Theory of Photon-Assisted Magnetoacoustic Resonance as a New Probe of Quadrupole Dynamics

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Motivated by the recent progress of phonon-mediated control in quantum spin devices, we propose a possibility of hybrid measurement using electron paramagnetic resonance (EPR) and a surface acoustic wave (SAW). Considering quadrupole-strain (QS) couplings suggested for silicon vacancies, we present a minimum model of the two-level system to investigate a magnetoacoustic resonance (MAR) coupled to various strain modes driven by the SAW. The longitudinal and transverse QS couplings can be changed by rotating a magnetic field, which depends on a combination of the strain modes. Using the Floquet theory, we elucidate each coupling effect on the time-averaged transition probability, especially focus on a single-phonon transition process. The important result is that the longitudinal QS coupling brings about a sharp photon-assisted resonance and leads to an abrupt change in the field-angle dependent transition probability. Since this photon transition process is always accompanied by the photon transition, the field angle for the sharp resonance peak can be detected by the EPR measurement. The hybrid EPR-MAR measurement is useful to confirm the existence of quadrupole degrees of freedom strongly coupled to elastic strains, and thus it is expected to be a complementary probe for the precise evaluation of quadrupole properties.

1. Introduction

Quadrupole degrees of freedom are important factors as well as spin degrees of freedom for high spin states possessing degenerate orbitals. A possibility of electromechanical control of nuclear spins has been reported by the recent experiment of a GaAs-based resonator.\textsuperscript{1} The emergence of sideband nuclear magnetic resonance peaks can be explained by the effect of dynamical quadrupole-strain (QS) coupling on the nuclear spin state. There is also a proposal of mechanically and electrically driven electron spin resonance for nitrogen-vacancy (NV) centers in diamond.\textsuperscript{2} Experimentally, for the NV centers, phonon induced orbital transitions were detected by photoluminescence excitation spectroscopy.\textsuperscript{3–5} Motivated by such progress of phonon-mediated control in spintronics, we theoretically investigate a possibility of hybrid measurement using electron paramagnetic resonance (EPR) and a surface acoustic wave (SAW).\textsuperscript{6} For solid-state electrons, the dynamical QS coupling can be driven by the SAW. By combination of photon and phonon transition processes, more precise analysis is expected to identify the symmetries of quadrupoles and also to reveal hidden quadrupole properties. We propose that the photon-assisted magnetoacoustic resonance (MAR) can be adopted as a complementary or an alternative probe to the conventional measurements of quadrupole such as elastic softening, nuclear quadrupole resonance, resonant x-ray diffraction, and polarized neutron diffraction.\textsuperscript{7}

In particular, we can apply the idea of the hybrid EPR-MAR measurement to confirm an extremely strong strain coupling with a quadrupole moment that possibly emerges in a silicon vacancy. This surprisingly strong QS coupling was first reported by bulk measurements of elastic softening in a boron-doped silicon wafer.\textsuperscript{8–11} The significance of an orbitally degenerate vacancy state was also suggested by early theoretical studies.\textsuperscript{12–14} The precise evaluation of vacancy concentration in the surface of a silicon wafer is inevitably required for information technology of semiconductor devices. In the SAW measurement for the silicon vacancy, the elastic strains are driven by oscillating strain modes such as selected quadrupole symmetries $2z^2 - x^2 - y^2$, $x^2 - y^2$, and $zx$ for the SAW propagating in the $x$ direction of the crystal axis.\textsuperscript{11} Owing to the magnetic-field-dependent multiplet, each QS coupling can be changed by rotating an applied magnetic field and can be evaluated by probing a coupled photon-phonon transition in the EPR measurement.

The Floquet theory is very powerful for examining the two-level system that interacts with a periodically oscillating field. In the Floquet formalism, a problem of solving a time-dependent Schrödinger equation is transformed to a time-independent eigenvalue problem based on an infinite-dimensional matrix form of the Floquet Hamiltonian.\textsuperscript{15–19} In the present study, there are both diagonal (longitudinal) and off-diagonal (transverse) couplings with the oscillating field related to various phonon modes. It is the most important to elucidate the former coupling effect on the photon-assisted transition in a single-phonon resonance process. The significance of the longitudinal coupling effect was previously studied in a different context of a superconducting quantum interference device driven by an ac field.\textsuperscript{16, 17} The key points are the energy shift of a sharp resonance peak and the peak broadening. Considering the transverse coupling effect as well, we extend the Floquet theory, which was frequently applied to multiphoton resonance processes in quantum devices,\textsuperscript{20} to investigate a more complicated problem of phonon-mediated transition processes in solid-state electronic spin systems.

This paper is organized as follows. In the first part of Sect. 2, we present a minimum model for the two-level system including QS couplings and derive this model from a realistic system suggested for the silicon vacancy. We compute the longitudinal and transverse QS couplings with the strain modes, which are given as a function of the rotation angle of the magnetic field. In the second part, the Floquet Hamiltonian is derived for the two-level system coupled to the SAW phonons.
For nearly degenerate Floquet states, it is useful to analyze an effective $2 \times 2$ matrix formulated by the Van Vleck perturbation theory for a multi-phonon resonance.\(^{16,17}\) In Sect. 3, we elucidate the longitudinal-mode and transverse-mode coupling effects on the time-averaged transition probability. Next, we show the field-angle dependence of the transition probability and how to evaluate the QS couplings with different symmetries. Finally, the conclusion and discussion are given by Sect. 4.

2. Model

2.1 Quadrupole-strain couplings in two-level system

To extract essential roles of QS couplings in the transition probabilities detected by the EPR, we study a minimum model of the two-level system described by the following Hamiltonian $H = H_0 + H_{QS} + H_{MW}$:\(^{5}\)

$$H_0 = \frac{1}{2} \hbar \omega_0 (-|g\rangle\langle g| + |e\rangle\langle e|),$$

$$H_{QS} = \frac{1}{2} g_L \varepsilon_L (t) (|g\rangle\langle g| - |e\rangle\langle e|) + \frac{1}{2} g_T \varepsilon_T (t) (|g\rangle\langle e| + |e\rangle\langle g|),$$

$$H_{MW} = \frac{1}{2} \hbar \Omega (|g\rangle e^{-i\omega_{MW} t} + |e\rangle g e^{i\omega_{MW} t}),$$

where $|g\rangle$ and $|e\rangle$ represent the wave functions of the ground and excited states of the two-level system, respectively, with the energy difference $\hbar \omega_0$. In Eq. (2), $g_L (g_T)$ is a QS coupling constant for the longitudinal (transverse) vibration modes between the two states, and the displacement $\varepsilon_L (\varepsilon_T)$ with the time $t$ dependence represents an elastic strain of an acoustic-wave $L$-mode ($T$-mode) coupled to quadrupoles. Equation (3) describes the absorption $|e\rangle\langle g|$ and emission $|g\rangle\langle e|$ processes of a microwave (MW) with frequency $\omega_m$ and the Rabi frequency $\Omega$ characterizes the periodic oscillation of the time-dependent transition probability. Here, the rotating wave approximation (RWA) has been considered.\(^{5}\) In the conventional manner, we introduce a unitary transformation $U = e^{-i\omega_{MW} t/2}$, where the pseudospin operator $I$ is defined by

$$I_L = \frac{1}{2} (|g\rangle\langle g| - |e\rangle\langle e|),$$

$$I_T = \frac{1}{2} (|g\rangle\langle e| + |e\rangle\langle g|) \equiv \frac{1}{2} (I_L + I_T),$$

and $I_y = [I_L, I_T]$. Transforming the original Hamiltonian $H$ into the rotating flame, we have

$$\hat{H} = UHU^\dagger - i\hbar U \frac{\partial U}{\partial t} = -\hbar (\omega_0 - \omega_I) + g_L \varepsilon_L (t) I_L + \frac{1}{2} g_T \varepsilon_T (t) [I_L e^{-i\omega_{MW} t} + L e^{i\omega_{MW} t}] + \hbar \Omega I_y.$$  

