Journal of Magnetics 17(1), 13-18 (2012) http://dx.doi.org/10.4283/JMAG.2012.17.1.013

Electron Spin Transition Line-width of Mn-doped Wurtzite GaN Film for the Quantum Limit

Jung-Il Park[1], Hyeong-Rag Lee[1], Su-Ho Lee[2]*, and Dong-Geul Hyun[3]

[1]Nano-Physics and Technology Laboratory, Department of Physics, Kyungpook National University, Daegu 702-701, Korea
[2]Department of Electrical Engineering, Dong-A University, Busan 602-714, Korea
[3]Department of Science Education, Teachers' College, Jeju National University, Jeju 690-781, Korea

(Received 26 October 2011, Received in final form 12 January 2012, Accepted 18 January 2012)

Starting with Kubo's formula and using the projection operator technique introduced by Kawabata, EPR line-profile function for a Mn^{2+}-doped wurtzite structure GaN semiconductor was derived as a function of temperature at a frequency of 9.49 GHz (X-band) in the presence of external electromagnetic field. The line-width is barely affected in the low-temperature region because there is no correlation between the resonance fields and the distribution function. At higher temperature the line-width increases with increasing temperature due to the interaction of electrons with acoustic phonons. Thus, the present technique is considered to be more convenient to explain the resonant system as in the case of other optical transition systems.

Keywords : electron paramagnetic resonance, projection operator, ^{55}Mn, GaN, line-width, spin Hamiltonian, wurtzite structure

1. Introduction

In recent years, it has become clear that wide band-gap semiconductors and oxides doped with transition metals constitute a new class of material systems exhibiting magnetic properties whose origin and methods of control are still not understood [1-4]. Information about the microscopic configuration of transition metal centers has been extracted from their paramagnetic resonance in many crystals, ranging from the purely covalent group IV semiconductors to the ionic group I-VII salts [5, 6]. The aim of this study is to determine the magneto-optical properties of the Mn ground state in epitaxial GaN film. Up to a doping level of about 10^{20} cm^{-3}, we investigate only isolated $S = 5/2$ centers by electron paramagnetic resonance (EPR). In GaN with a wurtzite lattice type, the Ga site has a tetrahedral symmetry that is axially distorted along the c-axis, resulting in C_{3V} symmetry. To good approximation, the crystal field can be considered to be equivalent to the perfect tetrahedral T_d symmetry present in the cubic crystals.

EPR spectroscopy is a technique based on the micro-wave absorption in unpaired electron spins in the presence of external radiation. It is a very powerful and sensitive tool for studying the electronic structures, lattice defects, and magnetic phases present in a material [7]. From a theoretical point of view, the studies performed thus far on a resonant system in the presence of external electromagnetic radiation have usually been based on the following methodologies: Green's function approach, the force-balance approach, Feynmann's path integral approach, the Wigner representation approach, and the projection operator technique. There have been numerous techniques [8, 9] for the calculation of resonance line-width. Suzuki presented a formula for electron systems using the resolvent superoperator method. Shibata and Ezaki developed a new type of expansion technique for determining time-correlation function, obtained kinetic coefficients using Mori's projection method, and applied the formula to calculate the half-width for an impurity system. Among the above mentioned techniques, we focus on the projection operator technique of Kawabata [10]. By using this method, we succeeded in formulating a response theory, which includes the Kubo [8] theory as the lowest-order approximation. The line-width derived is similar to those obtained by other methods. Furthermore, the amount of calculation steps involved in the technique

*Corresponding author: Tel: +82-51-200-7741
Fax: +82-51-200-7743, e-mail: leesuho@dau.ac.kr

of Kawabata is considerably less than that required for any other technique.

In the present study, first we will briefly review the response theory described earlier [11]. Next, we calculate the line-width and compare experimental data of Carlos *et al.* [12]. The scattering strength terms of the line-width is expanded using a conventional series expansion. Through numerical calculations, we analyze the absorption power by sweeping an external electromagnetic field and the temperature dependence of the line-width at a frequency of 9.49 GHz. Finally, we discuss the analysis for quantum limit and draw conclusions.

