The effects of a static magnetic field on the microwave absorption of hydrogen plasma in carbon nanotubes: a numerical study*

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We theoretically investigate the microwave absorption properties of hydrogen plasma in iron-catalyzed high-pressure disproportionation-grown carbon nanotubes under an external static magnetic field in the frequency range 0.3 GHz to 30 GHz, using the Maxwell equations in conjunction with a general expression for the effective complex permittivity of magnetized plasma known as the Appleton–Hartree formula. The effects of the external static magnetic field intensity and the incident microwave propagation direction on the microwave absorption of hydrogen plasma in CNTs are studied in detail. The numerical results indicate that the microwave absorption properties of hydrogen plasma in iron-catalyzed high-pressure disproportionation-grown carbon nanotubes can be obviously improved when the external static magnetic field is applied to the material. It is found that the specified frequency microwave can be strongly absorbed by the hydrogen plasma in iron-catalyzed high-pressure disproportionation-grown carbon nanotubes over a wide range of incidence angles by adjusting the external magnetic field intensity and the parameters of the hydrogen plasma.

Keywords: carbon nanotubes, hydrogen plasma, static magnetic field, microwave absorption

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1. Introduction

Carbon nanotubes (CNTs) have attracted much attention in recent years because of their one-dimensional nanostructure and unique chemical and physical properties.[1−13] The microwave absorbing ability of CNT materials has been extensively explored for potential applications in microwave shielding or absorbing materials.[14−34] The experimental data show that intrinsic CNTs display weak absorption to microwaves, but iron-catalyzed high-pressure disproportionation (HiPco) grown CNTs exhibit strong microwave absorption (around 2.45 GHz) with subsequent dramatic light emission and intense heat release in ultrahigh vacuum.[14−17] Some studies have been carried out to understand the possible microwave absorption mechanisms of HiPco CNTs.[14−25] The available research data indicate that the microwave absorption is related to the thermal activation release of H_2 in CNTs.[16,17] It has also been shown that the condensing capacity of hydrogen in CNTs increases with rising pressure and dropping temperature. The doping is favorable for hydrogen adsorption in CNTs.[20−35] The adsorption quantity of hydrogen in HiPco CNTs is in the range of 0.43–0.90 wt.% at room temperature[36−38] and can rise to $\sim$ 2.5 wt.% at 77 K.[38] Under microwave irradiation, the hydrogen can form plasma.[39−41] Microwave interaction with HiPco CNTs under high vacuum ($\sim 10^{-6}$ Torr) generates highly ionized hydrogen plasma. Since the plasma strongly couples with the microwave radiation, it provides an independent mechanism for absorbing the microwave energy.[16] Our group has investigated the microwave absorbing properties of non magnetized hydrogen plasma in CNTs by using double-fluid theory and the Wentzel–Kramers–Brillouin (WKB) approximation. It was theoretically revealed that the strong microwave absorption of HiPco CNTs is predominantly caused by the collisional absorption of the charge particle in the hydrogen plasma of HiPco CNTs. The strong microwave absorption (around 2.45 GHz) by HiPco CNTs has been well explained

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by our theory.[19,20]

An external static magnetic field can be used to effectively improve the physical properties of CNTs.[42,43] The objective of this work is to investigate in detail the microwave absorbing properties of hydrogen plasma in CNTs under an external static magnetic field, particularly the influences of the angle between the microwave propagation direction and the static background magnetic field direction on the microwave absorption of hydrogen plasma in CNTs. It is assumed that the density distribution of the free electron of hydrogen plasma in CNTs is homogeneous, and the sample is an absorbing isotropic material. The theoretical results show that the influences of external magnetic field intensity and the incident wave propagation direction on the microwave energy absorption are very significant.

