# Coherent single-spin electron resonance spectroscopy manifested at an exceptional-point singularity in doped polyacetylene

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Spin-dependent charge-transfer decay in an alkali atom-doped polyacetylene is studied in terms of the complex spectral analysis, revealing the single-spin Zeeman splitting influenced by the spin-orbit interaction. A non-Hermitian effective Hamiltonian has been derived from the total system Hermitian Hamiltonian using the Brillouin-Wigner-Feshbach projection method where the microscopic spin-dependent dissipation effect is correctly incorporated in the energy-dependent self-energy. Since the present method maintains the dynamical and chiral symmetries of the total system, we discovered two types of exceptional point (EP) singularities: The EP surface and the EP ring are attributed to the dynamical and chiral symmetry breaking, respectively. We reveal that the coherent single-spin electron resonance (SSESR) spectrum reflects the complex eigenenergy spectrum of the system. We formulate the SSESR spectrum in terms of the non-linear-response function in the Liouville-space pathway approach where we have constructed the Liouville-space basis using the complex eigenstates of the total Hamiltonian. We calculate the one-dimensional Fourier transform (1DFT) and two-dimensional Fourier transform (2DFT) SSESR spectra reflecting the spin-relaxation dynamics at the donor site. Whereas the 1DFT SSESR spectrum reflects the complex eigenenergy spectrum, the 2DFT gives detailed information about the quantum coherence in the spin-relaxation dynamics as a cross correlation between the two frequencies. We reveal a giant response in the coherent SSESR around the EP ring singularity due to the vanishing normalization factors at the EP ring and the resonance effect. We show this giant response is even more heavily pronounced in the 2DFT spectrum than in the 1DFT spectrum, which illustrates that the 2DFT SSESR can become a useful tool to observe the single-spin response of a molecule.

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# I. INTRODUCTION

The conventional idea that dissipation is just a minor perturbation causing population decay or quantum decoherence has been overturned in recent years. It has been recognized that the dissipation process has a significant impact on the quantum evolution of a state, especially when it is close to an accidental degeneracy point, such as a *diabolic point*. In contrast to ordinary perturbation, which removes the degeneracy in energy, dissipation gives rise to a peculiar singularity, called the *exceptional-point singularity* (EP) where the eigenvalues and the eigenstates coalesce [1–6].

Many intriguing phenomena attributed to the EP singularities have been reported, such as asymmetric mode conversion [7], unidirectional invisibility [8], dynamical control [9,10], higher-order phase transition with Fano resonance [11], a large amplification of the Fano absorption spectral component [12], amplification of lasing [13], pulse shortening [14], asymmetric localization [15], enhanced nonlinear response to a perturbation [16–21], and amplified spontaneous emission

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[22,23]. Since the system energy changes abruptly with a perturbation parameter around the EP singularity, the nonlinear response to an external field can be dramatically enhanced.

Recently, there have been many efforts to explore various materials exhibiting characteristic behaviors of the EP singularities [4]. As the number of the internal degrees of freedom of a subsystem increases, such as spin variables, polarizations in electronic, and/or photonic systems, the number of parameters characterizing the effective Hamiltonian increases so that we can expect higher-dimensional manifolds of the EP singularities to occur in the multidimensional parameter space. Recent studies have explored higher-dimensional manifold structures of the singularities, called *exceptional rings and exceptional surfaces* and associated phenomena in topological materials and photonic systems [24–32].

Although many theoretical analyses start with a non-Hermitian Hamiltonian as a fundamental time-evolution generator, they have paid less attention to the dynamical origin of the dissipation processes. Since the dissipation process is changed according to the motion of internal degrees of freedom of the subsystem as mentioned above, it becomes essential to consider the microscopic dissipation mechanism associated with each state. Meanwhile, we have developed a theory of the *complex spectral analysis* [12,33–35] in which

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the origin of the dissipation has been identified as the energy resonance in a Hermitian Hamiltonian for an open system. In the complex spectral analysis, the microscopic dynamics are renormalized into the non-Hermitian effective Hamiltonian written in terms of an energy-dependent self-energy function. As a result, the eigenvalue problem becomes nonlinear in the sense that the effective Hamiltonian itself depends on its own eigenvalue whereby the dissipation dynamics are reconciled with the microscopically reversible dynamics apparent in the original Hamiltonian.

The purpose of this paper is to propose a specific quantum system that exhibits the EP singularity manifold in a multidimensional parameter space and a method to observe its influence in a laboratory experiment. For this purpose, we consider an alkali atom-doped polyacetylene as a model system and study the electronic Zeeman spectrum influenced by the spin-dependent charge transfer decay. The charge-transfer process from the alkali metal to the polyacetylene conduction band has been studied mostly in terms of the induced structural changes of the interchain couplings [36-39] and soliton formation [40-44]. Recent studies have indicated that the spin-orbit coupling plays an important role in determining the electronic structures even in light element materials, such as graphene and polyacetylene [45,46]. Furthermore, the crucial role of the spin-orbit coupling for the emergence of the EP manifolds has been recently clarified in Weyl metals and in photonic systems [25,26,29,30,32]. We show that the EP manifolds appear in the electronic Zeeman spectrum due to the spin-dependent charge-transfer decay through the spin-orbit interactions where we observe an anisotropic EP ring and an isotropic EP surface.

As our second purpose, we demonstrate that coherent single-spin-resonance electron spectroscopy (SSESR) can be used to observe the EP singularities. The electron spin resonance (ESR) has been widely used to investigate the spin-relaxation process in polyacetylene and molecular chains [46-49]. However, conventional measurements of the ESR have required about 10<sup>10</sup> identical spins to obtain a good signal-to-noise ratio. In this context, significant progress has been made in the single-spin quantum control technique in the past two decades [50-53]. The spatial resolution and sensitivity of electron spin-resonance spectroscopy have now reached atomic-scale precision owing to recent advances in spin-polarized scanning tunneling microscopy (STM) and magnetic sensitivity in interaction with nitrogen vacancy centers in diamond [54-57] so that the ESR signal attributed to a microscopic target within a sample center can be distinguished. In this direction, we desire to find a way to further enhance the signal intensity from a single spin.

Here we study the coherent nonlinear single-electron spin resonance accompanied by the charge-transfer decay from the alkali donor to the one-dimensional conduction band of polyacetylene where we apply the pump-probe pulsed excitation to obtain the nonlinear response in addition to the static magnetic field. It has been recognized that the nonlinear magnetic resonance spectroscopy and its multidimensional Fourier transform spectroscopy, such as two-dimensional Fourier transform (2DFT) NMR or ESR, is a powerful tool to investigate the spin-relaxation process in a molecule [58–60]. We show that the EP singularity as a result of the cooperation between the Zeeman interaction and the spin-orbit coupling is well reflected in the coherent nonlinear 2DFT ESR spectrum.

It is known that the Liouville-space pathway approach in terms of the non-linear-response function is a useful tool to describe the nonlinear optical spectroscopy [59]. In the present paper, we construct a complete basis in the Liouville space to describe the proper Liouville pathway in terms of the solutions of the complex eigenvalue problem of the total Hamiltonian in which the energy dependence of the self-energy is essential [33,34,61]. We find that the time evolution following successive pulsed excitations becomes consistent with the entropy production only when we take into account the correct analytic continuation for the Liouville states.

One of our key findings is a giant response in the 2DFT ESR as a result of the vanishing normalization factor at the EP singularity. This is known as the Petermann effect, which acts in addition to the ordinary resonant enhancement from the Purcell effect in the signal enhancement of the spontaneous emission [20,22,62,63]. However, we have found that the signal enhancement becomes orders of magnitude larger in the coherent 2DFT ESR than in the one-dimensional Fourier transform (1DFT) ESR. To our knowledge, this is the first study to reveal such a giant response due to the EP singularity in a non-linear-response function.

In Sec. II, we present our model for the doped polyacetylene molecule in terms of the one-dimensional tight-binding model where the spin-orbit coupling associated with the charge transfer of the donor electron is taken into account as well as the local Zeeman effect. In Sec. III, we have solved the complex eigenvalue problem of the total system Hamiltonian in terms of the Brillouin-Wigner-Feshbach (BWF) projection method and obtained the complex eigenvalue spectrum in the three-dimensional parameter space of the external magnetic field. We find that the EP singularities appear depending on the relative angle of the external magnetic field to the intrinsic molecular field determined by the spin-orbit coupling. The coherent nonlinear 2DFT ESR spectrum as well as 1DFT ESR are formulated in terms of the Liouville-space representation in Sec. IV where the signal intensity exhibits strong anisotropy as the relative angle between the pump and the probe directions is changed. In addition, we find the giant response of the signal around the EP singularity, indicating that this method is a useful tool to observe the single electron spin-relaxation process. We conclude in Sec. V with some discussions.

## **II. MODEL**

We consider the spin-dependent charge-transfer decay of a heavy alkali atom binding to the end of a polyacetylene molecule as shown in Fig. 1 where the charge transfer occurs from a donor impurity level to the  $\pi^*$  conduction band of the molecule. In the present paper, we take the donor site as the origin of the Cartesian coordinate axis, i.e.,  $\mathbf{r}_{\rm D} = 0$ . In this paper, we consider the low-energy Zeeman excitation so that we neglect the effects of the  $\pi$  valence band and  $\sigma$  orbitals. We describe the conduction band in terms of the one-dimensional tight-biding model with spin variables under a simple Hückel approximation.

