

Multidimensional Spectroscopy of Time-Dependent Impurities in Ultracold Fermions

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We investigate the system of a heavy impurity immersed in a degenerated Fermi gas, where the impurity's internal degree of freedom (pseudospin) is manipulated by a series of radiofrequency (RF) pulses at several different times. Applying the functional determinant approach, we carry out an essentially exact calculation of the Ramsey-interference-type responses to the RF pulses. These responses are universal functions of the multiple time intervals between the pulses for all time and can be regarded as multidimensional (MD) spectroscopy of the system in the time domain. A Fourier transformation of the time intervals gives the MD spectroscopy in the frequency domain, providing insightful information on the many-body correlation and relaxation via the cross-peaks, e.g., the off-diagonal peaks in a two-dimensional spectrum. These features are inaccessible for the conventional, one-dimensional absorption spectrum. Our scheme provides a new method to investigate many-body nonequilibrium physics beyond the linear response regime with the accessible tools in cold atoms.

I. INTRODUCTION

Spectroscopy, which records the responses of materials to external electromagnetic fields, has long been and probably will always be an essential tool to investigate the structures, behaviors, chemical reactions, and physical processes in materials. Conventional spectroscopy, such as ordinary nuclear magnetic resonance (NMR) and optical spectroscopy, usually shows the responses as a function of a single variable, e.g., the frequency of the electromagnetic wave, and hence is called one-dimensional (1D). In contrast, multidimensional (MD) spectroscopy unfolds spectral information into several dimensions, which improves resolution and overcomes spectral congestion. In addition, MD spectroscopy carries rich information on the correlations between resonance peaks and provides insights into physics that 1D spectroscopy cannot access. One of the earliest and most widely successful MD spectroscopy is the two-dimensional (2D) NMR, first proposed by Jean Jeener and later demonstrated by Richard Ernst and collaborators [1, 2]. 2D NMR can help distinguish overlapping signals in complex molecules and unveil the couplings between different resonances, which revolutionize, e.g., molecular dynamics and structural biology [3, 4].

As an analog of its NMR counterparts, optical MD coherent spectroscopy (MDCS) [5, 6] adapts similar technology for the IR, visible, or UV regions and sheds new light on chemical kinetics and solid-state physics [7–15]. In particular, optical 2DCS reveals coherent and incoherent coupling dynamics between resonances near the energy of neutral and charged excitons in atomically thin transition metal dichalcogenides (TMD) [16, 17]. More recently, people have believed that these resonances are excitons dressed by Fermi sea electrons, i.e., quasiparticles named attractive or repulsive exciton-polarons [18–23]. Polaron, arguably the most celebrated quasiparticle [24, 25], has also attracted intensive interest in atomic physics experimentally [26–36] and theoretically [37–61]. A fundamental and quantitative understanding of these

nonequilibrium many-body dynamics between quasiparticles shown in 2D spectroscopy is fascinating but challenging. While a commonly adapted approach, the modified optical Bloch equation with phenomenological terms to include many-body effects [62–64], gives some intuitive interpretations, first-principal calculations of 2D spectroscopy are rare. Despite some progress [65–67], a quantitative but perturbative study of the complete 2D spectroscopy has only been carried out recently using the nonlinear (four-wave mixing) Golden Rule [17] and more recently with functional determinant approach [68], with parameters that can only be approximately obtained in a complex solid-state system.

By contrast, we perform an in-principal *exact* calculation in a much simpler but realistic system: a heavy impurity immersed in a degenerate Fermi gas. In such a system, a single parameter, scattering length, can fully describe the interaction between the impurity and the isolated and non-interacting Fermi gas at ultracold temperature and be accurately tuned by Feshbach resonance [69]. This system is closely related to the Fermi polaron problem, whose 1D spectroscopy shows singularities [70, 71] that are remnants of polaron resonances destroyed by the well-known Anderson's "orthogonality catastrophe" (OC) [72]. Our recent studies rigorously proved that these singularities could reduce back to polaron resonances if a mechanism exists to prevent OC, such as a superfluid pairing gap [73, 74]. However, as far as we know, the correlations and coherent dynamics between the Fermi singularities or polaron resonances in ultracold gases have never been investigated; our work here is the first numerically exact calculation of the MD spectroscopy of a polaron-like system in ultracold gases. Here, we apply the functional determinant approach (FDA) [75–78], a non-perturbative method that rigorously includes all high-order correlations and beyond mean-field many-body effects. Since exact solutions of many-body systems are rare, our results can give new insight, deepen our understanding of Fermi-edge singularity and polaron physics, and be regarded as a benchmark to access the accuracy of other approximation calcula-

tions of MD spectroscopy.

We also propose a realistic experimental scheme to measure the MD spectroscopy via a generalization of Ramsey spectroscopy. Ramsey spectroscopy is another technique similar to the NMR, which manipulates internal (e.g., pseudospin instead of spin) degrees of freedom and observes the interference determined by the surrounding many-body environment. Ramsey spectroscopy has found many vital applications in investigating many-body physics: characterizing quantum correlations [79–82], measuring topological invariance [83, 84], accessing many-body interactions and beyond mean-field effects [85–90], and studying impurity dynamics closely related to polaron physics [70, 71, 91, 92]. Ramsey spectroscopy has become a well-established experimental technique in ultracold atomic gases [30], thanks to the unprecedented controllability and rich toolbox atomic physics provides [69, 93]. However, to the best of our knowledge, all previous studies of Ramsey spectroscopies are 1D. Our work generalizes the Ramsey spectroscopy to multidimensional, opening the door to exploring high-order many-body correlations and beyond mean-field dynamics and providing a perfect meeting point for theoretical and experimental efforts to examine complex nonequilibrium responses that MD spectroscopy reveals.

The rest of this paper is organized as follows. In the following section, we establish our general formalism and show how to apply the exact FDA approach to calculate MD spectroscopy. Section III is devoted to presenting our numerical results. Finally, we conclude our paper by discussing the physics and proposing future extensions in section IV.

II. FORMALISM

The basic setup of our system is shown in Fig. 1 (a). We place a localized fermionic or bosonic impurity (the big black ball) with two internal pseudospins (hyperfine) states $|\uparrow\rangle$ and $|\downarrow\rangle$ (illustrated by the black arrow) in the background of a single-component ultracold Fermi gas (the red dots). The localization of impurity can be either achieved by confinement of a deep optical lattice or treated as an approximation to an impurity atom with heavy mass. At ultralow temperature, the background Fermi gas is considered non-interacting. We also assume the fermionic background atoms do not interact with $|\downarrow\rangle$, while s -wave interactions dominate interaction with $|\uparrow\rangle$. This interaction is characterized by the s -wave scattering length a and can be tuned via, e.g., Feshbach resonances [69]. The general spirit of our scheme is similar to the original Ramsey interferometry, where one uses radio-frequency (RF) pulses to manipulate the superposition of pseudospin states. Throughout this work, we assume the RF pulses to be infinitely fast rotations of pseudospin that do not perturb the background Fermi gas. After some time of evolution, dynamical phases accumulate for different pseudospin states, which reflects the many-body

responses to the different impurity-background interactions and can be measured by the interference. However, there is one crucial difference in our scheme: we use multiple pulses with different time delays in between to drive the pseudo-spin through many different quantum pathways that give nonlinear many-body responses in an MD spectroscopy.

One example of a three-pulse scheme is shown in Fig. 1 (a), which is similar to one of the most common 2D NMR pulse sequences, namely EXSY (EXchange SpectroscopyY). In this scheme, we prepare the impurity in the non-interacting state $|\downarrow\rangle$ initially, and apply the first $\pi/2$ pulse that rotates the pseudospin state to $(|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2}$. After some time τ , we apply the second pulse. Subsequently, we wait for another period of time, T , before applying the third pulse and carry out a detection some time t afterward. Following the same procedure as EXSY in NMR, we take the Fourier transformation with respect to both time variables τ and t to generate a 2D spectrum as a function of an absorption frequency ω_τ and an emission frequency ω_t , respectively, whose physical interpretation will become clear later (in the last paragraph of section II). The mixing time T allows many-body dynamical evolution between absorption and emission. Figure 1 (b) sketches a 2D spectrum in the box, where diagonal peaks (red crosses) on the dashed line mirror the singularities in the linear 1D spectrum (shown on the top and to the right of the box). Coupled resonances give rise to the off-diagonal cross-peaks (orange crosses) with the absorption frequency of one resonance and the emission frequency of the other, whereas uncorrelated resonances produce no cross-peaks. The cross peaks are thus the signature of correlations between resonances, which the 1D spectrum cannot distinguish. Figure 1 (c)-(g) shows several different pulse schemes investigated in this work. Most of the pulses in this work are $\pi/2$ pulses, except for the second pulse in the scheme of Fig. 1 (g), which is a $-\pi/2$ pulse. For convenience, we name (f) and (g) EXSY+ and EXSY−, respectively.

