Electron Spin Resonance Investigation of Fe^{3+} in Crystalline $LiNbO_3$ Under the Polarized External Radiation

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Abstract   We study the electron spin resonance line-width (ESRLW) of Fe^{3+} in crystalline $LiNbO_3$; the ESRLW is obtained using the projection operator method (POM) developed by Argyres and Sigel. The ESRLW is calculated to be axially symmetric about the c−axis and is analyzed by the spin Hamiltonian with an isotopic g factor at a frequency of $9.5 \ GHz$. The ESRLW increases exponentially as the temperature increases, and the ESRLW is almost constant in the high-temperature region ($T > 8000 \ K$). This kind of temperature dependence of the ESRLW indicates a motional narrowing of the spectrum when Fe^{3+} ions substitute the Nb^{5+} ions in an off-center position. It is clear from this feature that there are two different regions in the graph of the temperature dependence of the ESRLW.

Keywords   Electron spin resonance, Projection operator method, Magneto-optical transition, Absorption power, Line-width,   ion

Introduction

Electron spin resonance line-width (ESRLW) spectroscopy is a technique based on the microwave absorption in unpaired electron spins in the presence of the polarized external radiation field. It is a very powerful and sensitive tool for studying the electronic structures, lattice defects, and magnetic phases present in a material. The study of magneto-optical transitions of electron spin in crystals has almost always been restricted to a frequency range between far infrared radiation and visible light involving the quantum transitions in ESR[1-5].

$LiNbO_3$ is ferroelectric at room temperature and has a single phase transition to a paraelectric phase at $1473 \ K$, approximately $57 \ K$ below the melting temperature. It has attracted sustained scientific and technical interest, mainly due to its desirable nonlinear optical properties. Doping with transition metal and rare earth elements can enhance electro-optical coefficients and the photorefractive properties. Photorefractive materials are used for holographic data storage and promise high storage densities with short access times. Significant advances have been made toward overcoming problems of sensitivity and volatile readout using near stoichiomeritic $LiNbO_3$ doped with Fe. ESR studies on Fe-doped $LiNbO_3$ LiNbOcrystals have been reported by several authors: Taketa et al.[6],

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Petrov[7], and Rexford et al.[8]. In addition to these ESR studies, many other experiments such as NMR, NQR, the Mössbauer effect, optical absorption, and magnetic properties have been performed.

ESR can readily detect Fe^{3+} ions, the main features of the spectra can be analyzed in terms of a spin Hamiltonian of the form[9]

$$H = H_f + \beta \vec{B} \cdot \vec{g} \cdot \vec{S} + \sum_{k=2,4} \sum_{q=-k}^{k} f_k b_k^q \vec{O}_k^q (S_x, S_y, S_z)$$
$$= H_f + H_{sp},$$
$$O_2^0 = 3S_z^2 - S(S+1),$$
$$O_4^0 = 35S_z^4 - 30S(S+1)S_z^2 + 25S_z^2 - 6S(S+1) + 3S^2(S+1)^2,$$
$$O_4^3 = \{S_z(S_+^3 + S_-^3) + (S_+^3 + S_-^3)S_z\}/4, \quad \textbf{(1)}$$

containing electronic Zeeman and zero field splitting (ZFS) terms expressed using Stevens operators, the scaling factors relevant here take the values $f_2 = 1/3$. Where $\vec{B}$ is the applied static magnetic field, $\vec{g}$ is the spectroscopic splitting tensor, $\vec{S}$ is the electron spin operator, b_k^q denotes a ZFS parameter (k rank, q component) associated with the extended Stevens operator O_k^q. The spin and site symmetry of the paramagnetic center dictate which terms are nonzero. For $S = 5/2$ centers only terms $k = 2, 4$ with $-k \leq q \leq k$, are allowed. The Laue class of the center point group imposes further constraints. The two trigonal classes are relevant for the discussion of previous ESR studies, both allow nonzero terms b_2^0, b_4^0, b_4^{-3}, and b_4^3.

