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Electron Spin Resonance Line-widths of Carbon Nanotubes based on the Hyperfine Interaction

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Abstract The Kubo formalism and utilizing the projection operator technique (POT) introduced by Kawabata, the electron spin resonance (ESR) line-shape formula for carbon nanotubes through the hyperfine interaction introduced earlier in terms of POT. We can see that the line-width decreases exponentially as the temperature increases. The spin relaxation time show gradual decrease as magnetic field becomes larger. The analysis reveals the peculiarities in spin relaxation inherent to one dimensional system at low temperature and weak magnetic fields. Thus, the present technique is considered to be more convenient to explain the carbon nanotubes as in the case of other optical transitions.

Keywords Electron spin resonance, Projection operator technique, Carbon nanotubes, Line-widths

Introduction

Carbon nanotubes (CNTs) are fascinating low-dimensional objects that offer an outstanding playground to challenge the quantum theory at the nanoscale, manifesting novel physical phenomena. Electron spin resonance (ESR) spectroscopy has been used to determine the electronic properties of CNT. In ESR experiments, one applies a static magnetic field and measures the relative frequency dependence of the absorption power by sweeping an external electromagnetic field. Electron hyperfine interaction (HFI) effects are of great interest in the field of spintronics, and their detailed understanding is both of fundamental and of technological interest, e.g., for the coherent manipulation of spin qubits[1–4]. Since HFI are generally the leading terms breaking the special unitary invariance, deviations in the absorption power from delta peak, e.g., line-widths or line-shift, are directly connected to this interaction.

The objective of the present report is to theoretically investigate the electron spin relaxation properties in the CNTs, crucial piece of information for any spin related phenomena. Specifically, we consider the HFI with nuclear spins $I = 1/2$ of $^{13}\mathrm{C}$ isotopes. The HFI is thought to be one of the most important spin relaxation processes in the CNTs; strong radial confinement of electrons in the CNTs enhances electron-nuclear overlap and subsequently the hyperfine interaction compared to the bulk crystals.

From a theoretical point of view, the studies performed thus far on a resonant system in the presence of external electromagnetic field have

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usually been based on the linear response theories. Among them, we focus on the projection operator technique (POT) of Kawabata [5]. In this report, starting with the Kubo [6] identity and using the POT introduced by Kawabata, the line-shape formula for a single-welled carbon nanotube (SWNT) is derived [7–11]. We also discuss the analysis for quantum limit and draw conclusions.

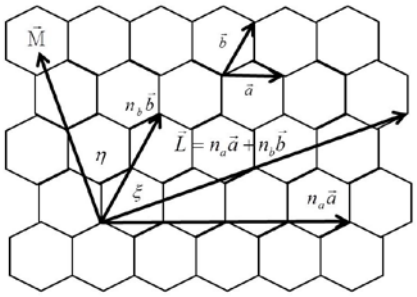

Figure 1. The lattice structure of graphene.

System

We begin with a few essential definitions concerning CNTs. The CNT is conveniently imagined as a spiral graphite sheet (graphene) rolled along the chiral vector $\vec{L}=n_a\vec{a}+n_b\vec{b}$. Here $\vec{a}=a_{cc}(1,0)$ and $\vec{b}=a_{cc}(1/2,\sqrt{3}/2)$ are the graphene lattice unit vectors with $a_{cc}=0.246$ nm and n_a and n_b are integers, which characterize the geometry of a CNT. The chiral angle is $\theta=\tan^{-1}\left[\sqrt{3}n_b/(2n_a+n_b)\right]$, and the tube radius is $\left|\vec{L}\right|=\sqrt{3}a_{cc}\sqrt{n_a^2+n_b^2+n_an_b}$, $d=\left|\vec{L}\right|/\pi$. The property of our interest is the longitudinal (τ_l) and transversal (τ_t) spin relaxation time of an electron with the radius vector $\vec{r}$ and spin $\vec{S}$ in a SWNT. The governing Hamiltonian caused by the Fermi contact HFI with N nuclear spins $\vec{I}^j$ located at lattice sites $\vec{R}_j$ can be expressed as [12–14]