2. Theoretical model

Under normal conditions, a plane wave propagating in a lossy medium, such as the plasma, obeys the following Maxwell's equations:

$$\boldsymbol{\nabla} \times \boldsymbol{E} = -\mathrm{i}\omega \boldsymbol{B}, \tag{1}$$

$$\boldsymbol{\nabla} \times \boldsymbol{B} = \mathrm{i}\omega \tilde{\varepsilon}_\mathrm{r} \varepsilon_0 \tilde{\mu}_\mathrm{r} \mu_0 \boldsymbol{E}, \tag{2}$$

where ω is the angular frequency, and $\tilde{\varepsilon}_\mathrm{r}$ and $\tilde{\mu}_\mathrm{r}$ are the relative complex permittivity and permeability, respectively.

Generally, the optical and dielectric properties of an absorbing isotropic medium, such as the plasma in CNTs, can be characterized by the relative complex permittivity

$$\tilde{\varepsilon}_\mathrm{r} = \varepsilon_\mathrm{r}' - \mathrm{i}\varepsilon_\mathrm{r}'' = \varepsilon_\mathrm{r}' - \mathrm{i}\frac{\sigma}{\omega \varepsilon_0}, \tag{3}$$

where σ is the direct-current (dc) conductivity of the medium.

The resulting wave equation is

$$\boldsymbol{\nabla}^2 \boldsymbol{E} = \frac{\tilde{\varepsilon}_\mathrm{r} \tilde{\mu}_\mathrm{r}}{c^2} \frac{\partial^2 \boldsymbol{E}}{\partial t^2}, \tag{4}$$

where c is the speed of light. We assume that the microwave traveling along the nanotube's x axis has a phase factor $\mathrm{e}^{(\mathrm{i}\omega t - \tilde{k}x)}$, then the solution of Eq. (4) is

$$\boldsymbol{E}(x) = \boldsymbol{E}_0 \, \mathrm{e}^{(\mathrm{i}\omega t - \tilde{k}x)}. \tag{5}$$

The $\tilde{k}$ is a complex propagation constant given by

$$\tilde{k} = \mathrm{i}\sqrt{\tilde{\varepsilon}_\mathrm{r} \tilde{\mu}_\mathrm{r}} \frac{\omega}{c} = k_\mathrm{r} + \mathrm{i}k_\mathrm{i}. \tag{6}$$

where the real part k_r of the complex wave number $\tilde{k}$ represents the propagation speed of the phasefront, and the imaginary part k_i reflects the attenuation absorption in the process of microwave propagation in the hydrogen plasma of CNTs.

In the presence of an additional uniform magnetic field B_0, the relative complex permittivity $\tilde{\varepsilon}_\mathrm{r}$ for the plane wave propagating through the plasma in CNTs at an arbitrary incidence angle is derived using the Appleton–Hartree formula[44] as

$$\tilde{\varepsilon}_\mathrm{r} = 1 - \frac{\nu_\mathrm{p}^2}{\nu^2} \left\{ \left[1 - \mathrm{i}\frac{\nu_\mathrm{e}}{\nu} - \frac{\nu_\mathrm{c}^2 \sin^2 \phi}{2(\nu^2 - \nu_\mathrm{p}^2 - \mathrm{i}\nu_\mathrm{e}\nu)} \right] \pm \left[\frac{\nu_\mathrm{c}^4 \sin^4 \phi}{4(\nu^2 - \nu_\mathrm{p}^2 - \mathrm{i}\nu_\mathrm{e}\nu)^2} + \frac{\nu_\mathrm{c}^2}{\nu^2} \cos^2 \phi \right]^{1/2} \right\}^{-1}, \tag{7}$$

