Dynamical orientation of carbon nanotubes by pulsed magnetic fields

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Abstract. We have studied the dynamical orientation of single-walled carbon nanotubes (SWNT) dissolved in D₂O induced by pulsed magnetic fields. A wave-form of the field was varied from micro-to mili-sec in time, and the field strength was varied from several Tesla up to 100 T. Optical transmission through the liquid sample was detected in a pulsed magnet at room temperature. The linear polarization degree of the optical transmission was used as a measure for the average alignment of the SWNTs in the liquid. The application of a 40 T pulse field aligned the tubes by 12 degrees on average. A harmonic oscillator model with simple geostatics was used for a simulation. Our model well explains all the investigated cases in which a magnetic-field-dependent magnetic moment of the system is taken into account. Our experimental results agreed qualitatively with those of Ajiki and Ando. The term induced by the flux change (dB/dt) became important in low magnetic fields and during short pulses, which require a dynamical part of the magnetic susceptibility.

1. Introduction
Carbon nanotubes have been the subject of different type of studies because of their potential applicability for industrial usage in a wide range of fields, from opto-electronics to chemi-physical quantum storage. One important challenge is to fabricate tractable nanotubes with a high orientation. Various techniques for the alignment of individual nanotubes have been under investigation, including mechanical stretching of gelatin films [1], electric field orientation [2], and magnetic field orientation [3] in addition to oriented deposition on tailored substrates. Among these, magnetic field orientation surpasses the others when the nanotubes are dispersed in a liquid solvent, since the field can be supplied in a relatively large space without electrodes or electric contacts. The magnetic field can also be used for the alignment during sample synthetization processes.

There have been several reports on the magnetic alignment of carbon nanotubes [3,4]. They have applied steady-state magnetic fields to determine whether the anisotropic magnetic susceptibilities are responsible for the effective alignment of the nanotubes. A field strength of 10 T was revealed to be sufficient for good alignment at room temperatures, where the magnetic energy overcomes the thermal energy causing Brownian motion. As the field became higher, the alignment became more effective. Given these results, a pulse magnet would be the most promising type of magnet for realizing a very high field in a sufficient space. The pulse magnet is more economical in terms of energy use and simpler to install than a steady magnet such as a superconducting, water-cooling magnet coil, etc. There has been, however, no systematic survey of the effect of the pulse magnetic field on the carbon nanotubes. There are two crucial parameters associated with the pulse magnetic field: the maximum
strength and the time-sweep rate of the magnetic field. In this report, we have studied the dynamical alignment of single-wall carbon nanotubes by use of various types of pulse magnets.

Figure 1. A sample cell for SWNTs in a liquid inserted into a pulse magnet with a Voigt geometry.

2. Experimental

The samples used in this study are SWNTs synthesized by the HiPCo technique that are dispersed in D$_2$O with sodium dodecyl sulphate surfactant. The typical size of the SWNTs is 1 nm in diameter and 100 nm in length. Various types of pulse magnets (inductance $L$) were employed with combinations of capacitor banks ($C$) and charging voltage in order to vary the maximum magnetic field $B_{\text{max}}$ (up to 41 T) and pulse duration in a wide range. The waveform of the field was varied by $L$ and $C$ in the relation of $B(t) \propto \sin\left(\frac{1}{LC} - \frac{R}{2L} t^2\right)$, where R denotes an electrical resistance of the whole the circuit system including a magnet. The pulse length in this report is defined as the time required to reach $B(t=\Pi)=B_{\text{max}}$. In this report, $\Pi$ was varied from micro sec to 20 msec. $\Pi=2.5 \mu$s was supplied by the single turn coil system conventionally used for megagauss generation.

Optical transmission of the liquid samples was measured via optical fiber cabling in and out of a magnet, and was connected to the image intensified CCD detector equipped to a polychromater, which was used in a streak mode, so as to detect the continuous change of the absorption spectra in magnetic fields [5]. The CCD detected the transmission spectra in wavelengths in the range of 600 nm – 760 nm,

Figure 2. (a) Streak image of absorption spectra at the peaks of chirality (7,6) and (8,7) in a pulse magnetic field. (b) The pulse field waveform and corresponding change of absorption intensity for $B//E$ and $B \perp E$ polarizations at the peak of (8,7).
which covers the second sub-band transitions of SWNTs [6]. The liquid sample was housed in a Teflon cell, and settled into a magnet with a Voigt configuration as shown in Fig. 1. A linear polarizer was inserted between the end of the incident light fiber and the cell quartz window. Optical anisotropy (the linear polarization degree) was obtained by the configuration of \( B \parallel E \) or \( B \perp E \), where \( E \) denotes the electric field component of incident light.