Taking the energy origin at the center of the conduction band, the charge-transfer Hamiltonian for a nonrelativistic



FIG. 1. Single-spin ESR setup for a heavy alkali donor atom binding to a one-dimensional polyacetylene. The orbital angular momentum (L) associated with the charge transfer is aligned in the z direction, and the external magnetic-field  $\mathbf{B}(\mathbf{r})$  acts on the spin at the donor site.

donor electron is represented by

$$\hat{H}_{\rm CT} = \varepsilon_D \sum_{\alpha} \hat{c}^{\dagger}_{0,\alpha} \hat{c}_{0,\alpha} + \sum_{\alpha} v(\hat{c}^{\dagger}_{0,\alpha} \hat{c}_{1,\alpha} + \hat{c}^{\dagger}_{1,\alpha} \hat{c}_{0,\alpha}) - J \sum_{n=1}^{N} \sum_{\alpha} (\hat{c}^{\dagger}_{n+1,\alpha} \hat{c}_{n,\alpha} + \hat{c}^{\dagger}_{n,\alpha} \hat{c}_{n+1,\alpha}),$$
(1)

where  $\hat{c}_{i,\alpha}^{\dagger}(\hat{c}_{i,\alpha})$  are the electron fermion operators with the site index *i* and the two-spinor indices  $\alpha$ . In  $\hat{H}_{CT}$ , the donor impurity atom is located at the 0th site with the energy  $\varepsilon_D$ , the nearest-neighbor transfer integral for the 1D tight-binding model is *J*, the spin-independent charge transfer from the donor to the band is represented by *v*, and *N* is the length of the polyacetylene molecule. With use of the wave-number representation,

$$\hat{c}_{k,\alpha} \equiv \sqrt{\frac{2}{N}} \sum_{n=1}^{N} \sin(kn) \hat{c}_{n,\alpha}, \quad \hat{c}_{k,\alpha}^{\dagger} \equiv \sqrt{\frac{2}{N}} \sum_{n=1}^{N} \sin(kn) \hat{c}_{n,\alpha}^{\dagger},$$
(2)

and taking the limit of infinite molecular length [12], the tightbinding molecular Hamiltonian [the last term of Eq. (1)] is diagonalized in the form

$$\hat{H}_{\rm mol} = \sum_{\alpha} \int_0^{\pi} dk \,\omega_k \hat{c}^{\dagger}_{k,\alpha} \hat{c}_{k,\alpha},\tag{3}$$

where  $\omega_k = -2J \cos kd$ . Hereafter, we will use the distance between the carbon atoms  $d \simeq 1.3$  A as a length unit. Then the charge-transfer Hamiltonian  $\hat{H}_{CT}$  is rewritten as

$$\hat{H}_{\rm CT} = \varepsilon_D \sum_{\alpha} \hat{c}^{\dagger}_{0,\alpha} \hat{c}_{0,\alpha} + \sum_{\alpha} \int_0^{\pi} dk \, v_k (\hat{c}^{\dagger}_{k,\alpha} \hat{c}_{0,\alpha} + \hat{c}^{\dagger}_{0,\alpha} \hat{c}_{k,\alpha}), \tag{4}$$

where  $v_k \equiv \sqrt{2/\pi} v \sin kd$ . In the present paper, we take  $\hbar = 1$ .

We consider a static local external magnetic field acting on the donor site applied by spin-polarized STM tip, for example. For the spin-density s(r), the Zeeman interaction Hamiltonian in the second quantized form is given by

$$\hat{H}_{Z}(\mathbf{B}) = -g_{B} \int \mathbf{s}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}) d^{3}\mathbf{r}$$
$$= -\frac{g_{B}}{2} \sum_{i=x,y,z} \sum_{\alpha,\beta} B_{i}(\sigma_{i})_{\alpha,\beta} \hat{c}^{\dagger}_{0,\alpha} \hat{c}_{0,\beta}, \qquad (5)$$

where  $g_B$  is the *g* factor,  $B_i$  is the magnetic field acting on the donor site, and  $\sigma_i$  (i = x, y, z) are the Pauli matrices. Hereafter, we rewrite  $B \equiv g_B |\mathbf{B}|/2$ , including  $g_B$  into the value of *B* so that *B* has dimensions of energy.

In addition, we consider the Rashba-type spin-orbit interaction [45,46,64] associated with the charge transfer from the impurity donor to the conduction band. The Hamiltonian is given by

$$\hat{H}_{\rm SO} = \xi_{\rm SO} \int \boldsymbol{s}(\boldsymbol{r}) \cdot [\mathbf{E}(\boldsymbol{r}) \times \boldsymbol{p}] d^3 \boldsymbol{r} , \qquad (6)$$

where we attribute the electric-field  $\mathbf{E}(\mathbf{r})$  to the charge-transfer polarization from the donor atom to the molecular chain, which is represented by  $\mathbf{E}(\mathbf{r}) = e\mathbf{r}/\chi$  with the electric susceptibility  $\chi$ . Then the spin-orbit interaction Hamiltonian reads

$$\hat{H}_{\rm SO} = \frac{e\xi_{\rm SO}}{2\chi} \int \hat{\boldsymbol{\sigma}} \cdot (\hat{\boldsymbol{r}} \times \hat{\boldsymbol{p}}) d^3 \boldsymbol{r} \equiv g_{\rm S} \int \hat{\boldsymbol{\sigma}} \cdot \hat{\boldsymbol{l}} d^3 \boldsymbol{r}$$
$$= g_{\rm S} \sum_{i=x,y,z} l_i(\sigma_i)_{\alpha,\beta} \hat{c}^{\dagger}_{0,\alpha} \hat{c}_{1,\beta} + \text{H.c.}, \tag{7}$$

where  $\hat{l}$  is the orbital angular momentum operator. We have evaluated the matrix element of the orbital angular momentum between the *ns* atomic orbital of the heavy alkali donor atom and the  $2p_y$  orbital of the carbon atom at the end of the molecule as

$$l_i = \langle 0, s | \hat{l}_i | 1, p_y \rangle = \int d^3 \mathbf{r} \, \varphi_s^*(\mathbf{r}) \hat{l}_i \varphi_{p_y}(\mathbf{r} - \mathbf{a}).$$
(8)

We neglect the small spin-orbit couplings between the carbon  $2p_y$  orbitals. We find from the symmetry that

$$l_z \neq 0, \quad l_x = l_y = 0, \tag{9}$$

as shown in Appendix A [45,65]. In the present paper, we take the z axis as a quantization axis with the spin-orbit coupling parameter  $L \equiv g_S l_z$  in (7) as shown in Fig. 1. It is seen from (7) that the orbital angular momentum associated with the charge transfer plays the role of an intrinsic molecular magnetic field acting on the two spinors, which we will call the *molecular field*,  $\mathbf{L} = L\hat{\mathbf{z}}$ , where L has dimensions of energy.

The half-width of the conduction band of polyacetylene has been estimated as  $2J \simeq 6$  eV [44]. In the present paper, we take J as the energy unit: 2J = 2. The charge-transfer strength v, the spin-orbit coupling strength  $g_s$ , the impurity donor energy-level  $\varepsilon_D$  are given for a specific donor atom, whereas the externally controllable parameter is the external magnetic-field **B**. Usually these parameter values satisfy  $2J \gg v \gtrsim g_s$ . In this paper, we take  $\varepsilon_D = 0$ , and we make a brief comment on the effect of  $\varepsilon_D$  in Appendix B. Since we focus on rapid motion of the donor electron, we neglect the effect of the slow motion of the structural change in the polyacetylene molecular backbone. In the next section, we study the complex spectra of the *total* Hamiltonian in terms of the external magnetic-field **B**-parameter space and reveal the EP singularities in this space.

## III. COMPLEX EIGENVALUE PROBLEM OF THE HAMILTONIAN AND THE EP SINGULARITIES

In this section we solve the complex eigenvalue problem of the total Hamiltonian. We first derive the effective Hamiltonian in terms of the BWF projection method and obtain the complex eigenspectrum of the Hamiltonian.

The complex eigenvalue problem of the total Hamiltonian for parameter **B** reads [12,33,66]

$$\hat{H}_{\text{tot}}(\mathbf{B})|\phi_{\xi}(\mathbf{B})\rangle = z_{\xi}(\mathbf{B})|\phi_{\xi}(\mathbf{B})\rangle, \qquad (10a)$$

$$\langle \tilde{\phi}_{\xi}(\mathbf{B}) | \hat{H}_{\text{tot}}(\mathbf{B}) = z_{\xi}(\mathbf{B}) \langle \tilde{\phi}_{\xi}(\mathbf{B}) |, \qquad (10b)$$

where the right- and the left-eigenstates  $|\phi_{\xi}(\mathbf{B})\rangle$  and  $\langle \tilde{\phi}_{\xi}(\mathbf{B})|$ , respectively, share the same eigenvalue  $z_{\xi}(\mathbf{B})$ . For the current problem, we restrict ourselves to a single-particle vector space consisting of  $\{|n, \alpha\rangle \ (n = 0, 1, ...; \alpha = \pm)\}$ , where  $|n, \alpha\rangle \equiv c_{n,\alpha}^{\dagger}|\text{vac}\rangle$  and  $|\text{vac}\rangle$  denotes the electron vacuum.

We will reduce the eigenvalue problem of the total Hamiltonian into the  $\{|0, +\rangle, |0, -\rangle\}$  space in terms of the projection operators,

$$\hat{P}_0 = |0, +\rangle \langle 0, +| + |0, -\rangle \langle 0, -|, \quad \hat{Q}_0 = 1 - \hat{P}_0.$$
(11)

Taking into account (9), we have derived the effective Hamiltonian matrix in the  $\hat{P}_0$  subspace as

$$H_{\text{eff}}(\mathbf{B};z) = -\begin{pmatrix} B_z & B_x - iB_y \\ B_x + iB_y & -B_z \end{pmatrix} + \Sigma(z) \begin{pmatrix} (v+L)^2 & 0 \\ 0 & (v-L)^2 \end{pmatrix}.$$
 (12)

The derivation of the effective Hamiltonian is shown in Appendix C. The microscopic dissipation effect into the continuum is renormalized into  $H_{\text{eff}}$  in terms of the self-energy function  $\Sigma(z)$  given by a Cauchy integral evaluated as

$$\Sigma^{\pm}(z) = \frac{2}{\pi} \int_0^{\pi} dk \frac{\sin^2 k}{(z - \omega_k)^{\pm}} = \frac{1}{2} (z \mp \sqrt{z^2 - 4}).$$
(13)

The self-energy has a branch cut in the region [-2:2] on the real *z* axis where the sign in the integral indicates the analytic continuation from the upper(+)- and lower(-)-half complex *z* plane corresponding to the retarded and advanced Green's functions, respectively [33]. By the definition (13), the self-energy is an odd function in terms of *z* as

$$\Sigma^{\pm}(-z) = -\Sigma^{\mp}(z), \tag{14}$$

reflecting the microscopic interaction with the continuum [68]. As will be shown later, this relation is crucial to ensure the symmetry (*chiral symmetry*) that gives rise to the EP ring structure.

We emphasize that the complex eigenvalues of the effective Hamiltonian are the same as those of the total Hermitian Hamiltonian only when we take into account the energy dependence of the self-energy [33,61,66]. As a result, the complex eigenvalues appear as complex-conjugate pairs, if they exist, corresponding to the resonance and antiresonance eigenstates, which we will call *the dynamical symmetry*. The solution pairs, thus, obtained are the fundamental building blocks to construct the Liouville space basis used in the formulation of the coherent ESR spectroscopy as shown in the next section.

It is seen that the Zeeman effect on the two-spinor at the donor site, the first term in (12), is modified by the dissipation effect of the second term. We emphasize that the spin-orbit coupling L effectively enhances or suppresses the



FIG. 2. Eigenvalues of the effective Hamiltonian as a function of **B** parallel to **L** when  $\theta_B = 0$  for v = 0.3 and L = 0.1. (a) Real part and (b) imaginary part. The solid and dashed lines in (b) correspond to the resonance and antiresonance state solutions in the resonant regime, respectively. In the off-resonant regime, the solid and dashed lines correspond to the bound and virtual bound states shown in (a). The gray lines in (a) are guidelines for the unperturbed energies of the two-spinors. The energy unit is taken as J = 1.

charge-transfer effect v when the spin direction is parallel or antiparallel to the molecular field **L**, respectively: The chargetransfer decay process becomes spin dependent due to the spin-orbit coupling.

