

Variational Approach for Impurity Dynamics at Finite Temperature

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We present a general variational principle for the dynamics of impurity particles immersed in a quantum-mechanical medium. By working within the Heisenberg picture and constructing approximate time-dependent impurity operators, we can take the medium to be in any mixed state, such as a thermal state. Our variational method is consistent with all conservation laws and, in certain cases, it is equivalent to a finite-temperature Green's function approach. As a demonstration of our method, we consider the dynamics of heavy impurities that have suddenly been introduced into a Fermi gas at finite temperature. Using approximate time-dependent impurity operators involving only one particle-hole excitation of the Fermi sea, we find that we can successfully model the results of recent Ramsey interference experiments on ^{40}K atoms in a ^6Li Fermi gas. We also show that our approximation agrees well with the exact solution for the Ramsey response of a fixed impurity at finite temperature. Our approach paves the way for the investigation of impurities with dynamical degrees of freedom in arbitrary quantum-mechanical mediums.

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The behavior of a quantum impurity immersed in a medium is a fundamental problem in physics, having relevance to phenomena ranging from the orthogonality catastrophe [1] to Fermi liquid theory [2]. In addition to displaying interesting effects in their own right, quantum impurity problems can be used to build up more complex many-body systems, such as a finite density of impurities [3]. They can also be used as a probe of correlations and entanglement in a quantum-mechanical medium [4].

Recent advances in cold-atom experiments have enabled a greater variety of quantum impurity problems to be investigated. The scenario of an impurity in a Bose Einstein condensate (the so-called “Bose polaron”) has been successfully realized in experiment [5–7] and could potentially provide insight into bosonic mediums in general [8]. For instance, there is the prospect of a universal Bose polaron in the regime where the boson-impurity scattering length is tuned to infinity [9]. Similarly, experimental investigations of impurities in a Fermi gas (i.e., “Fermi polarons”) [10–17] have deepened our understanding of quasiparticles in both quantum gases [18–26] and the solid state [27,28]. In particular, recent cold-atom experiments have observed the formation of Fermi polarons and their out-of-equilibrium dynamics [15], thus opening up an arena in which to explore nonequilibrium phenomena in a controlled manner.

However, a major theoretical challenge is how to include the effects of temperature when modeling the behavior of quantum impurities, since this introduces two complications. First, one must consider a medium that is in a mixed rather than a pure state. Second, one often needs to perform a thermal average over the impurity's dynamical degrees of

freedom (such as the initial impurity momenta in the case of mobile impurities). Therefore, theoretical works on Fermi and Bose polarons at finite temperature in three dimensions have, thus far, been restricted to pinned impurities [29–31], weak impurity-medium interactions [32,33], the virial expansion [34], and approximate diagrammatic approaches [35–37]. Most notably, there are no exact Monte Carlo approaches for such finite-temperature polarons, thus emphasizing the need for alternative methods.

In this Letter, we present a time-dependent variational principle for the quantum impurity problem that can, in principle, handle any mixed state of the medium. The key simplification is to construct approximate time-dependent impurity operators that are then applied to a static medium. We apply our variational approach to the dynamics of heavy impurities that are suddenly introduced into a Fermi gas at finite temperature. For the simplest approximation of the dynamics, our approach is equivalent to ladder diagrams within a finite-temperature Green's function approach. However, the variational calculation can be easily extended to describe more complex scenarios such as impurities that are initially entangled with the medium. Moreover, our method conserves the total probability of the system, which is not always the case in diagrammatic approximations. As a demonstration of the power and accuracy of our approach, we show that it reproduces exact results for a fixed impurity with minimal error, and it allows us to accurately model recent Ramsey interference experiments on ^{40}K atoms in a ^6Li Fermi gas [15], where no exact solution exists.

Variational principle.—To tackle finite temperature, we separate the time dependence of the impurity dynamics

from the thermal average over all states of the medium, and consider the impurity annihilation operator at time t within the Heisenberg picture

$$\hat{c}(t) = e^{i\hat{H}t}\hat{c}e^{-i\hat{H}t}. \quad (1)$$

Here, we assume a time-independent Hamiltonian \hat{H} for $t > 0$, and we work in units where \hbar , k_B , and the system volume are all set to 1. For the moment, we suppress the impurity's dynamical degrees of freedom (e.g., momentum) and we assume that the impurity is noninteracting with the medium at $t = 0$, i.e., $\hat{c}(0) \equiv \hat{c}$. However, the formalism can be easily generalized to include more complex initial states, as we will see below.