2. The Response Theory

It is convenient to write the expression of $\langle +m|\sigma_+(t)|-m\rangle$ in the second quantization formalism as $\langle +m|\sigma_+(t)|-m\rangle = Tr^{(e)}\{a^+_{-m}a_{+m}\sigma_+(t)\}$, where $\sigma_+(t)=\sigma_x+i\sigma_y$ on the right hand side is a second quantized operator, and "$Tr^{(e)}$" stands for trace in the space in which the total number of the electrons is restricted to one. Let us define the inner product of two operators X and Y by $(X,Y) = Tr^{(e)}(XY)$. Note that order of X and Y can be exchanged only if X and Y commute with the total number operator of electrons. Since the total Hamiltonian conserves the electron number, we have $(X, Y(t)) = (X(-t), Y)$. The time dependence of operators can be described by an operator L as $X(t) = \exp(iLt)X$, where L defined by $iLX = i[H, X]/\hbar$. Following Kawabata [10], we define two projection operators P_{+-} and Q_{+-} as

$$P_{+-}Y=\frac{(X_{+m-m},Y)}{(X_{+m-m},\sigma_+)}\sigma_+,\ Q_{+-}Y=\frac{(Y,\sigma_+)}{(X_{+m-m},\sigma_+)}X_{+m-m}, \qquad (1)$$

with $X_{+m-m}=a^+_{-m}a_{+m}$. We easily see that P_{+-} and Q_{+-} satisfy the condition imposed on projection operators, $P^2_{+-}=P_{+-}$, $Q^2_{+-}=Q_{+-}$ and $(Q_{+-}X,(1-P_{+-})Y)=((1-P_{+-})X,P_{+-}Y)$. We consider the equation of motion for

$$\Phi_{+m-m}(t)\equiv\frac{(X_{+m-m},\sigma_+(t))}{(X_{+m-m},\sigma_+)}. \qquad (2)$$

We separate $\tilde{X}_{+m-m}$ into two parts, parallel and orthogonal to $\tilde{X}_{+m-m}$. Then we obtain from Eq. (2)

$$\begin{aligned}\frac{d\Phi_{+m-m}(t)}{dt}&=-\frac{(\exp(-iLt)\tilde{X}_{+m-m},\sigma_+)}{(X_{+m-m},\sigma_+)}\\ &=i\omega_0\Phi_{+m-m}(t)-\frac{((1-K_{+m-m})X_{+m-m},\sigma_+(t))}{(X_{+m-m},\sigma_+)},\\ i\omega_0&=-\frac{(\tilde{X}_{+m-m},\sigma_+)}{(X_{+m-m},\sigma_+)}=\frac{(X_{+m-m},\tilde{\sigma}_+)}{(X_{+m-m},\sigma_+)}, \qquad (3)\end{aligned}$$

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

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

$$L_f\sigma_+=\omega\sigma_+,\ Q_{+-}L_f\sigma_+=0,\ (L_fQ_{+-}X)_{+m-m}0, \qquad (4)$$

since $P_{+-}\sigma_+=\sigma_+$, from Eq. (3) it follows that K_{+m-m} and Q_+ are orthogonal to each other, i.e., $(Q_{+-},\sigma_+)=0$. Then we obtain

$$\frac{d\Phi_{+m-m}}{dt}=i\omega_0\Phi_{+m-m}-\int_0^t d\tau\Phi_{+m-m}(\tau)\Gamma^{epr}_{+-}(t-\tau), \qquad (5)$$

where

$$\Gamma^{epr}_{+-}(t)=\frac{(K_{+m-m},R_{+m-m}(t))}{(X_{+m-m}\sigma_+)}. \qquad (6)$$