Using the unit $\hbar = 1$ hereafter, we write the many-body Hamiltonian as,

$$\hat{H} = \hat{H}_\downarrow |\uparrow\rangle\langle\uparrow| + \hat{H}_\downarrow |\downarrow\rangle\langle\downarrow|, \quad (1)$$

where the non-interacting ($\mathcal{H}_\downarrow$) and interacting ($\mathcal{H}_\uparrow$) Hamiltonian are given by $\hat{H}_\downarrow = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} c_{\mathbf{k}}^\dagger c_{\mathbf{k}}$ and $\hat{H}_\uparrow = \hat{H}_\downarrow + \sum_{\mathbf{k}, \mathbf{q}} \tilde{V}(\mathbf{k}-\mathbf{q}) c_{\mathbf{k}}^\dagger c_{\mathbf{q}} + \omega_s$. Here, ω_s denotes the energy differences between the two pseudospin levels. $c_{\mathbf{k}}^\dagger$ and $c_{\mathbf{k}}$ are creation and annihilation operators of the background fermions with momentum $\mathbf{k}$, respectively. $\epsilon_{\mathbf{k}} = k^2/2m$ is the single-particle kinetic energy of the background fermions with mass m . $\tilde{V}(\mathbf{k})$ is the Fourier transform of $V(\mathbf{r})$, the interaction potential between $|\uparrow\rangle$ and the background fermions. Initially, we prepare the impurity in $|\downarrow\rangle$ and the background fermions at some temperature T° . The background fermions can be described by a thermal density matrix $\rho_{\text{FS}} = \exp[-(\hat{H}_\downarrow - \mu\hat{N})/k_B T^\circ]/Z_{\text{FS}}$,

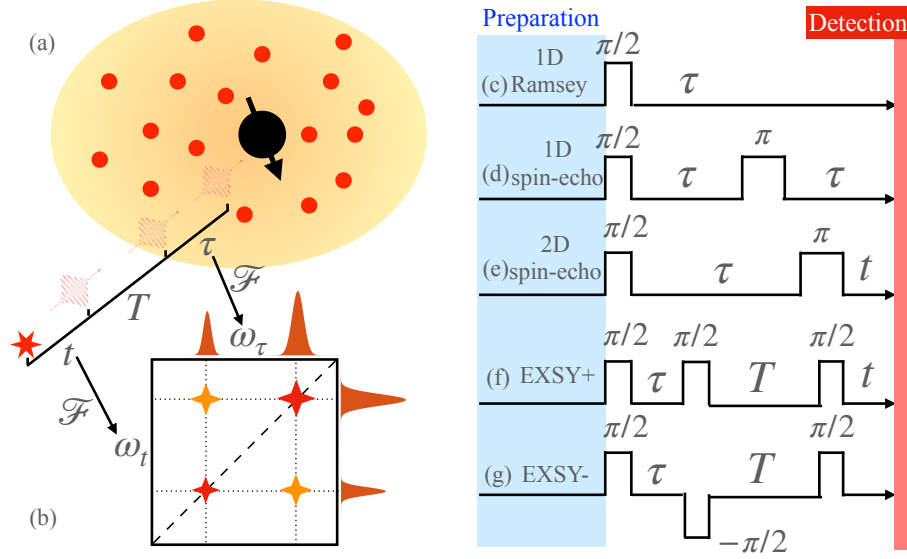


FIG. 1. (a) A sketch of system setup: A localized impurity (the big black ball with an arrow indicating pseudospin states) is immersed in a sea of host fermions (red dots) and is manipulated by a series of RF pulses with time intervals between pulses and detection (indicated by the red six-point star). (b) is an example of a 2D spectrum, where the absorption (ω_τ) and emission (ω_t) frequencies are obtained from the Fourier transformation for τ and t , respectively. The two red crosses on the diagonal (dashed line) mirror the two singularities in the 1D spectrum shown above and to the right of the 2D spectrum. The two orange crosses on the off-diagonal are called cross-peaks, revealing the correlations between the two singularities. (c) shows a pulse sequence for the 1D Ramsey scheme, (d) for the 1D spin-echo scheme, and (e) for the 2D spin-echo scheme. (f) and (g) are EXSY pulse schemes with different pulse phases, which we call EXSY+ and EXSY-, respectively.

where $\hat{N} = \sum_{\mathbf{k}} c_{\mathbf{k}}^\dagger c_{\mathbf{k}}$ is the number operator, Z_{FS} is a normalization constant, and k_B is the Boltzman constant. Here, $\mu \simeq E_F$ is the chemical potential determined by number density n , where E_F is the Fermi energy that also gives a typical many-body time-scale $\tau_F = E_F^{-1}$ and momentum scale $k_F = \sqrt{2mE_F}$.

We aim to investigate the dynamics of the system under multiple RF pulses with different time delays in between. As a concrete example, we focus on a three-pulse EXSY+ scheme, as illustrated in Fig. 1 (f). The RF pulses can manipulate the spin-state of the impurity within a much shorter time than the intrinsic time scales of the background fermions τ_F . As a result, one can neglect the evolution of the Fermi sea during the pulse and describe the pulse's effect as a rotation of the impurity's spin state. For example, a pulse that achieves a $\pi/2$ rotation can be defined as

$$R(\pi/2) \equiv \begin{pmatrix} R_{\uparrow\uparrow}^{(\pi/2)} & R_{\uparrow\downarrow}^{(\pi/2)} \\ R_{\downarrow\uparrow}^{(\pi/2)} & R_{\downarrow\downarrow}^{(\pi/2)} \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}. \quad (2)$$

The total time evolution in EXSY+ scheme is thus given by the unitary transformation

$$U(t, T, \tau) = U(t)R(\pi/2)U(T)R(\pi/2)U(\tau)R(\pi/2), \quad (3)$$

where

$$U(t') = \begin{pmatrix} e^{-i\hat{H}_\uparrow t'} & 0 \\ 0 & e^{-i\hat{H}_\downarrow t'} \end{pmatrix} \quad (4)$$

gives the time evolution in between pulses. We denote the initial state as $\rho_i = \rho_{\text{FS}} \otimes |\downarrow\rangle\langle\downarrow|$ and arrive at the final density matrix as $\rho_f = U\rho_i U^\dagger$. We can define a multidimensional response function in the time domain, $S(\tau, T, t)$, by measuring

$$\text{Re}[S(\tau, T, t)] = -\text{Tr}(\sigma_x \rho_f), \quad \text{Im}[S(\tau, T, t)] = -\text{Tr}(\sigma_y \rho_f), \quad (5)$$

where σ_x and σ_y are the usual Pauli matrices in the spin-basis. A tedious but straightforward manipulation of algebra can give a close form

$$S(\tau, T, t) = \sum_{i=1}^{16} S_i(\tau, T, t) \equiv \frac{1}{4} \sum_{i=1}^{16} \text{Tr}[I_i(\tau, T, t) \rho_{\text{FS}}]. \quad (6)$$

Here, $I_i(\tau, T, t)$, which we name as pathways, are a direct product of six operators in the form of $e^{\pm i\hat{H}t}$,

$$I_i(\tau, T, t) = c_{\vec{\sigma}_i} e^{i\mathcal{H}_{\sigma'_{1i}}\tau} e^{i\mathcal{H}_{\sigma'_{2i}}T} e^{i\mathcal{H}_{\uparrow}t} e^{-i\mathcal{H}_{\downarrow}t} e^{-i\mathcal{H}_{\sigma_{2i}}T} e^{-i\mathcal{H}_{\sigma_{1i}}\tau}, \quad (7)$$

where $\vec{\sigma}_i \equiv (\sigma_{1i}, \sigma_{2i}, \sigma'_{1i}, \sigma'_{2i})$ is a collective index that takes sixteen different combinations, and $c_{\vec{\sigma}_i} = -8R_{\uparrow\sigma_{2i}}^{(\pi/2)} R_{\sigma_{2i}\sigma'_{1i}}^{(\pi/2)} R_{\sigma'_{1i}\downarrow}^{(\pi/2)} R_{\downarrow\sigma_{2i}}^{(\pi/2)} R_{\sigma_{2i}\sigma_{1i}}^{(\pi/2)} R_{\sigma_{1i}\downarrow}^{(\pi/2)}$ are coefficients that take values of ± 1 . Here, we have applied the relation $R(\pi/2)^{-1} = R(-\pi/2) = R(\pi/2)^T$ for the derivation, where the superscript T denotes the transpose of a matrix (and should not be confused with mixing

time T). The sorting of the pathways can be arranged arbitrarily for convenience. Here, our first four pathways are chosen as

$$I_1(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (8)$$

$$I_2(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (9)$$

$$I_3(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (10)$$

and

$$I_4(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}. \quad (11)$$

The expressions for other twelve pathways can be found in Appendix A.