(6)

This means that the original Schrödinger equation is transformed as

$$i\hbar \frac{\partial}{\partial t} |\psi\rangle = \hat{H} |\psi\rangle \Rightarrow i\hbar \frac{\partial}{\partial t} U |\psi\rangle = \hat{H} U |\psi\rangle.$$  

(7)

For the propagating SAW, the elastic strain components are given by

$$\varepsilon_L (t) = a_L \cos(\omega_{mt} + \phi_L), \quad \varepsilon_T (t) = a_T \cos(\omega_{mt} + \phi_T),$$

(8)

where $\omega_m$ is the SAW frequency, $a_L (a_T)$ is the amplitude of the $L$-mode ($T$-mode), and the phase shift $\phi_L (\phi_T)$ depends on the positions in the direction of the propagating SAW. Notice that the phase difference $\phi_T - \phi_L$ is constant. Keeping in mind an ultrasonic measurement of gigahertz order, we assume $\omega_m \gg \omega_I$ ($\omega_m \pm \omega_I \approx \omega_m$) in the following discussion. For the high frequency SAW, the transformed Hamiltonian $\hat{H}$ is reduced to

$$\hat{H}(t) = -[\varepsilon_0 + \frac{1}{2} (A_L e^{i\omega_{MW} t} + A_T^* e^{-i\omega_{MW} t})] I_L + \frac{1}{2} [A_T e^{i\omega_{MW} t} + A_L^* e^{-i\omega_{MW} t}] I_T + \frac{1}{2} (\Delta^* I_L + \Delta L),$$

(9)

where

$$\varepsilon_0 = \hbar (\omega_0 - \omega_I), \quad A_L = -g_L a_L e^{i\phi_L}, \quad A_T = g_T a_T e^{i\phi_T}. \quad (10)$$

In the last term of $\hat{H}(t)$, we consider a relative phase difference of the coupled photon and phonon as $\Delta = |\Delta| e^{i\phi} = \hbar \chi e^{i\phi}$, and $\phi_I$ may take arbitrary values owing to random distributions of local quadrupoles (vacancies) in a crystal. For $|\Delta|/\hbar \omega_m \ll 1$, the calculated transition probability is not much dependent on $\phi_I$ as discussed later. Each term in Eq. (9) is sketched by Fig. 1. For understanding the present two-level system, it is helpful to express a matrix form of $\hat{H}(t)$ on the basis of $|g\rangle, |e\rangle$ as

$$\hat{H}(t) = \frac{1}{2} \left( \begin{array}{cc} -\varepsilon_0 & \Delta^* \\ \Delta & \varepsilon_0 \end{array} \right) + \frac{1}{2} \left( \begin{array}{cc} -A_L (\omega_{mt}) & A_T (\omega_{mt}) \\ A_L^* (\omega_{mt}) & A_T^* (\omega_{mt}) \end{array} \right),$$

(11)

and $A_{LT} (\omega_{mt}) = (A_{LT} e^{i\omega_{MW} t} + A_{LT}^* e^{-i\omega_{MW} t})/2$. For $\Delta = 0$, the time-dependent diagonal coupling with $A_L$ only changes the energy difference $\varepsilon_0 = \hbar \omega_0$ and does not contribute to the direct transition between $|g\rangle$ and $|e\rangle$. However, this coupling plays an important role in a photon-assisted transition process for a finite $\Delta$.

2.2 Derivation of two-level system in a realistic case

Here, considering the quartet ground state proposed for the silicon vacancy, we describe the QS interaction with elastic strains driven by the SAW. A combination of QS couplings with different symmetries is changed by rotating an applied

Fig. 1. Two-level system $|g\rangle$ and $|e\rangle$ coupled to strain modes driven by the SAW. The transition between $|g\rangle$ and $|e\rangle$ occurs through the transverse QS coupling $\langle A_T \rangle$ in addition to the MW photon coupling $\langle \Delta \rangle$. The transition is also due to the longitudinal QS coupling $\langle A_L \rangle$ in a photon-assisted transition process. Here, the detuning energy $\varepsilon_0$ is almost proportional to the magnetic field $H_z$ for the Zeeman splitting comparable to the single-phonon energy $\hbar \omega_m$. 

In the boron-doped silicon wafer, the elastic softening is well explained by the strong QS coupling with the vacancy orbital.\(^5\) It is expected that the electronic state with orbital \(L = 1\) and spin \(S = 1/2\) forms the \(\Gamma_8\) quartet ground and \(\Gamma_7\) doublet excited states, owing to the spin-orbit interaction in the \(T_d\) cubic crystal-field environment. We restrict ourselves to the \(\Gamma_8\) quartet described by the \(J = 3/2\) angular momentum operator. As illustrated in Fig. 2, we consider that the SAW propagates in the X[[100]] direction on the Z[[001]] surface of the silicon wafer in the presence of an external magnetic field rotating around Y[[010]] in the ZX plane. Under this experimental setting, the elastic strains coupled to the quadrupoles are given by the following three components:\(^11\)

\[
\begin{align*}
\epsilon_U &= \frac{1}{\sqrt{3}}(2\epsilon_{ZZ} - \epsilon_{XX} - \epsilon_{YY}), \\
\epsilon_V &= \epsilon_{XX} - \epsilon_{YY}, \\
\epsilon_{ZX} &= \frac{\partial u_X}{\partial Z} + \frac{\partial u_Z}{\partial X},
\end{align*}
\]

where \(u_i\) (\(i = X, Y, Z\)) is a displacement vector component and \(\epsilon_{ij} = \partial u_i/\partial x_i\) (\(x_i = i = X, Y, Z\)). Here, an irrelevant \(u_0\) has been considered for the SAW propagating along the X axis, and \(\epsilon_{YZ} = \epsilon_{XY} = 0.\(^11\)) As a consequence, the interaction of the \(\Gamma_8\) quadrupoles and local strains in the magnetic field can be described by \(H_{\text{local}} = H_{\text{Zeeman}} + H_{\text{QS}}:\)

\[
H_{\text{Zeeman}} = -gJ\mu_B J \cdot H,
\]

\[
H_{\text{QS}} = g_\Gamma (O_U \epsilon_U + O_V \epsilon_V) + g_{\Gamma 5} O_{ZX} \epsilon_{ZX}.
\]

Here, the quadrupole operators are represented by the second-rank tensor operators of \(J = (J_X, J_Y, J_Z)\) as

\[
O_U = \frac{1}{\sqrt{3}}(2J^2_U - J^2_X - J^2_Y),
\]

\[
O_V = J_X^2 - J_Y^2,
\]

\[
O_{ZX} = J_Z J_X + J_X J_Z.
\]

In Eq. (15) of the Zeeman term, \(g_J\) is the Landé’s g factor and \(\mu_B\) is the Bohr magneton. Equation (16) consists of the \(\Gamma_8\) and \(\Gamma_5\) irreducible representations in the cubic point group with the QS coupling constants \(g_\Gamma\) and \(g_{\Gamma 5}\), respectively.