$LiNbO_3$ structure at temperature below its ferroelectric Curie temperature (approximately $1210\ ^\circ C$) consist of planar sheet of oxygen atoms in a distorted hexagonal close-packed configuration. The octahedral interstices formed in this structure are one-third filled by lithium atoms, one-third filled by niobium atoms, and one-third vacant. In the c direction, the atoms occur in the interstices in the following order:..., Nb, vacancy, Li, Nb, vacancy, Li,.... In the paraelectric phase above the Curie temperature, the elastic force of the crystal become dominant and force the lithium and niobium ions into new positions, as illustrated in Fig. 1.
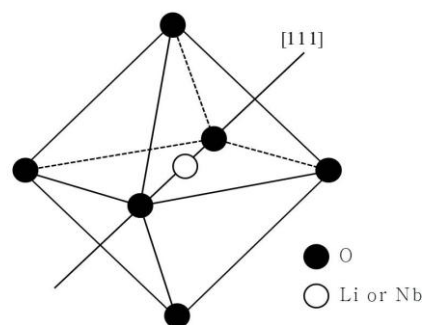
Figure 1. Schematic representation of ionic displacement in $LiNbO_3$ ferroelectric phase..

In this work, first we review the theory presented earlier and change the formula into an easy-to-deal-with form for ferroelectric materials. Next we shall calculate the ESRLW, for the ESR of Fe^{3+} in $LiNbO_3$, the ESRLW is obtained using the projection operator method (POM) developed by Argyres and Sigel. On the basis of numerical calculation, we will analyze the absorption power and the temperature dependence of the ESRLW at a frequency of $9.5\ GHz$ in polarized external radiation field. Finally we will give discussion and concluding remarks.

Review of theory

There have been numerous methods for the calculation of the ESRLW. Using the density operator method, Arora and Spector obtained a formula for various resonant systems including acoustic phonons. Suzuki presented a formula for electron systems using the resolvent superoperator method[12]. Peeters and Devreese presented a theory using the memory function and applied it to study various two- and three-dimensional electron systems[9–11]. Shibata and Ezaki developed a new type

of expansion method for determining time-correlation function, obtained kinetic coefficients using Mori's projection method[13]. The applied this method for studying magnetic impurity systems. Among the above mentioned methods, we focus on the POM approach of Argyres and Sigel[14]. By using this method, we succeeded in formulating a response theory, which includes the Kubo[15] theory as the lowest-order approximation[16–21]. The ESRLW derived is similar to those obtained by other methods. Furthermore, the amount of calculation steps involved in the method of Argyres and Sigel is considerably lesser than that required for any other method. For $LiNbO_3$ in the presence of a perpendicular static magnetic field, the ESR spectrum, up to a constant factor, can be expressed as

$$\frac{-\operatorname{Im}\Gamma_{+-}^{ESR}(\omega)}{\left[\omega-\omega_z-\operatorname{Re}\Gamma_{+-}^{ESR}(\omega)\right]^2+\left[\operatorname{Im}\Gamma_{+-}^{ESR}(\omega)\right]^2}, \quad (2)$$

where $\operatorname{Re}\Gamma_{+-}^{ESR}(\omega)$ and $\operatorname{Im}\Gamma_{+-}^{ESR}(\omega)$ are the real and the imaginary parts respectively, of the ESR spectrum. The correction to the resonance field can then be determined accurately from the equation $\omega-\omega_z-\operatorname{Re}\Gamma_{+-}^{ESR}(\omega)=0$ because $\operatorname{Im}\Gamma_{+-}^{ESR}(\omega)$ is slowly varying function of the angular frequency.

We introduce the annihilation and creation operators, $a_{\pm m}$ and $a_{\pm m}^+$, for an eigenstate of Hamiltonian. We can express the commutator in terms of these operators, as shown below

$$\left\langle\left[\sigma_-,\sigma_+(t)\right]\right\rangle_{EA}=\left\langle\sum_{\pm m}\left\langle-m\left|\sigma_+(t)\right|+m\right\rangle\left\{\left[\left\langle+m\left|\sigma_+(t)\right|+m\right\rangle-\left\langle-m\left|\sigma_+(t)\right|-m\right\rangle\right]a_{-m}^+a_{+m}\right.\right.$$

$$\left.\left.+\left\langle+m\left|\sigma_+(t)\right|-m\right\rangle\left(a_{-m}^+a_{-m}-a_{+m}^+a_{+m}\right)\right\}\right\rangle_{EA}$$

$$=\sum_{\pm m}\left\langle-m\left|\sigma_-\right|+m\right\rangle\left\langle+m\left|\sigma_+(t)\right|-m\right\rangle\left[f(\varepsilon_{-m})-f(\varepsilon_{+m})\right]. \quad (3)$$