To proceed, we consider the Heisenberg equation of motion for the impurity operator, $i\partial_t\hat{c}(t) = [\hat{c}(t), \hat{H}]$. The exact time-dependent operator will obey this equation; however, in general we will work with operators $\hat{c}(t)$ that only satisfy this approximately. Thus, inspired by other time-dependent variational principles [38,39], we define an “error” operator $\hat{\varepsilon}(t) \equiv i\partial_t\hat{c}(t) - [\hat{c}(t), \hat{H}]$, and then we minimize the average quantity

$$\Delta(t) = \text{Tr}[\hat{\rho}_0\hat{\varepsilon}(t)\hat{\varepsilon}^\dagger(t)]. \quad (2)$$

Here, the trace is over all realizations of the medium in the absence of the impurity, and $\hat{\rho}_0$ is the medium density matrix. Note that we are working in Fock space and thus the impurity operator can act directly on any particular medium state [40]. Importantly, our variational approach can be applied to any mixed state of the medium, but in the following we restrict ourselves to a thermal state at temperature T . Then we have $\hat{\rho}_0 = \exp(-\beta\hat{H}_0)/Z_0$, with \hat{H}_0 the medium-only Hamiltonian, $\beta \equiv T^{-1}$, and partition function $Z_0 = \text{Tr}[\exp(-\beta\hat{H}_0)]$. In the following, we define $\langle \dots \rangle_\beta \equiv \text{Tr}[\hat{\rho}_0 \dots]$.

We now expand $\hat{c}(t) \equiv \sum_j \alpha_j(t) \hat{O}_j$, where $\alpha_j(t)$ are time-dependent coefficients and \hat{O}_j are *time-independent* operators consisting of unique products of impurity and medium operators. In general, there is an infinite number of such operators, and the key is to limit ourselves to operators that form an appropriate variational basis for the problem at hand. Substituting the expansion into Eq. (2), imposing the minimization condition $\partial\Delta/\partial\alpha_j^* = 0$, and using the orthogonality of the operators \hat{O}_j , i.e., $\langle \hat{O}_j\hat{O}_l^\dagger \rangle_\beta = 0$ when $j \neq l$, we obtain [40]

$$i\dot{\alpha}_j(t) \langle \hat{O}_j\hat{O}_j^\dagger \rangle_\beta = \sum_l \alpha_l(t) \langle [\hat{O}_l, \hat{H}] \hat{O}_j^\dagger \rangle_\beta. \quad (3)$$

This key equation determines how the expansion coefficients $\alpha_j(t)$ in the approximate impurity operator $\hat{c}(t)$ vary in time. As opposed to the exact Heisenberg equation of motion, we see from Eq. (3) that the time dependence of the

impurity operator is controlled by the mixed state of the medium. This is a natural consequence of using a truncated basis of operators within our variational approach.

From Eq. (3), it is straightforward to demonstrate that our variational approach is conserving in the sense that the total probability $\langle \hat{c}(t)\hat{c}^\dagger(t) \rangle_\beta$ is constant (i.e., remains 1) throughout the time evolution [40]. Moreover, the proof of probability conservation makes no reference to the initial conditions, and it holds even when the Hamiltonian is explicitly time dependent.

For a time-independent \hat{H} , we can consider the stationary solutions of Eq. (3), i.e., $\alpha_j(t) \equiv e^{-iE_j t} \alpha_j$. In that case, we may solve a set of linear equations for the coefficients, resulting in the eigenvectors $\{\alpha_j^{(n)}\}$ with eigenvalues E_n . The associated stationary impurity operators $\hat{\phi}_n \equiv \sum_j \alpha_j^{(n)} \hat{O}_j$ may be chosen to be orthonormal under a thermal average, i.e., $\langle \hat{\phi}_m \hat{\phi}_n^\dagger \rangle_\beta = \delta_{mn}$. Because the total probability is conserved, these operators provide a complete basis for the approximate impurity operators and we thus have

$$\hat{c}(t) = \sum_n \langle \hat{c}(0) \hat{\phi}_n^\dagger \rangle_\beta \hat{\phi}_n e^{-iE_n t}, \quad (4)$$

where the thermal average allows us to take into account the boundary condition at time $t = 0$. We can then construct the relevant experimental observables by taking averages over products of the approximate impurity operators.

