Order Fractionalization

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Abstract

The confluence of quantum mechanics and complexity leads to the emergence of rich, exotic states of quantum matter which demand that we expand our ideas of quantum order. The twin concepts of spontaneously broken symmetry and off-diagonal long-range order (ODLRO) are fundamental to our understanding of phase transitions. In electronic matter it has long been assumed that Landau order parameters involve an even number of electron fields, with integer spin and even charge, that are bosons. On the other hand, in low-dimensional magnetism, operators are known to fractionalize so that the ground-state excitations carry spin-1/2. Motivated by experiment, mean-field theory and computational results, we extend the concept of ODLRO into the time domain, proposing that in a broken symmetry state the order parameter can fractionalize into half-integer objects. Using numerical renormalization group studies we show how such fractionalized order can be induced in quantum impurity models. We then conjecture that such order develops spontaneously in lattice quantum systems, due to positive feedback, with predictions for experiment.

A major theme in current condensed matter physics is the quest for new types of quantum matter such as high-temperature superconductors, topological insulators and spin liquids [1–8]. An important aspect of this research is the characterization of novel forms of order that emerge in these quantum materials; another concerns the new classes of excitation that accompany these orderings. Landau's theory of phase transitions [9] attributes the transformation in macroscopic properties to the development of an order parameter that breaks the microscopic symmetries of the system. Later, Yang observed [10] that such long-range order is manifested as an asymptotic factorization of spatial correlation functions at long distances into a product of order parameters $\langle\Psi(x)\Psi^\dagger(y)\rangle \xrightarrow{|x-y|\to\infty} \langle\Psi(x)\rangle\langle\Psi(y)\rangle^*$. The quantum operators Ψ are bosonic and condense into a state of "Off-Diagonal Long Range Order" (ODLRO).

In relativistic physics half-integer spin order parameters are prohibited by the spin-statistics theorem [11, 12], but in electronic condensed matter the absence of Lorentz invariance removes this restriction. Though half-integer order can be envisioned in Landau's theory of phase transitions, it is microscopically incompatible with ODRLO where the local operators that condense are bosons, formed from an even number of half-integer spin fermions. Conventional order parameters such as magnetization or pair density involve pairs of fermions and form part of the general paradigm of BCS/Hartree-Fock order parameters; more complicated "composite order parameters" involving four or more elementary fermion fields have also been envisioned [13, 14], but all have integer spin. This has led to the implicit assumption that in electronic quantum matter, order parameters satisfy an effective spin-statistics theorem, carrying integer spin and even charge.

Another important development in condensed matter physics is the discovery of "fractionalization", where the emergent excitations carry fractional quantum numbers [15–18]. A classic example is the one dimensional spin-1/2 Heisenberg antiferromagnet where a spin-flip, that changes the magnetic quantum number by an integer unit, creates a pair of spin-1/2 excitations called spinons [19, 20]. Higher dimensional examples include the fractional quantum Hall effect [17], and spin liquids like the Kitaev honeycomb model where the spin operators fractionalize into Majorana fermions [21]. Fractionalization has also been proposed to occur at continuous quantum phase transitions [22, 23] leading to "deconfined quantum criticality" where a fluctuating order parameter breaks up into new degrees of freedom. Whereas ODLRO is a ground-state property, fractionalization is associated with excitations, manifested in dynamical response functions and as correlations that are nonlocal in time. In this paper we explore the possible unification of ODLRO and fractionalization, proposing that quantum operators can fractionalize into half-integer order parameters. This order fractionalization conjecture (OFC) requires an extension of ODLRO into space-time, and suggests a new symmetry class of quantum order [24].

A key setting for our discussion is the Kondo lattice, a model describing an array of magnetic moments interacting via an antiferromagnetic exchange with a sea of conduction electrons. This model is widely used to describe the behavior of heavy fermion materials, where the screening of the magnetic moments by conduction electrons at low temperatures liberates their spins into the Fermi sea as delocalized heavy electrons (Fig. 1**A**, **B**), a process that enlarges the Fermi surface. Since a spin flip of a local moment creates a particle-hole pair of heavy fermions, we are led to interpret the expansion of the Fermi surface as a fractionalization of local moments into negatively charged fermions [25]. The origin of the moments is immaterial and their fractionalization into heavy fermions would even occur if they were of nuclear origin [26]. (Fig. 1 **C**).