where ν is the frequency of the incident electromagnetic wave, $\nu_\mathrm{p} = (1/2\pi)\sqrt{n_\mathrm{e} e^2/\varepsilon_0 m_\mathrm{e}}$ is the hydrogen plasma frequency (n_e is the free electron density of the hydrogen plasma. m_e and e are the mass and the charge of the electron, respectively), ν_e is the electronic effective collision frequency of the hydrogen plasma, the order of magnitude of ν_e is usually 10^9–10^{11} Hz based on the empirical value, $\nu_\mathrm{c} = (e/2\pi m_\mathrm{e})B_0$ is the electron gyrofrequency (B_0 is the magnetic induction intensity of the static background magnetic field), and ϕ is the angle of electromagnetic wave propagation with respect to the external static background magnetic field. The $\pm$ correspond to the left- and the right-hand polarization waves, respectively. By setting

$$r_1 = \sqrt{(\nu^2 - \nu_\mathrm{p}^2)^2 + (\nu_\mathrm{e}\nu)^2}, \tag{8}$$

$$\theta_1 = \mathrm{tg}^{-1} \frac{-\nu_\mathrm{e}\nu}{\nu^2 - \nu_\mathrm{p}^2} + 2k_1\pi, \tag{9}$$

the following equations are obtained

$$\frac{1}{\nu^2 - \nu_\mathrm{p}^2 - \mathrm{i}\nu_\mathrm{e}\nu} = (r_1 \, \mathrm{e}^{\mathrm{i}\theta_1})^{-1} = r_1^{-1} \, \mathrm{e}^{-\mathrm{i}\theta_1}, \tag{10}$$

$$\frac{1}{(\nu^2 - \nu_\mathrm{p}^2 - \mathrm{i}\nu_\mathrm{e}\nu)^2} = (r_1 \, \mathrm{e}^{\mathrm{i}\theta_1})^{-2} = r_1^{-2} \, \mathrm{e}^{-\mathrm{i}2\theta_1}. \tag{11}$$

For the right-hand polarization wave, from Eqs. (7) , (10), and (11), we have

$$\begin{aligned} \tilde{\varepsilon}_\mathrm{r} &= 1 - \frac{\nu_\mathrm{p}^2}{\nu^2} \left\{ \left[1 - \mathrm{i}\frac{\nu_\mathrm{e}}{\nu} - \frac{\nu_\mathrm{c}^2 \sin^2 \phi}{2} r_1^{-1} \, \mathrm{e}^{-\mathrm{i}\theta_1} \right] - \left[\frac{\nu_\mathrm{c}^4 \sin^4 \phi}{4} r_1^{-2} \, \mathrm{e}^{-\mathrm{i}2\theta_1} + \frac{\nu_\mathrm{c}^2}{\nu^2} \cos^2 \phi \right]^{1/2} \right\}^{-1} \\ &= 1 - \frac{\nu_\mathrm{p}^2}{\nu^2} \left\{ \left[1 - \mathrm{i}\frac{\nu_\mathrm{e}}{\nu} - \frac{\nu_\mathrm{c}^2 \sin^2 \phi}{2} r_1^{-1} \times (\cos\theta_1 - \mathrm{i}\sin\theta_1) \right] \right. \end{aligned}$$

$$-\left[\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}(\cos 2\theta_1-{\rm i}\sin 2\theta_1)+\frac{\nu_{\rm c}^2}{\nu^2}\cos^2\phi\right]^{1/2}\Bigg\}^{-1}, \tag{12}$$

where

$$\left[\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}(\cos 2\theta_1-{\rm i}\sin 2\theta_1)+\frac{\nu_{\rm c}^2}{\nu^2}\cos^2\phi\right]^{1/2} = \left[\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}\cos 2\theta_1+\frac{\nu_{\rm c}^2}{\nu^2}\cos^2\phi - {\rm i}\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}\sin 2\theta_1\right]^{1/2}. \tag{13}$$

Set

$$r_2 = \left[\left(\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}\cos 2\theta_1+\frac{\nu_{\rm c}^2}{\nu^2}\cos^2\phi\right)^2+\left(\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}\sin 2\theta_1\right)^2\right]^{1/2}, \tag{14}$$

$$\theta_2 = {\rm tg}^{-1}\left(\frac{-\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}\sin 2\theta_1}{\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}\cos 2\theta_1+\frac{\nu_{\rm c}^2}{\nu^2}\cos^2\phi}\right)+2k_2\pi. \tag{15}$$

