Dielectrophoresis force driven dynamics of carbon nanotubes in liquid crystal medium

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This work reports the translation motion of carbon nanotubes (CNTs) dispersed in nematic liquid crystal (NLC) under an ac electric field. This effect was studied for homogeneously and vertically aligned NLC cells driven by in-plane field and vertical electric field, respectively. Long axis of the CNTs is aligned along the liquid crystal director, and above the critical field, the director of the NLC is distorted due to translation motion of CNTs in NLC. The amplitude of this translation motion decreases with increasing frequency, whereas the critical field increases with increasing frequency. We present the mechanism of translation motion of CNTs and model this observed phenomenon based on dielectrophoretic force. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2769341]

I. INTRODUCTION

Carbon nanotubes (CNTs), due to their unique and useful characteristics, have been extensively studied for many nanotechnology applications.[1–8] CNTs with high aspect ratio (a few micrometers in length and 1–50 nm in diameter) provide high mechanical strength, chemical stability, and superb electrical characteristics. This material can be employed in devices such as field effect transistors,[9–12] memory storage devices,[6] sensors,[3,13] actuators,[14,15] and field emission sources.[2,8,16]

Different interactive forces such as electrostatic, van der Waals, and surface tension on carbon nanotubes are dependent on operating environment and material properties of host materials in which carbon nanotubes are dispersed. They have large induced polarization due to high aspect ratio of CNTs, and hence nanotubes with high aspect ratio are largely influenced by the dielectrophoretic (DEP) forces.[17] DEP force was studied in early work of Pohl[18] in the 1950s and a comprehensive review has been presented by Bruke.[19] The DEP force is defined as a force invoked by the dipole layer that is formed at the interface of two inhomogeneous materials, for instance, CNTs with no permanent charges in dielectric solvent. The induced CNT motion is determined by the dielectric properties of the CNT and host dielectric medium, whereas in electrophoresis the motion of the particle or nanotube is determined by the magnitude and polarity of net electric charge. There are two different effects associated with DEP forces: positive and negative DEP forces. Positive DEP force occurs when the CNTs are more polarized than the medium, causing the CNT to move forward to the maximum field gradient. The reverse is true for negative DEP force.

Recently, the CNT-doped liquid crystals have been proposed as a prospective material for the use in display devices.[20–23] Alignment of the liquid crystal molecules is one of the most important factors in the display devices. Long axis of the CNTs is aligned along the liquid crystal director,[20,24,25] and hence order parameter of liquid crystals can be improved by doping small amount of CNTs in liquid crystal (LC)[24,25] even in the absence of applied field. LC director can be oriented with an electric field[24,25] or magnetic field.[24] Mrozek *et al.* have reported that single-walled CNTs (SWCNTs) can be used to align the liquid crystal polymers (LCPs) via a kinetic seeding of homogeneous liquid crystal domains in cooling the LCP from its isotropic phase.[26] The dynamics of CNTs in LC medium under an external electric field is of theoretical and technological interest.[20–22,24,25] However, there are very few reports which depict the behavior of CNTs under ac electric field in the host dielectric medium. It has been recently reported by our group that the residual dc can be reduced with the doping of small amount of CNTs in LC medium.[20] It has been also reported that CNTs deform the LC director due to its translation motion[21,22] under an ac electric field. In this paper we examine the translation motion of SWCNTs in LC medium as a function of frequencies and critical fields. We modeled this observed translation motion based on dielectrophoretic force.

II. EXPERIMENTAL TECHNIQUES

SWCNTs of 5×10^{-3} wt % were doped in nematic LC medium. The HiPCO SWCNTs (Carbon Nanotechnology Inc.) were used after nitric acid treatment to remove catalysts. The diameter ranged from 4 to 10 nm with a wide distribution of CNT length from 0.1 to 0.5 μm, which was observed from atomic force microscopy (AFM) (Seiko SPA-400). For a homogeneous alignment, driven by in-plane field, the interdigitated opaque electrodes made from aluminum were placed on the bottom substrate only. The electrodes