$$H_{HF}=\Omega_0 a_{HF}\sum_{j=1}^{N}\vec{S}\vec{I}^{\,j}\delta(\vec{r}-\vec{R}_j)$$
$$\equiv\vec{\Theta}\vec{S}, \qquad (1)$$

where the HFI constant a_{HF} and the area of the graphene sheet Ω_0 are normalized per carbon atom. This Hamiltonian H_{HF} can also be expressed in terms of the fluctuating field operator $\vec{\Theta}$ that mediates spin relaxation. Field operator $\vec{\Theta}$ must be expressed in terms of electronic Bloch states of the relevant energy bands. In an effective mass approximation, the eigenstates for the conduction bands in the vicinity of the K point take the form

$$\left|k\right\rangle=\frac{1}{\sqrt{2A_0L}}\begin{pmatrix}\Psi_{\mu\nu}(k)\\1\end{pmatrix}\exp\left\{i\left(\varphi_{\mu\nu}\xi+k\eta\right)\right\},$$

$$\Psi_{\mu\nu}(k)=\frac{\varphi_{\mu\nu}-ik}{\sqrt{\varphi_{\mu\nu}^2+k^2}},$$

$$\varphi_{\mu\nu}=\frac{2\pi\left(\nu-\frac{\mu}{3}\right)}{L}, \qquad (2)$$

where A_0 denotes the length of the CNT, and the quantum number $\nu=0,\pm1,\pm2,...$ distinguishes the energy bands, while μ takes one of the three integers $-1,0,1$ that makes $(\nu_1-\nu_2-\mu)$ an integer multiple of 3. As shown in Fig. 1, ξ and η represent the coordinates for the axes directed along $\vec{L}$ (i.e., the circumference) and the CNT (i.e., $\vec{M}$), respectively. The corresponding dispersion relation for the $\left|k\right\rangle$ states reads

$$\varepsilon_{\mu k}=\gamma\sqrt{\varphi_{\mu\nu}^2+k^2}, \qquad (3)$$

where γ is a transfer matrix element. Assuming that only the lowest conduction band is occupied by electrons in CNT with $\nu = +1$ or -1, we restrict our consideration to the $\nu = 0$ case at a given temperature. Then, Eq. (3) in the vicinity of the K point can be approximated as [15]

$$\varepsilon_k = \frac{E_g}{2} + \frac{\hbar^2 k^2}{2m^*}, \quad (4)$$

with an effective mass $m^* = 2\pi\hbar^2 / 3L\gamma$ and the band gap $E_g = 4\pi\gamma / 3L$. In the K' valley, a similar dispersion relation can be obtained when k is substituted by k'. Although it is known that the external static magnetic field $\vec{B}$ modified the CNT electronic states, this effect is neglected as the relevant parameter $(d / 2a_H)^2$ (where d is the CNT diameter and $a_H = \sqrt{c\hbar / eB}$ the magnetic length) is practically very small. Hence, we only consider the influence of B on electron spin state through the Zeeman energy $\sigma\hbar\omega$; $\sigma = \pm 1/2$ is the spin projection on the $\vec{B}$ direction. Using the expressions given above, we can represent the fluctuating field operator in a second quantized form in terms of the electron creation-annihilation operators $a^\dagger_{k\sigma}$ and $a_{k\sigma}$,

$$\Theta_\alpha = \frac{a_{NT}}{A_0} \sum_{k_1,k_2,\sigma} \sum_{j=1}^{N} \exp\left\{ i\left(k_1 - k_2\right)\eta_j \right\} I^j_\alpha a^\dagger_{k_1\sigma} a_{k_2\sigma}, \quad (5)$$

where s denotes the coordinate for the spin states, the direction of the static magnetic field $\vec{B}$ is chosen as the z axis and two transversal directions as x and y ($\alpha = x, y, z$). In addition, $a_{NT} = a_{HF}\Omega_0 / L$ and η_j is the location of the $j-th$ nuclear spin on the CNT axis. As k_1 and k_2 are any two states in the Brillouin zone, Eq. (5) accounts for the effects of both intra- and inter-valley electron scattering on the nuclear spins.