There is considerable indirect experimental and theoretical support for spin fractionalization in the Kondo lattice. Using topology, Oshikawa has shown that in a Fermi liquid ground-state, the screened spins of a Kondo lattice contribute to an expansion of its Fermi surface volume [27]. Hall effect and de Haas van Alphen measurements subsequently detected jumps in the Fermi surface volumes at quantum phase transitions between antiferromagnetic and paramagnetic heavy fermion ground-states [28, 29]. The enlargement of the Fermi surface in the Kondo lattice indicates the formation of half-integer excitations from the lattice of local moments, a process that is most naturally interpreted as spin fractionalization.

Experimentally, there are important examples where Kondo spin screening appears coincident with the development of long range order. For instance, in both $NpPd_5Al_2$ and $CeCoIn_5$, singlet superconductivity develops directly from a Curie-Weiss paramagnet [30, 31] with a substantial loss of spin entropy. Similar phenomenon involving quadrupole degrees of freedom has been proposed for UBe_{13}, URu_2Si_2 and Pr X_2Al_{20} (X=Ti, Va) [32–34]. Theoretical evidence for spin fractionalization and broken symmetry is found from path-integral based, large-N treatments of Kondo lattices [35–38]. However, while these methods demonstrate the feasibility of order fractionalization in models with very large numbers of spin components, they are unable to demonstrate that this phenomenon extends to physical, spin-1/2

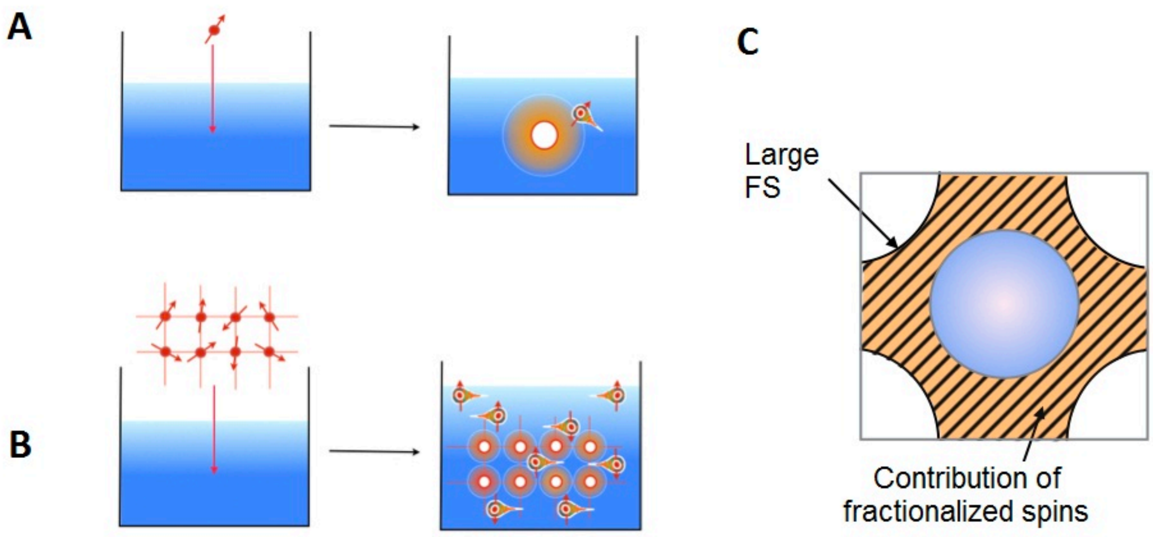

FIG. 1: Schematic illustrating the Kondo effect showing **(A)** the fractionalization of a single spin into a delocalized f-electron in a Kondo impurity model, **(B)** the fractionalization of local moments in a Kondo lattice, to form a fluid of heavy fermions; **(C)** the enlargement of the Fermi surface from small (blue) to large (hatched) due to the formation of heavy fermions, as predicted by Oshikawa's theorem [27].

Kondo lattices. Our motivation to seek new classes of broken symmetry derives from these experimental and theoretical considerations.