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have been separated at a distance of 30 μm with an electrode width of 10 μm. For a cell fabrication, a homogeneous alignment layer (AL-16139 from Japan Synthetic Rubber Co.) was first spin coated to a thickness of 800 Å on an electrode-patterned glass substrate. A rubbing process to the in-plane field direction was then performed on the substrate to align the nematic LC. The same alignment layer was coated on another glass substrate without electrode, and the similar rubbing process was then performed in antiparallel direction to the first one. The cell was assembled to give a cell gap (d) of 9 μm, where the plastic balls were used to maintain the cell gap. For a homeotropic alignment, the cell has been taken in the form of parallel plate capacitor, in which two indium tin oxide (ITO)-coated glass plates were separated by a cell gap of 60 μm with an active area of 3×3 cm^2. Alignment layer of AL00010 (Japan Synthetic Rubber Co.) has been coated on the glass electrodes. Superfluorinated LC mixtures purchased from Merck Co. (dielectric anisotropy $\Delta\varepsilon=+7.4$ at 1 kHz, birefringence $\Delta n=0.088$ at $\lambda=589$ nm, flow viscosity $\eta=18$ mm^2/s, at 20 °C) were filled at room temperature by the capillary action.

In both cells, the field direction is parallel to the LC director so that the LC director is not deformed at all due to the use of LCs having positive dielectric anisotropy but instead aligns more perfectly along the field direction. Nevertheless, the LC deformation is observed in both cells by motion of CNTs.[23,25] The fabricated test cells were observed under optical polarizing microscopy (Nikon DXM1200) by applying a sine wave voltage of different fields and frequencies.

III. THEORETICAL APPROACH

When the CNT-doped LC is placed in an ac electric field, the large dipole moment is induced in the CNTs due to their high aspect ratio[17] and CNTs experience the DEP force. Assuming damping or viscous forces on CNTs due to LC molecules to be negligible compared to dielectrophoretic force as CNTs have large induced dipole moment, the DEP force equation for long cylindrical nanotubes can be written as[27]

$$\mathbf{F}_{\mathrm{DEP}}=\Gamma\varepsilon_m\,\mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}},\varepsilon^*_m)\}\,\boldsymbol{\nabla}\,E^2, \tag{1}$$

where Γ is a geometrical factor and $\varepsilon^*_{\mathrm{CNT}}$ and ε^*_m are complex dielectric permittivities of the nanotube and LC medium, respectively. $K(\varepsilon_{\mathrm{CNT}},\varepsilon_m)$ represents the complex polarization factor (for spherical objects this is known as Claussius-Mossotti function). E denotes the applied ac field. For a nanotube of length l and radius r, the geometrical factor Γ is given by[27]

$$\Gamma=\frac{\pi r^2 l}{6} \tag{2}$$

for cylindrical object. The complex polarization factor is given by[27]

$$K(\varepsilon_{\mathrm{CNT}},\varepsilon_m)=\frac{\varepsilon^*_{\mathrm{CNT}}-\varepsilon^*_m}{\varepsilon^*_m} \tag{3}$$

or

$$\mathrm{Re}[K(\varepsilon_{\mathrm{CNT}},\varepsilon_m)]=\frac{\Delta\varepsilon\varepsilon_m+(\Delta\sigma\sigma_m)/\omega^2}{\varepsilon_m^2+\sigma_m^2/\omega^2}, \tag{4}$$

where $\Delta\varepsilon=(\varepsilon_{\mathrm{CNT}}-\varepsilon_m)$, $\Delta\sigma=(\sigma_{\mathrm{CNT}}-\sigma_m)$, and $\omega=2\pi f$, the angular vibrational frequency of the applied electric field. σ_{CNT} and σ_m are the conductivities of the CNTs and LC medium, respectively. For high and low frequency limits, Eq. (4) can be written as

$$\mathrm{Re}[K(\varepsilon_{\mathrm{CNT}},\varepsilon_m)]=\frac{\Delta\varepsilon}{\varepsilon_m}\quad(\omega\to\infty), \tag{5}$$

$$\mathrm{Re}[K(\varepsilon_{\mathrm{CNT}},\varepsilon_m)]=\frac{\Delta\sigma}{\sigma_m}\quad(\omega\to0). \tag{6}$$