We note that

$$[H_f,\sigma_+]=\hbar\omega_0\sigma_+ \qquad (7)$$

$$\begin{aligned}R_{+m-m}&=i(1-P_{+-})[H_{sp},\sigma_+]/\hbar,\\ K_{+m-m}&=i(1-P_{+-})[H_{sp},X_{+m-m}]/\hbar, \qquad (8)\end{aligned}$$

since $i(1-P_{+-})[H_f,\sigma_+]=0$ from Eq. (7) and similarly $i(1-Q_{+-})[H_f,X_{+m-m}]=0$. Then to calculate line-width to the second order in H_{sp}, we may replace L which determines the time dependence of $R(t)$ by L_f defined as

$$iL_fO=i[H_f,O]/\hbar \qquad (9)$$

for an arbitrary operator O. Generally the time dependence of $R_{+m-m}(t)$ is complicated, but if we make the above mentioned relations, the behavior of $R_{+m-m}(t)$ becomes simple,

$$R_{+m-m}(t)=\frac{1}{\hbar}i(1-P_{+-})\exp\left(it\frac{H_f}{\hbar}\right)[H_{sp},\sigma_+]\exp\left(it\frac{H_f}{\hbar}\right). \qquad (10)$$

Then we obtain [Appendix]

$$\begin{aligned}i\Gamma^{EPR}_{+-}[\omega]=&\\ -\frac{1}{2\hbar^2}\int_0^\infty([H_{sp},a^+_{-m}a_{+m}],\exp(iL_ft)[H_{sp},\sigma_+])\exp(i\omega t)dt&\\ +\frac{1}{2\hbar^2}\int_0^\infty(Q_{+-}[H_{sp},a^+_{-m}a_{+m}],\exp(iL_ft)[H_{sp},\sigma_+])\exp(i\omega t)dt. \qquad (11)&\end{aligned}$$

3. System

According to the crystal field model, the effect of a crystal medium on the electron levels of the interstitial ion is known as the intracrystalline Stark effect, which is due to the electrostatic field whose symmetry depends on the position of the ion in the crystal lattice. Within the framework of this model the Hamiltonian of a Mn^{2+} ion containing electrons can be expressed after taking the basic interaction between the electrons into account. For an external circularly polarized radiation $\vec{B}(t)=B_0(e^{-i\omega t}+\text{c.c})\hat{z}$ with angular frequency ω applied along the z-axis. We

consider the interaction between electrons, and describe the system in terms of a Hamiltonian. The expression consists of four parts, i.e., orbital energy, electronic Zeeman term, electron spin, and the hyperfine coupling, and is denoted as $H = H_f + H_{eZ} + H_S + H_{hf}$. Additionally, the orbital energy of approximately 10^5 cm^{-1} consists of four parts, i.e., $H_f = H_K + H_e + H_{ee} + H_{so}$, where H_K, H_e, H_{ee}, and H_{so} are the kinetic energy, Coulomb energy, energy of the Coulomb electrostatic interaction of the electrons of the ion, and spin-orbit coupling, respectively.

$$\hat{H}_f = -\sum_i \frac{\hbar^2}{2m_i}\nabla^2 - \frac{1}{4\pi\varepsilon_0}\sum_i \frac{Ze^2}{r_i} + \frac{1}{4\pi\varepsilon_0}\sum_{i<j}\frac{e^2}{|r_i - r_j|} + \sum_i \xi(r_i)\vec{L}_i \cdot \vec{S}_i, \quad (12)$$

where ξ is the spin-orbit coupling coefficient.

The Mn^{2+} ion having the electron configuration $3d^5$ is an S-state ion with $S = 5/2$ and a ground level of $^6S_{5/2}$. When this ion is embedded in a crystal, the spin is affected by the crystal field produced by the neighboring ions because the 3d shell of the Mn^{2+} ion is the outermost one. The electronic spin $S = 5/2$ of the five 3d electrons of the Mn^{2+} Kramers ion is conserved and typically combined to a $^6A_1$ ground-state configuration without orbital momentum. The EPR spectrum of Mn^{2+} ($S = 5/2$, $I = 5/2$) in wurtzite GaN film can be described by assuming the z-axis of the spin Hamiltonian parallel to the crystallographic c-axis and the y-axis parallel to C_2. The effective Mn^{2+} ground state manifold of 36 eigenstates can be described by the spin Hamiltonian that contains zero-field-splitting terms for an axial crystal field of the $^6S_{5/2}$ ground state. It can be characterized by [13]