The contribution of each pathway, $S_i(\tau, T, t)$, can be calculated exactly via FDA. To proceed, we define $\mathcal{H}_\downarrow \equiv \Gamma(h_\downarrow)$ and $\mathcal{H}_\uparrow \equiv \Gamma(h_\uparrow) + \omega_s$. Here $\Gamma(h) \equiv \sum_{\mathbf{k}, \mathbf{q}} h_{\mathbf{k}\mathbf{q}} c_{\mathbf{k}}^\dagger c_{\mathbf{q}}$ is a bilinear fermionic many-body Hamiltonian in the Fock space, and $h_{\mathbf{k}\mathbf{q}}$ represents the matrix elements of the corresponding operator in the single-particle Hilbert space. These matrix elements are explicitly given by $(h_\downarrow)_{\mathbf{k}\mathbf{q}} = \epsilon_{\mathbf{k}} \delta_{\mathbf{k}\mathbf{q}}$ and $(h_\uparrow)_{\mathbf{k}\mathbf{q}} = \epsilon_{\mathbf{k}} \delta_{\mathbf{k}\mathbf{q}} + \tilde{V}(\mathbf{k} - \mathbf{q})$. With these definitions, we can rewrite

$$S_i(\tau, T, t) = \frac{1}{4} \tilde{S}_i(\tau, T, t) e^{-i\omega_s f_i(t, T, \tau)}, \quad (12)$$

where $e^{-i\omega_s f_i(t, T, \tau)}$ gives a simple phase and $\tilde{S}_i(\tau, T, t)$ is a product of the exponentials of the bilinear fermionic operator, both of which can be calculated exactly. For example, we have $S_1(\tau, T, t) = \tilde{S}_1(\tau, T, t) e^{i\omega_s t} e^{-i\omega_s \tau} / 4$, where

$$\tilde{S}_1(\tau, T, t) = \text{Tr}[e^{i\Gamma(h_\downarrow)\tau} e^{i\Gamma(h_\uparrow)T} e^{i\Gamma(h_\uparrow)t} \times e^{-i\Gamma(h_\downarrow)t} e^{-i\Gamma(h_\uparrow)T} e^{-i\Gamma(h_\uparrow)\tau} \rho_{\text{FS}}]. \quad (13)$$

Applying Levitov's formula gives

$$\tilde{S}_1(\tau, T, t) = \det[(1 - \hat{n}) + R_1(\tau, T, t)\hat{n}], \quad (14)$$

with

$$R_1(\tau, T, t) = e^{ih_\downarrow\tau} e^{ih_\uparrow T} e^{ih_\uparrow t} e^{-ih_\downarrow t} e^{-ih_\uparrow T} e^{-ih_\uparrow\tau}, \quad (15)$$

and $\hat{n} = n_{\mathbf{k}} \delta_{\mathbf{k}\mathbf{k}'}$, where $n_{\mathbf{k}} = 1/(e^{\epsilon_{\mathbf{k}}/k_B T^\circ} + 1)$ denotes the single-particle occupation number operator. Calculations of other pathway contributions are similar, which are presented in Appendix A.

Numerical calculations are carried out in a finite system confined in a sphere of radius R . Keeping the density constant, we increase R towards infinity until numerical results are converged. Typically, we choose $k_F R = 250\pi$ in a calculation. We focus on the s -wave interaction channel between $|\downarrow\rangle$ and the background fermions near

a broad Feshbach resonance, which can be well mimicked by a spherically symmetric and short-range van-der-Waals type potential $V(r) = -C_6 \exp(-r^6/r_0^6)/r^6$. Here, C_6 determines the van-der-Waals length $l_{\text{vdW}} = (2mC_6)^{1/4}/2$, and we choose $l_{\text{vdW}} k_F = 0.01 \ll 1$, so the short-range details are unimportant. The low-temperature many-body physics can be determined by the s -wave energy-dependent scattering length $a(E_F) = -\tan \eta(k_F)/k_F$ at the Fermi energy E_F , with $\eta(E_F)$ being an energy-dependent s -wave scattering phase-shift tuned by adjusting r_0 . For the simplicity of notation, we denote $a \equiv a(E_F)$ hereafter. Consequently, $\tilde{S}_i(t, T, \tau)$ is a universal function of $k_B T^\circ/E_F$, $k_F a$, t/τ_F , and τ/τ_F in the whole time domain.

A summation of the contributions of all pathways gives the total response $S(t, T, \tau)$, and the spectrum in the frequency domain can be obtained via a double Fourier transformation

$$A(\omega_\tau, T, \omega_t) = \frac{1}{\pi^2} \int_0^\infty \int_0^\infty dt d\tau e^{i\omega_\tau \tau} S(\tau, T, t) e^{-i\omega_t t}, \quad (16)$$

where ω_t and ω_τ are interpreted as an absorption and emission frequency, respectively. On the other hand, the T -dependence of $A(\omega_\tau, T, \omega_t)$ can reveal the many-body coherent and incoherent dynamics. We notice that $A(\omega_\tau, T, \omega_t) = \sum_{i=1}^{16} A_i(\omega_\tau, T, \omega_t)$ can also be expressed as a summation of sixteen pathways, where the expression of each pathway is given by Eq. (16), with A and S replaced by A_i and S_i , respectively.

We emphasize that the MD spectroscopy contains all the information of 1D spectroscopy. For example, one can examine the $T = t = 0$ case, where the pulse scheme becomes the same as the original 1D Ramsey scheme shown in Fig. 1 (f). In this case, $S(\tau, T = 0, t = 0)$ reduces to the 1D Ramsey response function $S_a(\tau) = \text{Tr}(e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{-i\hat{\mathcal{H}}_\uparrow\tau} \rho_{\text{FS}})$, which is also called the time-dependent overlap function. Similarly, we have $S_e(t) \equiv S(\tau = 0, T = 0, t) = S_a^*(t)$, where the superscript $*$ denotes complex conjugate. Correspondingly, we have $\int d\omega_t A(\omega_\tau, T = 0, \omega_t) = A_a(\omega_\tau)$, where $A_a(\omega_\tau) = \int d\tau S_a(\tau) e^{i\omega_\tau \tau} / \pi$ is the 1D absorption spectrum. Similarly, we have $A_e(\omega_t) = \int d\omega_\tau A(\omega_\tau, T = 0, \omega_t) = A_a^*(\omega_t)$. Since $A_a(\omega_\tau)$ is the absorption spectrum, its complex conjugate $A_e(\omega_t)$ can thus be interpreted as an emission spectrum. These interpretations are consistent with the fact that the integration of $A(\omega_\tau, T = 0, \omega_t)$ over the emission frequency ω_t gives the 1D absorption spectrum $A_a(\omega_\tau)$ and vice versa. The physical process underlying $A(\omega_\tau, T, \omega_t)$ can be interpreted as follows: the system first gets excited by absorbing a photon with frequency ω_τ , after a period of mixing time T , and then emits a photon with frequency ω_t .