Developing this idea, we recall that the dynamics of an interacting fermion is determined by the Dyson self-energy, $\Sigma_{\alpha\beta}(2,1)$, an amplitude for the scattering of a single-particle state at space time $1 = (x_1, t_1)$ to a single-particle state at the space-time $2 \equiv (x_2, t_2)$ via intermediate many-body states. Here β and α are the internal quantum numbers of the incoming and outgoing fermions. The Hamiltonian that determines the time evolution is invariant under various global symmetry transformations such as spin rotation or global gauge invariance; at high temperatures the self-energy is also invariant under these symmetries. However if a phase transition occurs, the self-energy develops a symmetry-breaking component resulting from scattering off the order parameter. For example a ferromagnet develops a spontaneous Zeeman splitting driven by the internal Weiss field; a BCS superconductor develops a pairing field due to Andreev scattering off the condensate. In all these classical examples, the order parameter has an associated coherence length; for space/times larger than this coherence length, the (coarse-grained) self-energy can be regarded as a local, instantaneous symmetry-breaking potential, $\Sigma_{\alpha\beta}(2,1) = M_{\alpha\beta}(1)\delta(2-1)$, where the order parameter $M_{\alpha\beta}(1)$ transforms under an irreducible representation of the Hamiltonian symmetry group (Fig. 2A).

Fractionalization implies a factorization of quantum operators into two or more independent components. Similarly, we take order fractionalization to imply that at large space-time separations between 1 and 2, the self-energy factorizes into a product of fractional order parameters, $\Sigma_{\alpha\beta}(2,1) \sim \bar{V}_\alpha(2)V_\beta(1)$, where $V_\beta(1)$ and $\bar{V}_\alpha(2)$ describe a fractional, spinorial order parameter and its conjugate at locations 1 and 2, respectively. The independence of these two quantities requires that the intermediate fermionic state develops a bound-state that propagates without decay between 1 and 2, as shown schematically in Fig. 2**B**. Since V_β carries the quantum number of the incoming fermion, the intermediate bound-state fermion is neutral with respect to this quantum number. This establishes a link between order fractionalization and the the formation of neutral fermion bound-states.

Following the example of the Curie-Weiss theory of magnetism, here we develop support for the order fractionalization conjecture. There, the first step is to induce Curie magnetism with an external magnetic field acting on a single spin; next, one argues that in the bulk the interaction of one site on another provides a Weiss field that maintains a spontaneous magnetization [39]. Similarly here we seek to *induce* order fractionalization in Kondo impurity models by identifying an appropriate symmetry breaking field. This is a necessary pre-condition for us to argue that "fractionalizing Weiss fields" can spontaneously stabilize order fractionalization in lattices.

Induced Order Fractionalization. We first identify spin fractionalization in the single impurity Kondo model. Then we show we can induce order fractionalization in the two-channel Kondo impurity model where the local moment is screened by two separate conduction channels. The impurity Kondo model involves an antiferromagnetic spin exchange interaction between a local moment and the spin density of the conduction sea $H_I = J(\psi^\dagger\vec{\sigma}\psi)\cdot\vec{S}$, where $\psi^\dagger$ is a spinor that creates an electron at the site of the impurity. At low energies this model is a resonant level Fermi liquid where the interaction matrix elements behave as an effective (renormalized) Anderson model, $J\psi^\dagger(\vec{\sigma}\cdot\vec{S})\psi \to (\bar{V}\psi^\dagger f + V f^\dagger\psi)$. The equivalence of the effective Hamiltonian with its microscopic form implies that the operator combination of a spin and a conduction electron acts as a single, bound-state fermion

$$J(\vec{\sigma}_{\alpha\beta} \cdot \overbracket{\vec{S})\psi_{\beta}} = \bar{V}\hat{f}_{\alpha}. \tag{1}$$

Here the horizontal line contracting the spin and the fermion implies that at long times, this combination acts as a single composite fermion. Although this process has been amply demonstrated in large-N calculations [35, 40] and is implicitly guaranteed by the low energy equivalence of the Anderson and Kondo impurities models, we now present an explicit demonstration of its occurance in the spin-1/2 Kondo model using numerical renormalization group (NRG) methods [41].