The complex eigenvalues of the effective Hamiltonian are obtained by solving the characteristic equation,

$$\eta^{\pm}(z; \mathbf{B}) \equiv \{z - B_z - (v - L)^2 \Sigma^{\pm}(z)\} \\ \times \{z + B_z - (v + L)^2 \Sigma^{\pm}(z)\} - (B_x^2 + B_y^2) = 0.$$
(15)

Squaring out the root in the self-energy, we find the characteristic equation is a fourth-order polynomial equation, yielding two complex conjugate pair solutions in the resonant regime as mentioned above, and four real solutions in the off-resonant regime. With use of the spherical coordinate  $\mathbf{B} =$  $(B \sin \theta_B \cos \phi_B, B \sin \theta_B \cos \phi_B, B \cos \theta_B)$ , one can immediately see from (15) that the solutions depend only on the polar angle  $\theta_B$  as well as the field strength *B*.

First we show the results of the complex eigenvalues when the external magnetic field is parallel to the molecular field, i.e.,  $\mathbf{B} \parallel \mathbf{L} = L\hat{z}$ , i.e.,  $B_x = B_y = 0$ . It is immediately seen from (12) that the two-spinor states are decoupled in this case, and the characteristic equation (15) becomes a product of the two independent equations associated with  $|+\rangle$  and  $|-\rangle$  spin states. We show in Fig. 2 the complex eigenvalues of the effective Hamiltonian as a function of the magnetic-field strength *B* for  $\theta_B = 0$  (**B** || **L**) where the real and imaginary parts of the eigenvalues are shown in Figs. 2(a) and 2(b), respectively. The other parameters are taken as v = 0.3 and L = 0.1. The solutions for the different analytic continuations of the self-energies of  $\Sigma^+(z)$  and  $\Sigma^-(z)$  are depicted by the solid and dashed lines, respectively. The solutions for  $\Sigma^+(z)$ and  $\Sigma^{-}(z)$  correspond to the resonance and antiresonance states in the resonant regime, respectively.

As seen in Fig. 2(a), the real parts of the eigenvalues are split by the Zeeman effect: The real part of the energy corresponding to the parallel (antiparallel) spin states to **B** linearly decreases (increases) with *B*. Although this behavior is similar to the ordinary Zeeman effect on an isolated two-spinor, it should be noted that the eigenstates in the present case are



FIG. 3. Eigenvalues of the effective Hamiltonian as a function of **B** parallel to **L** when  $\theta_B = \pi/3$  for v = 0.3 and L = 0.1. (a) Real part and (b) imaginary part. The solid and dashed lines have the same meaning as in Fig. 2. The energy unit is taken as J = 1.

resonance (antiresonance) states which do not belong to the Hilbert space [33]. Indeed, the eigenstates have an imaginary part of the eigenvalues as long as the Zeeman split energies are in resonance with the conduction band as shown in Fig. 2(b). As mentioned above, the charge-transfer decay couplings become spin dependent due to the spin-orbit coupling so that in this case the parallel spin state  $|+\rangle$  has a larger decay rate than the antiparallel spin state  $|-\rangle$ .

As *B* increases and the Zeeman split energies come close to the conduction-band edge  $B \simeq 2$ , the resonance and antiresonance states coalesce at the EP singularity depicted by the arrow in Fig. 2(b). As *B* further increases, they are split into the bound and virtual-bound states [9,67]. The EP singularities near the band-edge  $B \simeq 2$  appear as a result of the energy resonance singularity between the discrete Zeeman states and the conduction continuum irrespective to the direction of **B**.

When we tilt  $\theta_B$  away from the polar axis of **L**, the two-spin states become coupled. We show the results of  $\theta_B = \pi/3$  in Fig. 3. The effect of  $\theta_B$  is prominent in the imaginary part of the eigenvalues; As  $\theta_B$  increases from 0, the gap of the imaginary parts of the complex eigenvalues of the two resonance states or the two antiresonance states is narrowed, whereas the values at **B** = 0 are unchanged. We see the EP singularities at the band edge still remain as indicated by the arrow. Since this EP singularity appears as a result of the energy resonance between the discrete Zeeman states and the conduction continuum irrespective of the polar angle  $\theta_B$ , the EP singularity forms an EP surface in the **B** space as shown in Fig. 4(a).

As mentioned above and shown in Appendix B, the total system Hamiltonian possesses the *chiral symmetry* expressed by (B3), which holds when  $B_z = 0$ . Since the effective Hamiltonian is derived from the total Hamiltonian and the microscopic spin-dependent charge-transfer decay is taken into account by the energy-dependent self-energy, the effective Hamiltonian also possesses the chiral symmetry: Owing to (14), the effective Hamiltonian at  $B_z = 0$  satisfies

$$\sigma_z \hat{H}_{\text{eff}}(z) \sigma_z = -\hat{H}_{\text{eff}}(-z), \qquad (16)$$

with the Pauli matrix  $\sigma_z$ . Note the correspondence with (B3), which indicates the derived effective Hamiltonian maintains the chiral symmetry of the total Hamiltonian.

Corresponding with (B5), it further follows from (16) that

$$\hat{H}_{\rm eff}(z)|\psi\rangle = z|\psi\rangle \Rightarrow \hat{H}_{\rm eff}(-z)\sigma_z|\psi\rangle = -z\sigma_z|\psi\rangle.$$
(17)



FIG. 4. The EP manifolds for v = 0.3 and L = 0.1. (a) EP surface in the **B**-space. (b) Real part of the complex eigenvalues on the  $B_x$ - $B_y$  plane where the EP ring appears at  $|B| = B_{\text{EP},-}$ . Note **B** has a dimension of energy with an energy unit of J = 1.

This relation ensures the appearance of pairs of solutions with opposite signs in addition to the complex-conjugate pair solutions due to the dynamical symmetry as mentioned just below (14). As a result of the combination of the dynamical symmetry and the chiral symmetry, the complex eigenvalues are obtained as a set of four solutions in the form

resonance 
$$\begin{cases} z_1 = \alpha - i\beta, \\ \bar{z}_1 = -\alpha - i\beta, \end{cases}$$
 antiresonance 
$$\begin{cases} z_1^* = \alpha + i\beta, \\ \bar{z}_1^* = -\alpha + i\beta, \end{cases}$$
(18)

with  $\alpha$ ,  $\beta > 0$ . Here we denote the upper bar and asterisk as taking the opposite sign of the real and imaginary parts, respectively, i.e.,  $-z = \overline{z}^*$ . The advantage of our theory is that we can deal with the two different types of symmetry breaking, the dynamical symmetry, and the chiral symmetry breaking in a unified manner. The effective Hamiltonian maintains the symmetry of the original Hamiltonian by correctly taking into account the energy dependence of the self-energy.

The characteristic equation is then given by

$$[(v - L)^{2} - 1]\{(v + L)^{2} - 1\}z^{4} + [4(v^{2} + L^{2})^{2} - 2(v^{2} - L^{2})^{2}(v^{2} + L^{2} + 1) - B^{2}\{(v^{2} - L^{2})^{2} - 2(v^{2} + L^{2} - 1)\}]z^{2} + \{B^{2} + (v^{2} - L^{2})^{2}\}^{2} = 0,$$
(19)

where the quartic polynomial equation in z reduces to a quadratic polynomial equation of  $z^2$ . This form is the same as studied in the context of parametric amplification in the dynamical Casimir system [68,69] where it was shown that the symplectic symmetry of the system is crucial to cause the EP singularity.

The EP singularities are determined by solving the discriminant equation of (19),

$$D(B; v, L) = B^{4} + 4(v^{2} + L^{2} - 1)B^{2} + 16v^{2}L^{2} = 0, \quad (20)$$

which gives the EP singularities at  $B = B_{\text{EP},\pm}$ , where

$$(B_{\rm EP,\pm})^2 = 2\{(1-v^2-L^2) \\ \pm \sqrt{(1-v^2-L^2)^2 - 4v^2L^2}\} > 0.$$
(21)

Taking into account that  $2J = 2 \gg v \gtrsim L$ , we approximately obtain  $B_{\text{EP},-} \simeq 2vL$  and  $B_{\text{EP},+} \simeq 2$ . We note that the



FIG. 5. Eigenvalues of the effective Hamiltonian as a function of **B** perpendicular to **L** when  $\theta_B = \pi/2$  ( $B_z = 0$ ) for v = 0.3 and L = 0.1. (a) Real part and (b) imaginary part. The gray lines in (a) are guidelines for the unperturbed energies. The spectra of the real and imaginary parts are shown in the expanded horizontal scale in (c) and (d), respectively. The arrows indicate  $B_{\text{EP},+}$  in (b) and  $B_{\text{EP},-}$  in (c) and (d). The energy unit is taken as J = 1.

appearances of the two EP singularities have different origins:  $B_{\rm EP,+}$  is due to the resonance singularity at which the discrete Zeeman levels are in resonance with the conduction band so that its emergence does not depend on the orientation of the external magnetic field. On the other hand, the emergence of  $B_{\rm EP,-}$  relies on the orthogonal orientation of the external magnetic field to the direction of the orbital angular momentum satisfying the chiral symmetry under the condition of the resonance and antiresonance splittings shown in Eq. (18). In this sense, we will call  $B_{\rm EP,+}$  and  $B_{\rm EP,-}$  the dynamical symmetry-breaking and the chiral symmetry-breaking points. Since the chiral symmetry-breaking point  $B_{\rm EP,-}$  appears only on the  $B_z = 0$  plane, it forms an EP ring structure, whereas  $B_{\rm EP,+}$  corresponds to the EP surface.

We show in Fig. 5 the complex eigenvalues of the effective Hamiltonian at  $B_z = 0$  where the real and imaginary parts are depicted in (a) and (b), respectively. The arrow in (b) indicates the dynamical symmetry-breaking point at  $B_{\rm EP,+}$ . We show the real and imaginary parts in Figs. 5(c) and 5(d), respectively, in the expanded scale around  $B_{\rm EP,-}$  where the arrows indicate the chiral symmetry-breaking points. Since this EP singularity appears only at  $B_z = 0$ , we see the EP ring structure on the  $B_x$ - $B_y$  plane as shown in Fig. 4(b).