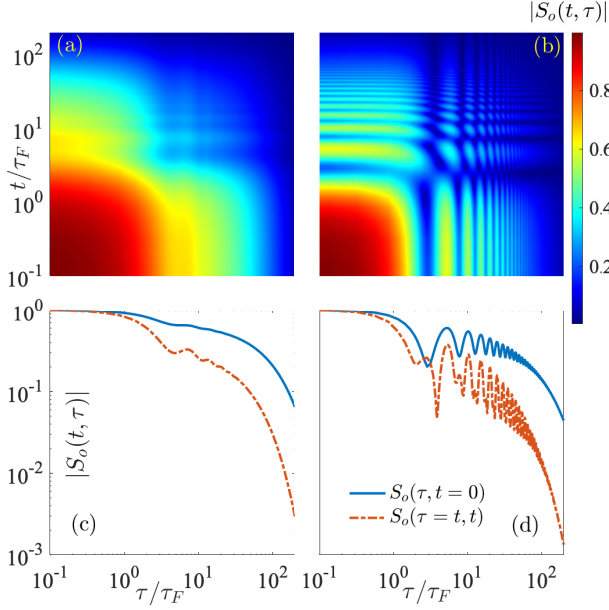


FIG. 2. Universal 2D spin-echo response's amplitude $|S_o(\tau, t)|$, as a function of τ and t , for (a) attractive interaction $k_F a = -0.5$ and (b) repulsive interaction $k_F a = 0.5$. The temperature is set as $k_B T^\circ = 0.03 E_F$. (c) shows $|S_o(\tau, t=0)|$ [the slice of (a) along the x -axis] and $|S_o(\tau, t=\tau)|$ [the slice of (a) along diagonal] as a function of τ in the blue solid and red dash-dotted curves, respectively. (d) shows the same slices for (b). Fig. 2 (c) indicates that $S_o(\tau, t=0)$ and $S_o(\tau, t=\tau)$ reduce to 1D Ramsey response $S_a(\tau)$ and 1D spin-echo signal $S_o(\tau)$, respectively.

III. RESULTS

A. Two-dimensional spin-echo response

Let us first investigate a relatively simple situation, $T = 0$, which is equivalent to the two-pulse scheme illustrated in Fig. 1 (e). The response function in the time domain is given by

$$S_o(\tau, t) \equiv S(\tau, T = 0, t) = \text{Tr}[I_o(\tau, t)\rho_{\text{FS}}], \quad (17)$$

where $I_o(\tau, t) = e^{i\mathcal{H}_\downarrow\tau} e^{i\mathcal{H}_\uparrow t} e^{-i\mathcal{H}_\downarrow t} e^{-i\mathcal{H}_\uparrow\tau}$. We notice that when $t = \tau$, the scheme is equivalent to 1D spin-echo scheme investigated in Refs. [70, 71] and illustrated in Fig. 1 (d), hence naming our scheme as a 2D spin echo scheme. We can examine that $S_o(t, t)$ reduce to the 1D spin-echo response $S_o(t) = \text{Tr}[e^{i\mathcal{H}_\downarrow t} e^{i\mathcal{H}_\uparrow t} e^{-i\mathcal{H}_\downarrow t} e^{-i\mathcal{H}_\uparrow t} \rho_{\text{FS}}]$.

While we can also calculate $S_o(\tau, t)$ by using $S(\tau, T, t)$ in Eq. (6) with $T = 0$, a direct calculation of Eq. (17) is more convenient. The expression of the 2D spin-echo response can be written as $S_o(\tau, t) = e^{i\omega_s t} \tilde{S}_o(\tau, t) e^{-i\omega_s \tau}$, where

$$\tilde{S}_o(\tau, t) = \text{Tr}[e^{i\Gamma(h_\downarrow)\tau} e^{i\Gamma(h_\uparrow)t} e^{-i\Gamma(h_\downarrow)t} e^{-i\Gamma(h_\uparrow)\tau} \rho_{\text{FS}}] \quad (18)$$

can be calculated exactly by applying Levitov's formula in the FDA

$$\tilde{S}_o(\tau, t) = \det[(1 - \hat{n}) + R_o(\tau, t)\hat{n}] \quad (19)$$

with

$$R_o(\tau, t) = e^{ih_\downarrow\tau} e^{ih_\uparrow t} e^{-ih_\downarrow t} e^{-ih_\uparrow\tau}. \quad (20)$$

Examples of this universal 2D response function $|S_o(t, \tau)| = |\tilde{S}_o(t, \tau)|$ with parameters $k_F a = -0.5$ and $k_F a = 0.5$ at a finite temperature $k_B T^\circ = 0.03 E_F$ are shown in Fig. 2 (a) and (b), respectively. The solid and dash-dotted curves in Fig. 2 (c) show $|S_o(\tau, t=0)|$ and $|S_o(\tau, t=\tau)|$ as a function of τ , i.e., the slice of (a) along the x -axis and diagonal, respectively. Figure 2 (d) shows the same slices for (b). Fig. 2 (c) indicates that $S_o(\tau, t=0)$ and $S_o(\tau, t=\tau)$ reduce to 1D Ramsey response $S_a(\tau)$ and 1D spin-echo signal $S_o(\tau)$, respectively.

B. 2D spin-echo spectrum

The 2D spin-echo spectrum in the frequency domain can be obtained by applying a double Fourier transformation, Eq. (16), with the relation $A_o(\omega_\tau, \omega_t) = A(\omega_\tau, T = 0, \omega_t)$. One can immediately observe that $A_o(\omega_\tau, \omega_t) = \int_0^\infty \int_0^\infty dt d\tau e^{i\tilde{\omega}_\tau \tau} \tilde{S}_o(\tau, t) e^{-i\tilde{\omega}_t t} / \pi^2$ with $\tilde{\omega}_\tau = \omega_\tau - \omega_s$ and $\tilde{\omega}_t = \omega_t - \omega_s$, i.e., the energy differences between two spin-states only give simple shifts of frequencies. Hereafter, unless specified otherwise, we denote $\tilde{\omega} = \omega - \omega_s$ for any frequency variable ω . Figure 3 shows our finite temperature ($k_B T^\circ = 0.03 E_F$) results for attractive $k_F a = -0.5$ and repulsive $k_F a = 0.5$ interactions in (a1)-(a4) and (b1)-(b4), respectively. We present the 2D contour of $\text{Re}[A_o(\omega_\tau, \omega_t)]$ as a function of $\tilde{\omega}_\tau$ and $\tilde{\omega}_t$ in Figs. 3 (a2) and (b2) and the corresponding 3D landscape in Figs. 3 (a3) and (b3). Here, we denote $\text{Re}[\mathcal{C}]$ and $\text{Im}[\mathcal{C}]$ as the real and imaginary parts of $\mathcal{C}$, respectively. For comparison, we also show the 1D spectra $\text{Re}[A_a(\omega_\tau)]$ in Figs. 3 (a1) and (b1) and $\text{Re}[A_e(\omega_t)] = \text{Re}[A_a(\omega_\tau)]$ in Figs. 3 (a4) and (b4). For completeness, we also show the imaginary part and amplitude of the corresponding 2D spectroscopy in Fig. 7 in Appendix B.

The absorption spectrum has been well studied before [70, 71]. When the interaction is attractive ($k_F a < 0$), only one power-law singularity $\text{Re}[A_a(\omega_\tau)] \sim \theta(\omega_\tau - \omega_{A-}) |\omega_\tau - \omega_{A-}|^{-a_{A-}}$ with exponent coefficient $a_{A-} > 0$ appears near ω_{A-} with a slight thermal broadening, as shown in Figs. 3 (a1) and (a4). In contrast, Figs. 3 (b1) and (b4) show two singularities for a repulsive interaction ($k_F a > 0$). These singularities are understood as manifestations of the well-known Anderson's orthogonality catastrophe (OC). Due to the existence of multiple particle-hole excitations of the background Fermi sea induced by the infinitely massive impurity, the many-particle states with and without impurity interactions