$$H_{sp} = \mu_B \vec{B}\cdot\vec{g}\cdot\vec{S} + D\left(S_z^2 - \frac{35}{12}\right) + \vec{S}\cdot\vec{A}\cdot\vec{I} + \frac{d}{6}\left(S_\alpha^4 + S_\beta^4 + S_\gamma^4 - \frac{707}{16}\right) + \frac{7F}{36}\left(S_z^4 - \frac{95}{14}S_z^2 + \frac{81}{16}\right) + P\left(I_z^2 - \frac{35}{12}\right) - \mu_B \vec{B}\cdot\vec{g}_I\cdot\vec{I}, \quad (13)$$

where $\vec{B}$ is the applied static magnetic field, $\ddot{g}$ is the spectroscopic splitting tensor, $\vec{S}$ is the electron spin operator, S_α, S_β, and S_γ are the projections of $\vec{S}$ onto the axis of the cubic crystal field. The hyperfine interaction A of the electronic spin with the nuclear spin of ^{55}Mn further splits the spin levels by energies on the order of 1.67×10^{-2} cm^{-1}. The fine-structure parameters d and F of the cubic crystal field for $S = 5/2$ states are smaller and on the order of 8.0×10^{-2} cm^{-1} only. Further contributions such as the nuclear Zeeman interaction characterized by $g_I \approx 0.0004g$ and nuclear quadrupolar interaction characterized by P give rise to even smaller energy corrections.

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 $\omega = \omega_z$. We note that $\chi''_{+-}(\omega)$ can be calculated as

$$\chi''_{+-}(\omega) = \frac{g_e^2\mu_B^2}{8\pi\Omega\hbar^2}\sum_{\mu,\nu,\pm m}\frac{\int_{-\infty}^{+\infty} dk_z\left[f(\varepsilon_{+m}) - f(\varepsilon_{-m})\right]\langle\mu|\sigma_-|\nu\rangle\langle\nu|\sigma_-|\mu\rangle}{i(\omega - \omega_z) + \Gamma_{+-}^{EPR}[\omega]}. \quad (14)$$

The power absorption delivered to the system is given by

$$P(\omega) = \frac{1}{2}H_0^2\,\mathrm{Re}\left[\chi''_{+-}(\nu)\right] = \frac{H_0^2 g_e^2\mu_B^2}{2\Omega h}\omega_z \frac{W_{+-}^{EPR}(\omega)\int_{-\infty}^{+\infty} dk_z\left[f(\varepsilon_{+m}) - f(\varepsilon_{-m})\right]}{\left[\omega - \omega_z - S_{+-}^{EPR}(\omega)\right]^2 + \left[W_{+-}^{EPR}(\omega)\right]^2}. \quad (15)$$

The $f(\varepsilon_{\pm m}) = \exp[\beta(\varepsilon_{\pm m} - \mu) + 1]^{-1}$ is the Fermi-Dirac distribution function of the electron state $|\pm m\rangle$. We consider the term $\Gamma_{+-}^{EPR}[\omega] \equiv iS_{+-}^{EPR}(\omega) + W_{+-}^{EPR}(\omega)$, where the line-shift in EPR spectra is $S_{+-}^{EPR}(\omega) = \mathrm{Im}\{\Gamma_{+-}^{EPR}[\omega]\}$ and the line-width is $W_{+-}^{EPR}(\omega) = \mathrm{Re}\{\Gamma_{+-}^{EPR}[\omega]\}$. The $S_{+-}^{EPR}(\omega)$ and $W_{+-}^{EPR}(\omega)$ terms in the denominator of the spin susceptibility are the line-profile functions of the absorption power. The line-profile is important when it comes to understanding microscopic properties of the electronic state in GaN:Mn^{2+} film. We can directly obtain the line-width and hence can clearly explain the temperature dependence of the line-width. 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 $\delta F_m = -[f(\varepsilon_{+m}) - f(\varepsilon_{-m})]/(\varepsilon_{+m} - \varepsilon_{-m})$, we obtain the line-profile function:

$$\begin{aligned}\Gamma_{+-}^{EPR}[\omega] \approx{}& \frac{1}{\hbar^2}\sum_{\mu\neq+m}\sum_{\Delta m\neq\pm1}\frac{\hbar(H_{sp})_{+m\mu}(H_{sp})_{\mu+m}}{\varepsilon_{\pm m} - \hbar\omega_z}\delta F_\mu \\ &+ \frac{1}{\hbar^2}\sum_{\mu\neq-m}\sum_{\Delta m\neq\pm1}\frac{\hbar(H_{sp})_{-m\mu}(H_{sp})_{\mu-m}}{\varepsilon_{\pm m} - \hbar\omega}\delta F_\mu \\ &+ \frac{1}{\hbar^2}\sum_{\Delta m\neq\pm1}\frac{2\hbar(\varepsilon_{\pm m} + \hbar\omega)(H_{sp})_{+m-m}^2}{\left[\varepsilon_{\pm m}{}^2 - \hbar^2(\omega - \omega_z)^2\right]}\delta F_\mu\left[1 - f(\varepsilon_{-m})\right].\end{aligned} \quad (16)$$

Eq. (16) is similar to Sawaki's result, which is based on the Stark ladder representation. The physical interpretation of Eq. (16) is as follows. The above mentioned terms represent the transition process of the electron spin from a state $+m(-m)$ to $-m(+m)$. Here, the distribution function represents as the condition for the transition process and $f(\varepsilon_{+m})[1 - f(\varepsilon_{-m})]$ represents that for the transition $+m \rightarrow -m$. We obtains the analytical eigenenergies [14]

$$\begin{aligned}\varepsilon_{\pm5/2} &= \pm g\mu_B B + \frac{D}{3} - \frac{d-F}{2} \pm \sqrt{\left[\pm\frac{3}{2}g\mu_B B + 3D + \frac{d-F}{6}\right]^2 + \frac{20}{9}d^2}, \\ \varepsilon_{\pm3/2} &= \pm\frac{3}{2}g\mu_B B - \frac{2}{3}D + (d-F), \\ \varepsilon_{\pm1/2} &= \mp g\mu_B B + \frac{D}{3} - \frac{d-F}{2} \pm \sqrt{\left[\pm\frac{3}{2}g\mu_B B - 3D + \frac{d-F}{6}\right]^2 + \frac{20}{9}d^2}.\end{aligned} \quad (17)$$

The line-width is calculated to be axially symmetric about the z-axis and analyzed in terms of the spin Hamiltonian with the parameters $g = 1.9994$, $A = -hc \times 65 \times 10^{-4}$ cm^{-1}, $d = 8.064 \times 10^{-4}$ cm^{-1}, $D = hc \times 220 \times 10^{-4}$ cm^{-1}, d–$F = hc \times 5 \times 10^{-4}$ cm^{-1}. According to the first-order approximation of large microwave energies $\hbar\omega >> |D| >> |d-F|$, the cubic crystal field parameters always occur in a combination d–F.

4. Line-profile Function for Quantum Limit

In this section, we calculate the line-width of a GaN:Mn^{2+} film for the quantum limit. We note that the characteristic feature of the line-profile function are determined by the functional dependence of $\Gamma^{EPR}_{+-}[\omega]$ on ω. In Eq. (14), the factor $f(\varepsilon_{+m})$–$f(\varepsilon_{-m})$ is not zero only for the states near Fermi level. But its dependence on $\pm m$ is immaterial because the statistical nature of Γ^{EPR}_{+-} is almost independent of them. From Eq. (16) we can easily see that $\Gamma^{EPR}_{+-}[\omega]$ has poles at $\omega = \pm\varepsilon_{\pm m}/\hbar$, $\omega = \pm\varepsilon_{\pm m}/\hbar + \omega_z$, and that if we let ω_l and ω_{l+1} be any two neighboring poles $(\omega_l < \omega_{l+1})$ such as Γ^{EPR}_{+-} goes to positive infinity when $\omega = \omega_l + 0$ and to negative infinity when $\omega = \omega_l - 0$. Therefore, as $i\Gamma^{EPR}_{+-}$ is monotonically decreasing in the region $(\omega_l < \omega < \omega_{l+1})$, the equation

$$\omega - \omega_z - i\Gamma^{EPR}_{+-}[\omega] = 0, \tag{18}$$

will have one and only one solution in this region. We call it $\Omega_{\pm m,l}$ and the line-profile function versus ω are illustrated in Fig. 1. Replacing ω by $\omega - i0$ we can rewrite Eq. (14) as