Next, we consider the rotation of the magnetic field and choose the \(z\) axis parallel to \(H/|H| = (\sin \theta, 0, \cos \theta)\) in the ZX plane. Accordingly, the \(x\) and \(y\) axes are set as \(x||(-\cos \theta, 0, \sin \theta)\) and \(y||(0, -\sin \theta, 0)\). The components of the \(\mathbf{J}\) operator in the \((xyz)\) coordinate are related to those in the \((XYZ)\) crystal coordinate as

\[
\begin{align*}
J_x &= -\sin \theta \cdot J_Y + \cos \theta \cdot J_Z, \\
J_y &= \sin \theta \cdot J_X - \cos \theta \cdot J_Z, \\
J_z &= \cos \theta \cdot J_X + \sin \theta \cdot J_Y.
\end{align*}
\]

Under the field parallel to the \(z\) axis, it is convenient to replace the quadrupole operators in Eqs. (17)-(19) by the linear combination of \(O_u = (2J_U^2 - J_X^2 - J_Y^2)/\sqrt{3}\), \(O_v = J_X^2 - J_Y^2\), and \(O_{zx} = J_J X + J_J Z\):

\[
\begin{align*}
O_U &\rightarrow -\frac{3}{2} \cos^2 \theta \cdot O_u + \frac{\sqrt{3}}{2} \sin^2 \theta \cdot O_v + \frac{\sqrt{3}}{2} \sin \theta \cos \theta \cdot O_{zx}, \\
O_V &\rightarrow \frac{\sqrt{3}}{2} \sin^2 \theta \cdot O_u + \frac{1 + \cos^2 \theta}{2} O_v - \sin \theta \cos \theta \cdot O_{zx}, \\
O_{ZX} &\rightarrow \frac{\sqrt{3}}{2} \sin \theta \cos \theta \cdot O_u - \sin \theta \cos \theta \cdot O_v - 2 \theta \cdot O_{zx}.
\end{align*}
\]

On the basis of the eigenstates of \(J_z (= \pm 1/2, \pm 3/2)\), the field-angle dependence of the QS interaction in Eq. (16) is explicitly written as \(H_{\text{QS}} = A_u O_u + A_v O_v + A_{\Gamma 5} O_{\Gamma 5}\), where

\[
A_u = g_\Gamma \left[ \left( \frac{1}{4} \epsilon_{XX} + \frac{\sqrt{3}}{4} \epsilon_{XY} \right) + \left( \frac{3}{4} \epsilon_{YY} - \frac{\sqrt{3}}{4} \epsilon_{XY} \right) \cos 2\theta \right] + g_{\Gamma 5} \left( \frac{\sqrt{3}}{2} \epsilon_{ZX} \right) \sin 2\theta, \\
A_v = g_\Gamma \left[ \left( \frac{\sqrt{3}}{4} \epsilon_{XX} + \frac{3}{4} \epsilon_{XY} \right) + \left( -\frac{\sqrt{3}}{4} \epsilon_{YY} + \frac{1}{4} \epsilon_{XY} \right) \cos 2\theta \right] + g_{\Gamma 5} \left( -\frac{\sqrt{3}}{2} \epsilon_{ZX} \right) \sin 2\theta, \\
A_{\Gamma 5} = g_{\Gamma 5} \left( \frac{\sqrt{3}}{2} \epsilon_{XX} - \frac{1}{2} \epsilon_{XY} \right) \sin 2\theta + g_{\Gamma 5} \left( -\epsilon_{ZX} \right) \cos 2\theta.
\]

For a strong magnetic field \(g_\mu_B H_z \sim h_0\omega_0\), we focus on the quadrupole-stain coupling between the ground state with \(J_z = 3/2\) denoted by \(|g\rangle\) and the first excited state with \(J_z = 1/2\) denoted by \(|e\rangle\) for \(H_{\text{Zeeman}}\). In the subspace of the two states, \(h_0\omega_0 = g_\mu_B H_z\) in Eq. (10), and the quadrupole operators \(O_u\) and \(O_{\Gamma 5}\) are reduced to

\[
O_u = \sqrt{3} (|g\rangle \langle g| - |e\rangle \langle e|) = 2 \sqrt{3} I_x, \\
O_{\Gamma 5} = \sqrt{3} (|e\rangle \langle g| + |g\rangle \langle e|) = 2 \sqrt{3} I_x
\]

and there is no \(O_v\) coupling between \(|g\rangle\) and \(|e\rangle\). For the local stain components of the propagating SAW,

\[
\epsilon_{i}(t) = a_i \cos(\omega_0 t + \phi_i)
\]

represents one of the strain modes \((i = U, V, ZX)\), where

\[
O_v = J_X^2 - J_Y^2,
\]

\[
O_{ZX} = J_Z J_X + J_X J_Z.
\]
The coefficient $C_{\mu,i}$ (i = 0, 1, 2) depend on $a_i$ and $\phi_i$ in Eq. (27) as

$$
C_{L,0} = \sqrt{3} g_{\mu} \left[ \frac{1}{2} a_U \left( \cos \frac{\phi_U}{\sin \phi_V} \right) + \frac{\sqrt{3}}{2} a_V \left( \cos \frac{\phi_V}{\sin \phi_V} \right) \right],
$$

$$
C_{L,1} = 3 g_{\mu} \left[ \frac{\sqrt{3}}{2} a_U \left( \cos \frac{\phi_U}{\sin \phi_V} \right) + \frac{1}{2} a_V \left( \cos \frac{\phi_V}{\sin \phi_V} \right) \right],
$$

$$
C_{L,2} = 3 g_{\mu} a_{ZX} \left( \cos \phi_{ZX} \sin \phi_{ZX} \right),
$$

$$
C_{T,1} = -2 \sqrt{3} g_{\mu} a_{ZX} \left( \cos \phi_{ZX} \sin \phi_{ZX} \right),
$$

$$
C_{T,2} = 2 \sqrt{3} g_{\mu} \left[ \frac{\sqrt{3}}{2} a_U \left( \cos \frac{\phi_U}{\sin \phi_V} \right) - \frac{1}{2} a_V \left( \cos \frac{\phi_V}{\sin \phi_V} \right) \right],
$$

and $C_{T,0}$ vanishes.

### 2.3 Floquet Hamiltonian

According to the Floquet theory, the time-dependent Schrödinger equation

$$
i\hbar \frac{\partial \psi(t)}{\partial t} = H(t) \psi(t),
$$

is solved by the following eigenvalue equation

$$[H(t) - i\hbar \frac{\partial}{\partial t}] \psi(t) = q \psi(t).
$$

The time-periodic wave function $\psi(t)$ is related to $\phi(t)$ as $\psi(t) = e^{-i\omega t} \psi(t)$ with the quasienergy $q$. The periodic time-dependent differential equation is transformed to a time-independent infinite-dimensional matrix eigenvalue problem through the Fourier expansion

$$H(t) = \sum_{m=-\infty}^{\infty} H^{[m]} e^{im\omega t}, \quad \phi(t) = \sum_{m=-\infty}^{\infty} \phi^{[m]} e^{im\omega t},$$

and Eq. (36) is reduced to

$$\sum_{m} [H^{[m]} + n\hbar \omega \delta_{mn}] \phi^{[m]} = q \phi^{[m]},$$

where $\delta_{mn}$ is the Kronecker delta. Let us introduce a trial eigenstate $|q_{\gamma}\rangle$ with the corresponding eigenvalue $q_{\gamma}$, which satisfies

$$\sum_{m} [H^{[m]} + n\hbar \omega \delta_{mn}] |q_{\gamma}\rangle = q_{\gamma} |q_{\gamma}\rangle,$$

and the integers $m$ and $n$ run from $-\infty$ to $\infty$. Using orthogonal states labeled by $\alpha$ and $\beta$ in the two-level system, Eq. (38) is rewritten as

$$\sum_{m} \langle \alpha | H^{[m]} | \beta \rangle \langle \beta | q_{\gamma} | \alpha \rangle = q_{\gamma} \langle \alpha | q_{\gamma} | \beta \rangle.
$$

Here, the eigenvector $\langle \alpha | q_{\gamma} \rangle$ is represented by a Floquet state $|\alpha\rangle = \alpha |q_{\gamma}\rangle$ which is considered as an oscillating-field dressed state $|\alpha\rangle$ is also related to $(|\gamma\rangle) \equiv e^{i\omega t}$. The matrix expression of the Floquet Hamiltonian

$$\langle \alpha | H^{[m]} | \beta \rangle = H^{[m]} + n\hbar \omega \delta_{mn}$$

is constructed by an infinite number of $|\alpha\rangle$. Indeed, $H^{[m]} |q_{\gamma}\rangle$ in Eq. (40) is extended to the generalized eigenvalue problem $H^{[m]} |q_{\gamma}\rangle = q_{\gamma} |q_{\gamma}\rangle$, where $q_{\gamma} = q_{\gamma} + i\hbar \omega + \langle \alpha \gamma | q_{\gamma} \rangle$ are satisfied. Owing to this translational invariance, it is sufficient to solve the essential eigenvalue $q_{\gamma}$ by diagonalizing $H^{[m]}$. In the following discussion, $\hbar = 1$ is used for simplicity.

