New oscillation in terahertz magneto-optical effect of single-walled carbon nanotubes film

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Received 17 November 2003; accepted 7 March 2004
Available online 15 April 2004

Abstract

The magneto-optical Kerr effect of single-walled carbon nanotubes (SWNTs) film in the Terahertz region is theoretically studied by means of the Drude model. The calculation shows a new oscillation occurring near the plasma frequency as the origin of the magneto-optical Kerr effect for the single-walled nanotubes film. We propose that the amplitude of this near plasma frequency oscillation is directly proportional to the external magnetic field and its period increases with increasing cyclotron frequency. We consider these features of this oscillation of SWNTs are related to the Landau energy of SWNTs. The reflectivity and figure of merit (FOM) spectrum also reveal that this oscillation is the dominant mechanism for the magneto-optical Kerr effect of SWNTs film. Meanwhile, the shift and large enhancement of Kerr rotation under different external magnetic fields is explained.

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Keywords: A. Carbon nanotubes; C. Modeling; D. Photoconductivity

1. Introduction

Since the discovery in 1991 by Iijima [1], carbon nanotubes have received considerable attention for their unique properties. In particular, single-walled carbon nanotubes (SWNTs) exhibit extraordinary electronic, mechanical and chemical properties that make them potentially numerous applications, such as nanoscopic quantum wires [2,3], molecular device [4] and single-molecule transistors [5], field-emission and electron guns [6], and so on. Among the properties of SWNTs, optical characteristic is one of the most interesting things for experimental and theoretical studies.

Depending on diameter and helicity, SWNTs behave as one-dimensional metals or as semiconductors. It is well known that the electronic structure of individual SWNT is specified by a pair of integers \((n, m)\), which represent a coordinate of the wrapping vector in the hexagonal lattice. Armchair SWNTs \((n = m)\) are gapless and should be metallic; while zigzag or chiral SWNTs \((n \neq m)\) have a gap and possess semiconducting or metallic properties [7,8]. The unique features of SWNTs are original from their wrapping vectors. In order to determine their electronic and optical properties, Raman spectroscopy [9], transmission electron microscopy (TEM), X-ray diffraction (XRD), infrared spectroscopy and ultraviolet photoemission spectroscopy (UPS) are applied [10].

Recently far-infrared Terahertz (THz) technology has become a very attractive research field. The interest in THz is steadily growing due to its prospering applications, which have involved semiconductor, label free genetic analysis, cellular level imaging, biological sensing and so on [11]. With the development of THz technology, it provides a new and powerful tool to study the optical properties of SWNTs in THz region. At present, however, just a few studies in regard to the properties of SWNTs in THz domain were performed [12,13]. The motivation of this work also comes from the recent advance in magneto-optical Kerr effect (MOKE) [14,15]. Shimano and his collaborators have developed THz technique which helps them to obtain the MOKE spectrum of n-type, undoped InAs in THz region [15].
Therefore, it becomes possible for ones to measure the MOKE spectrum of materials from 0.1 to 2.5 THz, which has become a standard and valuable technique to study various magnetic properties of electronic of materials, in particular, low dimensional systems. The MOKE spectrum of SWNTs film in far-infrared THz domain enlightens an effective method to study the optical and magnetic characteristics of SWNTs. In this paper, we report a theoretical analysis of magneto-optical Kerr effect (MOKE) of SWNTs film in far-infrared THz region.

2. Magneto-optical Kerr effect (MOKE)

In general, for most magneto-optical (MO) media, the dielectric tensor is symmetric in the absence of an external magnetic field. In this case, the dielectric tensor usually can be treated as a scalar. Applying an external magnetic field, the time reversal symmetry in the dielectric tensor is broken [16]. In the cartesian coordinate, in which z axis is normal to the sample surface, suppose a static magnetic field \( \mathbf{B} \), which is perpendicular to the sample surface, then from the Drude model the dielectric tensors are expressed as follows [15,17]:

\[
e_{xx} = e_{yy} = e_c \left[ 1 - \frac{\omega_p^2 (\omega^2 + \gamma \omega)}{(\omega^2 + \gamma \omega)^2 - \omega^2 \omega_c^2} \right]
\]

\[
e_{xy} = -e_{yx} = e_c \frac{\omega_o \omega_p^2}{(\omega^2 + \gamma \omega)^2 - \omega^2 \omega_c^2}
\]

\[
e_{zz} = e_c \left[ 1 - \frac{\omega_p^2}{(\omega^2 + \gamma \omega)} \right]
\]

where \( e_c \) represents the frequency-independent optical dielectric constant. The plasma frequency is defined as:

\[
\omega_p = \sqrt{N e^2 / \epsilon_0 m^*}, \quad \text{which depends on the carrier density} \ N \text{ and the effective mass} \ m^*. \quad \gamma \text{ is the damping constant and} \ \omega_c = eB/m^* \text{ denotes the cyclotron frequency.}
\]