Here we compare our results with a phenomenological parity-time (PT) Hamiltonian model which has been used to study the exceptional rings and surfaces [25,26,29,30,32]. In the phenomenological PT model, the dissipation term is represented by an imaginary constant, such as  $i\gamma$ . Indeed, when we approximate  $\Sigma(z)$  by -i in (13), then the effective Hamiltonian is approximated by

$$\hat{H}_{\text{eff}}(\mathbf{B}) \simeq -i(v^2 + L^2) - \begin{pmatrix} B_z - 2ivL & B_x - iB_y \\ B_x + iB_y & -B_z + 2ivL \end{pmatrix}.$$
(22)

For  $B_z = 0$ ,

$$\hat{H}_{\rm eff}(\mathbf{B}) = -i(v^2 + L^2) + \hat{H}_{\rm PT}(\mathbf{B}),$$
 (23)

where  $\hat{H}_{PT}(\mathbf{B})$  is a phenomenological PT-symmetric Hamiltonian,

$$\hat{H}_{\rm PT}(\mathbf{B}) = \begin{pmatrix} -2ivL & B_x - iB_y \\ B_x + iB_y & 2ivL \end{pmatrix}.$$
 (24)

The eigenvalues are given by

$$z_{\text{PT},\pm} = -i(v^2 + L^2) \pm \sqrt{\left(B_x^2 + B_y^2\right) - 4v^2L^2}.$$
 (25)

Therefore, this phenomenological treatment can also predict the emergence of the EP ring at  $B_{\text{PT,EP,-}} = 2vL$  for the  $B_z = 0$ case. However, the phenomenological model gives only the resonance eigenstates but not the antiresonance eigenstates that are also necessary to construct the Liouville-space basis in the calculation of the SSESR. On the other hand, in the present paper, we have obtained a set of four solutions (18) by taking into account the energy-dependent self-energy.

## IV. NONLINEAR COHERENT SSESR SPECTRUM IN THE LIOUVILLE SPACE

The EP singularities revealed in the preceding section significantly influence the ultrafast single-spin resonance spectroscopy. We consider the pump-probe single-spin free induction decay for the present system. In the pump-probe process, an electron with spin parallel to the external static magnetic-field **B** is transferred from the STM tip to the donor site by the pump electric field. The interaction with the pump field is described by

$$\hat{V}(t) = \mathcal{E}^*(t)|S, B_+\rangle\langle 0, B_+| + \mathcal{E}(t)|0, B_+\rangle\langle S, B_+|, \qquad (26)$$

where  $|S, B_+\rangle$  denotes an electronic state with the spin parallel to the external magnetic field at the STM tip, and  $\mathcal{E}(t)$  [ $\mathcal{E}^*(t)$ ] denotes the electric pump-pulse field.

After some delay time  $t_1$ , we introduce the probe microwave magnetic field to induce the transition between the Zeeman split states. The interaction of the electronic spin and the probe magnetic-field  $\mathbf{B}_1(\mathbf{r}, t)$  which is localized on the donor site is given by

$$\hat{W}(t) = -g_B \int \hat{s}(\mathbf{r}) \cdot \mathbf{B}_1(\mathbf{r}, t) d^3 \mathbf{r}.$$
 (27)

The probe magnetic impulsive pulse for the free induction decay is represented by

$$\mathbf{B}_{1}(\mathbf{r}_{D}, t) = \mathcal{B}_{1}(\mathbf{r}_{D}, t - t_{1}) + \mathcal{B}_{1}^{*}(\mathbf{r}_{D}, t - t_{1}), \qquad (28)$$

where  $\mathcal{B}_1(\mathbf{r}, t)$  and  $\mathcal{B}_1^*(\mathbf{r}, t)$  are the fields which are assumed to be localized at the donor site.

We measure the induced spin polarization at the donor site in terms of the Liouville-space representation [59,70–75]. The third-order induced spin polarization is given by

$$\begin{aligned} \boldsymbol{s}(\boldsymbol{r}_{D},t) &= (-i)^{3} \int_{t_{0}}^{t} d\tau_{3} \int_{t_{0}}^{\tau_{3}} d\tau_{2} \int_{t_{0}}^{\tau_{2}} d\tau_{1} \langle \langle \hat{\boldsymbol{s}}(\boldsymbol{r}_{D}) | e^{-i\mathcal{L}_{\text{tot}}(t-\tau_{3})} \\ &\times \mathcal{W}(\tau_{3}) e^{-i\mathcal{L}_{\text{tot}}(\tau_{3}-\tau_{2})} \mathcal{V}(\tau_{2}) e^{-i\mathcal{L}_{\text{tot}}(\tau_{2}-\tau_{1})} \\ &\times \mathcal{V}(\tau_{1}) e^{-i\mathcal{L}_{\text{tot}}(\tau_{1}-t_{0})} | \rho(0) \rangle \rangle, \end{aligned}$$

$$(29)$$



FIG. 6. Double-sided Feynman diagrams for the coherent ESR spectroscopy. The wavy line indicates the interaction with the pump excitation pulse, and the arrow indicates the interaction with the probe magnetic pulse field.

where  $\mathcal{L}_{tot}$  is the Liouvillian superoperator in the Liouville space defined by  $\mathcal{L}_{tot} := [\hat{H}_{tot}, \cdot]$ , and  $|\rho(0)\rangle\rangle$  is the initial-

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state density operator at an initial time  $t_0$ ,

$$|\rho(0)\rangle\rangle = |S, B_+; S, B_+\rangle\rangle. \tag{30}$$

In (29), the curly operators denote the superoperators corresponding to the Hilbert space operators. It is essential to describe the coherent spectroscopy in terms of the Liouvillespace representation in order to register the correct decay processes [34,61]. We have shown in Fig. 6 the double-sided Feynman diagram corresponding to the Liouville-space pathway of (32).

In the present paper, we consider the impulsive pulses of the electric pump and magnetic probe fields as

$$\mathcal{E}(t), \mathcal{E}^*(t), \mathcal{B}_1(\mathbf{r}_D, t), \mathcal{B}_1^*(\mathbf{r}_D, t) \propto \delta(t).$$
 (31)

We represent (29) in terms of the Liouville-space basis defined by (D3) where we have utilized the complex eigenstates of the total Hamiltonian defined by (10). The explicit representation of the complex eigenstates of the total Hamiltonian are given in Appendix C. Applying (D6) to (29), we have obtained the induced spin-polarization component as

$$s_{i}(\mathbf{r}_{D},t) \simeq (-i)^{3} \sum_{\boldsymbol{\xi},\boldsymbol{\xi}',\boldsymbol{\xi}''} e^{-i\Delta_{\boldsymbol{\xi}'',\boldsymbol{\xi}'}(t-t_{1})} e^{-i\Delta_{\boldsymbol{\xi},\boldsymbol{\xi}'}t_{1}} \langle \langle \hat{s}_{i}(\mathbf{r}_{D}) | \phi_{\boldsymbol{\xi}''}; \phi_{\boldsymbol{\xi}'} \rangle \rangle \langle \langle \tilde{\phi}_{\boldsymbol{\xi}'}; \tilde{\phi}_{\boldsymbol{\xi}'} | \mathcal{W}(t_{1}) | \phi_{\boldsymbol{\xi}}; \phi_{\boldsymbol{\xi}'} \rangle \rangle \langle \langle \tilde{\phi}_{\boldsymbol{\xi}}; \tilde{\phi}_{\boldsymbol{\xi}'} | 0, B_{+}; 0, B_{+} \rangle \rangle,$$
(32)

where  $\xi, \xi', \xi''$  denote the discrete resonance eigenstates  $\Delta_{\xi,\xi'} = z_{\xi} - z_{\xi'}^*$  and we neglect the contributions from the continuous states, assuming weak couplings of v and L.

With use of (D3), we obtain the non-linear-response function for  $s_i(\mathbf{r}_D, t)$  to the probe field component  $\mathbf{B}_{1,i}$  as

$$F_{i,j}(t_2, t_1) = 4g_B \sum_{\xi, \xi', \xi''} \operatorname{Im}[e^{-i\Delta_{\xi'', \xi'} t_2} e^{-i\Delta_{\xi, \xi'} t_1} \langle 0, B_+ | \tilde{\phi}_{\xi'} \rangle \langle \phi_{\xi'} | \hat{s}_i(\mathbf{r}_D) | \phi_{\xi''} \rangle \langle \tilde{\phi}_{\xi''} | \hat{s}_j(\mathbf{r}_D) | \phi_{\xi} \rangle \langle \tilde{\phi}_{\xi} | 0, B_+ \rangle],$$
(33)

where  $t_2 \equiv t - t_1$ . By the two-dimensional Fourier transformation, we obtain the 2DFT SSESR spectrum in the frequency domain as

$$\chi_{i,j}(\omega_{2},\omega_{1}) \equiv 4g_{B} \sum_{\xi,\xi',\xi''} \operatorname{Im} \left[ \int_{0}^{\infty} dt_{2} \int_{0}^{\infty} dt_{1} e^{i\omega_{2}t_{2} + i\omega_{1}t_{1}} e^{-i\Delta_{\xi'',\xi'}t_{2} - i\Delta_{\xi,\xi'}t_{1}} \langle 0, B_{+} | \tilde{\phi}_{\xi'} \rangle \langle \phi_{\xi'} | \hat{s}_{i}(\boldsymbol{r}_{D}) | \phi_{\xi''} \rangle \langle \tilde{\phi}_{\xi''} | \hat{s}_{j}(\boldsymbol{r}_{D}) | \phi_{\xi} \rangle \langle \tilde{\phi}_{\xi} | 0, B_{+} \rangle \right]$$

$$= -4g_{B} \sum_{\xi,\xi',\xi''} \operatorname{Im} \left[ \frac{1}{\omega_{2} - \Delta_{\xi'',\xi'}} \frac{1}{\omega_{1} - \Delta_{\xi,\xi'}} \langle 0, B_{+} | \tilde{\phi}_{\xi'} \rangle \langle \phi_{\xi'} | \hat{s}_{i}(\boldsymbol{r}_{D}) | \phi_{\xi''} \rangle \langle \tilde{\phi}_{\xi} | 0, B_{+} \rangle \right]$$

$$(34a)$$

$$\equiv -4g_B \sum_{\xi,\xi',\xi''} \operatorname{Im}[\chi_{i,j}^{(\xi,\xi',\xi'')}(\omega_2,\omega_1)],$$
(34b)

where the spectrum of  $\omega_1$  indicates the spin-polarization coherence with the pump process, whereas that of  $\omega_2$  indicates the induced polarization by the probe pulse. The selection rule for the allowed transition under the far-off resonant case is given by

$$\langle 0, B_{+} | \hat{s}_{i}(\mathbf{r}_{D}) \hat{s}_{j}(\mathbf{r}_{D}) | 0, B_{+} \rangle = \begin{cases} \frac{i}{2} \langle 0, B_{+} | \hat{s}_{k}(\mathbf{r}_{D}) | 0, B_{+} \rangle \epsilon_{ijk}, \\ \delta_{i,j}, \end{cases}$$
(35)

where  $\epsilon_{ijk}$  is the Levy-Civita symbol. It is seen from (34) that the quantum coherence between the resonance states is reflected in the cross correlation of  $\omega_1$  and  $\omega_2$  in the 2DFT SSESR spectrum [58,60].