Line-shape formula

The matrix Γ of the line-shape can be reduced to the Bloch-Redfield diagonal form with a leading diagonal composed of matrix elements $\Gamma_{xx} = 1/\tau_t$, $\Gamma_{yy} = 1/\tau_t$, $\Gamma_{zz} = 1/\tau_l$:

$$\frac{1}{\tau_l} = 2\pi n(\omega)\gamma_{xx}(\omega),$$

$$\frac{1}{\tau_t} = \pi[\gamma_{zz}(\omega) + n(\omega)\gamma_{xx}(\omega)], \quad (6)$$

where $n(\omega) = (1 + e^{-\hbar\omega/k_B T}) / 2$ and $\gamma_{\alpha\alpha}(\omega)$ is Fourier transformed correlation function of the operator Θ_α

$$\gamma_{\alpha\alpha}(\omega) = \frac{1}{2\pi\hbar^2} \int_{-\infty}^{\infty} \left\langle \Theta_\alpha(t)\Theta_\alpha \right\rangle e^{i\omega t} dt, \quad (7)$$

Evaluation of the line-shape can be reduced to finding relevant $\gamma_{\alpha\alpha}$. In Eq. (7), $\Theta_\alpha(t) = \exp(iH_{HF}t/\hbar)\Theta_\alpha \exp(-iH_{HF}t/\hbar)$, $\left\langle ... \right\rangle = Tr^{(e)}\left\{ e^{-H_{HF}/k_B T} ... \right\} / Tr^{(e)}\left\{ e^{-H_{HF}/k_B T} \right\}$, where H_{HF} is the Hamiltonian of the system, it take form [16,17]

$$H_{HF} = \sum_{k,\sigma} \varepsilon_k a^\dagger_{k\sigma} a_{k\sigma} + \sum_j \hbar\omega_\nu I^j_Z. \quad (8)$$

The nuclear spin operator I_x contained in the fluctuating field operator Θ_x is conveniently split into two parts $I_x = (I_+ + I_-)/2$ with the raising and lowering operators $I_\pm = I_x \pm iI_y$; correspondingly, $\Theta_\pm$ is defined from $\Theta_x = (\Theta_+ + \Theta_-)/2$ as a formal substitution for index α. Then, by averaging $e^{i(k_1 - k_2)\eta_j}$ over the random distribution of N nuclear isotopes ^{13}C, the Fourier transformation $\gamma_{\pm\mp}(\omega)$ of the correlation function $\left\langle \Theta_\pm(t)\Theta_\mp \right\rangle$ gives

$$\gamma_{\pm\mp}(\omega) = 2N \frac{a^2_{NT}}{\hbar A^2_0} \left\langle I_\pm I_\mp \right\rangle \sum_{k,k',\sigma} f_{k\sigma}(1 - f_{k'\sigma})\delta(\pm\hbar\omega_n + \varepsilon_k - \varepsilon_{k'} + \hbar\omega),$$

(9)

where $f_{k\sigma} = \left\langle a_{k\sigma}^{\dagger} a_{k\sigma} \right\rangle$ is the Fermi-Dirac distribution function for non-degenerate electron state $\left| k \right\rangle$. Since $\gamma_{++}(\omega) = \gamma_{--}(\omega) = 0$ from $\left\langle I_+ I_+ \right\rangle = \left\langle I_- I_- \right\rangle = 0$, Eq. (9) allows one to find $\gamma_{xx}(\omega) = [\gamma_{+-}(\omega) + \gamma_{-+}(\omega)]/4$ as well as $n(\omega)\gamma_{xx}(\omega)$ in the form

$$n(\omega)\gamma_{xx}(\omega) = \frac{1}{8}[\gamma_{+-} + (\omega) + \gamma_{-+}(\omega) + \gamma_{+-}(\omega) + \gamma_{-+}(\omega)].$$