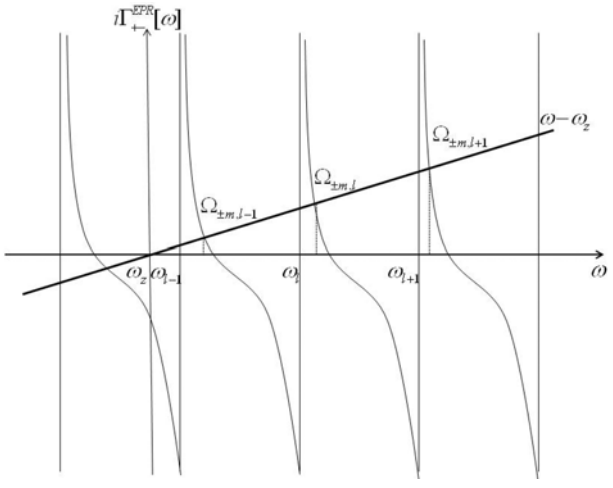

Fig. 1. The line-shape function $i\Gamma^{EPR}_{+-}$ versus ω. Its poles are denoted by ω_l's, and Ω_l's are the solutions of Eq. (18).

$$\chi''_{+-}(\omega) = \frac{g_e^2\mu_B^2}{8\Omega\hbar^2}\sum_{\mu,\nu,\pm m}\int_{-\infty}^{+\infty} dk_z [f(\varepsilon_{+m}) - f(\varepsilon_{-m})]\langle\mu|\sigma_-|\nu\rangle\langle\nu|\sigma_-|\mu\rangle \times \sum_l R_{+m-m,l}\,\delta(\omega - \Omega_{+m-m,l}), \tag{19}$$

where $R_{+m-m,l}$ is the reciprocal of the derivative of the left hand side of Eq. (18) at $\omega = \Omega_{+m-m,l}$ and is given by

$$\frac{1}{R_{+m-m,l}} = 1 + \frac{1}{\hbar}\sum_{\Delta m \neq \pm 1}\left[\frac{1}{(\Omega_{+m-m,l} - \varepsilon_{\pm m}/\hbar)^2} + \frac{1}{(\Omega_{+m-m,l} + \varepsilon_{\pm m}/\hbar)^2} + \frac{1}{(\Omega_{+m-m,l} - \omega_z - \varepsilon_{\pm m}/\hbar)^2} + \frac{1}{(\Omega_{+m-m,l} - \omega_z + \varepsilon_{\pm m}/\hbar)^2}\right], \tag{20}$$

here, we treat quantum limiting case $(\hbar\omega_z << 1)$.

Electron spin transition for the quantum limit can be realized when electron spin interaction is not too strong. Examining the nature of Γ^{EPR}_{+-} carefully, we find that there is one solution of Eq. (18) very near $\omega = \omega_z$, which will be called $\Omega_{+m-m,0}$. As a first order approximation we put $\Omega_{+m-m,0} = \omega_z i\Gamma^{EPR}_{+-}[\omega_z]$, for our calculation is meaningful only in the lowest order. Then we have $\Omega_{+m-m,0} = \omega_z\{1 - (2\hbar^2\omega_x^2)\Delta_{+m-m}\}$, where $\Delta_{+m-m} = (2\pi)^{-1}\Sigma\varepsilon_{\pm m}^{-1}$. The value of Δ_{+m-m} depend on the distribution of energy levels, but it is positive and in most cases of order of unity. Thus the absorption power is the ensemble of the infinitely sharp lines of which the resonance frequency is $\Omega_{+m-m,0}$ and the intensity is $[f(\varepsilon_{+m}) - f(\varepsilon_{-m})]R_{+m-m,0}$. At the same time, the center of the absorption peak will be shifted to lower frequency from ω_z. Through numerical calculations, in Fig. 2, we obtain the absorption power $P(B)$, for the spectrum of a GaN:Mn^{2+} film with the out-of-plane lattice