Let us apply the above matrix expression of the Floquet Hamiltonian in Eq. (41) to the two-level system coupled to the SAW and the MW in Eq. (9), where the SAW phonon frequency $\omega_m$ is replaced by $\omega$. First, the block Hamiltonian in the subspace of $|\gamma\rangle$ for the same integer $n$ can be written as

$$H^{[m]} = H^{[m]} + n\hbar \omega = \left( \begin{array} {cc} -\epsilon(0)/2 + \Delta & \Delta/2 \\ \Delta/2 & \epsilon(0)/2 \end{array} \right).$$

There are only finite matrix elements between $|g \rangle$ or $|e\rangle$, and $|g \rangle$ or $|e\rangle$, $m = n \pm 1$ in Eq. (41), and the corresponding block matrix parts are given by

$$H^{[m]} = \left( \begin{array} {cc} -A & A \\ A & -A \end{array} \right), \quad H^{[-m]} = \frac{1}{4} A^{T},$$

while $H^{[0]} = 0$ (|0\rangle \geq 2), where all elements are zeros in the $2 \times 2$ matrix $0$. Using these block matrices, the Floquet matrix is written as

$$H = \left( \begin{array} {ccc} H^{[0]} & H^{[-1]} & 0 & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ H^{[1]} & H^{[0]} & H^{[-1]} & 0 & 0 \\ H^{[-1]} & 0 & 0 & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots \end{array} \right).$$

Eigenvectors and the corresponding eigenvectors are solved by a finite number of blocks of the Floquet matrix. As reported by Ref. 16, we choose $n = 50$, namely, from $H^{[50]}$ to $H^{[0]}$ in Eq. (44), and check that the numerical results are sufficiently converged. Here, we devote ourselves to calculate the time-averaged transition probability between the two states $|g\rangle$ and $|e\rangle$.

$$\tilde{P}_{g \rightarrow e} = \sum_{m} \sum_{\gamma} |\langle \gamma | q_{\gamma} | g \rangle|^{2},$$

in the energy region $0 < \epsilon_0 < 2\omega$, and focus on a single phonon process at around $\epsilon_0 = \omega$.

Inclusion of T-mode phonons ($A_T \neq 0$) makes the numerical analysis more complicated than the case of $A_T = 0$ where
only L-mode phonons are coupled to photons. For the latter, sharp peaks of the transition probability are generated at around \( \epsilon_0 = \omega, 2\omega, \cdots \) for a strong oscillating field of the L-mode \(|A_L|/\omega \gg 1\). Owing to a finite amplitude \( A_T \), these peaks become broad and the nth peak positions shift away from \( \epsilon_0 = n\omega \). It is also marked that the appearance of the transition probability peaks is affected by the phase difference \( \theta_T - \theta_L \) between the two phonon modes as well as the amplitudes \(|A_L|/\omega|A_T|/\omega\). In addition, for the photon-phonon coupled process, it is necessary to consider the phase shift \( \phi_l \) of \( \Delta = |A|\epsilon_{0l}^* \) as well as the amplitude \(|A|\). As discussed in the previous subsection, \( A_L (\mu, L, T) \) is related to the QS coupling amplitude \( g_{\mu
u} \) and the phase shift \( \phi_{\mu} \) of the \( \mu \)-mode, which changes with the rotation angle \( \theta \) of the magnetic field as shown in Eq. (29).

2.4 Nearly degenerate Floquet states

It is very useful to consider the nearly degenerate Floquet states which are involved in a multiphonon process, and this is formulated by the Van Vleck perturbation theory. The derivation of an effective Hamiltonian for the almost degenerate levels was reported in a similar case of the two-level system coupled to an oscillating field.\(^{16,17}\) We extend the previous formulation including a direct off-diagonal coupling of the oscillating field between the two states, namely, \( A_T \) of the \( T \)-mode phonons in addition to \( A_L \) of the L-mode phonons.

Let us start from an unperturbed Hamiltonian \( H_{F0} = H_F(\Delta = 0, A_T = 0) \) in Eq. (44), and the eigenvalue equations are given by

\[
H_{F0} |\tilde{g}n\rangle = \left( -\frac{\epsilon_0}{2} + n\omega \right) |\tilde{g}n\rangle, \tag{46}
\]

\[
H_{F0} |\tilde{e}m\rangle = \left( \frac{\epsilon_0}{2} + m\omega \right) |\tilde{e}m\rangle. \tag{47}
\]

Equation (46) corresponds to the following original differential equation

\[
\frac{1}{2}\left[ -\epsilon_0 - |A_L| \cos(\omega t + \theta_L) \right] - \frac{\partial^2}{\partial t^2} \phi_\nu(t) = \lambda \phi_\nu(t), \tag{48}
\]

where \( A_L = |A_L|e^{i\theta_L} \) and \( \lambda = - (\epsilon_0/2) + n\omega \). The analytic solution is obtained as

\[
\phi_\nu(t) = e^{in\omega t}e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} = \sum_{k=-\infty}^{\infty} e^{ik\theta_L} J_k \left( \frac{|A_L|}{2\omega} \right) e^{i(n+k)\omega t}, \tag{49}
\]

which is expanded with the \( k \)th order Bessel functions \( J_k(z) \). This leads to the expression of \(|\tilde{g}n\rangle\) on the basis of the Floquet states \(|gk\rangle\) as

\[
|\tilde{g}n\rangle = \sum_{k=-\infty}^{\infty} e^{ik\theta_L} J_k \left( \frac{|A_L|}{2\omega} \right) |gk\rangle. \tag{50}
\]

In the same manner for Eq. (47),

\[
|\tilde{e}m\rangle = \sum_{k=-\infty}^{\infty} e^{ik\theta_L} J_k \left( -\frac{|A_L|}{2\omega} \right) |ek\rangle. \tag{51}
\]

Next, for finite \( \Delta \) and \( A_T \), the Floquet matrix elements are rewritten on the new basis set of \(|\tilde{g}n\rangle, |\tilde{e}m\rangle\) for any integer \( n \) and \( m \). Using \( J_n(2z) = \sum_m J_n-m(z)J_m(z) \) for \( z = |A_L|/2\omega \), the matrix element related to the transition \( e \to g \) is calculated as

\[
\langle \tilde{g}n| H_F |\tilde{e}m \rangle = \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} e^{ik\theta_L} \epsilon_{kl} \langle J_{n-l}| e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} |J_{m+l} \rangle \times e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} \langle gk | H_F | e \rangle \times e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} \delta_{mn}, \tag{52}
\]