This shows that for low and high frequency limits, the DEP force is independent of the frequency. CNTs in LC medium are initially aligned along the direction of LC director.[20,24,25] The DEP force causes the nanotube to move toward either of the electrodes depending on the relative permittivity of the nanotubes with respect to LC medium. Considering the translation motion of CNTs in nematic liquid crystal (NLC) medium along x axis and applied electric field as a sinusoidal wave, the instantaneous force at a time t can be written as

$$F_{\mathrm{DEP}}=\Gamma\varepsilon_m\,\mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}},\varepsilon^*_m)\}\frac{d}{dx}E_0^2\sin^2(\omega t). \tag{7}$$

Note that we have ignored the vector notation in the above expression, since CNTs' motion is only one dimensional. Differentiating $\sin^2\omega t$,

$$F_{\mathrm{DEP}}=\Gamma\varepsilon_m\,\mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}},\varepsilon^*_m)\}E_0^2\omega\sin(2\omega t)\frac{dt}{dx}, \tag{8}$$

and the equation of motion,

$$m\frac{dv}{dt}=\frac{\Gamma\varepsilon_m\,\mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}},\varepsilon^*_m)\}E_0^2\omega\sin(2\omega t)}{v}. \tag{9}$$

Integrating the above equation and applying the initial condition ($t=0$, $v=0$), the velocity of CNTs in LC medium at a time t can be written as

$$v=E_0\sqrt{\frac{2\Gamma\varepsilon_m\,\mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}},\varepsilon^*_m)\}}{m}}\sin(\omega t). \tag{10}$$

Experimentally it has been observed that CNTs perturbed the LC director after the certain field E_{cr} (defined as critical field) of applied frequency, i.e.,

$$v=0\quad\text{if } E_0\leqslant E_{\mathrm{cr}},$$

$$v>0\quad\text{if } E_0>E_{\mathrm{cr}}. \tag{11}$$

Therefore Eq. (10) can be modified as

$$v=(E_0-E_{\mathrm{cr}})\sqrt{\frac{2\Gamma\varepsilon_m\,\mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}},\varepsilon^*_m)\}}{m}}\sin(\omega t). \tag{12}$$

As $v=dx/dt$, therefore the displacement of CNTs in LC medium at a time t can be written as

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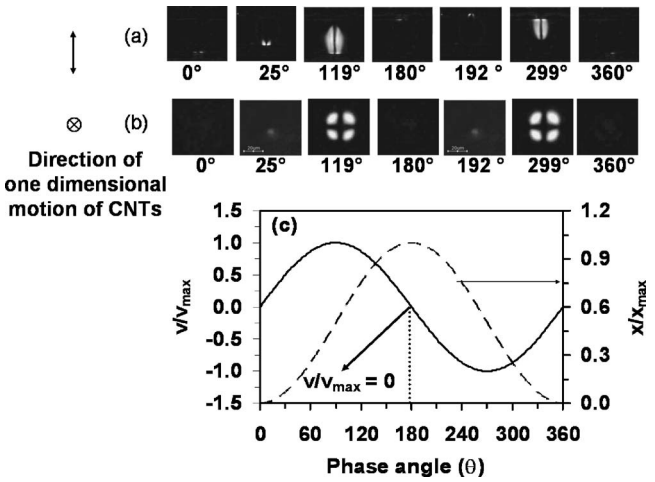

FIG. 1. Position of CNTs captured at different times in both the cells at the frequency of 1 Hz. (a) Positions of CNTs captured at 0°, 25°, 119°, 180°,192°, 299°, and 360° phase angles of input sine wave in homogeneous aligned cell driven by in-plane field. The direction of the motion of the CNTs in homogeneous aligned cell driven by in-plane field is in the plane of the paper. (b) Positions of CNTs captured at 0°, 25°, 119°, 180°, 192°, 299°, and 360° in homeotropic aligned cell. The direction of the motion of the CNTs in homeotropic aligned cell is perpendicular to the plane of the paper. (c) Theoretically plotted graph for velocity (continuous line) and displacement (dashed line) of CNTs in LC medium as a function of time. The magnitude of velocity of the CNTs in the dielectric medium is maximum, at the phase angles of 90° and 270° and zero at 0°, 180°, and 360°.