In NRG the conduction bath is discretized logarithmically, mapping the model to an impurity spin coupled to a tight-binding (Wilson) chain with exponentially decaying tunneling amplitude. This produces the imaginary part of the Green's function at a set of discrete frequencies, which are then interpolated to produce a continuous, analytic function satisfying the necessary sum rules. We then transform the T-matrix of the conduction electrons $T(z)$ so obtained, to the irreducible self-energy $\Sigma(z)$ using the relation

$$\Sigma(z) = \frac{T(z)}{1 + g(z)T(z)}, \tag{2}$$

where $g(z)$ is the bare local Green's function of the conduction electrons at the position of the impurity. The unitary single-particle scattering generated by a Kondo singlet at low energies implies that $g(0)T(0) = -1$ at the Fermi energy and hence a singular structure in $\Sigma(z \sim 0)$, making the extraction of $\Sigma(z)$ sensitive to interpolation errors in the NRG. We employ a limiting procedure in which $g(z) \to (1 - \epsilon)g(z)$, keeping ϵ larger than the interpolation errors induced in $T(z)$ (c.f. Supplementary Materials).

Figure Fig. 2**C** shows the result of a NRG calculation of the one-particle irreducible electron self-energy in the spin-1/2 Kondo model, indicating that it contains a sharp, resolution-limited pole at zero energy, with the asymptotic form $\Sigma_{\alpha\beta} \sim \delta_{\alpha\beta}\bar{V}V/\omega$. The pole demonstrates the development of a many-body fermionic bound-state at the Fermi energy carrying $S = 1/2$ and charge e; the sharpness of the pole confirms that the spectral decomposition of the emergent f-electron field has no overlap with one-particle excitations of the conduction sea, i.e it is an emergent fermion, described by the Lagrangian $\mathcal{L}_f = f^{\dagger}_{\sigma}(-i\partial_t)f_{\sigma}$. The background to the peak can be fit with ω^2 at low energies and is due to Fermi-liquid interactions (c.f. Supplementary Materials).

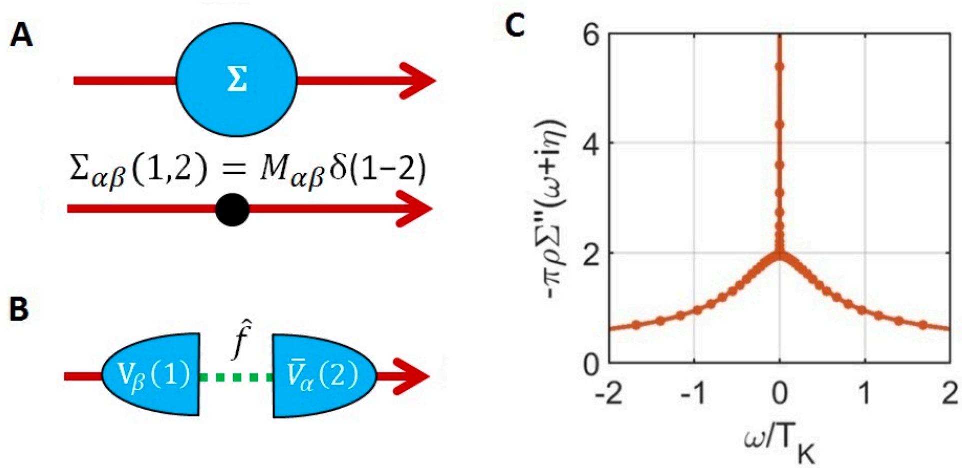

FIG. 2: **(A)** In a conventional broken symmetry state, the coarse-grained electronic self-energy (top) is instantaneous and local. **(B)** Order fractionalization leads to a factorization of the self-energy into two spinorial components, linked by a low energy fermionic bound-state (bottom). **(C)** The irreducible self-energy in the single-channel single-impurity Kondo model, computed using NRG, displaying a sharp fermionic pole, on top of a Fermi liquid background (see supplementary material).

To confirm that the local moment fractionalizes into a pair of fermions, we apply a small external magnetic field, under which the Lagrangian acquires a term proportional to the magnetization $\vec{M} = \frac{1}{2}\psi^\dagger\vec{\sigma}\psi + \vec{S}$, i.e. $\mathcal{L} \to \mathcal{L} - \vec{B}.\vec{M}$. Equivalently, we can employ a Gallilean transformation into a reference frame rotating with angular velocity $\vec{\omega} = (g\mu_B)\,\vec{B}$. In the rotating reference frame, $\psi \to U\psi$ and $f \to Uf$ where $U = e^{-it\vec{\omega}\cdot\vec{\sigma}/2}$, and under this transformation,

$$\mathcal{L}_f \to f^\dagger\left(-i\partial_t - \vec{\omega}\cdot\vec{\sigma}/2\right)f = \mathcal{L}_f - (g\mu_B)\vec{B}\cdot\vec{S}_f \tag{3}$$

where $\vec{S}_f \equiv f^\dagger_\alpha\left(\frac{\vec{\sigma}}{2}\right)_{\alpha\beta} f_\beta$. Comparing this to the original $\vec{M}$, we can identify $\vec{S}_f$ as the spin, fractionalized into a product of Dirac (i.e. complex) fermions. We note that, unlike in a path integral approach that assumes stable fractionalized excitations, here we have demonstrated their stability without approximation from the sharp pole structure of the self-energy.