In the same manner, \(|\tilde{e}m|H_F |\tilde{g}n \rangle \equiv \delta_{nm} \equiv (\delta_{nm})^2 \) is obtained for the transition \( g \to e \). For the matrix element related to the transition \( g \to g \),

\[
\langle \tilde{g}n| H_F |\tilde{g}m \rangle = \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} e^{ik\theta_L} \epsilon_{kl} \langle J_{n-l}| e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} |J_{m+l} \rangle \times e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} \langle gk | H_F | g \rangle \times e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} \delta_{mn}. \tag{53}
\]

Using \( J_{n-l}(z) + J_{n+l}(z) = 2J_n(2z) \) and the summation \( \sum_m J_m(z)J_{m-n}(z) = \delta_{mn} \), we obtain

\[
\langle \tilde{g}n| H_F |\tilde{g}m \rangle = \epsilon_{nm} \sum_{k=-\infty}^{\infty} J_{n-k}(z)J_{m+k}(z) \times e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} \delta_{nm}, \tag{54}
\]

In the same manner, for the transition \( e \to e \),

\[
\langle \tilde{e}m| H_F |\tilde{e}n \rangle = \epsilon_{nm} \sum_{k=-\infty}^{\infty} J_{n-k}(z)J_{m+k}(z) \times e^{i|A_L|/2\omega \sin(\omega t + \theta_L)} e^{i(n+l)\omega t} \delta_{nm}. \tag{55}
\]

In the above calculations, the following matrix elements have been used:

\[
\langle gk| H_F |e \rangle = \delta_{kl} \left( \frac{|A_L|}{2} \right) J_{n-l}(z)J_{m+l}(z), \tag{56}
\]

\[
\langle ek| H_F |g \rangle = \delta_{kl} \left( \frac{|A_L|}{2} \right) J_{n-l}(z)J_{m+l}(z), \tag{57}
\]

\[
\langle gk| H_F |g \rangle = \left( \frac{|A_L|}{2} \right) \delta_{kl} \left( \frac{|A_L|}{2} \right) J_{n-l}(z)J_{m+l}(z), \tag{58}
\]

\[
\langle ek| H_F |e \rangle = \left( \frac{|A_L|}{2} \right) \delta_{kl} \left( \frac{|A_L|}{2} \right) J_{n-l}(z)J_{m+l}(z), \tag{59}
\]

Let us consider that the nearly degenerate states \(|\tilde{g}0\rangle\) and \(|\tilde{e}, -n \rangle\) are coupled to each other through \( v^\delta_{\tilde{g}e} \) defined in
Eq. (52), where $-\varepsilon_0/2 \approx \varepsilon_0/2 - n\omega$. The infinite-dimensional matrix of $H_F$ on the basis of $|\{\bar{g}n\}, \{\bar{e}m\}\rangle$ is reduced to an effective $2 \times 2$ matrix within $|\bar{g}0\rangle$ and $|\bar{e}n\rangle$ using the Van Vleck perturbation theory. The perturbation parameters are $|A|$ and $|A_T|$. Using a similar matrix form in Refs. 16 and 17, we present an effective matrix of

$$
\tilde{H}_F = \begin{pmatrix}
-\frac{\varepsilon_0}{2} + \delta & i\tilde{g}_n^b \\
\tilde{g}_n^b & -\frac{\varepsilon_0}{2} - \delta - n\omega
\end{pmatrix},
$$

(60)

where $\delta$ is the energy shift that corresponds to the ac Stark shift,\(^\text{16,17}\) and the off-diagonal matrix element is represented by the first order of the perturbation parameters as

$$
\tilde{g}_n^b = \left(\tilde{v}_k^{\bar{g}}\right)^* = e^{-i\delta t} \frac{\Delta}{2} J_k \left(\frac{|A_L|}{\omega}\right) + \frac{|A_T|}{4} e^{-i(i\theta_i + \theta_j)} J_{k+1} \left(\frac{|A_L|}{\omega}\right)
$$

(61)

The leading term of $\delta$ is given by

$$
\delta = -\sum_{l=0}^{\infty} \frac{|\tilde{v}_k^{\bar{g}}|^2}{\varepsilon_0 + k\omega},
$$

(62)

which indicates that an $n$-phonon resonance occurs at $\varepsilon_0 = n\omega + 2\delta$. Indeed, the eigenvalues of $\tilde{H}_F$ are

$$
\tilde{q}_n = \frac{n\omega}{2} \pm \sqrt{\left(n\omega - \varepsilon_0 + 2\delta\right)^2 + \left|\tilde{g}_n^b\right|^2},
$$

(63)

which lead to the time-dependent transition probability from $|\bar{g}\rangle$ to $|\bar{e}\rangle$ mediated by $n$ phonons

$$
p^{(n)}_{\bar{g} \rightarrow \bar{e}}(t) = \frac{|\tilde{v}_n^{\bar{g}}|^2 \sin^2 \tilde{q}t}{4} = \frac{|\tilde{v}_n^{\bar{g}}|^2}{4} - \cos 2\tilde{q}t,
$$

(64)

where $\tilde{q} = \sqrt{(n\omega - \varepsilon_0 + 2\delta)^2 + |\tilde{g}_n^b|^2}$. In the long-time limit, we obtain the $n$-phonon time-averaged transition probability

$$
p^{(n)}_{\bar{g} \rightarrow \bar{e}} = \frac{1}{2} \frac{|\tilde{v}_n^{\bar{g}}|^2}{4} + \frac{(n\omega - \varepsilon_0 + 2\delta)^2}{4}.
$$

(65)

The broadening of the transition peaks $4|\tilde{v}_n^{\bar{g}}|^2\ell$ depends on the following model parameters

$$
\Delta \equiv |A|^{\bar{g}}|A_L|, \quad \theta_{TL} \equiv \theta_T - \theta_L, \quad J_k \equiv J_k \left(\frac{|A_L|}{\omega}\right),
$$

(66)

and is explicitly written as

$$
|\tilde{v}_n^{\bar{g}}|^2 = \frac{1}{4} \left(\left|J_+ \left(\cos \phi - n\omega \frac{|A_T|}{|A_L|} \cos \theta_{TL}\right)^2 + J_{-1} |A_L| \sin \phi \right|^2 + \frac{|A_T|^2}{2} (J_{-1} - J_{-2}) \sin \theta_{TL}\right)^2.
$$

(67)

3. Results

First, the $A_L$ and $A_T$ dependence of the transition probability in the two-level system is investigated by solving Eqs. (42)–(44). When $|A_L|$ and $|A_T|$ are fixed, the transition probability depends on the phase difference $\theta_{TL}$ between the $L$-mode and $T$-mode phonons. Although it is also dependent on the photon coupling represented by $|A| e^{i\phi}$, the phase $\phi$ effect on the transition probability is negligibly small for $|A|/\omega \ll 1$, where $\omega$ is the energy of a single phonon.

Next, the roles of QS couplings in the transition probability are elucidated for the two basis states split by a magnetic field and coupled to each other through the quadrupole operators in Eqs. (25) and (26). As described in Sect. 2.2, $\varepsilon_0$ corresponds to the Zeeman energy of the level splitting. The QS couplings $A_L$ and $A_T$ in Eq. (10) depend on the rotation angle $\theta$ of the magnetic field. Both couplings are determined from the combination of the three strain modes in Eq. (29). For a quartet system such as the $\Gamma_6$ state in the silicon vacancy, we have assumed that the magnetic field is strong enough ($\varepsilon_0/\omega \sim 1$) to neglect the contribution from the higher excited states, namely, the $O_v$ coupling between the ground and second excited states. Although we focus on a single phonon process near $\varepsilon_0/\omega = 1$, we show the results for the two-level system including the $\varepsilon_0/\omega \ll 1$ parameter region to elucidate the roles of longitudinal and transverse QS couplings in the field-dependent transition probability. Indeed, inclusion of the higher excited states in the present model is required for a quantitatively analysis of the quartet case, especially for a small magnetic field.