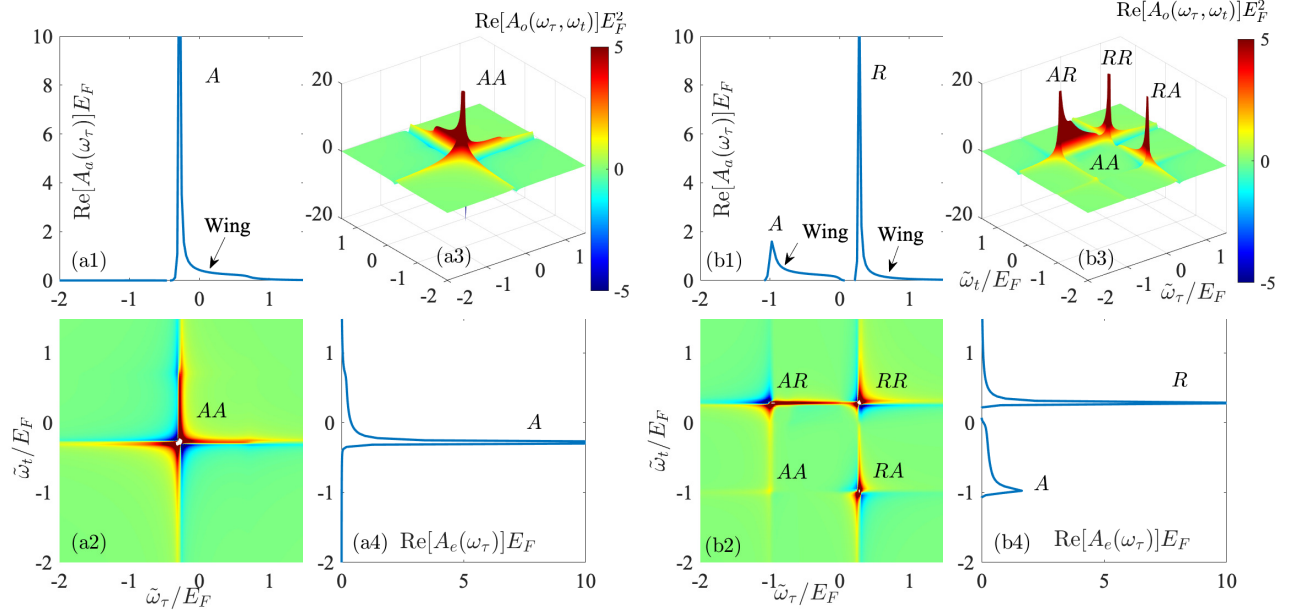


FIG. 3. (a1) and (a4) shows the 1D absorption spectrum for attractive interaction $k_F a = -0.05$ and finite temperature $k_B T^\circ = 0.03 E_F$. The absorption singularity is denoted as A . (a2), and (a3) shows the contour and 3D landscape of the 2D spin-echo spectrum $\text{Re}[A_o(\omega_\tau, \omega_t)]$, where the diagonal peak is denoted as AA . (b1)-(b4) are the same as (a1)-(a4), correspondingly, but for repulsive interaction $k_F a = 0.5$. There are two singularities in (b1), the absorption spectrum, namely repulsive and attractive singularities, which are denoted as R and A . The corresponding diagonal peaks in (b2) and (b3) are denoted as AA and RR , while the off-diagonal cross-peaks are denoted as AR and RA .

are orthogonal, which leads to a vanishing quasiparticle residue. Our recent studies further examined the scenario where a mechanism, such as a superfluid gap or finite impurity mass, suppresses those multiple particle-hole excitations [73, 74]. In this case, OC can be prevented, and the singularities reduce back to the so-called attractive or repulsive Fermi polarons. At the same time, the “wings” attached to the singularity, indicated in Figs. 3(a1) and (b1), separate from the polaron signal and reduce to the so-called molecule-hole continuums. Because of their close relations to polaron resonances, we name these singularities as attractive and repulsive singularities and denote them by A and R , respectively, in Figs. 3 (a1), (a4), (b1), and (b4).

The 2D spectrum in Figs. 3 (a2) and (a3) shows a double dispersion lineshape commonly found in 2D NMR around $(\tilde{\omega}_\tau, \tilde{\omega}_t) \approx (\tilde{\omega}_{A-}, \tilde{\omega}_{A-})$, which is called a diagonal peak denoted as AA . For attractive interaction $k_F a = -0.5$, the attractive singularity appears at $\tilde{\omega}_{A-} \approx -0.28 E_F$ in the absorption spectrum. We have numerically verified that the integration of 2D spectroscopy over emission frequency ω_t gives the 1D absorption spectrum $A_a(\omega_\tau)$ (not shown here). Interestingly, we can observe that there is no diagonal spectral weight corresponding to the wing. Rather, the spectral weight on the off-diagonal $A_o(\omega_\tau, \omega_t \approx \omega_{A-})$ and $A_o(\omega_\tau \approx \omega_{A-}, \omega_t)$ is significant and resembles the lineshape of the wing. This is a non-trivial manifestation of OC in the 2D spectroscopy: the inhomogeneous and homogeneous line-

shape does not have the OC characteristic. Here, the inhomogeneous and homogeneous lineshape refer to the lineshape near a singularity along the diagonal or the direction perpendicular to the diagonal, which is better illustrated in the amplitude of 2D spectroscopy shown in Fig. 7 (c) in Appendix B. As we can observe, the widths of the singularity are much sharper along these two directions, which might help experimental identification of the singularity, especially at finite temperatures. The homogeneous and inhomogeneous broadenings in MD spectroscopy also have their own experimental significance, similar to their NMR or optical counterpart. In a realistic experiment, the ensemble average of the impurity signal can give rise to a further inhomogeneous broadening induced by the disorder of the local environment (such as spatial magnetic field fluctuation). However, these disorders are usually non-correlated and would not introduce homogeneous broadening [6, 15, 16].

For repulsive interaction $k_F a = 0.5$, there are two singularities, the attractive and repulsive singularities, in the 1D absorption spectrum. These singularities appear at $\tilde{\omega}_{A+} \approx -0.98 E_F$ and $\tilde{\omega}_{R+} \approx 0.28 E_F$ in Figs. 3 (b1) and (b4). As shown in Fig. 3 (b2) and (b3), there are two diagonal peaks, AA and RR , in the 2D spectroscopy that mirror the attractive and repulsive singularities. In addition, there are also two significant cross-peaks, AR and RA , which indicate a strong many-body quantum correlation between the attractive and repulsive singularity. As far as we know, this is the first prediction

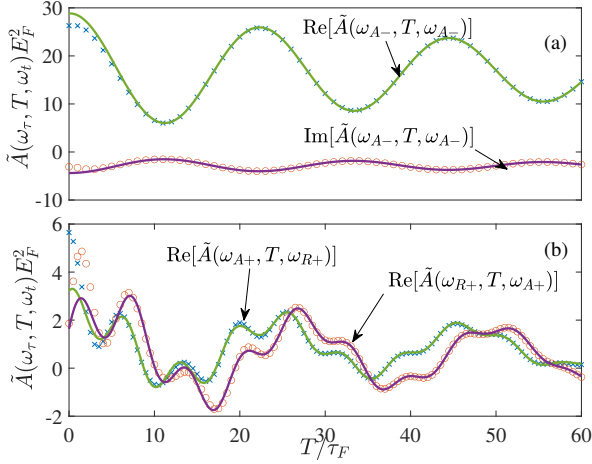


FIG. 4. T -dependence of EXSY+ spectrum $\tilde{A}(\omega_\tau, T, \omega_t)$ for (a) attractive interaction $k_F a = -0.5$ and (b) repulsive interaction $k_F a = 0.5$, with temperature $k_B T^\circ = 0.03 E_F$. The cross and circle symbols denote the real and imaginary parts for the diagonal peak $\tilde{A}(\omega_\tau = \omega_{A-}, T, \omega_t = \omega_{A-})$ in (a) and the real part of cross-peaks $\tilde{A}(\omega_\tau = \omega_{A+}, T, \omega_t = \omega_{R+})$ and $\tilde{A}(\omega_\tau = \omega_{R+}, T, \omega_t = \omega_{A+})$ in (b). The curves use the fitting formula in the main text to fit numerical results at the late time $T \geq 10\tau_F$. We, however, also show the fitting curve at $T < 10\tau_F$ to emphasize the differences in the early time behaviors.

of many-body correlations between Fermi singularities in cold atom systems. If the impurity has a finite mass or the background Fermi gas is replaced by a superfluid with an excitation gap, we believe these cross-peaks would remain and represent the correlations between attractive and repulsive polarons.