(10)

Using Eqs. (9) and (10) and identity $\left\langle I_{\pm} I_{\mp} \right\rangle = \left\langle I_x^2 \right\rangle + \left\langle I_y^2 \right\rangle \pm \left\langle I_z \right\rangle \cong 2\left\langle I_x^2 \right\rangle$, one can derive relaxation parameters in Eq. (6). Under the assumption that the nuclear spin splitting ω_n is negligible compared to ω, it takes the form

$$\pi n(\omega)\gamma_{xx}(\omega) = N \frac{a_{NT}^2}{\hbar A_0^2} \sum_{k,k',\sigma} \left\langle I_x^2 \right\rangle [f_{k\sigma}(1 - f_{k'\sigma}) + f_{k'\sigma}(1 - f_{k\sigma})] \delta(\hbar\omega + \varepsilon_k - \varepsilon_{k'}).$$

(11)

Applying inequalities $f_{k\sigma} << 1$, $f_{k'\sigma} << 1$, Eq. (11) non-degenerate electrons reduces to

$$\pi n(\omega)\gamma_{xx}(\omega) = 2N \frac{a_{NT}^2}{\hbar A_0^2} \left\langle I_x^2 \right\rangle \sum_{k,k',\sigma} f_{k\sigma} \delta(\hbar\omega + \varepsilon_k - \varepsilon_{k'}).$$

(12)

Similarly, we find

$$\pi\gamma_{zz}(0) = 2N \frac{a_{NT}^2}{\hbar A_0^2} \left\langle I_z^2 \right\rangle \sum_{k,k',\sigma} f_{k\sigma} \delta(\varepsilon_k - \varepsilon_{k'}). \qquad (13)$$

Note that in the case of $I = 1/2$, $I_\alpha^2 = 1/2$ that lead to $\left\langle I_x^2 \right\rangle = \left\langle I_z^2 \right\rangle = 1/4$.

Following Kawabata [5], we define two projection operators $P_{\pm}$ and $Q_{\pm}$ as

$$P_{\pm} Y \equiv \frac{(X_{\pm}, Y)}{(X_{\pm}, \sigma_+)} \sigma_+, \; Q_{\pm} Y \equiv \frac{(Y, \sigma_+)}{(X_{\pm}, \sigma_+)} X_{\pm}, \qquad (14)$$

with $X_{\pm} = a^{+} a$. We easily see that $P_{\pm}$ and $Q_{\pm}$ satisfy the condition imposed on projection operators, $P_{\pm}^2 = P_{\pm}$, $Q_{\pm}^2 = Q_{\pm}$ and $(Q_{\pm} X, (1 - P_{\pm}) Y) = ((1 - P_{\pm}) X, P_{\pm} Y) = 0$. We consider the equation of motion as below

$$\Xi_{\pm}(t) \equiv \frac{(X_{\pm}, \sigma_+(t))}{(X_{\pm}, \sigma_+)}. \qquad (15)$$

We separate $\tilde{X}_{\pm}$ into two parts, parallel and orthogonal to $\tilde{X}_{\pm}$. Then we obtain from Eq. (15)

$$\frac{d\Xi_{\pm}(t)}{dt} = -\frac{(\exp(-iLt)\tilde{X}_{\pm}, \sigma_+)}{(X_{\pm}, \sigma_+)} = i\omega_0 \Xi_{\pm}(t) - \frac{((1 - K_{\pm})X_{\pm}, \sigma_+(t))}{(X_{\pm}, \sigma_+)},$$

$$i\omega_0 = -\frac{(\tilde{X}_{\pm}, \sigma_+)}{(X_{\pm}, \sigma_+)} = \frac{(X_{\pm}, \tilde{\sigma}_+)}{(X_{\pm}, \sigma_+)}, \qquad (16)$$

where $\tilde{X}_{\pm} = iLX_{\pm}$ and $K_{\pm} = (1 - Q_{\pm})X_{\pm}$.