We have derived the explicit form of the discrete eigenstates of the total Hamiltonian in terms of the projection method given by (C11) and (C12) as

$$\begin{aligned} |\phi_{\xi}\rangle &= \frac{1}{\mathcal{N}_{\xi}^{1/2}} \Bigg[ \Bigg\{ |0,+\rangle + (v+L)\sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} |k,+\rangle \Bigg\} \\ &+ \frac{B_{x} + iB_{y}}{B_{z} + (v-L)^{2} \Sigma^{+}(z_{\xi}) - z_{\xi}} \Bigg\{ |0,-\rangle + (v-L)\sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} |k,-\rangle \Bigg\} \Bigg], \end{aligned}$$
(36a)

$$\begin{split} \langle \tilde{\phi}_{\xi} | &= \frac{1}{\mathcal{N}_{\xi}^{1/2}} \Bigg[ \Bigg\{ \langle 0, + | + (v+L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} \langle k, + | \Bigg\} \\ &+ \frac{B_{x} - iB_{y}}{B_{z} + (v-L)^{2} \Sigma^{+}(z_{\xi}) - z_{\xi}} \Bigg\{ \langle 0, - | + (v-L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} \langle k, - | \Bigg\} \Bigg], \end{split}$$
(36b)

where the normalization factor is given by

$$\mathcal{N}_{\xi} = \left(1 - (v+L)^2 \frac{d}{dz} \Sigma^+(z_{\xi})\right) + \left(1 - (v-L)^2 \frac{d}{dz} \Sigma^+(z_{\xi})\right) \frac{z_{\xi} + B_z - (v+L)^2 \Sigma^+(z_{\xi})}{z_{\xi} - B_z - (v-L)^2 \Sigma^+(z_{\xi})}.$$
(37)

With use of the explicit forms of the transition matrix elements given in (C19), we find that each term of the two-dimensional Fourier transform spectrum is proportional to the product of the normalization factors,

$$\chi_{i,j}^{(\xi,\xi',\xi'')}(\omega_2,\omega_1) \propto \frac{1}{\mathcal{N}_{\xi}\mathcal{N}_{\xi''}\mathcal{N}_{\xi'}^*} \frac{1}{\omega_2 - \Delta_{\xi'',\xi'}} \frac{1}{\omega_1 - \Delta_{\xi,\xi'}} \,. \tag{38}$$

The first factors (the normalization constants) represent the Peterman effect, and the second and third resonance factors (the propagators) represent the Purcell effect [20,22].

We have also calculated the 1DFT for the sudden excitation of the probe pulse at  $t_1 = 0$  [60], which reads

$$\mathcal{S}_{i,j}(\omega_2) = 4g_B \sum_{\xi',\xi''} \operatorname{Re}\left[\frac{1}{\omega_2 - \Delta_{\xi'',\xi'}} \langle 0, B_+ | \tilde{\phi}_{\xi'} \rangle \langle \phi_{\xi'} | \hat{s}_i(\mathbf{r}_D) | \phi_{\xi''} \rangle \langle \tilde{\phi}_{\xi''} | \hat{s}_j(\mathbf{r}_D) | 0, B_+ \rangle\right]$$
(39)  
$$\equiv 4g_B \sum \operatorname{Re}\left[\mathcal{S}_{i,\xi''}^{(\xi',\xi'')}(\omega_2)\right].$$
(40)

$$\equiv 4g_B \sum_{\xi',\xi''} \operatorname{Re}\left[\mathcal{S}_{i,j}^{(\xi',\xi'')}(\omega_2)\right]. \tag{40}$$

Each term of  $S_{i,j}^{(\xi',\xi'')}$  is also composed of a normalization factor and a resonance enhancement factor as

$$\mathcal{S}_{i,j}^{(\xi',\xi'')}(\omega_2) \propto \frac{1}{\mathcal{N}_{\xi'}^* \mathcal{N}_{\xi''}} \frac{1}{\omega_2 - \Delta_{\xi'',\xi'}}.$$
(41)

It is seen from comparison of (38) and (41) that the 2DFT spectrum is relatively more enhanced than the 1DFT spectrum.

We show in Fig. 7 the 1DFT SSESR spectrum when the external magnetic field is directed parallel to the molecular field: (a)  $B_z = 0.5$ , (b)  $B_z = 0.1$ , and (c)  $B_z = 0.02$ , whereas  $B_x = B_y = 0$ . In this case, since the spin states are decoupled into the up- and down-spin subspaces, it is easy to evaluate the 1DFT SSESR spectrum. Here we evaluate the  $S_{x,x}$  component shown in Fig. 7(a) in the resonance pole approximation as

$$S_{x,x}(\omega_2) = 4g_B |\langle 0, B_+ | \tilde{\phi}_+ \rangle|^2 \operatorname{Re} \left[ \frac{\langle \phi_+ | \hat{s}_x | \phi_- \rangle \langle \dot{\phi}_- | \hat{s}_x | \phi_+ \rangle}{\omega_2 - \Delta_{\phi_-,\phi_+}} \right]$$
(42a)

$$\simeq 4g_B \frac{\omega_2 - 2B_z}{(\omega_2 - 2B_z)^2 + 4\gamma^2 (v^2 + L^2)^2},$$
 (42b)

with  $\gamma$  defined by

$$\gamma \equiv |\operatorname{Im} \Sigma(B_z)| = \frac{1}{2}\sqrt{4 - B_z^2} .$$
(43)

In (42a), we have used  $\Delta_{\phi_-,\phi_+} \simeq 2B_z + 2i\gamma(v^2 + L^2)$ ,  $\langle \phi_+ | \hat{s}_x | \phi_- \rangle \simeq \langle \tilde{\phi}_- | \hat{s}_x | \phi_+ \rangle \simeq 1$ , and  $|\langle 0, B_+ | \tilde{\phi}_+ \rangle|^2 \simeq 1$  as evaluated in (C19), which are good approximations for  $B_z \ll 2$ . Therefore, the  $S_{x,x}$  component is represented by a dispersiontype function with the node at  $\omega_2 = 2B_z$  and the width of  $2\gamma(v^2 + L^2)$ . As shown in the top row of Fig. 7 as  $B_z$  decreases, the node position of  $S_{x,x}$  shifts from (a)  $\omega_2 \simeq 1.0$ , (b) 0.2, to (c) 0.04, which corresponds to the decrease in the separation of the real parts of the eigenvalues shown in Fig. 2(a). On the other hand, the spectral width of  $S_{x,x}$  becomes slightly broadened, reflecting the very slight increase in the separation of the imaginary part of the eigenvalues in Fig. 2(b).

Similarly, the  $S_{y,x}$  component is given by

$$S_{y,x}(\omega_2) = 4g_B |\langle 0, B_+ | \tilde{\phi}_+ \rangle|^2 \operatorname{Re} \left[ \frac{\langle \phi_+ | \hat{s}_y | \phi_- \rangle \langle \phi_- | \hat{s}_x | \phi_+ \rangle}{\omega_2 - \Delta_{\phi_-,\phi_+}} \right]$$
(44a)

$$\simeq -8g_B \frac{2\gamma(v^2 + L^2)}{(\omega_2 - 2B)^2 + 4\gamma^2(v^2 + L^2)^2},$$
 (44b)

i.e., a negative Lorentzian with the peak at  $\omega_2 = 2B$  and the width  $2\gamma(v^2 + L^2)$  where we have used  $\langle \phi_+ | \hat{s}_y | \phi_- \rangle \simeq i$  in (44a). As shown in the middle row of Fig. 7, the negative shift of the peak position and the slight increase in the width also correspond to the changes in the real and imaginary parts of the eigenvalues in Figs. 2(a) and 2(b), respectively. The  $S_{z,z}$  component is also given by the dispersion-type function with the node at  $\omega_2 = 0$  and the width of  $2\gamma(v + L)^2$  as

$$S_{z,z}(\omega_2) = 4g_B |\langle 0, B_+ | \tilde{\phi}_+ \rangle|^2 \operatorname{Re} \left[ \frac{\langle \phi_+ | \hat{s}_z | \phi_+ \rangle \langle \phi_+ | \hat{s}_z | \phi_+ \rangle}{\omega_2 - \Delta_{\phi_+,\phi_+}} \right]$$
(45a)

$$\simeq 4g_B \frac{\omega_2}{\omega_2^2 + 4\gamma^2 (\upsilon + L)^4}.$$
(45b)

as shown in the bottom row of Fig. 7, where we have used  $\langle \phi_+ | \hat{s}_z | \phi_+ \rangle \simeq 1$  and  $\Delta_{\phi_+,\phi_+} \simeq -2i\gamma(v+L)^2$  in (45a). Due to the symmetry, the other components of  $S_{x,z}$ ,  $S_{y,z}$ ,  $S_{z,x}$ , and  $S_{y,z}$  vanish. We show the 2DFT SSESR spectrum in Fig. 8 for the same conditions. The cross correlation between  $\omega_1$  and  $\omega_2$  provides us with detailed information about the spin relaxation at the impurity site.



FIG. 7. The 1DFT SSESR spectrum  $S_{i,j}(\omega_2)/g_B$  for (a)  $B_z = 0.5$  and (b)  $B_z = 0.1$ , and (c)  $B_z = 0.02$ , where  $B_x = B_y = 0$ . The parameters are taken as v = 0.3 and L = 0.1. The spectral components  $S_{x,x}$ ,  $S_{y,x}$ , and  $S_{z,z}$  are shown in the top, middle, and bottom rows, respectively. The horizontal axis denotes the frequency of  $\omega_2$  with a unit of J.

As shown in Figs. 3 and 5, the spin states are coupled through the spin-orbit interaction when the external magnetic field is tilted away from the polar axis of L. The change in the spin states is reflected in the 2DFT SSESR as an anisotropy in terms of the relative angle between the probe field and the detection direction. We show in Fig. 9 the 2DFT SSESR spectrum when the external magnetic field is perpendicular to the molecular field: (a)  $B_x = 0.5$ , (b)  $B_x = 0.1$ , and (c)  $B_x = 0.02$  for  $B_y = B_z = 0$  where the first-fourth rows are  $\chi_{y,x}$ ,  $\chi_{z,x}$ ,  $\chi_{x,y}$ , and  $\chi_{x,z}$  components, respectively. This figure corresponds to the case of Fig. 5. Although these spectral components are forbidden by the selection rule for the far-offresonant case (35), they become allowed by the introduction of the spin-orbit coupling whose effect becomes prominent due to the resonance factor. The cross correlation between  $\omega_1$ and  $\omega_2$  provides detailed information on the spin-relaxation process at the donor site.