A particular scenario of interest is where an initially noninteracting impurity is suddenly coupled to the medium through a quench of the system parameters. The many-body response to the introduction of the impurity can be probed via Ramsey interferometry [15], which yields the time-dependent overlap [29,30] $S(t) \equiv \langle \hat{c} e^{i\hat{H}_i t} e^{-i\hat{H} t} \hat{c}^\dagger \rangle_\beta = e^{iE_i t} \langle \hat{c}(t) \hat{c}^\dagger(0) \rangle_\beta$, where \hat{H}_i is the initial noninteracting Hamiltonian and E_i is the initial energy of the impurity. This so-called Ramsey response is also intimately connected to the energy spectrum of the system, since it corresponds to the Fourier transform of the impurity spectral function [29,30].

Finally, we emphasize that our approach may be naturally extended to systems evolving under a series of time-independent Hamiltonians (or a time-dependent Hamiltonian via Trotterization). Within each such interval, we solve for the expansion coefficients using Eq. (3), and then impose the boundary conditions arising from the previous evolution via the thermal average in Eq. (4).

Impurity in a Fermi sea.—To demonstrate the utility and accuracy of our finite-temperature variational approach, we consider the quench dynamics of a spin- \downarrow impurity immersed in a spin- \uparrow Fermi sea. We model the interactions using a two-channel Hamiltonian [43]

$$\hat{H} = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} \hat{c}_{\mathbf{k}\sigma}^\dagger \hat{c}_{\mathbf{k}\sigma} + \sum_{\mathbf{k}} \epsilon_{\mathbf{k}M} \hat{b}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + g \sum_{\mathbf{k}_1 \mathbf{k}_2} (\hat{b}_{\mathbf{k}_1}^\dagger \hat{c}_{\mathbf{k}_2 \uparrow} \hat{c}_{\mathbf{k}_1 - \mathbf{k}_2, \downarrow} + \hat{c}_{\mathbf{k}_1 - \mathbf{k}_2, \downarrow}^\dagger \hat{c}_{\mathbf{k}_2 \uparrow} \hat{b}_{\mathbf{k}_1}). \quad (5)$$

Here, $\hat{c}_{\mathbf{k}\sigma}^\dagger$ and $\hat{b}_{\mathbf{k}}^\dagger$ respectively create spin- σ fermions and closed-channel molecules with momentum \mathbf{k} , while $\epsilon_{\mathbf{k}\sigma} = k^2/(2m_\sigma)$ and $\epsilon_{\mathbf{k}M} = k^2/[2(m_\uparrow + m_\downarrow)] + \nu$, where m_σ is the spin- σ fermion mass and ν is the bare detuning of the closed channel. g is the strength of the coupling between the open and closed channels. From the low-energy scattering amplitude at relative momentum \mathbf{k} , $f(k) = -1/(a^{-1} + R^*k^2 + ik)$, the range parameter is $R^* = \pi/(m_r^2 g^2)$, while the s -wave scattering length a is obtained via the prescription $m_r/(2\pi a) = -\nu/g^2 + \sum_{\mathbf{k}} 1/(\epsilon_{\mathbf{k}\uparrow} + \epsilon_{\mathbf{k}\downarrow})$, where $m_r = (1/m_\uparrow + 1/m_\downarrow)^{-1}$ is the reduced mass and Λ is an ultraviolet cutoff that should not affect the low-energy dynamics. The relevant dimensionless quantities that parametrize the system are $1/(k_F a)$, $k_F R^*$, T/T_F , and t/τ_F , with k_F the Fermi wave number of the spin- \uparrow Fermi sea, while the Fermi temperature $T_F = k_F^2/(2m_\uparrow)$ and the Fermi time $\tau_F = 1/T_F$.