3. Results
The streak image of the absorption spectra in a pulse magnetic field is shown in Fig. 2(a). The chirality-assigned (7,6) and (8,7) absorption peaks change their spectra with an application of the field. Fig. 2(b) is a plot of the time evolution of the absorption intensity for the peak (8,7) in magnetic fields. Reflecting the optical anisotropy of the SWNTs, the intensity grew for \( B \parallel E \) and decreased for \( B \perp E \) [6]. A gradual decay of the intensity was observed after the pulse application.

The nematic order parameter defined by \( S = 1/2 \left[ \langle \cos^2 \theta \rangle - 1 \right] \) is correlated to the optical anisotropy \( A = (\alpha_\parallel - \alpha_\perp)/(\alpha_\parallel + 2\alpha_\perp) \) in the present case, where \( \theta \) is the average angle between the nanotube and the direction of \( B \), and \( \alpha_\parallel \) and \( \alpha_\perp \) are the absorption intensity in the case of \( B \parallel E \) and \( B \perp E \) [7]. In the case when a pulse magnet with \( \Pi = 20 \) msec was used, the optical anisotropy (or \( \theta \)) increased almost linearly with \( B_{\text{max}} \). \( S \) increased up to 0.3 (\( \Delta \theta = 11.6^\circ \)) at \( B_{\text{max}} = 42 \) T. The pulse width dependence was investigated under \( B_{\text{max}} = 10 \) T, the results of which are summarized in Fig. 3. Upon increasing \( \Pi \), \( \Delta \theta \) increased until it was saturated around 15 msec. This result indicates that the pulse width \( \Pi = 20 \) msec possesses almost equivalent effect to that by the steady-state magnetic fields.

![Figure 3. The change of optical anisotropy and the corresponding orientation angle of SWNTs as a function of pulse widths, \( \Pi \) with \( B_{\text{max}} \) set at 10 T.](image3.png)

![Figure 4. The change of optical anisotropy subject to a pulse magnetic field of \( B_{\text{max}} = 41 \) T and \( \Pi = 20 \) msec, and the result of the calculation by eq. (1).](image4.png)

4. Analyses
In order to understand the dynamics for the orientational response of SWNTs against the pulse field, a harmonic oscillator model with simple geostatics was undertaken. The equation of motion is expressed as,
\[ I_{\text{SNW}} \frac{d^2\theta(t)}{dt^2} + \gamma \frac{d\theta(t)}{dt} + k(\theta(t) - \theta_{eq}) = -NB^2(\chi_\parallel - \chi_\perp)\sin 2\theta(t), \quad (1) \]

where \( I_{\text{SNW}} \) is a moment of inertia assuming SNWT to be a solid bar, and \( \gamma \) is a damping term incorporating the viscosity of the host liquid. \( \theta_{eq} \) is defined as the thermal equilibrium averaged angle of SWNTs. The harmonic oscillator model has been successfully applied to diamagnetic materials, and used for detection of small diamagnetic anisotropy \[8\]. A calculated result is compared with the observed optical anisotropy taken at \( B_{\text{max}} = 41 \text{T} \) in Fig.4. The calculation could reproduce the data almost completely during the time when the field was applied. There is little discrepancy in the region of slow decay.

We found that magnetic-field-dependent magnetic susceptibility both for \( \chi_\parallel \) and \( \chi_\perp \) should be taken into account for the overall agreement of all of the data taken at various \( B_{\text{max}} \). \( \Delta \chi \), which is defined as the variation of \( (\chi_\parallel - \chi_\perp) \) from that at 41 T (normalized values at 41 T), was obtained by fitting the data at changing values of \( B_{\text{max}} \), and is shown in the inset of Fig.4. The drastic increase of \( \Delta \chi \) at very low magnetic fields is indicative of the theory of Ajiki and Ando \[9\], and is an example of the Aharonov-Bohm effect observed via the magnetic susceptibility.

As for \( \Pi \) (a pulse width) dependence, when \( B_{\text{max}} \) becomes smaller and \( \Pi \) shorter, the calculated curve tends to deviate from that of the experiments at the last part of the response. We have therefore included the time derivative of the magnetic field \( dB/dt \) in eq.(1). This modification almost completely corrected the reproduction of the data even including the decayed variation. The necessity of this \( dB/dt \) term implies the importance of the dynamical magnetic susceptibility in a region of low magnetic fields.

4. Conclusion

We found that a 20 msec pulse width is sufficient for the full orientation achieved by a steady field at the same strength. The application of 40 T aligned the tubes by 12 degrees. The orientation was simulated by a harmonic oscillator model with simple geostatics. Our model well explains all of the investigated cases if the magnetic-field-dependent magnetic moment of the system is taken into account. The values of the magnetic moment obtained from our experiments agreed qualitatively with those calculated theoretically by Ajiki and Ando. The term induced by the flux change \( (dB/dt) \) became remarkable at the region of low magnetic fields and fast pulses, a result that may stem from the dynamical magnetic susceptibility.

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