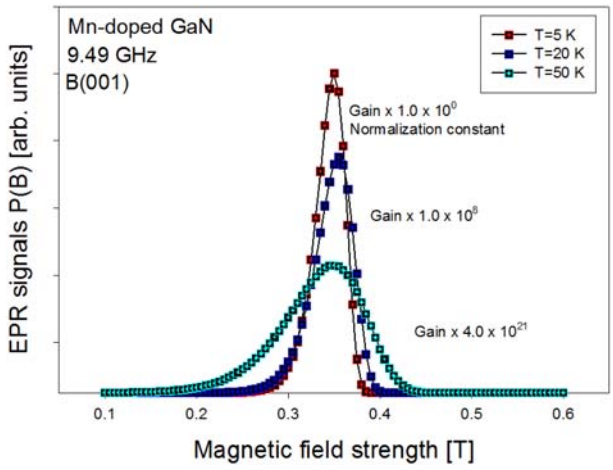

Fig. 2. (Color online) Magnetic field dependence of the absorption power $P(B)$ by sweeping an external electromagnetic field at $T = 5$ K, 20 K, and 50 K with a frequency of 9.49 GHz.

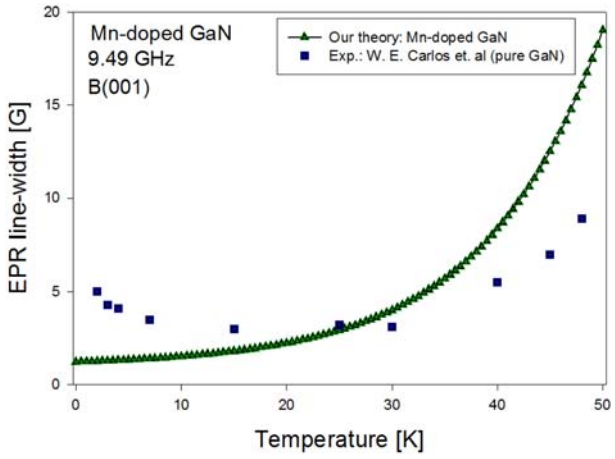

Fig. 3. (Color online) The temperature dependence of the line-width in Mn-doped wurtize GaN film at a frequency of 9.49 GHz. The deltas denote the theoretical results and the squares denote the experimental data of Carlos *et al*.

constant 5.187 Å and the bulk value 3.187 Å at $T = 5$ K, $T = 20$ K, and $T = 50$ K. From the magnetic field dependence of the absorption power by sweeping an external electromagnetic field, we can see the broadening effect near the resonance peak, which exhibits increase as the temperature increases. The analysis of the absorption power is very important for understanding the magnetic properties of materials [15, 16]. In Fig. 3, the temperature dependence of the line-width obtained compare experimental data of Carlos *et al.* [12]. The line-width increases with increasing temperature due to the interaction of electrons with acoustic phonons.

5. Concluding Remarks

So far we have investigated the theory of EPR in Mn^{2+}-doped wurtzite structure GaN film introduced earlier in terms of projection operator technique. The theory was applied to examination of temperature dependence of the line-width for the quantum limit. We can see that the line-width increases exponentially as the temperature increases, and the line-width is almost constant in the low-temperature region. This feature clearly shows that there are two different regions in the graph of the temperature dependence of the line-width. The line-width is barely affected in the low-temperature region because there is no correlation between the resonance fields and the distribution function. At higher temperatures the line-width increases with increasing temperature due to the interaction of electrons with acoustic phonons. The temperature at which this change occurs seems to be approximately 10 K. Therefore, we wish to emphases that projection operator technique provides a useful method for the analysis presented here, as compared to other methods, owing to the reduction in the number of calculation steps. We conclude that the calculation process presented in this work is useful for studying the resonant system.