To apply our variational approach, we take the approximate time-dependent operators to be of the form

$$\hat{c}_{\mathbf{q}\downarrow}(t) \simeq \alpha_{\mathbf{q};0}(t) \hat{c}_{\mathbf{q}\downarrow} + \sum_{\mathbf{k}} \alpha_{\mathbf{q};\mathbf{k}}(t) \hat{c}_{\mathbf{k}\uparrow}^\dagger \hat{b}_{\mathbf{q}+\mathbf{k}} + \sum_{\mathbf{k}_1 \neq \mathbf{k}_2} \alpha_{\mathbf{q};\mathbf{k}_1 \mathbf{k}_2}(t) \hat{c}_{\mathbf{k}_2 \uparrow}^\dagger \hat{c}_{\mathbf{k}_1 \uparrow} \hat{c}_{\mathbf{q}-\mathbf{k}_1+\mathbf{k}_2, \downarrow}, \quad (6)$$

where \mathbf{q} specifies the initial impurity momentum. The form of Eq. (6) contains the lowest order terms one would obtain if one took $\hat{c}_{\mathbf{q}\downarrow}(t) = e^{i\hat{H}t} \hat{c}_{\mathbf{q}\downarrow} e^{-i\hat{H}t}$ and performed an expansion in \hat{H} . Additional terms in Eq. (6) can similarly be obtained by considering higher order terms in the expansion, and since the Hamiltonian preserves the particle number, all operators generated in this fashion have one and only one impurity annihilation operator (either in the open or the closed channel configuration) [40]. However, note that the approximate impurity operator in Eq. (6) features time-dependent variational parameters, in contrast to the simple perturbative expansion. This is similar in spirit to the zero-temperature variational approach to the impurity wave function first introduced in Ref. [19] and applied to impurity dynamics in Refs. [15,44]. Taking the stationary condition for the operator in Eq. (6) then yields the equations [40]

$$(E - \epsilon_{\mathbf{q}\downarrow}) \alpha_{\mathbf{q};0} = g \sum_{\mathbf{k}} \alpha_{\mathbf{q};\mathbf{k}} \langle \hat{c}_{\mathbf{k}\uparrow}^\dagger \hat{c}_{\mathbf{k}\uparrow} \rangle_\beta, \quad (7a)$$

$$(E - \epsilon_{\mathbf{q};\mathbf{k}}) \alpha_{\mathbf{q};\mathbf{k}} = g \alpha_{\mathbf{q};0} + g \sum_{\mathbf{k}_1} \alpha_{\mathbf{q};\mathbf{k}_1 \mathbf{k}} \langle \hat{c}_{\mathbf{k}_1 \uparrow}^\dagger \hat{c}_{\mathbf{k}_1 \uparrow} \rangle_\beta, \quad (7b)$$

$$(E - \epsilon_{\mathbf{q};\mathbf{k}_1 \mathbf{k}_2}) \alpha_{\mathbf{q};\mathbf{k}_1 \mathbf{k}_2} = g \alpha_{\mathbf{q};\mathbf{k}_2}, \quad (7c)$$

where we have defined $\epsilon_{\mathbf{q};\mathbf{k}_1 \mathbf{k}_2} = \epsilon_{\mathbf{k}_2 - \mathbf{k}_1 + \mathbf{q}, \downarrow} + \epsilon_{\mathbf{k}_1 \uparrow} - \epsilon_{\mathbf{k}_2 \uparrow}$, $\epsilon_{\mathbf{q};\mathbf{k}} = \epsilon_{\mathbf{q}+\mathbf{k}, M} - \epsilon_{\mathbf{k} \uparrow}$, and we will take $\langle \hat{c}_{\mathbf{k}\uparrow}^\dagger \hat{c}_{\mathbf{k}\uparrow} \rangle_\beta \equiv f_{\mathbf{k}\uparrow}$ to be the Fermi-Dirac distribution of the \uparrow Fermi sea.