There may be various methods for evaluating Eq. (3) that yield similar results; however, the evaluation is more effectively achieved using the POM proposed by Argyres and Sigel, which is a method based on the equation of motion. Their theory seems to be quite general in the sense that it is based on rigorous formalism and the self-consistent projection technique. Following Argyres and Sigel, we define the projection operators by use of the following selection rule

$$P_{+-}X=\frac{X_{+m-m}}{(\sigma_+)_{+m-m}}\sigma_+, \quad \tilde{P}_{+-}=1-P_{+-} \quad (4)$$

where $\tilde{P}_{+-}$ is an Abelian inverse. We note that $P_{+-}\sigma_+=\sigma_+$, $\tilde{P}_{+-}\sigma_+=0$, $P_{+-}^2=P_{+-}$, and $P_{+-}\tilde{P}_{+-}=0$, while we isolate $R(\omega)=P_{+-}R(\omega)+\tilde{P}_{+-}R(\omega)$ and then operate with P_{+-} and $\tilde{P}_{+-}$ separately to obtain with the use of relations

$$(\omega-P_{+-}L)P_{+-}R(\omega)-P_{+-}L\tilde{P}_{+-}R(\omega)=\sigma_+,$$
$$(\omega-\tilde{P}_{+-}L)\tilde{P}_{+-}R(\omega)-\tilde{P}_{+-}LP_{+-}R(\omega)=0. \quad (5)$$

Thus, we obtain the expression for the quantity of interest $R_{+m-m}(\omega)$ as follows

$$\omega-\frac{R_{+m-m}}{(\sigma_+)_{+m-m}}-\frac{\left\{L\left[1+G_{\tilde{P}}(\omega)\tilde{P}_{+-}L\right]\frac{R_{+m-m}}{(\sigma_+)_{+m-m}}\sigma_+\right\}_{+m-m}}{(\sigma_+)_{+m-m}}=1. \quad (6)$$

Because we are interested in the spin Hamiltonian, it is convenient to introduce the Liouville operators L_f and L_{sp} corresponding to H_f and H_{sp}, respectively, i.e., $L=L_f+L_{sp}$ with $L_fX\equiv[H_f,X]$ and $L_{sp}X\equiv[H_{sp},X]$. We then note that $L_f\sigma_+=\omega\sigma_+$, $\tilde{P}_{+-}L_f\sigma_+=0$, and $(L_f\tilde{P}_{+-}X)_{+m-m}=0$, following the definitions of L_f and $\tilde{P}_{+-}$. We make use of the following relations

$$\sum_\mu(\tilde{P}_{+-}X)_{\mu-m}=\sum_{\mu\neq+m}(X)_{\mu-m},$$
$$\sum_\mu(\tilde{P}_{+-}X)_{+m\mu}=\sum_{\mu\neq-m}(X)_{+m\mu},$$
$$(L_f\sigma_+)_{+m-m}=\hbar\omega(\sigma_+)_{+m-m}. \quad (7)$$