$$x = \frac{(E_0 - E_{cr})}{\omega}\sqrt{\frac{2\Gamma\varepsilon_m \mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}}, \varepsilon^*_m)\}}{m}}[1 - \cos(\omega t)]. \tag{13}$$

For ωt is odd multiple of π, the displacement of the CNTs in LC medium will be maximum ($x_{\max}$). Therefore amplitude (Ampl), i.e., maximum displacement, of the translation motion of the CNTs in LC medium can be written as

$$\mathrm{Ampl} = \frac{2(E_0 - E_{cr})}{\omega}\sqrt{\frac{2\Gamma\varepsilon_m \mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}}, \varepsilon^*_m)\}}{m}}. \tag{14}$$

Equation (12) represents the velocity of CNT in LC medium at a time t, whereas Eq. (13) represents the displacement of CNT as a function of time. Equation (14) shows the amplitude of the translation motion of CNT in LC medium.

IV. RESULTS AND DISCUSSION

We have recently reported that under high electric field, CNTs perturbed the LC molecules due to their translational motion which distorted the LC director, resulting in the light leakage under the cross polarizing microscope.[21] In a homogeneously aligned sample driven by in-plane field, the light leakage in a form of vertical strips is observed, where the strips move back and forth between electrodes, as shown in Fig. 1(a). The top view of this light leakage in a homeotropically aligned cell is observed in a form of four-lobe textures [see Fig. 1(b)]. This light leakage altered periodically with time in both cells. According to Eq. (12), the velocity of CNTs varies sinusoidally with time, as shown in Fig. 1(c).

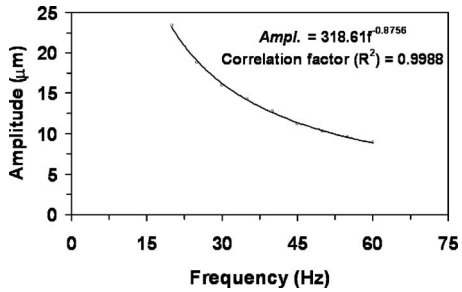

FIG. 2. Variation of amplitude of translation motion of CNTs in LC medium with frequency in homogeneous aligned cell driven by in-plane field at the field of 1 V/μm. The circles represent the experimental curve, whereas continuous line on the circles is the best fitting according to the Eq. (15).

Positions of light leakage due to the translational motion of CNTs were captured at different times for both cells. Experimentally at phase angles of 0° and 360°, no light leakage was observed in both cells. This can be understood by zero velocity of CNTs such that there is no perturbation in LC molecules due to the CNT motion and hence no light leakage was observed. On the other hand, at phase angles of 119° and 299°, the velocities of CNTs are close to the maximum which in turn perturbs the adjacent LC molecules, resulting in large light leakage. It is important to note that these patterns were never visible in pure NLC, assuring that these patterns originated from the translational motion of CNTs. Since CNTs move to the positive x direction during the first half cycle (0-π) of a sine wave and they return during the next half cycle, the positive dielectrophoretic force is applied to CNTs.

We also observed that the amplitude of the translational motion of CNTs between electrodes decreased with increasing frequency at a constant voltage. Assuming that $K(\varepsilon^*_{\mathrm{CNT}}, \varepsilon_m)$ is constant in the frequency range of 10–60 Hz, the following equation (15) has been fitted to the experimental curve, as shown in Fig. 2,

$$\mathrm{Ampl} = \frac{A(E_0 - E_{cr})}{f^n}, \tag{15}$$

where n and $A = (1/\pi)\sqrt{2\Gamma\varepsilon_m \mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}}, \varepsilon^*_m)\}/m}$ are fitting parameters. The value of n obtained from the fitting is 0.88, which is slightly less than the expected value of 1. This difference may be due to a diminutive effect of frequency dependent term $\mathrm{Re}\{K(\varepsilon^*_{\mathrm{CNT}}, \varepsilon^*_m)\}$. Moreover in low frequency region the dielectrophoretic force is dominated by the conductivity of CNTs in addition to liquid crystal medium. In general, the conductivity of the dielectric materials (CNTs as well as LCs) decreases with decreasing frequency. Therefore, the value of n, which is slightly less than 1, is quite reasonable. The value of fitting parameter $A(E_0 - E_{cr})$ is found to be 318.6. The dielectric properties of CNTs can be determined from this parameter, if the physical parameters such as the dimension and mass/density of CNTs are known.