Lastly, we show the most striking feature due to the presence of the EP ring, *the giant response* from the single-spin resonance. For the illustration, we show in Fig. 10 the  $S_{z,x}$ component of the 1DFT SSESR for various values of  $B_x$  as we cross the EP ring singularity, corresponding to Figs. 5(c) and 5(d). As the value of  $B_x$  decreases from (1) to (i), the negative peak position shifts to the lower-frequency side. This peak position reflects the real Zeeman splitting as  $2B_x$  as shown in Fig. 5(c). As  $B_x$  further decreases and comes close to the  $B_{EP,-}$ singularity, the signal amplitude becomes large and positive as seen in (f). When  $B_x$  crosses over  $B_{EP,-}$ , the sign of the signal flips whereas keeping the large signal amplitude as shown in (e). Then as  $B_x$  decreases, the signal amplitude becomes small with a dispersion-type function with a node at  $B_x = 0$  as shown in (d) to (a).

The giant response at the EP ring singularity is much more pronounced in the 2DFT SSESR spectrum. We show in Fig. 11 the 2DFT SSESR spectral components at  $B_x =$ 0.063 27 corresponding to Fig. 10(e). We find a huge signal amplitude in the spectrum, due to the enhancement from the normalization constant and the resonance factor as mentioned in (38). This giant response of the SSESR at the EP singularity may be very useful for observing the single-spin resonance, which is very weak under ordinary conditions. Therefore, this method can potentially provide a powerful tool to observe the spin-relaxation process with an atomic scale precision.

#### V. CONCLUDING REMARKS

We have studied the complex eigenenergy structure associated with the charge-transfer decay of an alkali-doped polyacetylene molecule under an external static magnetic field. Starting with the Hermitian Hamiltonian of the total system, we have derived the non-Hermitian effective Hamiltonian where the microscopic spin-dependent decay processes are incorporated in terms of the energy-dependent self-energy. The dynamical and chiral symmetries of the total Hamiltonian are maintained in the effective Hamiltonian.

We have found that the spin-orbit interaction influences the Zeeman splitting of the spin states at the donor site in the charge-transfer decay. As a result, the complex eigenenergy structure exhibits a strong anisotropy in the external magnetic field: The EP singularity due to the chiral symmetry breaking



FIG. 8. The 2DFT SSESR spectrum for (a) $B_z = 0.5$ , (b)  $B_z = 0.1$ , and (c)  $B_z = 0.02$ , where  $B_x = B_y = 0$ . The parameter values are the same as in Fig. 7. The spectral components of  $\chi_{x,x}$ ,  $\chi_{y,x}$ , and  $\chi_{z,z}$  are shown in the top, middle, and bottom views, respectively, where the spectral intensities are depicted by the color indicators of ( $\chi_{i,j}/g_B$ ) (a.u.). The horizontal and vertical axes denote the frequency of  $\omega_1$  and  $\omega_2$  in an energy unit of J = 1, respectively.

appears when the magnetic field is perpendicular to the molecular field and forms an EP ring. By contrast, the EP singularity due to the dynamical symmetry breaking appears irrespective of the field direction yielding an EP surface.

We have revealed that the coherent single-spin-resonance spectrum is a powerful tool to observe the complex eigenenergy spectrum of the system with atomic precision by using its sensitivity to the directions of the probe and detection pulses. For the formulation of the SSESR, we have utilized the non-linear-response function in the Liouville-space pathway approach where we have constructed the Liouville-space basis in terms of the complex eigenstates of the total Hamiltonian. We emphasize the importance of the energy dependence of the self-energy in the Liouville-space formalism. In the Liouville-space formalism, we need to take into account pairs of resonance and antiresonance states with complex conjugate eigenvalues so that the Liouville basis is consistent with the second law of thermodynamics, i.e., entropy production [33,34,61]. The present method properly deals with the dynamical symmetry of the total Hermitian Hamiltonian in the effective Hamiltonian by taking into account the energy dependence of the self-energy.

We have demonstrated that the 1DFT and 2DFT SSESR spectra probe the spin-relaxation dynamics at the donor site. Whereas the 1DFT SSESR spectrum reflects the complex eigenenergy spectrum, the 2DFT gives detailed information on the quantum coherence in the spin-relaxation dynamics as a cross correlation between the two frequencies. We found a giant response in the coherent SSESR around the EP ring singularity due to the vanishing normalization factors at the EP ring and the resonance effect. We have discovered that the giant response is much larger in magnitude in the 2DFT spectrum than in the 1DFT spectrum. Therefore, the 2DFT SSESR spectroscopy becomes a promising tool to observe the



FIG. 9. The 2DFT SSESR spectrum for (a)  $B_x = 0.5$ , (b)  $B_x = 0.1$ , and (c)  $B_x = 0.02$ , where  $B_z = B_y = 0$  for the same parameters as in Fig. 5, where the spectral intensities are depicted by the color indicators of  $(\chi_{i,j}/g_B)$  (a.u.). The horizontal and vertical axes denote the frequency of  $\omega_1$  and  $\omega_2$  in an energy unit of J = 1, respectively. The first–fourth rows are  $\chi_{y,x}$ ,  $\chi_{z,x}$ ,  $\chi_{x,y}$ , and  $\chi_{x,z}$  components, respectively, which are only allowed by the introduction of the spin-orbit coupling.

single-spin response in a molecule in contrast to the ordinary linear response, such as absorption or spontaneous emission spectroscopies [22,23].

Several issues remain to be explored further. In this paper, we have focused on the spin-polarized charge-transfer decay in a polyacetylene molecule due to the spin-orbit coupling. The charge transfer in polyacetylene has also been investigated in terms of the soliton or polaron formation accompanied by the structural deformation [44]. Although we do not take into account the dynamical change in the



FIG. 10. The  $S_{z,x}/g_B$  component of the 1DFT SSESR around the EP ring singularity, which occurs at  $B_x = B_{EP,-} = 0.063\,2808$ . The double line between (e) and (f) indicates the edge of the EP ring. The horizontal axis denotes the frequency of  $\omega_2$  with a unit of J.

molecular structure in this paper because it is considered to be much slower than the electronic motion, it would be worth considering the influence of the soliton or polaron motion on the single-spin-resonance spectroscopy.

We have revealed in Sec. III the appearance of the two EP singularities: EP surface and EP ring. If the spin-orbit interaction is large or the conduction bandwidth is small, it can be shown that the EP ring and EP surface will merge. It would be interesting to study the response at the merged singularities where non-Markovian effects might be enhanced [9,76].

For the detailed study very close to the EP, it would be better to use a basis that takes into account the pseudoeigenstate representations [5]. Otherwise the resonant and antiresonant state representations may increase numerical error very close to the EP singularity because the coalesced state at an EP is self-orthogonal and cannot be normalized. Recently, Hashimoto and Kanki *et al.* have invented the *extended Jordan block basis* which is continuously connected to the Jordan block exactly at the exceptional point. The extended Jordan block basis provides a useful way to describe the observable quantities continuously across the exceptional point [77–80]. This representation may remove some difficulties in calculating the ESR spectrum close to the EP singularity. Moreover, an interesting aspect in the Liouville space representation is the order of the EP singularity. Since, as shown in (D3), the eigenstate of the Liouvillian is given by a dyadic product of the eigenstates of the Hamiltonian which tend to the coalesced states of the Hamiltonian at an EP, there are four eigenstates of the Liouvillian coalescing at the EP, leading to the EP4 singularity in the Liouville space. This fourth-order pole structure may be reflected at the EP ring singularity. Detailed analysis of this EP4 will be performed in the future.

In the present paper, we have adopted the impulsive pulse limit in the SSESR spectrum to examine the effect of the EP singularity. Meanwhile, recent advances in the pulse control technique with the use of phase-locked laser pulse, such as carrier-envelope-phase control [81], can enable us to study the single-spin-relaxation process in more detail, such as quantum decoherence as well as population decay.



FIG. 11. The 2DFT SSESR spectrum at  $B_x = 0.063\,27 \simeq B_{\rm EP,-}$  where the spectral intensities are depicted by the color indicators of  $(\chi_{i,i}/g_B)$  (a.u.). The horizontal and vertical axes denote the frequency of  $\omega_1$  and  $\omega_2$  in an energy unit of J = 1, respectively.

For the SSESR calculation, we have used the timedependent perturbation method in terms of the probe magnetic pulse field, assuming a weak probe field intensity. However, this treatment could be reexamined close to the EP singularity because the interaction energy with the probe pulse becomes very large even though the field intensity is weak. For the strong time-dependent external field, the Floquet method could be applied to the present system in the Liouville space [82–84].

Lastly, we comment on some applications of the present methods. In the present paper, we have demonstrated that the spin-dependent dissipation effect is crucial to the giant response of the single-spin ESR in mesoscopic materials. Replacing the impurity atom with a quantum dot, we may apply the present model to the electron transfer through a single quantum dot under the external magnetic field [50]. We can also expect that the giant nonlinear response is useful to control the spin-dependent electronic transport through a quantum dot used as a qubit in quantum information technology. Therefore, the present results can provide new insight to the research field of spintronics [85].

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## APPENDIX A: MATRIX ELEMENTS OF THE ORBITAL ANGULAR MOMENTUM BETWEEN THE ATOMIC ORBITALS

In this Appendix, we evaluate the matrix elements of the orbital angular momentum between the *ns* orbital of the heavy alkali donor atom and the  $2p_y$  orbital of the carbon atom at the end of the molecule. The matrix elements are given by

$$l_i \equiv \langle 0, s | \hat{l}_i | 1, p_y \rangle = \int d^3 \boldsymbol{r} \, \varphi_s^*(\boldsymbol{r}) \hat{l}_i \varphi_y(\boldsymbol{r} - \boldsymbol{a}), \qquad (A1)$$

where we locate the *ns* donor atomic orbital  $\varphi_z$  at the origin, and the  $2p_y$  carbon atomic orbital  $\varphi_y$  is centered at  $\mathbf{r} = \mathbf{a}$  as shown in Fig. 1. The wave functions are given by

$$\varphi_{y}(\mathbf{r}) = R_{21}(r)\{Y_{1,1}(\theta,\phi) + Y_{1,-1}(\theta,\phi)\},$$
 (A2)

$$Y_{1,1}(\theta,\phi) + Y_{1,-1}(\theta,\phi) = \sqrt{\frac{3}{8\pi}} \sin \theta \sin \phi,$$
 (A3)

and the alkali atom ns orbital as

$$\varphi_s(\mathbf{r}) = \varphi_s(r) = \frac{1}{\sqrt{4\pi}} R_{ns}(r) e^{-r}.$$
 (A4)

Substituting (A2) into (A1), we have after simple calculation,

$$l_{z} = \sqrt{\frac{3}{4\pi}} \int d^{3}\boldsymbol{r} \, \varphi_{s}^{*} \left(\boldsymbol{r}\right) e^{-ia\hat{\rho}_{s}} \frac{1}{i} (a \, \sin^{2}\theta \, \sin^{2}\phi \frac{\partial}{\partial r} R_{21}(r) + a \frac{\cos^{2}\theta \, \sin^{2}\phi - \cos^{2}\phi}{r} R_{21}(r) - R_{21}(r) \sin\theta\cos\phi \bigg).$$
(A5)

Similarly, we obtain for  $\hat{l}_x$  and  $\hat{l}_y$  elements as

$$l_x = \sqrt{\frac{3}{\pi}} \int d^3 \boldsymbol{r} \, \varphi_s^*(\boldsymbol{r} + \boldsymbol{a}) R_{21}(r) \cos \theta, \qquad (A6)$$

 $(0, -| | -B_x - i)$ 

$$l_{y} = \sqrt{\frac{3}{4\pi}} a \int d^{3}r \, \varphi_{s}^{*}(r+a) \frac{1}{i} \sin \theta \, \cos \theta \, \sin \phi$$
$$\times \left( \frac{\partial}{\partial r} R_{21}(r) - \frac{1}{r} R_{21}(r) \right). \tag{A7}$$

Substituting (A4) into (A5)–(A7), and executing the solid angle integral, we find

$$l_{z} = \frac{3}{16} \sqrt{\frac{3}{2}} \pi a \int dr \, r^{2} R_{ns}(r+a) e^{-(r+a)} \frac{1}{i} \frac{\partial}{\partial r} R_{21}(r), \quad (A8)$$

whereas

$$l_x = l_y = 0. \tag{A9}$$

The radial integral can be performed once the radial wave function of the atomic orbitals is given.