3.1 Effects of longitudinal and transverse phonon couplings on time-averaged transition probability

Let us consider the $L$-mode for $A_T = 0$ at first. The detailed study was already reported by a similar model used for the multiphoton quantum interference.\(^\text{16}\) Here, the role of photons is replaced by that of $L$-mode phonons, while the photon is involved in the transition probability at $\varepsilon_0 = 0$ if there is no contribution from the phonons. For $A_L = 0$, the transition probability shows the maximum $1/2$ only at $\varepsilon_0 = 0$ that corresponds to a low magnetic field $H_L \sim \omega_t \ll \omega$. On the other hand, in the absence of the photon coupling ($\Delta = 0$), the transition cannot be caused by the $L$-mode phonons for $A_L \neq 0$. Thus, the photon-assisted process is
In particular, for $|A_T|/\omega \leq 1$, the transition probability shows narrow peaks near $\varepsilon_0 = 3\omega, 5\omega, \cdots$ and a relatively broad peak near $\varepsilon_0 = \omega$. When $|\Delta|$ is finite, additional narrow peaks emerge near $\varepsilon_0 = 2\omega, 4\omega, \cdots$ and the peak widths become broader with increasing $|\Delta|$ as shown in Fig. 4. For a relatively large $|\Delta|$, the peak at $\varepsilon_0 = \omega$ merges into the peak at $\varepsilon_0 = 0$, and the transition probability shows an almost constant value close to $1/2$ in $0 < \varepsilon_0 < \omega$. This indicates that the contribution from the higher excited states may not be negligible in the quartet case discussed in Sect. 2.2.

To elucidate the roles of the phonon coupling with the $L$- and $T$-modes together in the transition probability, we devote ourselves to a small $|\Delta|/\omega < 0.1$ for the photon coupling, where the effect of the photon phase shift $\phi_i$ is negligible. When both $A_L$ and $A_T$ are finite, the phase difference $\theta_{TL}$ of the two phonon modes affects the transition probability to some extent. This is an important point for the MAR assisted by photons. Here, we examine a single phonon resonance for $|A_T|/|A_L| < 1$ to show explicitly a key role of a finite $A_T$. The $A_T$ effect brings about the broadness of a narrow peak in the transition probability at $\varepsilon_0 = \omega$ and also shifts the peak position $\varepsilon_0$ towards a lower value from $\omega$. It is specially marked that the sharpness of the peak becomes prominent for $|A_L|/\omega \approx 1.8$. This is explained by Eq. (67) in which the difference of the Bessel functions ($J_0$–$J_2$) almost vanishes for $n = 1$. In addition, the resonance peak becomes narrower when $\theta_{TL}$ is adjusted to satisfy $\cos \theta_{TL} = (|\Delta|/\omega|A_T|) = 1.8|\Delta|/|A_T| < 1$, where $|\delta_{TL}|$ is almost close to zero. The $\theta_{TL}$ dependence is shown for $|\Delta|/\omega = 0.01, |A_L|/\omega = 1.8$ and $|A_T|/\omega = 0.8$ in Fig. 5. According to Eq. (29), the QS couplings depend on the rotation angle $\theta$ of the magnetic field. Thus, $A_L = -g_L a_t e^{i\theta} \varepsilon$ and $A_T = g_T a_t e^{i\theta} \varepsilon$ can be controlled by the field angle for the appearance of such a sharp transition probability peak as demonstrated in the next subsection.

![Fig. 4](image_url) (Color online) Transition probability for the $T$-mode phonons plotted as a function of $\varepsilon_0/\omega$ for various $\Delta$, where $A_T/\omega$ is chosen for the $T$-mode coupling and $A_L = 0$.

![Fig. 5](image_url) (Color online) Transition probability for the two-level system coupled to both $L$- and $T$-mode phonons plotted as a function of $\varepsilon_0/\omega$ for $\Delta/\omega = 0.01$. The amplitudes of the $L$-mode ($A_L = |A_L|e^{i\phi_i}$) and $T$-mode ($A_T = |A_T|e^{i\phi_i}$) couplings are fixed as $|A_L|/\omega = 1.8$ and $|A_T|/\omega = 0.8$. The broadness of the peak near $\varepsilon_0/\omega = 1$ is sensitively dependent on the phase difference of the two modes $\theta_{TL} = \theta_T - \theta_L$ around $\theta_{TL}/\pi = 0.5$.

required for a finite transition probability coupled to the $L$-mode phonons, and the transition probability peaks reach $1/2$ at nearly $\varepsilon_0 = \omega, 2\omega, \cdots$. This indicates a possibility of MAR with the use of the $L$-mode phonons. Figure 3 shows that the transition probability peaks become sharp at $\varepsilon_0 = n\omega$ for $|\Delta|/\omega \ll 1$. As $|\Delta|$ increases, each peak broadens and the $n$th peak position $\varepsilon_0$ shifts towards a lower value from $\varepsilon_0 = n\omega$. In particular, the single phonon process at $\varepsilon_0 = \omega$ shows the largest shift. This result is inferred from Eqs. (62), (65) and (67). The broadness of the peaks are represented approximately by $|\delta_{TL}| = 2|\Delta|J_0(|\Delta|/\omega)$ and the peak shifts also obey $|\delta_{TL}|^2$ for $|\Delta|/\omega \ll 1$. The calculated transition probability for $A_T = 0$ does not depend on the phase shifts $\phi_i$ and $\theta_L$.

Next, we show the role of the $T$-mode phonon in the absence of the $L$-mode for $A_L = 0$. This was originally investigated by Shirley, formulating the Floquet theory for the two-level system with an oscillating field, and corresponds to $\Delta \to 0$ in our model.\textsuperscript{15} In particular, for $|A_T|/\omega \leq 1$, the transition probability shows narrow peaks near $\varepsilon_0 = 3\omega, 5\omega, \cdots$ and a relatively broad peak near $\varepsilon_0 = \omega$. When $|\Delta|$ is finite, additional narrow peaks emerge near $\varepsilon_0 = 2\omega, 4\omega, \cdots$ and the peak widths become broader with increasing $|\Delta|$ as shown in Fig. 4. For a relatively large $|\Delta|$, the peak at $\varepsilon_0 = \omega$ merges into the peak at $\varepsilon_0 = 0$, and the transition probability shows an almost constant value close to $1/2$ in $0 < \varepsilon_0 < \omega$. This indicates that the contribution from the higher excited states may not be negligible in the quartet case discussed in Sect. 2.2.

3.2 Effects of quadrupole-strain couplings on time-averaged transition probability

On the basis of the QS couplings described by Eqs. (28)–(34), we change the quadrupole coupling constants ($g_T$ and $g_{T_1}$), the local strain amplitudes ($a_t, a_v, a_{2X}$) and the relative time-independent phase shifts of the SAW modes ($\phi_{T1}$ and $\phi_{2X}$ measured from $\phi_{TX} = 0$). Here, $a_t = a_v$ is set for simplicity, so that the weight of the $\Gamma_3$ and $\Gamma_5$ QS couplings in the two-level system is represented by the ratio of $g_T a_t$ and $g_T a_{2X}$. The energy difference between the two states is changed as $\varepsilon_0 = \varepsilon$, and the $z$ axis of the field orientation is rotated as $(\sin \theta, 0, \cos \theta)$ in the $ZX$ plane of the crystal (see Fig. 2). First, we compute $|A_L|, |A_T|$ and $\theta_{TL}$, as a function of the field angle $\theta$ for given $g_T a_t, g_T a_{2X}, \phi_{TX}$, and $\phi_3$. Next, we solve the eigenvalue problem of $H_2$ in Eq. (44) to obtain the time-averaged transition probability in Eq. (45) as a function of $\theta$ for the fixed $\varepsilon_0 = \omega$ and as a function of $\varepsilon_0$ for various values of $\theta$. In the following, a single phonon process is intensively studied to elucidate a new aspect of the quadrupoles.