System

The ESRLW is characterized by the imaginary part of the equation for electron spin susceptibility

$$\chi_{+-}''(\omega)=\frac{g_e^2\mu_B^2}{4Vh}\lim_{a\to+0}\int_0^\infty dt\ \exp(-i\omega t-at)\left\langle[\sigma_-(0),\sigma_+(t)]\right\rangle_{EA}, \quad (8)$$

where $\sigma_{\pm}(t) = \exp(iLt)\sigma_{\pm}$, with L being the Liouville operator corresponding to the Hamiltonian of the system. We denote the sum of the spin matrix of the total electrons in the system as $\sigma_{\pm} = \sigma_x \pm i\sigma_y$. For a polarized external radiation field and angular frequency ω applied along the z direction, the spin susceptibility delivered to the ferroelectric $LiNbO_3$ crystals under the influence of a static magnetic field B is given by

$$\chi_{+-}^{"}(\omega) = \frac{g_e^2 \mu_B^2}{4Vh}\lim_{a\to 0}\text{Re}\sum_{\pm m}\{f(\varepsilon_{+m})-f(\varepsilon_{-m})\}\langle-m|\sigma_-|+m\rangle\int_0^\infty dt\ \exp(-i\omega t-at)\langle-m|\sigma_+(t)|+m\rangle, \tag{9}$$

where "Re" denotes "the real part of", $f(\varepsilon_{\pm m})$, which is the Fermi-Dirac distribution function of the electron state $|\pm m\rangle$. We note that $\chi_{+-}^{"}(\omega)$ can be calculated as

$$\chi_{+-}^{"}(\omega) = \frac{g_e^2 \mu_B^2}{4V\hbar}\left\langle\sum_{\mu,\nu,\pm m}\frac{\int_{-\infty}^{+\infty}dk_z\left[f(\varepsilon_{+m})-f(\varepsilon_{-m})\right]\langle\mu|\sigma_-|\nu\rangle\langle\nu|\sigma_-|\mu\rangle}{\varepsilon_{+m}-\varepsilon_{-m}-\hbar\omega+H_{+m-m}-H_{-m+m}-i\hbar\Gamma_{+-}^{ESR}(\omega)}\right\rangle_{EA}. \tag{10}$$

We consider the term $\Gamma_{+-}^{ESR}(\omega) \equiv iS_{+-}^{ESR}(\omega) + W_{+-}^{ESR}(\omega)$, where the line-shift in ESR spectra is $S_{+-}^{ESR}(\omega) = \text{Im}\left[\Gamma_{+-}^{ESR}(\omega)\right]$ and the ESRLW is $W_{+-}^{ESR}(\omega) = \text{Re}\left[\Gamma_{+-}^{ESR}(\omega)\right]$. Then we obtain the spin susceptibility as

$$\text{Re}\left[\chi_{+-}^{"}(\omega)\right] = \frac{\omega_z}{4V\hbar}g_e^2\mu_B^2\frac{W_{+-}^{ESR}(\omega)\int_{-\infty}^{+\infty}dk_z\left[f(\varepsilon_{+m})-f(\varepsilon_{-m})\right]}{\left[\omega-\omega_z-S_{+-}^{ESR}(\omega)\right]^2+\left[W_{+-}^{ESR}(\omega)\right]^2}. \tag{11}$$

The absorption power delivered to the system is given by

$$P_{ESR}(\omega) \propto H_0^2\,\text{Re}\left[\chi_{+-}^{"}(\omega)\right]. \tag{12}$$

The amplitude H_0 can be treated as the driving force of ESR. It determines the external frequency and the amplitude of the oscillating component of magnetization. The square of H_0^2, which is the modulus of the ESR signals, controls the absorption power. In order to obtain the ESRLW, the energy

terms can be expressed as $\Delta E_{\nu\mu}^{\pm} = \left[(\varepsilon_\mu-\varepsilon_\nu)\pm\hbar\omega\right]$ and here,

$$\delta F_m = -\frac{f(\varepsilon_{+m})-f(\varepsilon_{-m})}{\varepsilon_{+m}-\varepsilon_{-m}}. \tag{13}$$