## **APPENDIX B: CHIRAL SYMMETRY** OF THE TOTAL HAMILTONIAN

In this Appendix we show the chiral symmetry of the total system Hamiltonian in the **B**-parameter space. For this purpose, we represent the total Hamiltonian in terms of the alternate site basis which is aligned as

$$\{|0, +\rangle, |1, -\rangle, |2, +\rangle, \dots, |0, -\rangle, |1, +\rangle, |2, -\rangle\}.$$
 (B1)

The total Hamiltonian is represented by

$ 0,+\rangle$	$ 1, -\rangle$	$ 2,+\rangle$		0,- angle	$ 1,+\rangle$	$ 2, -\rangle$		
$\varepsilon_D - B_z$	0	0	• • •	$-B_x + iB_y$	v + L	0		
0	0	0		v-L	0	-J		
0	0	0		0	-J	0		
÷	÷	÷	÷	÷	÷	÷	÷	
$-B_x - iB_y$	v-L	0		$\varepsilon_D + B_z$	0	0		
v + L	0	-J		0	0	0		
0	-J	0	•••	0	0	0		

For  $\varepsilon_D = 0$ , we see that the total Hamiltonian satisfies a chiral symmetry [86] when  $B_z = 0$ ,

 $\langle 0, + |$ 

 $\langle 1, - |$ 

(2, +)

(1, +)

(2, -)

 $\hat{H}_{\text{tot}} =$ 

$$\boldsymbol{\sigma}_{z}\hat{H}_{\text{tot}}\boldsymbol{\sigma}_{z}=-\hat{H}_{\text{tot}},\tag{B3}$$

where  $\sigma_z$  is an infinite-dimensional matrix for alternate site basis,

$$\boldsymbol{\sigma}_{z} \equiv \begin{pmatrix} I & 0\\ 0 & -I \end{pmatrix}. \tag{B4}$$

It follows from (B3) that

$$\hat{H}_{\text{tot}}|\psi\rangle = z|\psi\rangle$$
, (B5a)

$$\Rightarrow \boldsymbol{\sigma}_{z} \hat{H}_{\text{tot}} \bar{\boldsymbol{\sigma}}_{z} \bar{\boldsymbol{\sigma}}_{z} |\psi\rangle = z \boldsymbol{\sigma}_{z} |\psi\rangle, \qquad (B5b)$$

$$\Rightarrow \hat{H}_{\text{tot}}\bar{\boldsymbol{\sigma}}_{z}|\psi\rangle = -z\bar{\boldsymbol{\sigma}}_{z}|\psi\rangle , \qquad (B5c)$$

which suggests that the eigenvalues of the total Hamiltonian are obtained as a pair of z and -z at  $B_z = 0$ . We note that when  $\varepsilon_D \neq 0$  or  $B_z \neq 0$ , the chiral symmetry is not satisfied so that the EP ring does not appear. We have shown that the

total Hamiltonian has a property that it exhibits the chiral symmetry at the special chiral configuration in the **B** space, i.e., at  $B_{z} = 0.$ 

## APPENDIX C: COMPLEX EIGENVALUE PROBLEM OF THE TOTAL HAMILTONIAN

In this Appendix, we derive the effective Hamiltonian in terms of the BWF projection method. We consider the projection operator onto the two-spinor at the donor site given by

$$\hat{P}_0 \equiv \sum_{\alpha=\pm} |0,\alpha\rangle \langle 0,\alpha|, \qquad (C1)$$

and its complement,

$$\hat{Q}_0 \equiv 1 - \hat{P}_0 = \sum_{\alpha=\pm} \int dk |k, \alpha\rangle \langle k, \alpha|.$$
(C2)

Acting these projection operators on (10) from the left, we have

$$\hat{P}_0\hat{H}_{\rm tot}\hat{P}_0|\phi_{\xi}\rangle + \hat{P}_0\hat{H}_{\rm tot}\hat{Q}_0|\phi_{\xi}\rangle = z_{\xi}\hat{P}_0|\phi_{\xi}\rangle,\tag{C3a}$$

$$\hat{Q}_0 \hat{H}_{\text{tot}} \hat{P}_0 |\phi_{\xi}\rangle + \hat{Q}_0 \hat{H}_{\text{tot}} \hat{Q}_0 |\phi_{\xi}\rangle = z_{\xi} \hat{Q}_0 |\phi_{\xi}\rangle. \tag{C3b}$$

The effective Hamiltonian is given by

$$\hat{H}_{\rm eff}(\mathbf{B};z) = \hat{P}_0 \hat{H}_{\rm tot} \hat{P}_0 + \hat{P}_0 \hat{H}_{\rm tot} \hat{Q}_0 \frac{1}{z - \hat{Q}_0 \hat{H}_{\rm tot} \hat{Q}_0} \hat{Q}_0 \hat{H}_{\rm tot} \hat{P}_0.$$
(C4)

Evaluating this for the present model gives Eq. (12).

Then the complex eigenvalue problem of the effective Hamiltonian reads

$$\hat{H}_{\rm eff}(\mathbf{B};z)\hat{P}_0|\phi_{\xi}\rangle = z_{\xi}\hat{P}_0|\phi_{\xi}\rangle, \quad \langle \tilde{\phi}_{\xi}|\hat{P}_0\hat{H}_{\rm eff}(\mathbf{B};z) = z_{\xi}\langle \tilde{\phi}_{\xi}|\hat{P}_0.$$
(C5)

The explicit form of the eigenvalue problem then reads

$$\begin{pmatrix} -B_z + (v+L)^2 \Sigma^+(z_{\xi}) & -B_x + iB_y \\ -B_x - iB_y & B_z + (v-L)^2 \Sigma^+(z_{\xi}) \end{pmatrix} \begin{pmatrix} \langle 0, + |\phi_{\xi} \rangle \\ \langle 0, - |\phi_{\xi} \rangle \end{pmatrix} = z_{\xi} \begin{pmatrix} \langle 0, + |\phi_{\xi} \rangle \\ \langle 0, - |\phi_{\xi} \rangle \end{pmatrix}$$
(C6)

for the right eigenstates, and

$$\begin{pmatrix} \langle \tilde{\phi}_{\xi} | 0, + \rangle \\ \langle \tilde{\phi}_{\xi} | 0, - \rangle \end{pmatrix}^{T} \begin{pmatrix} -B_{z} + (v+L)^{2} \Sigma^{+}(z_{\xi}) & -B_{x} + iB_{y} \\ -B_{x} - iB_{y} & B_{z} + (v-L)^{2} \Sigma^{+}(z_{\xi}) \end{pmatrix} = z_{\xi} \begin{pmatrix} \langle \tilde{\phi}_{\xi} | 0, + \rangle \\ \langle \tilde{\phi}_{\xi} | 0, - \rangle \end{pmatrix}^{T}$$
(C7)

for the left eigenstates. From these eigenvalue problems, we see the relations,

$$\frac{\langle 0, -|\phi_{\xi}\rangle}{\langle 0, +|\phi_{\xi}\rangle} = \frac{B_x + iB_y}{B_z + (v - L)^2 \Sigma^+(z_{\xi}) - z_{\xi}},$$
(C8a)

$$\frac{\langle \tilde{\phi}_{\xi} | 0, -\rangle}{\langle \tilde{\phi}_{\xi} | 0, +\rangle} = \frac{B_x - iB_y}{B_z + (v - L)^2 \Sigma^+(z_{\xi}) - z_{\xi}},$$
(C8b)

which leads to

$$\frac{\langle \tilde{\phi}_{\xi} | 0, -\rangle \langle 0, -| \phi_{\xi} \rangle}{\langle \tilde{\phi}_{\xi} | 0, +\rangle \langle 0, +| \phi_{\xi} \rangle} = \frac{B_x^2 + B_y^2}{(B_z + (v - L)^2 \Sigma^+ (z_{\xi}) - z_{\xi})^2} .$$
(C9)

The eigenstates of the total Hamiltonian are obtained by adding the  $\hat{Q}_0$  component as

$$|\phi_{\xi}\rangle = \hat{P}_{0}|\phi_{\xi}\rangle + \hat{Q}_{0}|\phi_{\xi}\rangle = \hat{P}_{0}|\phi_{\xi}\rangle + \frac{1}{z_{\xi} - \hat{Q}_{0}\hat{H}_{\text{tot}}\hat{Q}_{0}}\hat{Q}_{0}\hat{H}_{\text{tot}}\hat{P}_{0}|\phi_{\xi}\rangle.$$
(C10)

In the present system, we have obtained

$$|\phi_{\xi}\rangle = \langle 0, + |\phi_{\xi}\rangle \left\{ |0, +\rangle + (v+L)\sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})^{+}_{z_{\xi}}} |k, +\rangle \right\} + \langle 0, - |\phi_{\xi}\rangle \left\{ |0, -\rangle + (v-L)\sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})^{+}_{z_{\xi}}} |k, -\rangle \right\}$$
(C11)

for the discrete right eigenstate with the complex eigenvalue of  $z_{\xi}$ . Similarly, the discrete left eigenstates are obtained as

$$\langle \tilde{\phi}_{\xi} | = \langle \tilde{\phi}_{\xi} | 0, + \rangle \left\{ \langle 0, + | + (v+L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})^{+}_{z_{\xi}}} \langle k, + | \right\} + \langle \tilde{\phi}_{\xi} | 0, - \rangle \left\{ \langle 0, - | + (v-L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})^{+}_{z_{\xi}}} \langle k, - | \right\}.$$
(C12)

The normalization factors are determined by the biorthonormality between the right and the left eigenstates as

$$\langle \tilde{\phi}_{\xi} | \phi_{\xi'} \rangle = \delta_{\xi,\xi'}. \tag{C13}$$