For the simplest quench scenario, where an impurity at momentum \mathbf{q} is initially noninteracting with the Fermi sea, we have the Ramsey response

$$S_{\mathbf{q}}(t) = e^{i\epsilon_{\mathbf{q}\downarrow}t} \langle \hat{c}_{\mathbf{q}\downarrow}(t) \hat{c}_{\mathbf{q}\downarrow}^\dagger \rangle_\beta \simeq \sum_n |\alpha_{\mathbf{q};0}^{(n)}|^2 e^{i(\epsilon_{\mathbf{q}\downarrow} - E_{\mathbf{q};n})t}, \quad (8)$$

where we have used Eq. (4) to obtain the approximate expression in terms of variational parameters. In practice, we will initially have a finite density of impurities in thermal equilibrium with the medium. Because the momentum of the impurity operator is preserved during the dynamics, we can thermally average over the initial momenta at the end of the calculation, yielding $S(t) = (2\pi\beta/m_\downarrow)^{3/2} \sum_{\mathbf{q}} e^{-\beta\epsilon_{\mathbf{q}\downarrow}} S_{\mathbf{q}}(t)$. However, we stress that since we are not explicitly including correlations between impurities, the validity of such an approach is limited to a low density of impurities.

Dynamics of a fixed impurity.—In the particular case of a fixed (infinitely heavy) impurity in a Fermi sea, the quantum dynamics can be solved exactly [30,45,46] since it reduces to solving for the single-particle states of the Fermi sea in the presence of a fixed potential [40]. Because the exact solution formally involves an infinite number of excitations of the Fermi sea, this provides a highly non-trivial benchmark for our theory. Moreover, since there is no impurity momentum, we can ignore \mathbf{q} in Eq. (8) [i.e., $S(t) \equiv S_0(t)$] and we only need to consider the effects of temperature on the medium.

Figure 1 displays a comparison of our variational results with the exact solution for three different interaction regimes: repulsive ($1/a > 0$), attractive ($1/a < 0$), and the unitary limit ($1/a = 0$). We see that our approach captures the short-time Ramsey response exactly, as expected from perturbative calculations [44], and it only noticeably deviates from the exact result for times $t \gtrsim 10\tau_F$ at very low temperatures. Note that our single-excitation approximation cannot describe the orthogonality catastrophe [1], which governs the long-time behavior of the fixed impurity at $T = 0$ [29,30]. However, our approximation will match the exact result when $T \gg T_F$ since it contains the leading order contributions to the virial expansion. In general, thermal effects lead to an exponential decay of the amplitude $|S(t)|$ at long times [14,47,48], while the phase of $S(t)$ is determined by the dominant quasiparticle peaks in the energy spectrum, which are less sensitive to temperature. These features are all well described within the variational approach [40].

Comparison with experiment.—For the case of heavy ^{40}K impurities in a ^6Li gas [15], the quench dynamics involved a preparation sequence during which the impurities were weakly interacting with the Fermi gas. We