$$i\Gamma_{+-}^{ESR}(\omega) = -\frac{i}{\omega\hbar^2}\int_{-\infty}^{\infty}dk_z\int_0^\infty dt\left\{\left[\frac{\Delta E_{\nu+m}^-H_{\mu\nu}\delta F_\mu}{\hbar\omega-\Delta E_{\nu+m}^-}\right]_{\beta\neq\alpha} - \left[\frac{\Delta E_{\nu+m}^-H_{\nu\mu}\delta F_\nu}{\hbar\omega-\Delta E_{\nu+m}^-}\right]_{\mu\neq\nu} - \left[\frac{\Delta E_{\mu+m}^-H_{\mu\nu}\delta F_\nu}{\hbar\omega-\Delta E_{\mu+m}^-}\right]_{\mu\neq\nu}\right.$$
$$+\left[\frac{\Delta E_{\mu\nu}^-H_{+m\nu}\delta F_\mu}{\hbar\omega-\Delta E_{\mu\nu}^-}\right]_{\nu\neq+m} + \left[\frac{\Delta E_{\nu+m}^-H_{\mu\nu}\delta F_\mu}{\hbar\omega-\Delta E_{\nu+m}^+}\right]_{\mu\neq\nu} - \left[\frac{\Delta E_{\mu+m}^-H_{\mu\nu}\delta F_\nu}{\hbar\omega-\Delta E_{\nu+m}^+}\right]_{\mu\neq\nu}$$
$$\left.-\left[\frac{\Delta E_{\mu\nu}^-H_{\mu\nu}\delta F_\nu}{\hbar\omega-\Delta E_{\mu+m}^+}\right]_{\mu\neq\nu} + \left[\frac{\Delta E_{\mu\nu}^-H_{+m\nu}\delta F_\mu}{\hbar\omega-\Delta E_{\beta\nu}^+}\right]_{\nu\neq+m}\right\}\left\{\int_{-\infty}^{\infty}dk_z\delta F_\mu\right\}^{-1}. \tag{14}$$

We separate the energy terms in the denominator of the ESRLW, and rewrite the elements of the ESRLW as Eq. (14) is similar to Sawaki's result [22], which is based on the Stark ladder representation. However, our formula uses more terms for expressing the Fermi-Dirac distribution. The physical interpretation of Eq. (14) 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 $f(\varepsilon_{+m})\left[1-f(\varepsilon_{-m})\right]$ represents that for the transition $+m\to-m$. The energy term $\Delta E_{\mu\nu}^{\pm}$ represents enforces energy conservation.

The ESRLW is calculated to be axially symmetric about the z-axis and analyzed in terms of the spin Hamiltonian with the paramet $g_{\square} = 1.996$, $g_{\perp} = 2.004$ $b_2^0/h = 4.932$ GHz , $b_4^0/h = -0.155$ GHz , $b_4^3/h = 3.5$ GHz, and $b_4^{-3}/h = -0.84$ GHz [23]. The most important features are six intense resonances corresponding to the central transitions, a splitting of the six central resonances that increases with increasing magnetic field strength. Six strong absorptions, corresponding to the $\langle-\frac{1}{2},\pm m| \to \langle+\frac{1}{2},\pm m|$ transitions, are observed. Hence, the $m = -5/2$ component appears at low magnetic fields and the $m = +5/2$ component appears at high fields. A spreading of the other allowed transitions $|+\frac{3}{2},\pm m\rangle \to |+\frac{5}{2},\pm m\rangle$, $|+\frac{1}{2},\pm m\rangle \to |+\frac{3}{2},\pm m\rangle$, $|-\frac{3}{2},\pm m\rangle \to |-\frac{1}{2},\pm m\rangle$, and $|-\frac{5}{2},\pm m\rangle \to |-\frac{3}{2},\pm m\rangle$.

We obtain the analytical eigenenergies

$$\varepsilon_{\pm5/2,\pm5/2} = \pm\frac{5}{2}g\beta B_z + 10b_2^0 + 60b_4^0, \quad \varepsilon_{\pm3/2,\pm3/2} = \pm\frac{3}{2}g\beta B_z - 2b_2^0 - 180b_4^0,$$

$$\varepsilon_{\pm1/2,\pm1/2} = \pm\frac{1}{2}g\beta B_z - 8b_2^0 + 120b_4^0, \quad \varepsilon_{\pm3/2,\pm5/2} = \frac{\sqrt{5}}{2}g\beta B_x \pm i\frac{\sqrt{5}}{2}g\beta B_y,$$