By using Eqs. (14) and (15), equation (13) becomes

$$\left[\frac{\nu_{\rm c}^4\sin^4\phi}{4}r_1^{-2}(\cos 2\theta_1-{\rm i}\sin 2\theta_1)+\frac{\nu_{\rm c}^2}{\nu^2}\cos^2\phi\right]^{1/2} = r_2^{1/2}\left(\cos\frac{\theta_2}{2}+{\rm i}\sin\frac{\theta_2}{2}\right). \tag{16}$$

By substituting Eq. (16) into Eq. (12), the following equation is obtained

$$\tilde{\varepsilon}_{\rm r} = 1-\frac{\nu_{\rm p}^2}{\nu^2}\left[1-\frac{\nu_{\rm c}^2\sin^2\phi}{2}r_1^{-1}\cos\theta_1-r_2^{1/2}\cos\frac{\theta_2}{2}+{\rm i}\left(-\frac{\nu_{\rm e}}{\nu}+\frac{\nu_{\rm c}^2\sin^2\phi}{2}r_1^{-1}\sin\theta_1-r_2^{1/2}\sin\frac{\theta_2}{2}\right)\right]^{-1}. \tag{17}$$

We set

$$r_3 = \left[\left(1-\frac{\nu_{\rm c}^2\sin^2\phi}{2}r_1^{-1}\cos\theta_1-r_2^{1/2}\cos\frac{\theta_2}{2}\right)^2+\left(-\frac{\nu_{\rm e}}{\nu}+\frac{\nu_{\rm c}^2\sin^2\phi}{2}r_1^{-1}\sin\theta_1-r_2^{1/2}\sin\frac{\theta_2}{2}\right)^2\right]^{1/2}, \tag{18}$$

$$\theta_3 = {\rm tg}^{-1}\left(\frac{-\frac{\nu_{\rm e}}{\nu}+\frac{\nu_{\rm c}^2\sin^2\phi}{2}r_1^{-1}\sin\theta_1-r_2^{1/2}\sin\frac{\theta_2}{2}}{1-\frac{\nu_{\rm c}^2\sin^2\phi}{2}r_1^{-1}\cos\theta_1-r_2^{1/2}\cos\frac{\theta_2}{2}}\right)+2k_3\pi. \tag{19}$$

From Eqs. (17)–(19), the equivalent relative complex permittivity of the hydrogen plasma in the CNT film under the static magnetic field can be obtained as

$$\tilde{\varepsilon}_{\rm r} = 1-\frac{\nu_{\rm p}^2}{\nu^2}r_3^{-1}\cos\theta_3+{\rm i}\frac{\nu_{\rm p}^2}{\nu^2}r_3^{-1}\sin\theta_3. \tag{20}$$

At a certain wavelength λ, the electromagnetic wave absorption coefficient α for the normal incidence into a thin semitransparent film of thickness d can be determined by

$$\alpha = \frac{4\pi k_{\rm i}d}{\lambda}. \tag{21}$$

Taking into account $\tilde{\varepsilon}_{\rm r}=\tilde{k}^2$, from Eqs. (3), (6), and (21), we obtain the following expression for the microwave absorption coefficient of the hydrogen plasma in the CNT film under the static magnetic field:

$$\alpha = \frac{4\pi d}{\lambda}\left[\frac{(\varepsilon'^2_{\rm r}+\varepsilon''^2_{\rm r})^{1/2}-\varepsilon'_{\rm r}}{2}\right]^{1/2} = \frac{2\sqrt{2}\pi dv}{c}\left[\left(1+\frac{\nu_{\rm p}^4}{\nu^4}r_3^{-2}-\frac{2\nu_{\rm p}^2}{\nu^2}r_3^{-1}\cos\theta_3\right)^{1/2}+\frac{\nu_{\rm p}^2}{\nu^2}r_3^{-1}\cos\theta_3-1\right]^{1/2}, \tag{22}$$

where d is the thickness of the CNT film.