C. 2D EXSY spectrum

In this section, we focus on the 2D spectrum, $A(\omega_\tau, T, \omega_t)$, of the EXSY+ pulse scheme illustrated by Fig. 1 (f), which can be exactly calculated by Eqs. (6), (12) and (16) with the FDA. As mentioned above, in a 2D spin-echo spectrum (and the 1D spectra), the trivial energy difference between $|\downarrow\rangle$ and $|\uparrow\rangle$, ω_s , only introduces a frequency shift of the spectra as $(\omega_\tau, \omega_t) \rightarrow (\tilde{\omega}_\tau, \tilde{\omega}_t)$. In contrast, the scenario is a bit more complicated for the EXSY+ spectrum, where the spectrum can be expressed as a summation of sixteen pathway contributions, and each pathway is associated with a different phase $e^{-i\omega_s f_i(t, T, \tau)}$ in Eq. (12) (see Appendix A for details). Consequently, each pathway contribution $A_i(\omega_\tau, T, \omega_t)$ has shifted to different centers in the frequency domain accordingly. The features of Fermi singularities, in general, lie within a frequency range of a few Fermi energy E_F around $(\tilde{\omega}_\tau, \tilde{\omega}_t) = (0, 0)$ in the 2D spectrum $A(\omega_\tau, T, \omega_t)$. In addition, for a typical ultracold experiment, ω_s is usually much larger than the Fermi energy

E_F . As a result, only the first four pathways associated with $e^{i\omega_s t} e^{-i\omega_s \tau}$ would give a non-negligible contribution, i.e., $A(\omega_\tau, T, \omega_t) \approx \tilde{A}(\omega_\tau, T, \omega_t) = \sum_{i=1}^4 A_i(\omega_\tau, T, \omega_t)$, within the frequency range in interest. A comparison between $A(\omega_\tau, T, \omega_t)$ and $\tilde{A}(\omega_\tau, T, \omega_t)$ for $\omega_s/E_F = 2\pi$ is shown in Appendix A, where perfect agreement is observed. Such reduction of pathways not only allows a faster calculation but also helps us to identify the important pathways and further separate them using a so-called “phase cycling” technique detailed in the next section.

We find that the general landscape of the 2D spectrum $\tilde{A}(\omega_\tau, T, \omega_t)$ also shows strong off-diagonal contributions and cross-peaks (see Fig. 6, for example), similar to the 2D spin-echo spectrum $A_o(\omega_\tau, \omega_t)$. However, the dependency of $\tilde{A}(\omega_\tau, T, \omega_t)$ on the mixing time T can give us further information on the many-body coherent and incoherent dynamics. There is one additional complication, though: we observe a fast oscillation with frequency ω_s in the T -dependency of $\tilde{A}(\omega_\tau, T, \omega_t)$, which originates from the interferences between the contribution of I_3 and I_4 , that is proportional to $e^{-i\omega_s T}$ and $e^{i\omega_s T}$ respectively. We are not interested in this trivial oscillation. Instead, we would like to investigate the dynamic in the time scale of τ_F and choose to study the signals at $T_M \omega_s = 2\pi M$, where M is an integer. Notice that since $\omega_s \gg E_F$, T_M/τ_F can be considered to be almost continuous. As we will see later, this choice of T_M is not necessary if we apply the phase cycling to separate the pathways.

Figure 4 (a) shows the real and imaginary part of $A(\omega_\tau \approx \omega_{A-}, T, \omega_t \approx \omega_{A-})$ for $k_F a = -0.5$ as cross and circle symbols, showing a damping oscillation behavior at a late time. This long-time behavior can be fitted perfectly with a formula $F(T) = A_a \cos(\omega_a T + \varphi_a) \exp(-T/T_a) + B$ for the real and imaginary parts separately, both of which give $\omega_a \approx 0.28 E_F$ and $T_a \approx 80\tau_F$. We also find this damping oscillation behavior with the same ω_a and T_a at other parts of the spectrum. One can recognize $\omega_a \approx |\omega_{A-}|$ and the damping lifetime T_a reflects a non-coherent many-body dynamic, which might be related to the finite temperature $k_B T^\circ \approx 0.03 E_F$. Figure 4 (b) shows the T -dependence of $\text{Re}[\tilde{A}(\omega_\tau, T, \omega_t)]$ with $k_F a = 0.5$ for AR and RA cross-peaks as cross and circle symbols, which can be fitted by a combination of two damping oscillations $F(T) = A_a \cos(\omega_a T + \varphi_a) \exp(-T/T_a) + A_r \cos(\omega_r T + \varphi_r) \exp(-T/T_r) + B$ illustrated by the solid curves. The numerical fitting gives $\omega_a \approx |\omega_{A+}|$, $\omega_r \approx |\omega_{R+}|$, $T_a \approx 30\tau_F$ and $T_r \approx 80\tau_F$. These long-time damping oscillations indicate the non-trivial relaxation process during the mixing time T , induced by multiple particle-hole excitations. However, at a very early time, probably only a few particle-hole pairs have been excited, and higher order correlation has not been built up, which explains the deviation between the fitting results and numerical calculation. It is also interesting to notice that T_a and T_r are different, which implies there might be an intrinsic dynamical process between the attractive and repulsive singularities.

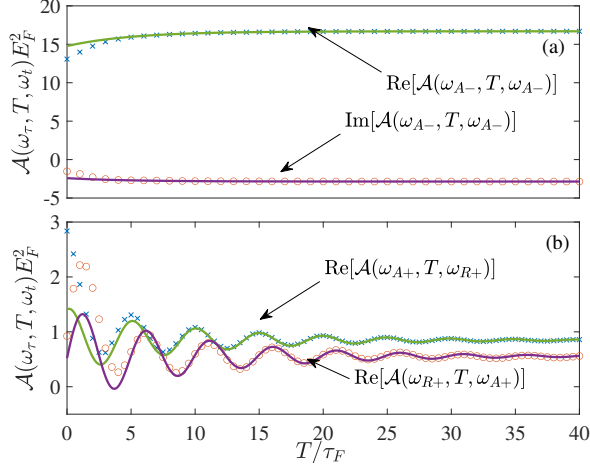


FIG. 5. The same as Fig. 4, but for a phase cycling spectrum $\mathcal{A}(\omega_\tau, T, \omega_t)$.

D. Phase cycling

Since the total spectrum is a summation of multiple pathway contributions, it is sometimes important to be able to separate and measure one or some of the pathway contributions. For example, we notice that the third and fourth pathways, $I_3(\tau, T, t) = \dots e^{i\mathcal{H}_\downarrow T} \dots e^{-i\mathcal{H}_\uparrow T} \dots$ and $I_4(\tau, T, t) = \dots e^{i\mathcal{H}_\uparrow T} \dots e^{-i\mathcal{H}_\downarrow T} \dots$, are the ones responsible for a fast oscillation in the mixing time T with the trivial frequency ω_s . In addition, we have $I_3(\tau, T, t) = I_o(\tau + T, t)$ and $I_4(\tau, T, t) = I_o(\tau, T + t)$, implying these two pathways give the same information as the 2D spin-echo sequence. Therefore, it would be interesting to be able to eliminate the contributions of I_3 and I_4 , which can be achieved by following the same spirit as phase cycling, an important technique in NMR.

Phase cycling is a technique that uses a linear combination of signals (with possibly different weights) from different pulse schemes to select the contribution of one or few coherent pathways. As a concrete example, we define $\mathcal{A}(\omega_\tau, T, \omega_t) \equiv \tilde{A}(\omega_\tau, T, \omega_t) - \tilde{A}^-(\omega_\tau, T, \omega_t)$, where $\tilde{A}^-(\omega_\tau, T, \omega_t)$ is the 2D Ramsey response for the EXSY-pulse scheme indicated in Fig. 1 (g). A manipulation of algebra gives (see Appendix A)

$$\mathcal{A}(\omega_\tau, T, \omega_t) = 2A_1(\omega_\tau, T, \omega_t) + 2A_2(\omega_\tau, T, \omega_t), \quad (21)$$

which only includes the first two pathways. One can immediately notice that both pathways, $I_1(\tau, T, t)$ and $I_2(\tau, T, t)$, are independent of ω_s , where we are no longer restricted to measuring signals at $T = T_M$.