$$\varepsilon_{\pm1/2,\pm3/2} = \sqrt{2}g\beta B_x + i\sqrt{2}g\beta B_y, \quad \varepsilon_{\mp1/2,\pm1/2} = \frac{3}{2}g\beta B_x \pm i\frac{3}{2}g\beta B_y,$$

$$\varepsilon_{\mp1/2,\pm5/2} = 3\sqrt{10}b_4^3. \tag{15}$$

Discussion and conclusions

We have reviewed the theory concerning the POM for magneto-optical transitions in crystalline $LiNbO_3$, which was introduced earlier in terms of a conventional series representation. The theory was applied to examine the temperature dependence of the ESRLW of the $LiNbO_3$. Through numerical calculations, we studied the dependence of absorption power (peak-to-peak) on the magnetic field for -5/2 allowed transitions, as shown in Fig. 2.
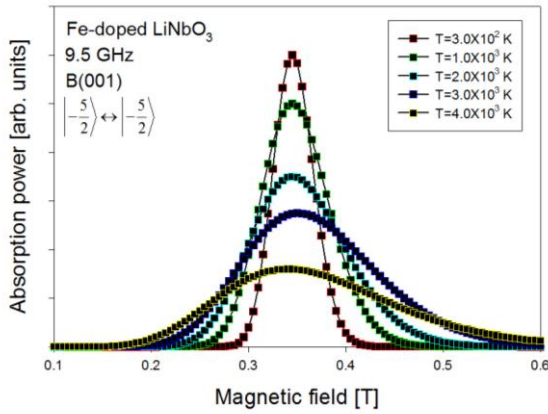

Figure 2. Magnetic field dependence of the absorption power of the Fe^{3+} ion for allowed transition from $-5/2$ to $-5/2$ at a frequency of $9.5\ GHz$.

The spectrum is inhomogeneous broadened by the magnetic dipole-dipole interaction. It should be noted that the absorption power are Lorentzian.
We can see from Fig. 3 that the ESRLW increases exponentially as the temperature increases, and the ESRLW is almost constant in the high-temperature region ($T > 8000\ K$). This feature clearly shows that there are two different regions in the graph of the temperature dependence of the ESRLW: (i) a low-temperature region, in which an exponential increase in the ESRLW is observed with increasing temperature, and (ii) a high-temperature region, in

which a sudden increase in the ESRLW is observed. The ESRLW is barely affected in the high-temperature region because there is no correlation between the resonance fields and the distribution function. This kind of temperature behavior of the EPRLW indicates a motional narrowing of the spectrum, when Fe^{3+} ions substitute the Nb^{5+} ions in an off-center position, and thus, there can be fast jumping of dipoles between several symmetrically equivalent configurations.
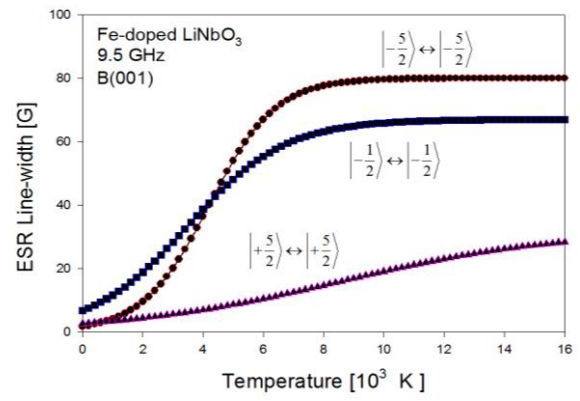
Figure 3. Temperature dependence of the ESRLW of Fe^{3+} at a frequency of $9.5\ GHz$.

We believe that the temperature dependence ESRLW is caused by the spin lattice relaxation time (τ).
Further, $\tau \propto T^{-1}$ implies that the relaxation occurs through transfer of energy from a single spin to a single excitation mode of the lattice, which has essentially the same frequency as the splitting of the levels. This result is quite unexpected because at these high-temperatures, two phonon processes (i.e., Raman process) are known to dominate.
We conclude that the calculation process presented in this work is useful for studying the magneto-optical transitions in $LiNbO_3$ crystals. Furthermore, if the continued fraction methods are used in formulating the ESRLW function, better numerical results may be obtained

Acknowledgements

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