Commonly, the free electronic density in plasma is 10^{17}–10^{20} m^{-3}.[45,46] Considering the hydrogen adsorption content of HiPco CNTs and the mass of the hydrogen plasma divided by the total mass of hydrogen under the microwave irradiation, we take $n_{\rm e}$ to be of the order of 10^{17} m^{-3}.

3. Results and discussion

For different external magnetic field intensities, the microwave absorption coefficients of the hydrogen plasma embedded inside HiPco CNTs have been calculated based on Eqs. (8), (9), (14), (15), (18), (21), and (22) in the frequency range from 0.3 GHz to 30.0 GHz. For $n_{\rm e}=1.181\times10^{17}$ m^{-3}, $\nu_{\rm e}=22.0$ GHz, and $\phi=0$, it can be seen in Fig. 1 that when $\nu_{\rm c}=2.0$ GHz, the maximum absorption coefficient $\alpha_{\max}$ is 16.71 at $\nu=2.60$ GHz. When $\nu_{\rm c}=0$ GHz, $\nu_{\rm c}=12.0$ GHz, and $\nu_{\rm c}=22.0$ GHz, $\alpha_{\max}$ is 16.65, 14.98, and 12.06 at ν, equaling 2.45 GHz, 11.3 GHz, and 21.7 GHz, respectively. Obviously, the maximum absorption peak shifts towards the high frequency with the electron gyrofrequency increasing. When $\nu_{\rm c}$ is changed from 0 GHz to 2.0 GHz, $\alpha_{\max}$ is increased by 0.36%. However, when $\nu_{\rm c}$ is changed from 2.0 GHz to 22.0 GHz,

$\alpha_{\max}$ is reduced by 27.78%. For $\nu_c = 12.0$ GHz and $\nu_c = 22.0$ GHz, two absorption peaks are observed in each case.

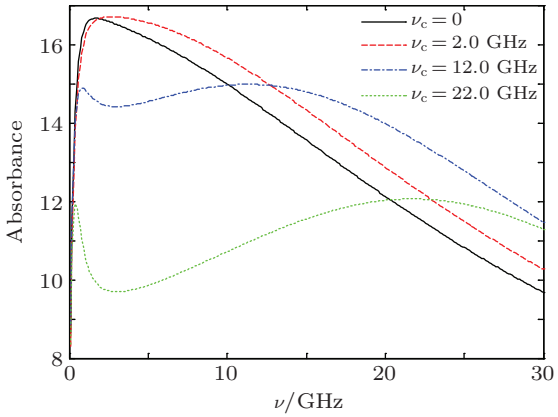


Fig. 1. (colour online) Microwave absorption coefficients of hydrogen plasma in CNT film versus ν under a static magnetic field for different values of ν_c, where $n_e = 1.181 \times 10^{17}$ m^{-3}, $\nu_e = 22.0$ GHz, and $\phi = 0$.