The corresponding double Fourier transformation $\mathcal{A}(\omega_\tau, T, \omega_t)$ is studied in Fig. 5. For the attractive ($k_F a = -0.5$) interaction case shown in Fig. 5 (a), a numerical fitting with formula $F(T) = A \exp(-T/T_a) + B$ indicates a pure exponential relaxation with lifetime $T_a \approx 5\tau_F$. On the contrary, for the repulsive ($k_F a = +0.5$)

interaction case shown in Fig. 5 (a), a damping oscillation $F(T) = A_\Delta \cos(\omega_\Delta T_\Delta + \varphi_\Delta) \exp(-T/T_\Delta) + B_\Delta$ of $\omega_\Delta \approx |\omega_{R+} - \omega_{A+}|$ and damping lifetime $T_\Delta \approx 10\tau_F$. This damping oscillation indicates the intrinsic coherent and incoherent many-body dynamics between the attractive and repulsive Fermi singularity. We notice that a similar damping oscillation between exciton-polarons in TMDs has previously been observed in the non-rephasing signal of an optical 2D spectroscopy [16] and explained by the nonlinear Golden Rule [17]. More recently, this behavior has also been observed in a FDA study [68]. To our knowledge, our result is the first prediction of the coherent and incoherent dynamic process between the two Fermi-edge singularities in ultracold gases. We also believe the same procedure can be applied to study the many-body dynamical process between attractive and repulsive polarons.

IV. CONCLUSION

In summary, we have investigated how to extend the 1D Ramsey spectrum to multidimensional, which goes beyond the linear response regime and can reveal correlations between many-body singularities and resonances. Multidimensional spectroscopy also allows us to investigate the many-body coherent and relaxation dynamics that are not accessible in 1D spectra. Such a scheme is especially suitable and accessible in the clean and controllable systems of ultracold gases.

As a concrete example, we investigate the Fermi singularity problem and present a numerical exact many-body formalism for the simulation of the multidimensional Ramsey spectrum of a heavy impurity in a Fermi gas, both in the time domain and frequency domain. We believe this is the first investigation of the nonlinear responses in such systems and the first prediction of many-body correlations between attractive and repulsive singularities, remnants of polaron resonances destroyed by Anderson's orthogonal catastrophe. For the first time, we also predict the many-body coherent dynamic and relaxation between the two Fermi singularities.

We believe these many-body correlations and dynamics should also exist between attractive and repulsive polarons, which can be calculated exactly if the background gas is a Bardeen-Cooper-Schrieffer superfluid [73, 74]. Another approach would be to investigate mobile impurity with a Chevy ansatz. Although this is an approximated approach, it might lead to intuitive understanding. Finally, we argue that the application of multidimensional Ramsey spectroscopy should not be limited to impurity systems, and the same spirit can be generalized to other ultracold atom systems [79–90].

V. ACKNOWLEDGMENTS

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Appendix A: Pathway contributions

In the main text, Eq. (6) indicates that $S(\tau, T, t)$ can be written as a summation of sixteen different pathway contributions $S(\tau, T, t) = \sum_{i=1}^{16} S_i(\tau, T, t)$ where $S_i(\tau, T, t) = \text{Tr}[I_i(\tau, T, t)\rho_{\text{FS}}]/4$. The sixteen pathways $I_i(\tau, T, t)$ can be written out explicitly:

$$I_1(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1a})$$

$$I_2(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1b})$$

$$I_3(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1c})$$

$$I_4(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}. \quad (\text{A1d})$$

$$I_5(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\downarrow\tau}, \quad (\text{A1e})$$

$$I_6(\tau, T, t) = -e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\downarrow\tau}, \quad (\text{A1f})$$

$$I_7(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\downarrow\tau}, \quad (\text{A1g})$$

$$I_8(\tau, T, t) = -e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\downarrow\tau}, \quad (\text{A1h})$$

$$I_9(\tau, T, t) = e^{i\hat{\mathcal{H}}_\uparrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1i})$$

$$I_{10}(\tau, T, t) = -e^{i\hat{\mathcal{H}}_\uparrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1j})$$

$$I_{11}(\tau, T, t) = -e^{i\hat{\mathcal{H}}_\uparrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1k})$$

$$I_{12}(\tau, T, t) = e^{i\hat{\mathcal{H}}_\uparrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1l})$$

$$I_{13}(\tau, T, t) = e^{i\hat{\mathcal{H}}_\uparrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\uparrow\tau}, \quad (\text{A1m})$$

$$I_{14}(\tau, T, t) = e^{i\hat{\mathcal{H}}_\downarrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\downarrow\tau}, \quad (\text{A1n})$$

$$I_{15}(\tau, T, t) = -e^{i\hat{\mathcal{H}}_\uparrow\tau} e^{i\hat{\mathcal{H}}_\downarrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\uparrow T} e^{-i\hat{\mathcal{H}}_\downarrow\tau}, \quad (\text{A1o})$$

$$I_{16}(\tau, T, t) = -e^{i\hat{\mathcal{H}}_\uparrow\tau} e^{i\hat{\mathcal{H}}_\uparrow T} e^{i\hat{\mathcal{H}}_\uparrow t} e^{-i\hat{\mathcal{H}}_\downarrow t} e^{-i\hat{\mathcal{H}}_\downarrow T} e^{-i\hat{\mathcal{H}}_\downarrow\tau}. \quad (\text{A1p})$$

Since the dimensions of the many-body operator $\mathcal{H}_\downarrow$ and $\mathcal{H}_\uparrow$ grows exponentially with respect to the number of particle N , a direct calculation of $S_i(\tau, T, t)$ is not accessible. However, by applying Levitov's formula in FDA, we can show that $S_i(\tau, T, t)$ reduces to a determinant in a single-particle Hilbert space that grows only linearly to N , allowing an in-principle exact calculation. For this purpose, we rewrite $\mathcal{H}_\downarrow \equiv \Gamma(h_\downarrow)$ and $\mathcal{H}_\uparrow \equiv \Gamma(h_\uparrow) + \omega_s$, where $\Gamma(h) \equiv \sum_{\mathbf{k}, \mathbf{q}} h_{\mathbf{k}\mathbf{q}} c_{\mathbf{k}}^\dagger c_{\mathbf{q}}$ is a bilinear fermionic many-body operator, and $h_{\mathbf{k}\mathbf{q}}$ represents the matrix elements corresponding to the single-particle operator. As a result, each pathway's contribution can be written as $S_i(\tau, T, t) = \tilde{S}_i(\tau, T, t) e^{-i\omega_s f_i(\tau, T, t)}/4$, where $\tilde{S}_i(\tau, T, t) = \text{Tr}[\tilde{I}_i(\tau, T, t)\rho_{\text{FS}}]$. Here, $\tilde{I}_i(\tau, T, t)$ has the same expression as $I_i(\tau, T, t)$ but with $\mathcal{H}_\downarrow$ and $\mathcal{H}_\uparrow$ replaced by $\Gamma(h_\downarrow)$ and $\Gamma(h_\uparrow)$, respectively. Since both $\Gamma(h_\downarrow)$ and $\Gamma(h_\uparrow)$ are fermionic bilinear operators, applying Levitov's formula gives

$$\tilde{S}_i(\tau, T, t) = \det[(1 - \hat{n}) + R_i(\tau, T, t)\hat{n}], \quad (\text{A2})$$

where $R_i(\tau, T, t)$ has the same expression as $I_i(\tau, T, t)$ but with $\mathcal{H}_\downarrow$ and $\mathcal{H}_\uparrow$ replaced by $h_\downarrow$ and $h_\uparrow$, respectively. The phase factor $e^{-i\omega_s f_i(\tau, T, t)}$ can also be obtained by replacing $e^{\pm i\mathcal{H}_\downarrow t'}$ and $e^{\pm i\mathcal{H}_\uparrow t'}$ with 1 and $e^{\pm i\omega_s t'}$ in the expression of $I_i(\tau, T, t)$, where t' can be τ , T , or t . This expression is now ready for numerical calculation as mentioned in the main text.