Next we separate $\sigma_+(t)$ into two parts, parallel and orthogonal to σ_+, and using the useful relations

$$L_f \sigma_+ = \omega_0 \sigma_+, \qquad Q_{\pm} L_f \sigma_+ = 0, \qquad (L_f Q_{\pm} X)_{+m-m} 0,$$

since $P_{+-}\sigma_+ = \sigma_+$, from Eq. (11) it follows that $K_{\pm}$ and σ_+ are orthogonal to each other, i.e., $(Q_{\pm}, \sigma_+) = 0$.

When a external electromagnetic radiation with angular frequency ω and amplitude H_0 is incident upon a system along the $z-$axis, a electron spin transition occurs at around $\omega = \omega_z$. Then we obtain

$$\chi_{\pm}^{"}(\omega) = \frac{N a_{NT}^2}{4\pi A_0^2 \hbar^2} \sum_{k.k',\sigma} \frac{\int_{-\infty}^{+\infty} dk_z (f_{k\sigma} - f_{k'\sigma}) \left\langle k \left| I_- \right| k' \right\rangle \left\langle k' \left| I_- \right| k \right\rangle}{i(\omega - \omega_z) + \Gamma_{\pm}^{CNT}[\omega]}.$$

(17)

The power absorption delivered to the system is given by

$$P_{CNT}(\omega) = \frac{1}{2} H_0 \, \mathrm{Re}\left[\chi_{\pm}^{"}(\omega)\right] = N \frac{H_0^2 a_{NT}^2}{A_0^2 h} \omega_z \frac{W_{+-}^{CNT}(\omega) \int_{-\infty}^{+\infty} dk_z [f_{k\sigma} - f_{k'\sigma}]}{\left[\omega - \omega_z - S_{\pm}^{CNT}(\omega)\right]^2 + \left[W_{\pm}^{CNT}(\omega)\right]^2}.$$

(18)

We consider the term $\Gamma_{\pm}^{CNT}[\omega] \equiv iS_{\pm}^{CNT}(\omega) + W_{\pm}^{CNT}(\omega)$ where the

line-shift in ESR spectra is $S_{\pm}^{CNT}(\omega) = \mathrm{Im}\left\{\Gamma_{\pm}^{CNT}[\omega]\right\}$ and the ESR line-width is $W_{\pm}^{CNT}(\omega) = \mathrm{Re}\left\{\Gamma_{\pm}^{CNT}[\omega]\right\}$. The $S_{\pm}^{CNT}(\omega)$ and $W_{\pm}^{CNT}(\omega)$ terms in the denominator of the spin susceptibility are the line-shape formula of the absorption power. The line-shape is important for understanding the working in CNT. The absorption power caused by external radiation can be expressed by a term of the spin susceptibility that is proportional to the imaginary part of the spin susceptibility and the square of the amplitude of the external radiation. The distribution function can be expressed as $\delta F_{k,k',\sigma} = -(f_{k\sigma} - f_{k'\sigma})/(\varepsilon_{k\sigma} - \varepsilon_{k'\sigma})$, we obtain the line-widths formula:

$$\Gamma_{+-}^{EPR}[\omega] \approx \frac{\pi}{\hbar^2} n(\omega)\gamma_{xx}(\omega) \sum_{k,k',\sigma} \frac{\hbar(H_{HF})_{k\sigma}(H_{HF})_{k'\sigma}}{\varepsilon_{k\sigma} - \hbar\omega_z} \delta F_{k,k',\sigma}$$

$$+ \frac{\pi}{\hbar^2}\gamma_{zz}(0) \sum_{k,k',\sigma} \frac{\hbar(H_{HF})_{k\sigma}(H_{HF})_{k'\sigma}}{\varepsilon_{k'\sigma} + \hbar\omega_z} \delta F_{k,k',\sigma}$$

$$+ \frac{\pi}{\hbar^2} n(\omega)\gamma_{xx}(\omega) \sum_{k,k',\sigma} \frac{2\hbar(\varepsilon_{k\sigma} + \hbar\omega)(H_{HF})_{k'\sigma}^2}{\left[\varepsilon_{k'\sigma}{}^2 - \hbar^2(\omega - \omega_z)^2\right]} \delta F_{k,k'\sigma}\left[1 - f_{k\sigma}\right]. \quad (19)$$