The real component of diagonal elements usually displays the ordinary optical absorption. \( e_{xx} \) and \( e_{zz} \) are mainly determined by \( e_c, \omega_p \) and \( \gamma \). \( e_{xy} \) is identical with the dielectric function \( e \) without the external magnetic field and is best measured before applying the magnetic field. The imaginary component of the off-diagonal element \( e_{xy} \) represents the magneto optical absorption, which arises from interband and intraband transition. It is responsible for the magneto-optical effects [18,19].

The Kerr rotation \( \theta \) and ellipticity \( \varphi \) are related with the dielectric tensor. They can be defined by the formula [15]:

\[
\Psi = \theta + \varphi = e_{xy}/(1 - e_{xx}) \sqrt{e_{xx}}.
\]

It is known that the Kerr effect involves reflected light, so we can define the average reflectivity \( \mathcal{R} \) [20]:

\[
\mathcal{R} = \frac{|r_+|^2 + |r_-|^2}{2}
\]

Here \( r_\pm = (1 - \sqrt{e_\pm})/(1 + \sqrt{e_\pm}) \) are the reflection coefficients for the left and right circular polarizations respectively, in which \( e_\pm = e_{xx} \pm i e_{xy} \) are the dielectric tensor's eigenvalues. Moreover, the complex index of refraction can be got as:

\[
\hat{n}_\pm = e_\pm [19].
\]

3. Numerical analysis

The details of the fabrication process of the SWNTs film have been given by the previous reports. The SWNTs film is a composite consisting of a random mixture of tubes of different types, oriented rather randomly at least within the plane of the film [12,13,21]. At room temperature, the MOKE spectrum of the SWNTs film can be calculated within the Drude model as described above. In [21], Ugawa and his collaborators investigated experimentally and theoretically the far-infrared optical properties of the SWNTs film. Employing the Drude model and Drude–Lorentzian model and using the following optical parameters: \( \omega_p/2\pi = 0.102 \text{ eV (154.8 THz)}, \omega_c/2\pi = 0.092 \text{ meV (0.14 THz)}, \gamma/2\pi = 0.04 \text{ eV (60.0 THz)} \) and \( \epsilon_c = 8.5 \), they could give good fit for the experimental data. In the present work, we take the above theoretical parameters according to Ref. [21], in which we suppose the external magnetic field \( B = 0.5 \text{ T} \), and then the numerical results are obtained by putting these data into the Drude model.

Fig. 1 shows the calculated diagonal \( xx \) component of the dielectric tensor, the Kerr rotation \( \theta \) and ellipticity \( \varphi \) of the SWNTs film. A significant enhancement of MOKE spectra is observed near 154.8 THz, where both the real part of the diagonal component \( \text{Re}(e_{xx}) \) and imaginary part \( \text{Im}(e_{xx}) \) are equal to zero, as seen from Fig. 1. The origin of the large enhancement of MOKE still remains under dispute. Especially, the large Kerr rotation becomes the focus of attention because of its importance in the optical readout of magnetically stored information and other useful applications. In the investigation of Ref. [22], it was suggested that the large enhancement of MOKE spectra occurs where \( \text{Re}(e_{xx}) \approx 1.0 \). A recent report pointed out that this enhancement should take place at \( \text{Re}(e_{xx}) \approx 1.0 \) [20]. Although the viewpoints are not consistent with each other, all of them stressed that the plasma frequency \( \omega_p \) plays an important role in the MOKE. In fact, we also notice in our calculations that the enhanced MOKE spectra appear near the plasma frequency. However, from Fig. 1, we can find that the large enhancement of the MOKE spectrum occurs around 154.8 THz, where \( \text{Re}(e_{xx}) \sim 0.0 \). The further calculation also finds \( \text{Re}(e_{xx}) \) are not equal to 1.0. Therefore, the previous interpretations are not fully valid to describe our case.