While $\tilde{S}_i(\tau, T, t)$ are universal functions of $k_B T^\circ/E_F$, $k_F a$, t/τ_F , and τ/τ_F , the total response $S(\tau, T, t)$ involves the interference of phase factors $e^{-i\omega_s f_i(\tau, T, t)}$ between each contribution. The resulting oscillation in frequency ω_s is not very interesting. Nevertheless, we find that only a few pathways contribute to the singularities of $A(\omega_\tau, T, \omega_t)$ we are interested in. Our numerical results show that $\tilde{S}_i(\tau, T, t)$ oscillates at frequency $\sim E_F$, much slower than $\omega_s \gg E_F$ in usual ultracold experiments. Consequently, if we focus on the Fermi singularity features that appear at $|\omega_\tau - \omega_s|, |\omega_t - \omega_s| \sim E_F$ in the 2D spectrum $A(\omega_\tau, T, \omega_t)$, only the pathways I_1 , I_2 , I_3 , and I_4 associated with the phase factor $e^{i\omega_s t} e^{-i\omega_s \tau}$ contribute. The phase $e^{i\omega_s t} e^{-i\omega_s \tau}$ only gives rise to a simple frequency shift of $\omega_t \rightarrow \tilde{\omega}_t$ and $\omega_\tau \rightarrow \tilde{\omega}_\tau$. Figure 6 compares $A(\omega_\tau, T, \omega_t)$ and $\tilde{A}(\omega_\tau, T, \omega_t) = \sum_{i=1}^4 A_i(\omega_\tau, T, \omega_t)$ that only includes contributions from the first four pathways, for a set of chosen parameters: $k_F a = 0.5$, $k_B T^\circ = 0.03 E_F$, $\omega_s/E_F = 2\pi$, and $T = 5\tau_F$. Figure 6 (a) and (b) shows the whole 2D spectrum for $A(\omega_\tau, T, \omega_t)$

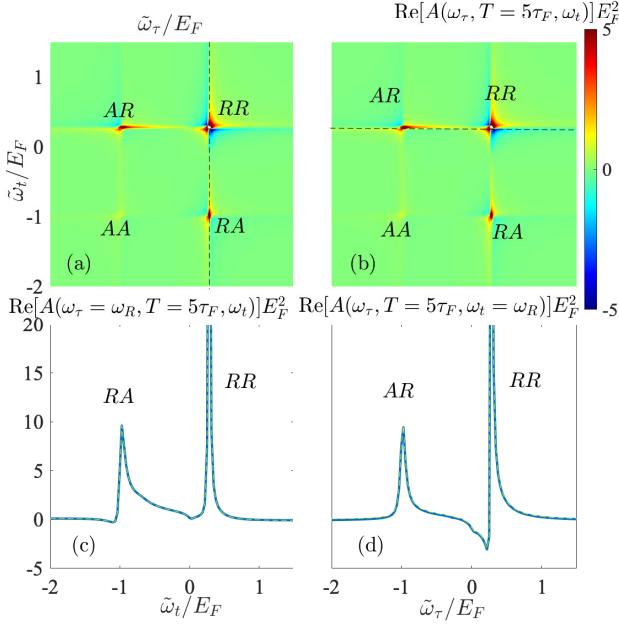


FIG. 6. (a) shows the 2D spectroscopy $A(\omega_\tau, T, \omega_t)$ that includes pathway contributions from all sixteen pathways, and (b) $\tilde{A}(\omega_\tau, T, \omega_t)$ only includes the first four. Solid and dashed curves in (c) compare $A(\omega_\tau = \omega_R, T, \omega_t)$ [the slice along the dashed line in (a)] and $\tilde{A}(\omega_\tau = \omega_R, T, \omega_t)$, respectively, and show excellent agreement by an essentially overlapping. (d) shows the same comparison for $A(\omega_\tau, T, \omega_t = \omega_R)$ and $\tilde{A}(\omega_\tau, T, \omega_t = \omega_R)$ [the slice along the dashed line in (b)]. Other parameters are $k_F a = 0.5$, $k_B T^\circ = 0.03 E_F$, $\omega_s/E_F = 2\pi$, and $T = 5\tau_F$.

and $\mathcal{A}(\omega_\tau, T, \omega_t)$, respectively. The solid and dashed curves in Fig. 6 (c) show the spectrum $A(\omega_\tau, T, \omega_t)$ and $\tilde{A}(\omega_\tau, T, \omega_t)$ along $\omega_\tau = \omega_R$, the slice indicated by the dashed line in Fig. 6 (a). Fig. 6 (d) shows the same comparison for $\omega_t = \omega_R$, the slice indicated by the dashed line in Fig. 6 (b). All comparison gives perfect agreement, e.g., the solid and dashed curves essentially overlap in Fig. 6 (c) and (d).

Among the first four pathways, there is still a phase dependence on $\omega_s T$. To be specific, while $A_1(\omega_\tau, T, \omega_t)$ and $A_2(\omega_\tau, T, \omega_t)$ are independent of ω_s , $A_3(\omega_\tau, T, \omega_t)$ and $A_4(\omega_\tau, T, \omega_t)$ has a phase dependence of $e^{-i\omega_s T}$ and $e^{i\omega_s T}$, respectively. To investigate the dynamics in the time scale of τ_F , we can study the signals at $T_M \omega_s = 2\pi M$, where M is an integer. Notice that since $\omega_s \gg E_F$, T_M/τ_F can be considered to be almost continuous. Such T -dependence is shown in Fig. 4 in the main text.

Another way to observe the T -dependence that is independent of ω_s is by using the so-call phase cycling, which uses a linear combination of results from different pulse schemes to eliminate some of the pathway contributions. For example, in the EXSY- pulse scheme illustrated in Fig. 1 (g), we can carry out the same calculation as

EXSY+ with the middle pulse replaced by a $-\pi/2$ rotation

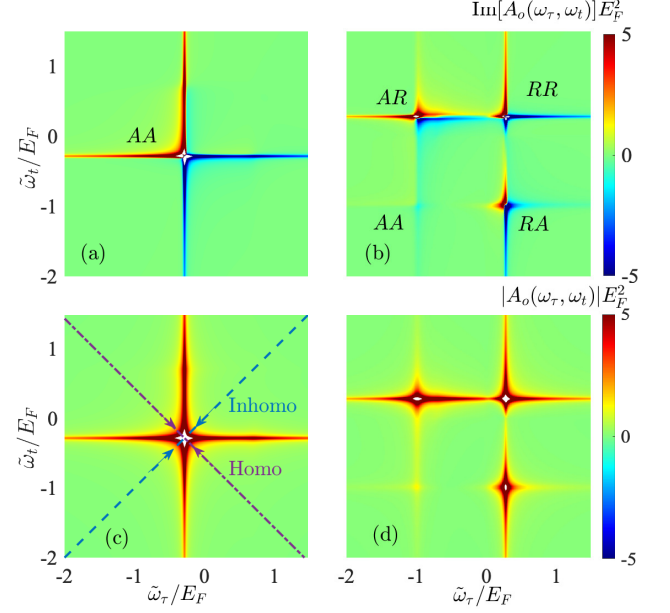


FIG. 7. (a) and (b) shows contour plots of the imaginary part of the 2D spin-echo spectrum $A_o(\omega_\tau, \omega_t)$ for attractive $k_F a = -0.5$ and repulsive $k_F a = 0.5$ interactions, respectively. (c) and (d) show the amplitude for $k_F a = -0.5$ and $k_F a = 0.5$, respectively. The temperature is set as $k_B T^\circ = 0.03 E_F$.

$$R(-\pi/2) = R(\pi/2)^T = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix}. \quad (\text{A3})$$

We also only need to focus on the first four pathways and can find that $I_1^- = -I_1$, $I_2^- = -I_2$, $I_3^- = I_3$, and $I_4^- = I_4$, where the superscript “-” indicates the quantities for EXSY- scheme. Consequently, we have $\tilde{A}^- = -A_1 - A_2 + A_3 + A_4$. As a result, in the differences between the EXSY+ and EXSY-, $\mathcal{A}(\omega_\tau, T, \omega_t) \equiv \tilde{A}(\omega_\tau, T, \omega_t) - \tilde{A}^-(\omega_\tau, T, \omega_t)$, only the contributions from the first two pathways remain, i.e.,

$$\mathcal{A}(\omega_\tau, T, \omega_t) = 2A_1(\omega_\tau, T, \omega_t) + 2A_2(\omega_\tau, T, \omega_t) \quad (\text{A4})$$

which no longer depends on ω_s .

Appendix B: Imaginary part and Amplitude of the 2D spin-echo spectrum

For completeness, we show in Fig. 7 the imaginary part and amplitude of the 2D spin-echo spectrum for the same parameters in Fig. 3 in the main text. In particular, we indicate the homogeneous and inhomogeneous lineshape in Fig. 7 (c).

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