Magneto-optical effect of single-wall carbon nanotubes in ultra-high magnetic fields

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Among nano-carbon materials, the single-walled carbon nanotube (SWNT) attracts much attention in this decade as a material for the industrial application. Electronic properties are characterized by its intrinsic pseudo-1D structure and by chiral-selectivity of metal and semiconductor. In the optical properties of SWNT, the excitonic effect is of particular importance. Because of the presence of K and K' valley degeneracy in the Brillouin zone, there exist 16 exciton states, and by considering Coulomb interactions of intra- and inter-valley, one of these is optically active (bright) and others are inactive (dark). The degeneracy of K and K' valley is solved by an external magnetic field parallel to the tube direction, and hence, the bright and dark exciton change into two bright excitons [1]. As a result, complicated exciton states have important role in the bend edge optical properties. In addition, the energy band of SWNTs is substantially modulated by Aharonov-Bohm effect under finite magnetic fields, which is called the Ajiki-Ando (AA) splitting [2]. This indicates that we can discuss the exciton structures in SWNT from the detailed analysis of the AA splitting. Though the AA splitting should be observable in the photoluminescence and the optical absorption as magneto-optical effects in high magnetic fields, the observation of a clear AA absorption spectral splitting is difficult. Previous attempt on the magneto-optical study conducted up to 45 T [3] showed only the appearance of shoulder structure in the absorption peaks but not a clear AA splitting. Therefore, the application of ultra-high magnetic fields above 100 T is of necessary for clarification of the exciton AA splittings.

Our group has investigated the AA splitting of SWNT in ultra-high magnetic fields. Using the electromagnetic flux compression (EMFC) method [4] we measured the magneto-absorption spectrum parallel to the tube direction in the E//B (Voigt) configuration in magnetic fields of up to 300 T. We observed the splitting of the second subband absorption peaks of HiPco-SWNTs dispersed in D₂O (Figure 1). Similar behavior was also observed for SWNTs in different growth condition (PFO-ACCVD/*d*-toluene) [5]. However, SWNTs dispersed in liquid are randomly oriented, which causes decrease of the effective magnetic field parallel to the tube direction, and broadening of the absorption peaks. These effects are unfavorable for the observation of the clear AA splitting in ultra-high magnetic fields.

Therefore, we attempt to synthesize a stretched polyvinyl alcohol (PVA) film with SWNTs, to prepare highly-oriented SWNTs. SWNTs were dispersed in the polymer sodium

deoxycholate (DOC), then, mixed with PVA and subjected to be dried to form a film [6]. We devised a stretching machine for good controllability. Figure 2 shows the magneto-absorption spectra of stretched PVA film with (7, 7) metallic SWNTs, measured by the non-destructive pulse magnet. The observed splitting width, Δ , of the second subband absorption peak is consistent with those expected from the AA splitting without any additional broadening. It is expected from these results that the AA splitting can be observed more distinctly by the EMFC method. We will also show the results for the (10, 2) semiconducting SWNTs for comparison.



Figure 1: Absorption spectra of SWNTs dispersed in D_2O in the E//B (Voigt) configuration. EMFC method was used to generate ultra-high magnetic fields up to 300 T. O.D. stands for the optical density of the spectra. The vertical lines show the peak positions as a result of Gaussian curve fitting for each chiral SWNTs.



Figure 2: Absorption spectra (E//B) of a stretched PVA film with (7, 7) metallic SWNTs, measured by the non-destructive pulse magnet. The black solid lines are the fitting curves for each spectrum, which are the superposition of two Gaussian curves (broken lines).

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