The normalization condition requires

$$1 = \langle \tilde{\phi}_{\xi} | \phi_{\xi} \rangle = \langle \tilde{\phi}_{\xi} | 0, + \rangle \langle 0, + | \phi_{\xi} \rangle \left\{ \left( 1 - (v+L)^2 \frac{d}{dz} \Sigma^+(z) \Big|_{z=z_{\xi}} \right) + \frac{\langle \tilde{\phi}_{\xi} | 0, - \rangle \langle 0, - | \phi_{\xi} \rangle}{\langle \tilde{\phi}_{\xi} | 0, + \rangle \langle 0, + | \phi_{\xi} \rangle} \left( 1 - (v-L)^2 \frac{d}{dz} \Sigma^+(z) \Big|_{z=z_{\xi}} \right) \right\}.$$
(C14)

Substituting (C9) into (C14), we obtain the normalization factor as

$$\mathcal{N}_{\xi} \equiv (\langle \tilde{\phi}_{\xi} | 0, + \rangle \langle 0, + | \phi_{\xi} \rangle)^{-1} = \left( \left\{ 1 - (v+L)^2 \frac{d}{dz} \Sigma^+(z_{\xi}) \right\} \right) + \left( 1 - (v-L)^2 \frac{d}{dz} \Sigma^+(z_{\xi}) \right) \frac{\left( B_x^2 + B_y^2 \right)}{\left[ B_z + (v-L)^2 \Sigma^+(z_{\xi}) - z_{\xi} \right]^2}$$
(C15a)

$$= \left(\left\{1 - (v+L)^2 \frac{d}{dz} \Sigma^+(z_{\xi})\right\}\right) + \left(1 - (v-L)^2 \frac{d}{dz} \Sigma^+(z_{\xi})\right) \frac{z_{\xi} + B_z - (v+L)^2 \Sigma^+(z_{\xi})}{z_{\xi} - B_z - (v-L)^2 \Sigma^+(z_{\xi})}.$$
(C15b)

which is the same as (37). Taking

$$\langle \tilde{\phi}_{\xi} | 0, + \rangle = \langle 0, + | \phi_{\xi} \rangle = \mathcal{N}_{\xi}^{-1/2}, \tag{C16}$$

the right and left eigenstates for the resonance states are represented by

$$\begin{split} |\phi_{\xi}\rangle &= \frac{1}{\mathcal{N}_{\xi}^{1/2}} \Bigg[ \Bigg\{ |0,+\rangle + (v+L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} |k,+\rangle \Bigg\} + \frac{B_{x} + iB_{y}}{B_{z} + (v-L)^{2} \Sigma^{+}(z_{\xi}) - z_{\xi}} \\ &\times \Bigg\{ |0,-\rangle + (v-L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} |k,-\rangle \Bigg\} \Bigg], \end{split}$$
(C17a)

$$\begin{split} \langle \tilde{\phi}_{\xi} | &= \frac{1}{\mathcal{N}_{\xi}^{1/2}} \Biggl[ \Biggl\{ \langle 0, + | + (v+L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} \langle k, + | \Biggr\} + \frac{B_{x} - iB_{y}}{B_{z} + (v-L)^{2} \Sigma^{+}(z_{\xi}) - z_{\xi}} \\ & \times \Biggl\{ \langle 0, - | + (v-L) \sqrt{\frac{2}{\pi}} \int_{0}^{\pi} dk \frac{\sin k}{(z-\omega_{k})_{z_{\xi}}^{+}} \langle k, - | \Biggr\} \end{split}$$
(C17b)

It should be noted that since the exceptional point is given by the simultaneous solution of the equations,

$$\eta^+(z_{\xi}) = 0, \quad \frac{d}{dz}\eta^+(z) = 0,$$
 (C18)

where  $\eta^+(z) = 0$  is the characteristic equation given in (15),  $\mathcal{N}_{\xi} = 0$  at the exceptional point. This indicates that the we cannot normalize the two independent resonance states at the exceptional point [5,77,78,80].

With use of the explicit form of the eigenstates of the total Hamiltonian, the transition matrix elements in the spectrum are evaluated as

$$\begin{split} \langle \tilde{\phi}_{\xi} | 0, B_{+} \rangle &= \frac{1}{\mathcal{N}_{\xi}^{1/2}} \bigg\{ \langle 0, + | 0, B_{+} \rangle + \langle 0, - | 0, B_{+} \rangle \frac{B_{x} - iB_{y}}{B_{z} + (v - L)^{2} \Sigma^{+}(z_{\xi}) - z_{\xi}} \bigg\}, \end{split}$$
(C19a)  
$$\langle \tilde{\phi}_{\xi''} | \hat{s}_{j} | \phi_{\xi} \rangle &= \frac{1}{\mathcal{N}_{\xi''}^{1/2}} \frac{1}{\mathcal{N}_{\xi''}^{1/2}} \bigg\{ \langle 0, + | \hat{s}_{j} | 0, + \rangle + \langle 0, + | \hat{s}_{j} | 0, - \rangle \frac{B_{x} + iB_{y}}{B_{z} + (v - L)^{2} \Sigma^{+}(z_{\xi}) - z_{\xi}} \\ &+ \langle 0, + | \hat{s}_{j} | 0, - \rangle \frac{B_{x} - iB_{y}}{B_{z} + (v - L)^{2} \Sigma^{+}(z_{\xi''}) - z_{\xi''}} \\ &+ \langle 0, - | \hat{s}_{j} | 0, - \rangle \frac{B_{x} - iB_{y}}{(B_{z} + (v - L)^{2} \Sigma^{+}(z_{\xi''}) - z_{\xi''})(B_{z} + (v - L)^{2} \Sigma^{+}(z_{\xi}) - z_{\xi})} \bigg\},$$
(C19b)  
$$\langle \phi_{\xi'} | \hat{s}_{i} | \phi_{\xi''} \rangle &= \frac{1}{\mathcal{N}_{\xi''}^{*^{1/2}}} \frac{1}{\mathcal{N}_{\xi''}^{*^{1/2}}} \bigg\{ \langle 0, + | \hat{s}_{i} | 0, + \rangle + \langle 0, + | \hat{s}_{i} | 0, - \rangle \frac{B_{x} + iB_{y}}{B_{z} + (v - L)^{2} \Sigma^{+}(z_{\xi''}) - z_{\xi''}} \\ &+ \langle 0, + | \hat{s}_{i} | 0, - \rangle \frac{B_{x} - iB_{y}}{B_{z} - (v - L)^{2} \Sigma^{+}(z_{\xi''}) - z_{\xi''}} \bigg\}$$

$$+\langle 0, -|\hat{s}_{i}|0, -\rangle \frac{B_{z}^{2} + (v - L)^{2} \Sigma^{-}(z_{\xi'}^{*}) - z_{\xi'}^{*}}{(B_{z} + (v - L)^{2} \Sigma^{+}(z_{\xi''}) - z_{\xi''})(B_{z} + (v - L)^{2} \Sigma^{-}(z_{\xi'}^{*}) - z_{\xi'}^{*})} \Bigg\}, \quad (C19c)$$

$$\langle 0, B_{+} | \tilde{\phi}_{\xi'} \rangle = \frac{1}{\mathcal{N}_{\xi'}^{*1/2}} \bigg\{ \langle 0, B_{+} | 0, + \rangle + \langle 0, B_{+} | 0, - \rangle \frac{B_{x} + iB_{y}}{B_{z} + (v - L)^{2} \Sigma^{-}(z_{\xi'}^{*}) - z_{\xi'}^{*}} \bigg\}.$$
(C19d)

# APPENDIX D: ANALYTIC CONTINUATION FOR THE LIOUVILLE PATHWAYS

In this section, we note the direction of the analytic continuation of the Liouville pathway in the double-sided Feynman diagram where we choose the pathway which is decaying in the future. Now we consider the transition probability from state  $|a\rangle$  to  $|b\rangle$  as

$$P_{ba}(t) = |\langle b|e^{-i\hat{H}t}|a\rangle|^2.$$
 (D1)

In terms of the Liouville pathway representation, we write this as

$$P_{ba}(t) = \langle \langle b; b | e^{-i\mathcal{L}t} | a; a \rangle \rangle.$$
 (D2)

We choose the analytic continuation so that the probability decays for the future. Therefore, we take the Liouville basis given by [34]

$$|\phi_{\xi};\phi_{\xi'}\rangle\rangle \equiv |\phi_{\xi}\rangle\langle\phi_{\xi'}|, \quad \langle\langle\tilde{\phi}_{\xi};\tilde{\phi}_{\xi'}| \equiv |\tilde{\phi}_{\xi'}\rangle\langle\tilde{\phi}_{\xi}|, \qquad (D3)$$

where  $\langle \phi_{\xi'} |$  and  $\langle \tilde{\phi}_{\xi} |$  are Hermitian conjugates of  $|\phi_{\xi'}\rangle$  and  $|\tilde{\phi}_{\xi}\rangle$ , respectively. Whereas  $|\phi_{\xi}\rangle$  and  $\langle \tilde{\phi}_{\xi} |$  are the right- and the

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left-complex eigenstates of the Hamiltonian, and the Liouville bases are the complex eigenstates of the Liouvillian,

$$\mathcal{L}|\phi_{\xi};\phi_{\xi'}\rangle\rangle = \Delta_{\xi,\xi'}|\phi_{\xi};\phi_{\xi'}\rangle\rangle, \quad \langle\langle\tilde{\phi}_{\xi};\tilde{\phi}_{\xi'}|\mathcal{L} = \Delta_{\xi,\xi'}\langle\langle\tilde{\phi}_{\xi};\tilde{\phi}_{\xi'}|,$$
(D4)

where

$$\Delta_{\xi,\xi'} = z_{\xi} - z_{\xi'}^*.$$
 (D5)

This basis satisfies the bicompleteness and the biorthonormality in the Liouville space as

$$1 = \sum_{\xi,\xi'} |\phi_{\xi};\phi_{\xi'}\rangle\rangle\langle\langle\tilde{\phi}_{\xi};\tilde{\phi}_{\xi'}|, \quad \langle\langle\tilde{\phi}_{\xi};\tilde{\phi}_{\xi'}|\phi_{\xi''};\phi_{\xi'''}\rangle\rangle = \delta_{\xi,\xi''}\delta_{\xi',\xi'''}.$$
(D6)

Substituting (D6) into (D2) we find

$$P_{ba}(t) = \sum_{\xi,\xi'} e^{-i(z_{\xi} - z_{\xi'}^{*})t} \langle b|\phi_{\xi}\rangle \langle \phi_{\xi'}|b\rangle \langle \tilde{\phi}_{\xi}|a\rangle \langle a|\tilde{\phi}_{\xi'}\rangle, \quad (D7)$$

which decays with the rate  $\text{Im}(z_{\xi} - z_{\xi'}^*)$  from the contributions of the resonance and antiresonance eigenstates, as required.

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