Figure 3 shows the optical micrographs at different electric fields in in-plane switching cell at a frequency of 60 Hz and the extracted amplitudes of the translational motion of CNTs. Clearly, the amplitude of translational motion increases linearly with increasing electric field at a constant

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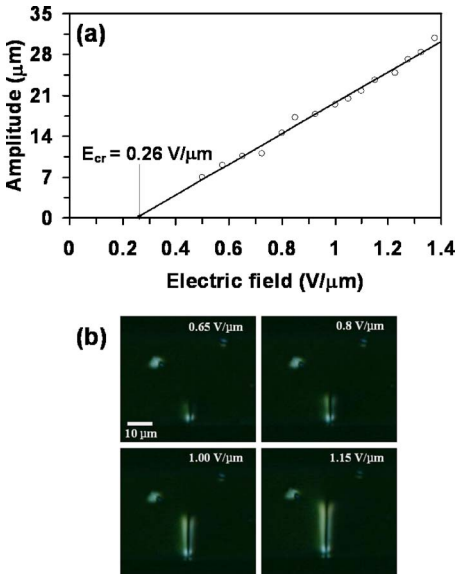

FIG. 3. (a) Amplitude of translation motion of CNTs vs electric field at the frequency of 60 Hz for homogeneously aligned cell, driven by inplane field. (b) Optical micrographs of the SWNT-doped LC cell describing the amplitude of the translation motion of the CNTs at different fields of 0.65, 0.80, 1.0, and 1.15 V/μm at the frequency of 60 Hz in the homogeneously aligned cell driven by in-plane field.

frequency, and continuous light leakage has been observed in high field region, as expected from Eq. (14). Thus, our theoretical model supports the experimental observations. If Fig. 3(a) is extrapolated, it intercepts the electric field axis at 0.26 V/μm, which corresponds to the critical electric field as, at this field, the amplitude of translation motion becomes zero. Experimentally we could not determine the amplitudes of translation motion having less than 5 μm, their corresponding electric fields, and hence the exact critical electric field above which the CNTs start translation motion. However, we have determined the electric field corresponding to the minimum measured amplitude of 6 ± 1 μm for in-plane cell at different frequencies and is given in Fig. 4. We also determined the electric field in homeotropic aligned cell at minimum observed amplitude of translation motion (see Fig. 4). According to Eq. (14), electric fields would vary almost linearly with frequency if Ampl is constant [with the assumption that $K(\varepsilon_{\mathrm{CNT}},\varepsilon_m)$ shows the static behavior in the frequency range of 10–60 Hz]. Our experimental results showed that the electric filed is increased linearly with increase in frequency of the applied field in both the cells (see Fig. 4). Thus from the above it is clear that our experimental results follow the theoretical model. This dynamic behavior of CNTs in liquid crystal medium suggests that the motion of CNTs can be also controlled by changing the dielectric constant of host material, which can be useful for many micro- or nanoelectromechanical devices.

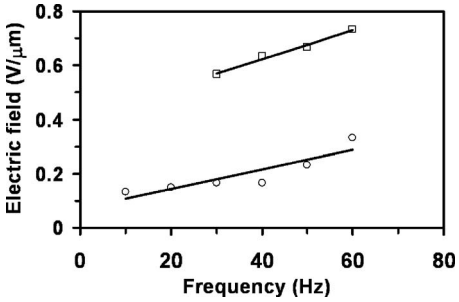

FIG. 4. Variation of electric field with frequency in homogeneously aligned sample at constant amplitude (6 ± 1 μm) of translation motion of CNTs driven by in-plane field (squares) and vertically aligned samples (circles).

V. CONCLUSIONS

We have investigated the dynamics of CNTs driven by positive dielectrophoretic force in nematic liquid crystal medium. Expressions for velocity and displacement of translational motion of CNTs as a function of time have been formulated in terms of frequency and applied electric field. The amplitude of the translational motion of CNTs is nearly inversely proportional to the frequency, whereas it is directly proportional to the applied electric field. The critical field of the translational motion of CNTs increases linearly with increasing frequency. Our theoretical model explains well the experimental observations.

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