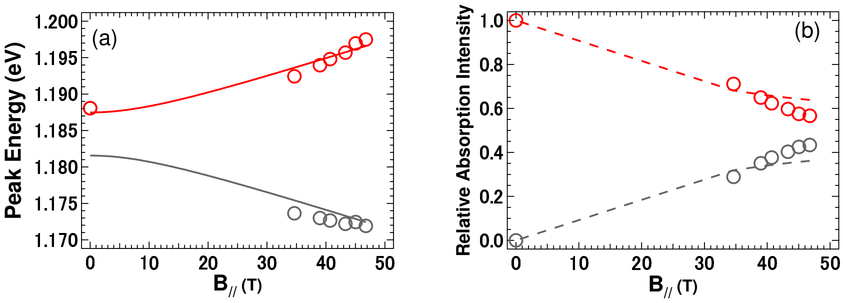

Fig. 3 (Color online) (a) Peak energy shifts of the (7,5) SWNT. The results of the fitting are shown by solid lines. (b) Relative absorption intensity of the (7,5) SWNT. The dashed line is calculated.

Table 1 The AB splitting per unit field, μ and the dark and bright exciton splitting at zero field, Δ_{bd} obtained from the spectral peak fitting of each chirality.

(n,m)	d_t (nm)	μ_{theory}	μ(meV/T)	Δ_{bd}(meV)
(7,5)	0.83	0.47	0.52±0.13	7.0±5.4
(7,6)	0.89	0.50	0.55±0.20	7.2±7.2

solvent is d-toluene[7]. The liquid sample was held in a miniature glass cell of which lid was sealed by Teflon tape and stycast black resin in order to prevent the active d-toluene from leaking (see Fig. 1). The pulse magnet is operated in liquid nitrogen. The sample space is devised to hold a room temperature by heating of an inner wall of a tube isolated from the ambience of liquid nitrogen by a thin vacuum space layer. The manganin thin wire capable of providing 50 watt was wounded around the innermost tube as shown in Fig. 1.

3 Results and Discussion

Figure 2 shows absorption spectra measured up to 54 T at 290 K. Respective peaks arise from the chirality (7,5), (7,6), (8,6), and (8,7). The peak of (10,5) appears at a high energy shoulder of the peak (8,7). For each chirality, one sharp peak at 0 T splits gradually into two peaks upon increasing the field. It is evident that upon applying a magnetic field, new peak appeared at a low energy side of the main peak for the case of the chirality (7,5), (7,6), and (8,6). This clearly indicates lower energy location of the dark excitons according to the spectral calculation by Ando[6]. This result is consistent with those reported by other groups[3,4,5]. As for the chiralities (8,7), it is rather difficult to judge which energy side the new peak emerged due to the ambiguity caused by an overlap with (10,5).

In order to track the peak shifts and splitting, the spectrum from each chirality was deconvoluted by a Gaussian waveform. The results in the case of (7,5) are shown Fig. 3 after correcting an effective magnetic field parallel ($B_{//}$) to the tube axis. Since the SWNTs dispersed in a liquid, the effective $B_{//}$ should be corrected taking into account of random orientation with the mean orientation directed by the external magnetic field toward the direction of the magnetic field[8]. The peak splitting is insufficiently resolved for reliable spectral deconvolution until 35 T. Peak positions of the split spectra are fitted by the relation similar to those treated by other group[4,5], and the calculated lines were presented by a solid line in Fig.3. The values of the dark and bright exciton splitting Δ_{bd}, and of coefficient μ representing the AB splitting in magnetic field, given by $\Delta_{bd} = \mu B_{//}$ are, summarized in Table 1. Using these parameters the absorption relative intensities of each peak were reproduced quite well by the calculation (dotted lines) as seen in Fig.3 (b). It can be noticed that as for the values of the experimental $\mu_{exp.}$, a fairly good agreement was obtained with those by the theory[1]. The results of Δ_{bd} which stands for the bright-dark exchange splitting are also consistent with those by the other groups[4,5].

4 Summary

Magneto-absorption measurements were carried out on PFO-SWNTs, of which absorption peaks were well defined according to the selected chirality. Spectral splittings were well resolved above 35 T, and the AB splitting and the exchange energies were determined. The band-edge dark excitons were found to be located at a lower energy side of that of the bright one, in agreement with those results from PL spectra reported by the other groups.

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