The MOKE is a manifestation of many phenomena: spin-polarization, spin-orbit interaction, plasma edge effect, joint density of states, momentum matrix ele-
ments and so on [18, 23]. From the microscopic point of view, the external magnetic field causes the splitting of Landau energy levels. Under the magnetic field, the density of states is no longer the continuum function and there will exist many odd points at the places, where the energy $E = (n + \frac{1}{2})\hbar\omega_c$. These discrete energy levels lead to a series of oscillations and these oscillations obviously induce many magneto-optical effects.

Carbon materials show some unique properties due to their special structures. The aforementioned tight bonding calculation and the original Landau level calculation found that the Landau energy level of two-dimensional graphite is in direct proportion to $\sqrt{n}$, here $n$ is integers. According to this result, we note that the level spacing is not equal. Furthermore, an interesting thing was found that the free energy of two-dimensional graphite increases with increasing the magnetic field. For carbon nanotube, since the magnetic field that is applied to the curved surface of a carbon is not uniform, the energy band of SWNT exhibits oscillations with a period which is determined by the symmetry of the nanotube. When the magnetic field parallels to the nanotube axis, the energy gap oscillates like a triangular chopped wave and as a result, a semiconducting carbon can become metallic and a metallic nanotube would become a semiconducting nanotube. While the magnetic field is perpendicular to the nanotube axis, the mixing of Bloch orbits also displays oscillation periodically with the magnetic field. Consequently, in this case, a metallic Fermi surface appears and disappears irregularly [24]. Based on these facts, we put forward the interpretation of the MOKE of the SWNTs film as following.

We consider that a certain oscillation should exist around the plasma frequency $\omega_p$, and the enhancement of Kerr rotation is surely induced by this oscillation. This oscillation usually occurs near the plasma frequency $\omega_p$. At the two sides of $\omega_p$, the phase of the oscillation is opposite. When $\omega < \omega_p$, the oscillation has the positive maximum; while $\omega > \omega_p$, the negative maximum is presented. An obvious and interesting thing is that the oscillation vanishes when the frequency $\omega$ equals $\omega_p$. We also denote that the amplitude of this oscillation is in proportion to the magnitude of the external magnetic field, and its period is proportional to the cyclotron frequency $\omega_c$. The enhancement of Kerr rotation can be traced from this oscillation. In the following discussion, for the convenience, we name this oscillation near plasma frequency (NPF) as NPF oscillation.

As seen from Fig. 1, the Kerr rotation $\theta$ has the positive maximum at 126.3 THz, while the negative peak presents at 171.5 THz. When $\omega < \omega_p$, the Kerr rotation $\theta$ becomes zero. These features are consistent with the properties of NPF oscillation. NPF oscillation could be acted as the origin of large Kerr rotation. In order to demonstrate this more clearly, the calculated Kerr rotation under different external magnetic fields is represented by Fig. 2. At the same time, in order to illustrate the problem clearly and compare with Ref. [21], here and hereafter we take cm$^{-1}$ as the unit of frequency, in which the plasma frequency $\omega_p$ is equal to 5166.7 cm$^{-1}$. In Fig. 2, the external magnetic field varies between 0.1 and 4.0 T. It can be seen that the marked positive peak of Kerr rotation is shifted towards lower frequency with increasing incident photo frequency. At the same time, the negative peak is shifted to higher frequency. When the external magnetic field is 0.5 T, the maximum Kerr rotation occurs at 4178.0 cm$^{-1}$, and the minimum of Kerr rotation is
When the value of B is 4.0 T, however, the positive peak appears at 4069.6 cm\(^{-1}\), and the negative peak is at 5814.3 cm\(^{-1}\).

Furthermore, it can be noted obviously that the amplitude of Kerr rotation shows a pronounced increase as the external magnetic field increases. The most interesting phenomenon shown in Fig. 2 is that the Kerr rotations are equal to zero for all different magnetic field when the frequency is near the plasma frequency. All of these features are associated with, and can be explained by, the NPF oscillation. As mentioned previously, we consider that the NPF oscillation would be enhanced due to increase of the external magnetic field and this brings on the increase of amplitude of Kerr rotation. On the other hand, because the cyclotron frequency \(\omega_c\) increases as the magnetic field increasing, the period of NPF oscillation becomes longer. Consequently, both peaks of Kerr rotation are shifted along the lower and higher frequency shown as Fig. 2.