In the vicinity of each valley, that leads to the electron distribution function in the form [18]

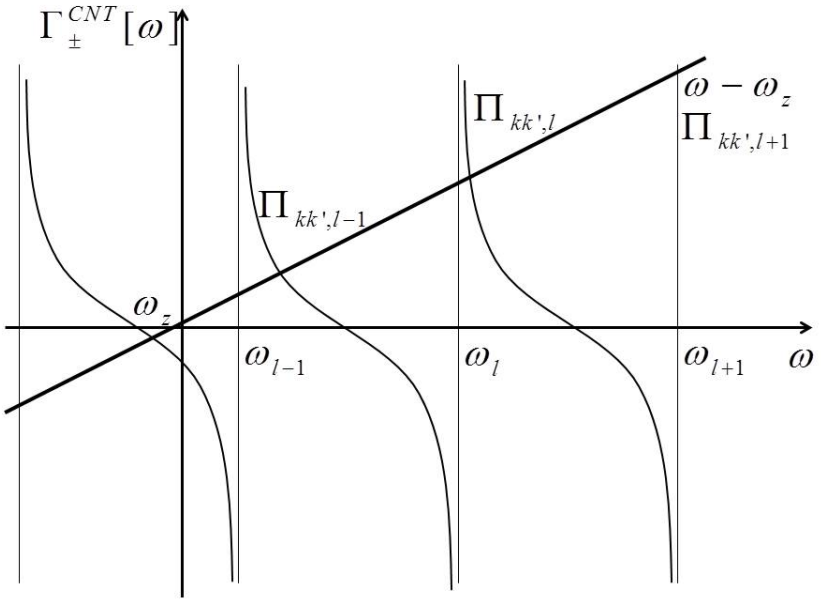

Figure 2. The line-profile function versus ω. Its poles are denoted by ω_l's, and $\Pi_{kk'}$'s are the solutions of Eq. (19).

$$\sum_{k,\sigma} f_{k\sigma} = \frac{\hbar}{2A_0}\sqrt{\frac{2\pi}{m^* k_B T}} \exp(-\varepsilon_{k\sigma}/k_B T). \quad (20)$$

We calculate the line-widths of CNT for the quantum limit. We note that the characteristic feature of the line-widths are determined by the functional dependence of $\Gamma_{\pm}^{CNT}[\omega]$ on ω. In Eq. (16), the factor $f_{k\sigma} - f_{k'\sigma}$ is not zero only for the states near

Fermi level. But its dependence on k is immaterial because the statistical nature of $\Gamma_{\pm}^{CNT}$ is almost independent of them. From Eq. (19) we can easily see that $\Gamma_{\pm}^{CNT}[\omega]$ has poles at $\omega = \pm\varepsilon_k / \hbar$, $\omega = \pm\varepsilon_k / \hbar + \omega_z$, and that if we let ω_l and ω_{l+1} be any two neighboring poles $(\omega_l < \omega_{l+1})$ such as $\Gamma_{\pm}^{CNT}$ goes to positive infinity when $\omega = \omega_l + 0$ and to negative infinity when $\omega = \omega_l - 0$. Therefore, as $i\Gamma_{\pm}^{CNT}$ is monotonically decreasing in the region $\omega_l < \omega < \omega_{l+1}$, the equation $\omega - \omega_z - i\Gamma_{\pm}^{CNT}[\omega] = 0$, will have one and only one solution in this region. We called it $\Pi_{kk',0}$ and the line-profile formula versus ω are illustrated in Fig. 2. Electron spin resonance for the quantum limit can be realized when hyperfine interaction of electrons is not too strong. Examining the nature of $\Gamma_{\pm}^{CNT}$ carefully, we find that there is one solution of Eq. (19) very near $\omega = \omega_z$, which will be called $\Pi_{kk',0}$. As a first order approximation we put $\Pi_{kk',0} = \omega_z i\Gamma_{\pm}^{CNT}[\omega_z]$, for our calculation is meaningful only in the lowest order. Then we have $\Pi_{kk',0} = \omega_z \left\{1 - (2\hbar^2\omega_z^2)\Delta_{kk'}\right\}$, where $\Delta_{kk'} = (2\pi)^{-1}\sum \varepsilon_k^{-1}$. The value of $\Delta_{kk'}$ depend on the distribution of energy levels, but it is positive and in most cases of order of unity.