Figure 6 (a) shows a $\Gamma_3$-component dominant case ($g_T a_{2X} > g_T a_t$), choosing $\phi_0 = \phi_T = \phi_{2X} = 0$ and fixing the two-level splitting at $\varepsilon_0 = \omega$. The transition probability is plotted as a function of the field angle $\theta$, and it shows very narrow dips at $\theta \approx \pm \pi/4$. In Eq. (29), $|A_T| = g_T a_t \approx |C_T,1| \cos 2\theta$ owing to $|C_T,1| \gg |C_T,2|$, and $|A_T|$ almost vanishes at $\theta \approx \pm \pi/4$, namely, $|A_T| \ll |A_L| = g_L a_t$. Both $g_L a_t$ and $g_T a_t$ are also

\[ \begin{align*}
\Delta \to 0 & \text{ in our model.}^{15} \text{ In particular, for } |A_T|/\omega \leq 1, \text{ the transition probability shows narrow peaks near } \\
& \varepsilon_0 = 3\omega, 5\omega, \cdots \text{ and a relatively broad peak near } \varepsilon_0 = \omega. \text{ When } |\Delta| \text{ is finite, additional narrow peaks emerge near } \\
& \varepsilon_0 = 2\omega, 4\omega, \cdots \text{ and the peak widths become broader with increasing } |\Delta| \text{ as shown in Fig. 4. For a relatively large } |\Delta|, \text{ the peak at } \\
& \varepsilon_0 = \omega \text{ merges into the peak at } \varepsilon_0 = 0, \text{ and the transition probability shows an almost constant value close to } 1/2 \text{ in } 0 < \varepsilon_0 < \omega. \text{ This indicates that the contribution from the higher excited states may not be negligible in the quartet case discussed in Sect. 2.2.}
\end{align*} \]
plotted as a function of $\theta$ in Fig. 6 (b) and indicates that the $L$-mode coupling is dominant at $\theta \approx \pm \pi/4$. The origin of the dips is explained by the appearance of a sharp transition probability peak as shown in Fig. 7. Notice that the energy of the peak position shifts towards a lower value from $\varepsilon_0 = \omega$. When $\theta$ is varied from the above value for the sharp peak ($\theta/\pi = 0.225$ in Fig. 7), the peak broadening causes the abrupt increase in the $\theta$-dependent transition probability $P_{\varepsilon_0 = \omega}$ at $\varepsilon_0 = \omega$ in Fig. 6 (a). This is a key to the microscopic measurement of the quadrupole components. The $L$-mode-phonon-mediated transition is always accompanied by the photon absorption, and the corresponding resonance can be detected by the EPR measurement. For the same phase shifts $\phi_U = \phi_Y = \phi_{ZX}$, the narrow dips in the $\theta$-dependent $P_{\varepsilon_0 = \omega}$ become invisible as the photon coupling $|\Delta|$ decreases. It is because the resonance peak appears exactly at $\varepsilon_0 = \omega$. Instead, the similar narrow dips can be found when $\varepsilon_0$ is slightly shifted from $\omega$ (for instance, $\varepsilon_0/\omega = 1.01$).

In generic, the difference of phase shifts $\phi_U$, $\phi_Y$, and $\phi_{ZX}$ ($= 0$) has to be considered for the $\theta$ dependence of the transition probability, which can be controlled by the SAW experiment. Here, we show the data for $|\phi_U - \phi_Y| = \pi$ taking account of the phase difference between two strains $\varepsilon_U$ and $\varepsilon_Y$ reported by the previous SAW experiment of the silicon wafer.\(^{11}\) The photon coupling is fixed at $\Delta = 0.01$, which is small enough to treat $\Delta$ as a real number. In Fig. 8 (a), the two dips are also found in the $\theta$-dependent $P_{\varepsilon_0 = \omega}$. Since $(g_L a_{Z})/(g_T a_{Z}) > 1$, the field angle $\theta$ for the minimum transition probability slightly shifts towards a lower value from

\[ \theta \]
π/4. This is also explained by the existence of a sharp transition probability peak at a lower value of $\epsilon_0$ shifted from $\omega$ as shown in Fig. 9. The resonance near $\epsilon_0 = \omega$ is owing to the $L$-mode dominant contribution. In Fig. 8 (b), $|A_L| = |g_{r2}a_l|$ almost reaches a local maximum ($\approx 1.8\omega$) when $|A_T| = |g_{rT}a_T|$ equals a local minimum. Let us consider the field angle $\theta_{\text{tip}}$ for the minimum $P_{\text{tip}}$. This $\theta_{\text{tip}}$ almost equals the value for the minimum $|A_L|$, which can be evaluated by Eq. (29). Using $\xi \equiv (g_{rT}a_U)/(g_{rT}a_ZX)$, $|A_L|$ is rewritten as

\[
\frac{|A_L|^2}{6(g_{rT}a_{ZX})^2} = 1 + \xi^2\left(1 + \frac{\sqrt{3}}{2}\right) + A_{\phi_T}\cos(4\theta + \alpha),
\]

and

\[
A_{\phi_T} = \left[1 - \xi^2\left(1 + \frac{\sqrt{3}}{2}\right)^2\right]^{\frac{1}{2}}\xi^2(1 + \sqrt{3})^2\cos^2 \phi_U,
\]

\[
\tan \alpha = -\frac{\xi(1 + \sqrt{3})}{1 - \xi^2\left(1 + \frac{\sqrt{3}}{2}\right)^2}\cos \phi_U.
\]

When the $\Gamma_3$ component is dominant as $\xi \ll 1$ and $A_{\phi_T} > 0$, one of the conditions for the minimum $|A_L|$ is given by $\theta = (\pi - |a|)/4$ for $0 < |\phi_U| < \pi/2$. This equation also indicates that for $\pi/2 < |\phi_U| < \pi$, the transition probability shows a similar dip near the negative $\theta = -(\pi - |a|)/4$. On the other hand, for $\xi \gg 1$ and $A_{\phi_T} < 0$ which correspond to the $\Gamma_3$-dominant case, one can find that these values are replaced by $|a|/4$ and $-|a|/4$, respectively. When $\theta$ differs from $\theta_{\text{tip}}$ in Fig. 9, the transition probability peak broadens keeping the peak position $\epsilon_0$ at almost the same value. This broadening comes from the $T$-mode contribution. In the quartet case discussed in Sect. 2.2, the contribution from the higher excited states may not be negligible when the peak at $\epsilon_0/\omega = 1$ merges into the peak at $\epsilon_0 = 0$ for $\theta/\pi = 0.1$ or $\theta/\pi = 0.25$.

We conclude that it is essential to find the field angle $\theta_{\text{tip}}$ for the sharp resonance peak. Owing to the photon-assisted transition dominated by the $L$-mode phonon, the resonance peak can be probed by the EPR measurement. This $\theta_{\text{tip}}$ is related to the ratio $\xi$ of the QS couplings with the different symmetries $\Gamma_3$ and $\Gamma_5$. In addition, the ratio of the quadrupole coupling constants $g_{rT}$ and $g_{rT}$ can be evaluated from the measurement of the local strain amplitudes $a_i$ ($i = U, V, ZX$).