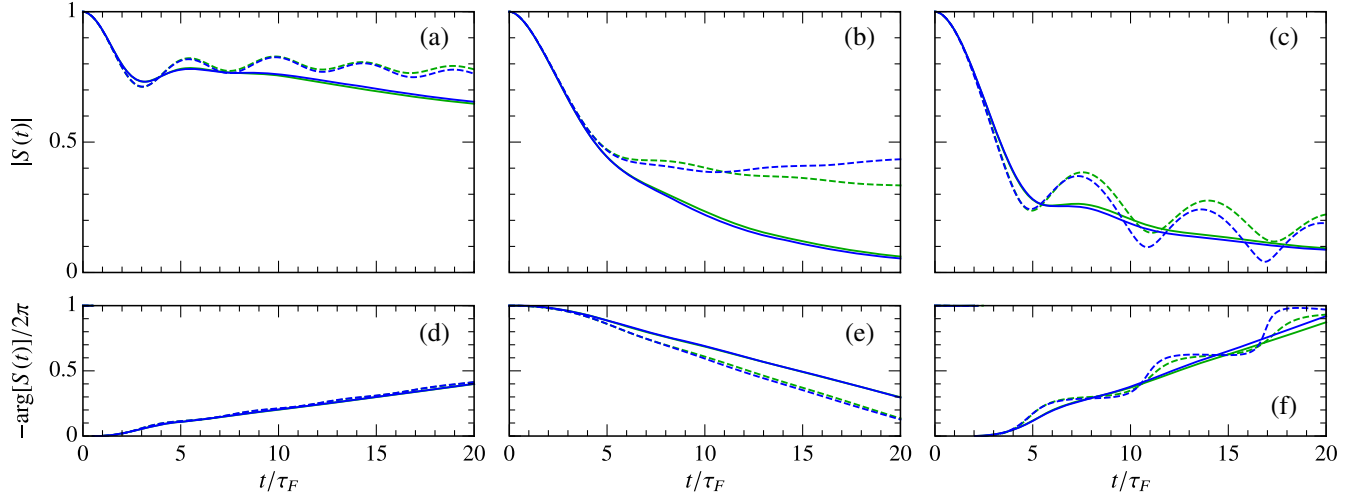


FIG. 1. Ramsey response of a fixed impurity from the variational approach (blue) and exact solution (green) for range parameter $k_F R^* = 1$. Dashed and solid lines correspond to temperatures $T = 0$ and $T = 0.2T_F$, respectively. The interactions during the time evolution are (a),(d) repulsive with $1/(k_F a) = 1$; (b),(e) attractive, $1/(k_F a) = -1$; and (c),(f) at unitarity $1/a = 0$.

model this using a two-step quench as in Ref. [15], which modifies the Ramsey response in Eq. (8) to

$$S'_q(t) = e^{ic_{q\downarrow}(t+2t_1)} \langle \hat{c}_{q\downarrow,t_1}(t) \hat{c}_{q\downarrow,-t_1}^\dagger(0) \rangle_\beta, \quad (9)$$

where we define operators $\hat{c}_{q\downarrow,t_1}(0) = e^{i\hat{H}_1 t_1} \hat{c}_{q\downarrow} e^{-i\hat{H}_1 t_1}$, $\hat{c}_{q\downarrow,t_1}(t) \equiv e^{i\hat{H} t} \hat{c}_{q\downarrow,t_1} e^{-i\hat{H} t}$, and \hat{H}_1 is the Hamiltonian (with

associated scattering length a_1) applied for a time t_1 just before and after the time evolution governed by \hat{H} . Equation (9) can easily be evaluated within our variational approach by modifying the initial condition in Eq. (4) [40].

In Figs. 2(a)–2(f), we see that the calculated Ramsey response agrees remarkably well with the experimental data from Ref. [15], without the use of any fitting parameters. In particular, we find that we require the thermal average over