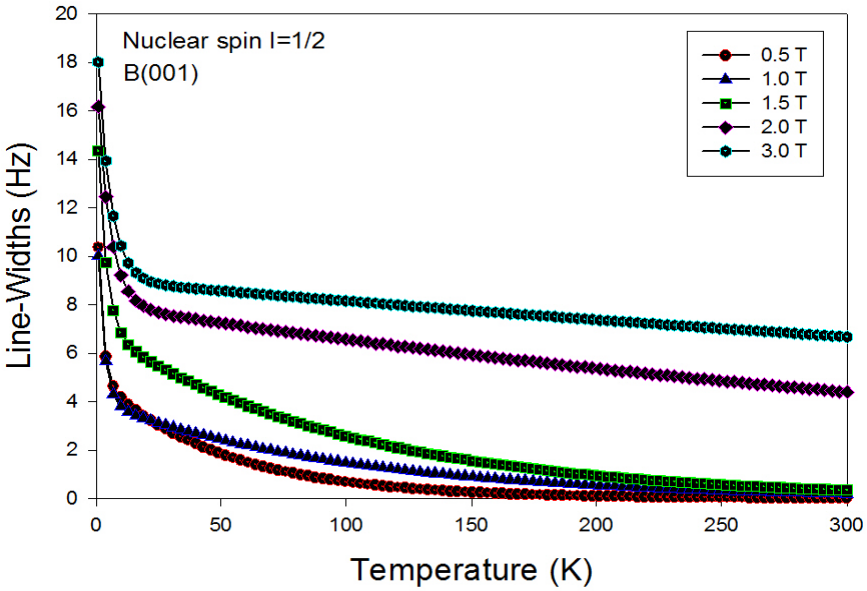

Figure 3. Calculated line-widths in a (8,0) zigzag SWNT as a function of temperature for different values of magnetic field strength.

We consider a zigzag SWNT with (8,0) and assume $\Gamma = 1\ \mu\text{eV}$. Other parameters are known to be: $x = 0.011$, $\Omega_0 = \sqrt{3}b^2/4$, $b = 0.249\ \text{nm}$, $\gamma = \gamma_0\Omega_0 / b$, and $a_{hf} / 2\pi\hbar = 22.5\ \text{MHz}$. Through numerical calculations, Fig. 3 presents the calculated line-width as a function of temperature at various magnetic field strengths. The line-width decreases with increasing temperature due to the hyperfine

interaction of electrons. From the line-widths, on the order of 0.5, 1.0, 1.5, 2.0, and 3.0 T, we can see the broadening effect of line-widths near the resonance peak. Line-widths also show gradual decrease as B becomes larger.

Concluding remarks

So far we consider ESR in a SWNT through the HFI with nuclear spins $I = 1/2$ of ^{13}C isotopes introduced earlier in terms of POT. The theory was applied to examination of temperature dependence of the line-widths for the quantum limit. We can see that the line-width decreases exponentially as the temperature increases. The spin relaxation time show gradual decrease as magnetic field B becomes larger. The analysis reveals the peculiarities in spin relaxation inherent to one dimensional system at low temperature and weak magnetic fields. The line-width is barely affected in the high-temperature region because there is no correlation between the resonance fields and the distribution function. Therefore, we wish to emphases that POT provides a useful method for the analysis presented here, as compared to other methods.

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