Owing to the penetration of the SAW into the solid, the strain amplitudes $a_i$ show the same exponential decay along the $Z$ axis\(^{11}\) and cause a significant decrease in $|A_L|$ and $|A_T|$. Since the ratio of $g_{rT}a_U$, $g_{rT}a_V$, and $g_{rT}a_{ZX}$ is unchanged, $|A_T|/|A_L|$ keeps the $\theta$ dependence obtained on the surface ($Z = 0$) during the decay of $a_i$. Accordingly, the local minimum of $P_{\text{tip}}$ appears at almost the same values $\theta$ as shown in Fig. 10, where $\eta$ is defined as the value of $(g_{rT}a_{ZX})/\omega$ keeping $g_{rT}a_U = g_{rT}a_V = 0.5g_{rT}a_{ZX}$. The origin of the left narrow dip at $\theta = -0.34\pi$ for $\eta = 0.5$ is also explained by the local maximum $|A_L| = g_{rL}a_L \approx 1.8\omega$. This is evaluated from the value of $(5/6)g_{rL}a_L$ in Fig. 8 (b) plotted for $\eta = 0.6$. For small $a_i$ ($\eta < 0.2$), $P_{\text{tip}}$ shows almost constant values $\approx 1/2$ in the entire region $-\pi/2 \leq \theta \leq \pi/2$. Thus, the decay of $a_i$ does not affect the appearance of the dips in the $\theta$-dependent transition probability. In the single phonon process at $\epsilon_0 = \omega$, $|A_L|/\omega \approx 1.8$ is the most appropriate condition for detecting the photon-assisted $L$-mode phonon resonance, which is explained by Eq. (67) as discussed in Sect. 3.1. This condition of $|A_L|$ can be accessed by tuning the frequency $\omega$ and the amplitudes $a_i$ of the SAW strain modes.

4. Conclusion and Discussion

In this paper, we have studied a minimum model of the two-level system for the hybrid EPR-MAR measurement and have demonstrated how the dynamical QS couplings can be evaluated by the magnetic field angle dependence of the time-averaged transition probability. Here, the photon frequency is negligibly small compared to the phonon frequency of gigahertz order. The longitudinal ($L$-mode, $A_L$) and transverse ($T$-mode, $A_T$) couplings of the two states are related to the three phonon modes of elastic strains ($\epsilon_{UU}$, $\epsilon_{VV}$, and $\epsilon_{ZX}$). These are driven by the SAW propagating in the principal axis ($X$ axis). The key point is that both couplings are changed by rotating the magnetic field since the combination of the three QS couplings depends on the rotation angle.

Using the Floquet theory, we have investigated the effects of $L$-mode and $T$-mode couplings on the transition probability

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**Fig. 9.** (Color online) Transition probability plotted as a function of $\epsilon_0/\omega$ for various $\theta$ around $0/\pi = 0.167$ for the minimum $P_{\text{tip}}$ in Fig. 8.

**Fig. 10.** (Color online) Transition probability $P_{\text{tip}}$ at $\epsilon_0/\omega = 1$ plotted as a function of the magnetic-field angle $\theta$. The phase shifts of the three SAW strain modes are chosen as $\phi_U/\pi = 0.25$, $\phi_V/\pi = -0.75$, and $\phi_{ZX} = 0$. Here, $g_{rT}a_U = g_{rT}a_V = 0.5g_{rT}a_{ZX}$, and the data are plotted for various values of $\eta \equiv (g_{rT}a_{ZX})/\omega \leq 0.6$. The data for $\eta = 0.6$ is the same as $P_{\text{tip}}$ in Fig. 8.
as a function of $\varepsilon_0/\omega$, where $\varepsilon_0$ and $\omega$ are the energy difference of the two states and the SAW phonon frequency, respectively. It is essential to focus on the single phonon process for $\varepsilon_0/\omega \lesssim 1$. The sharpness of the transition probability peak originates from the $L$-mode coupling effect assisted by the phonon with a small coupling $A$, while the peak broadening at $\varepsilon_0/\omega \approx 1$ is owing to the $T$-mode coupling. The appearance of the transition probability peaks also depends on the phase difference between the $L$-mode and $T$-mode couplings.

On the basis of this result, we have shown the effects of the QS couplings on the transition probability at $\varepsilon_0/\omega = 1$ as a function of the field angle $\theta$ for a $\Gamma_3$-quadrupole dominant case. The important finding is the appearance of the two narrow dips in the transition probability owing to the vanishing of $|A_T|$ and the approach of $|A_L|$ to a local maximum. The origin of the dips is explained by the lower energy shift of a sharp resonance peak from $\varepsilon_0/\omega = 1$ and the strong $\theta$ dependence of the peak broadening. Experimentally, the field angle for the sharp resonance can be probed by the EPR measurement since the photon-assisted transition becomes more significant for $|A_T| > |A_L|$. This is important for evaluating the ratio of QS couplings with different symmetries.

In the hybrid EPR-MAR measurement as described above, the $L$-mode phonon plays an important role in the appearance of a sharp resonance peak in the field-angle-dependent transition probability. Since the $L$-mode coupling is usually inactive in the transition process, we have introduced the idea of photon-assisted MAR and demonstrated how to extract a dominant $L$ mode for the MAR from the various phonon modes. The sharpness of the resonance peak becomes more prominent for a weak photon filed, which may allow high sensitivity in the conventional optical measurements.

For the silicon vacancy presented in Sect. 2.2, the ratio of the $\Gamma_3$ and $\Gamma_5$ QS coupling constants $g_{\Gamma_3}/g_{\Gamma_5} = 1.6$ was reported by the low-temperature ultrasonic measurement.\(^9\) This indicates the $\Gamma_3$-quadrupole dominant case as discussed in Sect. 3.2 when the amplitudes of elastic strains are almost the same in magnitude. It is expected that the field angle for a sharp resonance appears in $1/8 < \theta/\pi < 1/4$. Indeed, the $\theta$-dependent transition probability also depends on the phase differences between the strain modes driven by the SAW. Thus, the evaluation of the QS coupling constants requires precise measurement of $a_i$ and $b_i$ of the strains represented by $\varepsilon_i = a_i \cos(\omega t + \phi_i)$ ($i = U, V, ZX$).

The idea of the hybrid EPR-MAR measurement can also be applied to reveal hidden quadrupole properties in the $S = 1$ ground state of the NV center. For the NV spin state in the $C_{5v}$ crystal-field environment, electronic dipole levels in the same point-group character as electric quadrupoles represented by the second-rank spin tensors $Q_{xx} = S_x^2 - S_y^2$ and $Q_{xy} = S_xS_y + S_yS_x$ for the $x$ component of the dipole; $Q_{yy} = S_y^2 - S_x^2$ and $Q_{yx} = S_xS_y + S_yS_x$ for the $y$ component. Among them, there has been a lack of information on the relevance of $Q_{xx}$ and $Q_{xy}$ to electric-field control of the NV spin.\(^23\)\(^,\)\(^28\) A possibly significant role of these quadrupoles has been pointed out by the recent theoretical proposal of mechanically and electrically driven electron spin resonance as a new application of spin-strain interactions in the NV center.\(^2\)

More detailed investigations of the unknown quadrupole couplings are required to realize a promising platform of spin-controlled devices for quantum information processing and sensing applications.\(^26\)

It will be also intriguing to apply our idea to revisit various quadrupole properties in well-localized $f$-electron systems such as CeB$_6$.\(^7\)\(^,\)\(^27\) It is expected that the antiferroquadrupole ordering transition in CeB$_6$ affects the local $f$-electron level structure with the $\Gamma_3$ symmetry, which can modify a photon-phonon coupling process in the EPR measurement under a propagating SAW of gigahertz order. According to the resonant x-ray diffraction experiment, a linear combination of quadrupole order parameters is continuously controlled by controlling the direction of an applied magnetic field.\(^28\) This is a new aspect of quadrupole dynamics that is detectable in various orbitally degenerate electron systems.

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