It has been indicated previously that the dispersion of Landau energy level of SWNT is related with the magnetic field and the energy band shows periodical oscillating as a function of magnetic field [24]. The NPF oscillation can be ascribed to Landau energy level of SWNTs. But how the change of Landau level makes a direct influence on NPF oscillation is unknown. A possible process might be that the energy gap or free energy of SWNTs increases with the magnetic field and this causes the enhancement of the amplitude of NPF oscillation. But as a macroscopic and experiential model, the Drude model can not give the original mechanism and this needs more detailed and deep investigation in the future work. Here our purpose is to
clarify that the NPF oscillation can give a good interpretation for the MOKE of SWNTs film and the NPF oscillation is related to the Landau energy level of nanotubes.

Till now, it has been illustrated that the enhancement of MOKE spectrum of the SWNTs film are on account of NPF oscillation. The plasma frequency plays a crucial role in this oscillation and the MOKE signals are mainly determined by this oscillation.

The diagonal and off-diagonal components of optical conductivity tensor \( \sigma_{xx} \) and \( \sigma_{xy} \) have been calculated from the Kramers–Kronig (KK) transformation [23,25], as shown in the Fig. 3. This was done at \( B = 0.5 \) T at room temperature. When \( \omega \) is below 300.0 cm\(^{-1} \), the real part of the diagonal optical conductivity approximates to a constant. Then it decreases with increasing frequency. This feature exhibits a typical metal or semiconductor characters in low frequency, and it arises from interband transitions [26]. The minimum of the imaginary part of off-diagonal optical conductivity is observed around 1157.5 cm\(^{-1} \), which comes from the intraband transitions, and it means that there maybe exists a strong absorption near this frequency.

We have further investigated the nature of the SWNTs film in far-infrared frequency domain. Fig. 4 depicts the average reflectivity \( \bar{R} \) and the figure of merit (FOM), which is usually used to characterize the performance of a certain material. \( \bar{R} \) can be obtained using Eq. (4) and \( \text{FOM} = \sqrt{\bar{R}^4} \) [20]. It can be seen that the reflectivity has a similar behavior as in [21]. The reflectivity is monotonically reduced with respect to the frequency when \( \omega \) is below the plasma frequency 5166.7 cm\(^{-1} \). Then a small peak presents around the plasma frequency. But, when the frequency is above the plasma frequency, it is found that the reflectivity increases with the increase of frequency. The FOM spectrum exhibits oscillating intensely around the plasma frequency \( \omega_p \). These behaviors are undoubtedly associated with NPF oscillation, and these are further evidence of the MOKE resulting from NPF oscillation.

Finally, we denote that even at low temperatures, the MOKE spectrum of the SWNTs film would not change a lot because the previous experiment found that the temperature dependence for the SWNTs film was quite small and no vibrational structures were observed at low temperature [21]. Hence, it can be regarded NPF oscillation for the SWNTs film would only have small change with temperature decreasing. This is a fascinating feature of SWNTs compared with other materials.

4. Conclusions

The MOKE of the SWNTs film in far-infrared THz region has been theoretically calculated employing the Drude model and obvious MOKE spectra are obtained. We have shown that the significant enhancement of Kerr rotation is caused by a certain oscillation, NPF oscillation, which occurs near the plasma frequency. The amplitude of NPF oscillation is directly proportional to the external magnetic field and its period increases with increasing the cyclotron frequency. We argued that NPF oscillation could adequately describe all the features of the calculated MOKE of the SWNTs film in THz domain and give a simple interpretation of the results. The shift of calculated Kerr rotations and change of their amplitude under different magnetic fields all demonstrate that NPF oscillation is the origin of MOKE. Furthermore, this explanation was confirmed by the average reflectivity and the FOM spectrum, which exist clearly visible behaviors when the frequency is around the plasma frequency. As a conclusion, we consider that the NPF oscillation is an essential factor for the MOKE of the SWNTs film, and large enhancement of Kerr rotation is mainly due to this oscillation.

Acknowledgements

This project was supported by the major project of knowledge innovation project of Chinese Academy of Sciences (No. KJCX2-SW-N02).
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