Figure 2 shows the dependence of the microwave absorption coefficient of hydrogen plasma in CNTs on ϕ for different incident microwave frequencies, where $n_e = 1.181 \times 10^{17}$ m^{-3}, $\nu_e = 22.0$ GHz, and $\nu_c = 22.0$ GHz. When $\nu = 4.18$ GHz, the maximum absorption coefficient $\alpha_{\max}$ is 26.32 at ϕ, equaling 1.24 (or 1.90) rad, and α is more than 20 in the ϕ range of 0.95–2.19 rad, i.e., the ϕ width of the absorption coefficient over 20 is 1.24 rad. It is found that the specified frequency microwave (such as 4.18 GHz) can be strongly absorbed by the hydrogen plasma in HiPco CNTs with a wide range of incidence angles. When $\nu = 3.08$ GHz and 4.08 GHz, $\alpha_{\max}$ is 25.17 and 26.24 at ϕ, equaling 1.18 (or 1.96) rad and 1.23 (or 1.91) rad, respectively. When $\nu \leq 4.18$ GHz, two symmetric absorption peaks relative to $\phi = \pi/2$ are observed for each microwave frequency. The absorption peaks approach $\phi = \pi/2$ as ν increases. The theoretical curves show an inflexion frequency of about 4.18 GHz. The absorption coefficient has a positive correlation with the incident microwave frequency before the inflexion frequency and the absorption coefficient dependence reverses after the inflexion. When ν is changed from 3.08 GHz to 4.18 GHz, $\alpha_{\max}$ is increased by 4.57%. When ν is close to 4.28 GHz, the absorption coefficient does not increase but decreases suddenly when the incident microwave frequency increases. For $\nu = 4.28$ GHz, no absorption peak is found, and the absorption coefficient α is less than 12.

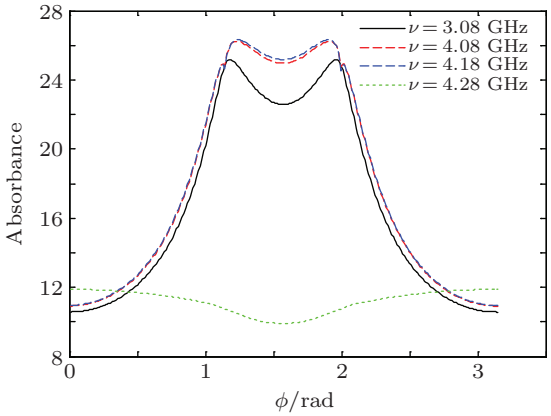


Fig. 2. (colour online) Microwave absorption coefficients of hydrogen plasma in CNT film versus ϕ under a static magnetic field for different values of ν, where $n_e = 1.181 \times 10^{17}$ m^{-3}, $\nu_e = 22.0$ GHz, and $\nu_c = 22.0$ GHz.

4. Conclusions and discussion

We develop a theoretical model for the microwave absorption of hydrogen plasma in HiPco CNTs under an external static magnetic field. The effects of the external magnetic field intensity and the angle of microwave propagation with respect to the external magnetic field direction on the microwave absorption are discussed in detail. For $n_e = 1.181 \times 10^{17}$ m^{-3}, $\nu_e = 22.0$ GHz, and $\phi = 0$, the numerical results indicate that as the external magnetic field intensity increases, the maximum absorption peak shifts towards the high frequency. When $\nu_c = 12.0$ GHz and $\nu_c = 22.0$ GHz, two absorption peaks are observed in each case, which favor the microwave absorption of the material.

For $n_e = 1.181 \times 10^{17}$ m^{-3}, $\nu_e = 22.0$ GHz, and $\nu_c = 22.0$ GHz, when $\phi \leq 4.18$ GHz, two symmetric absorption peaks are observed for each microwave frequency. When $\nu = 4.18$ GHz, the maximum absorption coefficient $\alpha_{\max}$ is 26.32, and the ϕ width of the absorption coefficient over 20 is 1.24 rad.

It is concluded that the microwave absorption properties of the material can be obviously improved when an external static magnetic field is applied to the plasma in CNTs. It is theoretically found that the specified frequency microwave can be strongly absorbed by the hydrogen plasma in HiPco CNTs over a wide range of incidence angles by adjusting the external magnetic field intensity and the parameters of the hydrogen plasma. In addition, it can be noted that when no external magnetic field presents (by setting $\nu_c = 0$), the absorption coefficient in Eq. (22) reduces to the absorption Eq. (28) in Ref. [31], which is in good agreement with the experimental result.

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