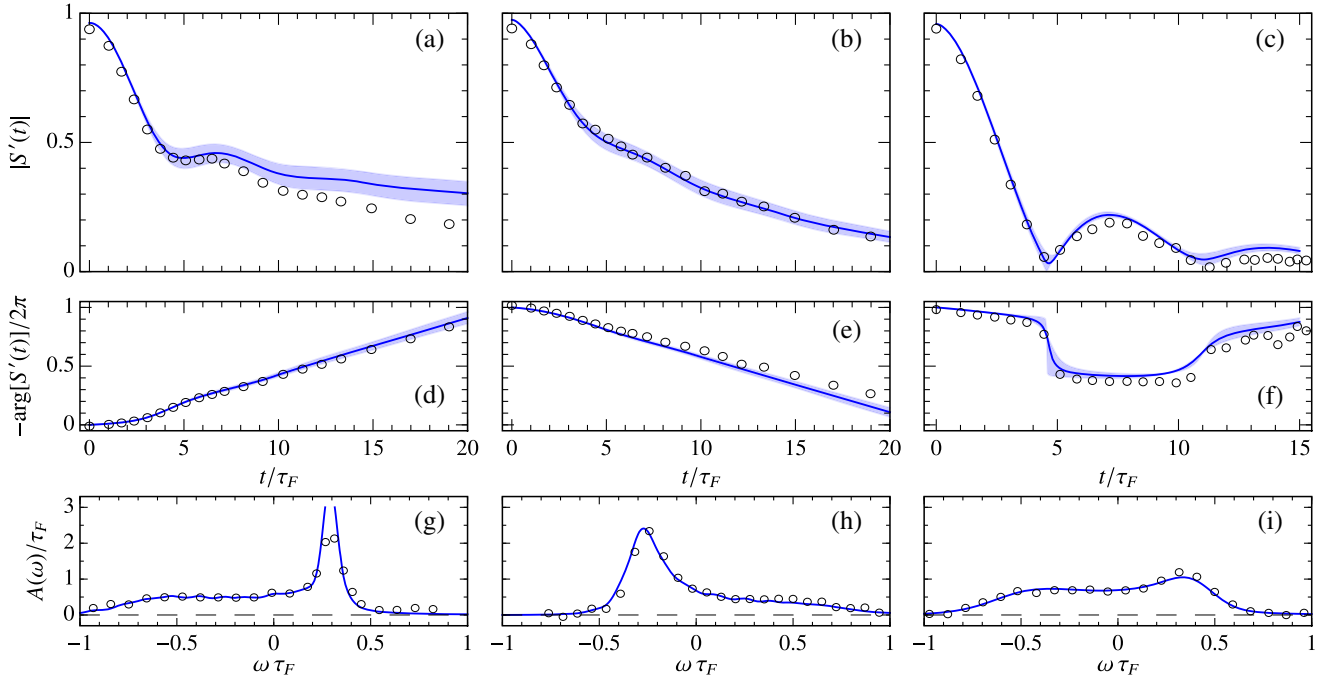


FIG. 2. (a)–(f) Ramsey response and (g)–(i) radio-frequency spectra of ^{40}K impurities in a ^6Li Fermi gas. We show the experimental results from Ref. [15] (circles) together with results from our variational approach (solid lines), thermally averaged over impurity momentum. In (a)–(f), the shading corresponds to the experimental uncertainty of the scattering length [15]. From left to right, the experimental parameters are $1/(k_F a) = \{0.23, -0.86, -0.08\}$, $T/T_F = \{0.17, 0.16, 0.18\}$, and $1/(k_F a_1) = \{3.9, -5.8, -4.8\}$. In all panels, $k_F R^* = 1.1$ and $t_1 = 4.0\tau_F$.

impurity momentum, as well as the thermal state of the Fermi gas, in order to accurately model the response [40]. Our approach also reproduces the impurity spectral function $A(\omega) = \text{Re} \int_0^\infty (dt/\pi) e^{i\omega t} S(t)$ [40] measured in radio-frequency spectroscopy, as shown in Figs. 2(g)–2(i). The discrepancy at long times in Fig. 2(a) suggests that the approximation does not fully capture the decoherence rate of the repulsive branch, which is consistent with the small difference in the repulsive peak between theory and experiment in Fig. 2(g). This intriguing result suggests that there is an additional decay channel for the repulsive branch of the heavy impurity that is not present in the infinite-mass case [40].

Relationship to diagrammatic approaches.—Solving Eq. (7) for the energy yields the expression

$$E = \epsilon_{\mathbf{q}\downarrow} + \sum_{\mathbf{k}_2} f_{\mathbf{k}_2\uparrow} \left(\frac{E - \epsilon_{\mathbf{q};\mathbf{k}_2}}{g^2} - \sum_{\mathbf{k}_1} \frac{1 - f_{\mathbf{k}_1\uparrow}}{E - \epsilon_{\mathbf{q};\mathbf{k}_1\mathbf{k}_2}} \right)^{-1},$$

which corresponds to the pole of the impurity Green's function, $E = \epsilon_{\mathbf{q}\downarrow} + \Sigma(\mathbf{q}, E)$, where $\Sigma(\mathbf{q}, E)$ is the impurity self-energy calculated using ladder diagrams at finite temperature [35]. Therefore, our variational method is equivalent to a finite-temperature Green's function approach—indeed, the Ramsey response in Eq. (8) is simply proportional to the time-dependent impurity Green's function. However, our formulation has the advantage that it can be readily adapted to describe more complex dynamics, such as the two-step quench in Eq. (9) or Rabi oscillations [44].

Conclusions.—We have developed a general variational approach for impurity dynamics, and we have used it to successfully model a heavy impurity in a Fermi gas at finite temperature. Our results suggest that the dynamics observed in experiment is well described by approximations with a single excitation of the Fermi sea, and that the effect of the impurity mass is masked by residual interactions during the preparation, as well as by thermal fluctuations. Our method paves the way for further investigations of quantum impurities, involving different variational operators (e.g., derived from coherent states) or other scenarios, such as the Bose polaron.

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