Appendix

We introduce the annihilation and creation operators, $a_{\pm m}$ and $a_{\pm m}^{+}$, for an eigenstate of H_f. Thus we obtain useful relations as below:

$$\frac{S_{+m}}{\hbar}=a_{+m}^{+}a_{-m},\ \frac{S_{-m}}{\hbar}=a_{-m}^{+}a_{+m},\ \frac{2}{\hbar}S_z=a_{+m}^{+}a_{+m}-a_{-m}^{+}a_{-m}=N_{+m}-N_{-m}, \tag{A1}$$

$$\frac{S_{+m}S_{-m}+S_{-m}S_{+n}}{\hbar^2}=a_{+m}^{+}a_{-m}a_{-n}^{+}a_{+n}+a_{-m}^{+}a_{+m}a_{+n}^{+}a_{-n}$$
$$=-a_{+m}^{+}a_{-n}^{+}a_{-m}a_{+n}-a_{+m}^{+}a_{+n}^{+}a_{+m}a_{+n} \tag{A2}$$

$$-\frac{1}{2}[(N_{+m}+N_{-m})(N_{+n}+N_{-n})+(N_{+m}-N_{-m})(N_{+n}-N_{-n})]$$
$$=a_{+m}^{+}a_{+n}^{+}a_{+m}a_{+n}+a_{-m}^{+}a_{-n}^{+}a_{-m}a_{-n}=-N_{+m}N_{+n}-N_{-m}N_{-n}=-\frac{1}{2}-\frac{2S_m^zS_n^z}{\hbar^2} \tag{A3}$$

Acknowledgments

This research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology (NRF-2011-0027409). The authors thank the Dong-A University Research Fund.

References

[1] T. Dietl and H. Ohno, MRS Bull. 714 (2003).
[2] T. Dietl, in Proceedings 27th International Conference on the Physics of Semiconductors, edited by C. G. V. d. W. J. Menendez, AIP, Melville, New York (2005) p. 56.
[3] C. Liu, F. Yun, and H. Morkoc. J. Mater. Sci. **16**, 555 (2005).
[4] S. J. Pearton, C. R. Abernathy, M. E. Overberg, G. T. Thaler, and L. A. Boather, J. Appl. Phys. **93**, 1 (2003).
[5] G. Cong, Y. Lu, W. Peng, X. Liu, X. Wang, and Z. Wang, J. Cryst. Growth **276**, 381 (2005).
[6] K. Engl, M. Beer, N. Gmeinwieser, U. T. Schwarz, J. Zweck, W. Wegscheider, S. Miller, A. Miler, H. J. Lugauer, G. Bruderl, A. Lell, and V. Harle, J. Cryst. Growth **289**, 6 (2006).
[7] Electron Paramagnetic Resonance of Transition Ions, A. Abragam and B. Bleaney, Clarendon Press, Oxford (1970).

[8] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[9] H. Mori, Prog. Theor. Phys. **34**, 399 (1965).
[10] A. Kawabata, J. Phys. Soc. Jpn. **29**, 902 (1970).
[11] J. I. Park, J. Y. Sug, and H. R. Lee, J. Kor. Phys. Soc. **51**, 623 (2007).
[12] J. I. Park, J. Y. Sug, and H. R. Lee, J. Kor. Phys. Soc. **58**, 1644 (2011).
[13] J. Y. Sug, Phys. Rev. B **64**, 235210 (2001).
[14] J. Y. Sug, Phys. Rev. E **55**, 314 (1997).
[15] W. E. Carlos, J. A. Freitas, Jr., and J. N. Kuznia, Phys. Rev. B **24**, 17878 (1993).
[16] T. Graf, M. Gjukic, M. Hermann, M. S. Brandt, and M. Stutzmann, Phys. Rev. B **67**, 165315 (2003).
[17] J. Schneider and Z. Naturforsch. A **17A**, 189 (1962).
[18] J. I. Park, H. K. Lee, and H. R. Lee, J. Magnetics **16**, 108 (2011).
[19] T. W. Kim and J. K. Oh, J. Magnetics **13**, 43 (2008).