An important aspect of fractionalization is the emergence of an internal gauge symmetry. Typically, fractionalized excitations carry an internal gauge charge [22, 42–46]. The fractionalized spin is invariant under gauge transformations of the emergent f-excitations, $f_\alpha \to e^{i\theta(t)}f_\alpha$. Moreover the composite fermion $(\vec{\sigma}\cdot\vec{S})\psi$ involves a product of the hybridization and the f-electron, $\bar{V} f_\sigma$ that is is invariant under $U(1)$ transformations of both fields $f_\sigma \to e^{i\theta(t)} f_\sigma$, $V \to e^{i\theta(t)} V$. When these transformed fields are substituted into the action, it becomes $\mathcal{L}_f \to f^\dagger(i\partial_t + A_0) f$, where $A_0 = \dot{\theta}$ is an emergent gauge field coupled to the number operator of the f-electrons. The path-integral approaches suggest that the right action for the f-electrons contains an additional topological term that controls the irreducible representation of the spin of the form $\mathcal{L}_f = f^\dagger i\partial_t f + A_0(n_f - Q)$ where $Q = 1$ for the $SU(2)$ Kondo model. With this formulation we can always choose a gauge where $V(t)$ is real and the phase fluctuations are entirely absorbed into $A_0(t)$. The NRG results suggest that the mean-field saddle point describing a fractionalized ground-state in which $A_0 = 0$ captures the essential physics of the excitations of the $S = 1/2$ SU(2) Kondo model. Fourier transforming the conduction electron self-energy into the time-domain, we see it exhibits long-range temporal correlations

$$\Sigma(t_1 - t_2) \xrightarrow{|t_1 - t_2| \to \infty} |V|^2 \mathrm{sgn}(t_1 - t_2)/2. \tag{4}$$

In the single-channel Kondo model these correlations do not break any physical symmetry but they will be important for our subsequent discussion.

To address our original question regarding order fractionalization, we next turn to the two-channel Kondo (2CK) model where the channels of screening electrons are indexed by $\lambda = 1, 2$. The channel-symmetric two channel Kondo model has a quantum critical ground-state [47–49] , which can be loosely interpreted as a resonant-valence-bond (RVB) state of singlet between the spin and each of the two conduction channels (Fig. 3**A**). As we now show, breaking this channel symmetry induces order fractionalization. The 2CK exchange interaction is

$$H_I = J\psi^\dagger_\lambda(\vec{\sigma} \cdot \vec{S})\psi_\lambda + \delta J \hat{\Psi}, \tag{5}$$

where the channel asymmetry δJ couples to the composite operator $\Psi = \left[\psi^\dagger_1(\vec{\sigma}\cdot\vec{S})\psi_1 - \psi^\dagger_2(\vec{\sigma}\cdot\vec{S})\psi_2\right]$, inducing an asymmetric Kondo coupling $J \pm \delta J$ in the two channels. δJ plays the role of an external field that induces composite order $\langle\Psi\rangle$. Renormalization group studies tell us that a finite channel asymmetry $\delta J > 0$ destabilizes the quantum critical point, stabilizing a Kondo singlet in the strongest channel [50], here associated with the $\lambda = 1$, loosely interpreted as a valence bond solid (VBS) state (Fig. 3**B**). As in the one-channel Kondo model, this implies the formation of a fermionic bound-state

$$J(\vec{\sigma}_{\alpha\beta} \cdot \overbracket{\vec{S})\psi_{\lambda\beta}} = \bar{V}_\lambda \hat{f}_\alpha, \qquad (\lambda = 1, 2), \tag{6}$$

but with a channel-dependent amplitude $\bar{V}_\lambda = (\bar{V}, 0)$ that projects into the strongest channel. The quantum numbers of the composite fermion divide into two: a c-number spinor $\bar{V}_\lambda$ that carries the channel quantum number and a residual fermion with spin and charge but no channel index.

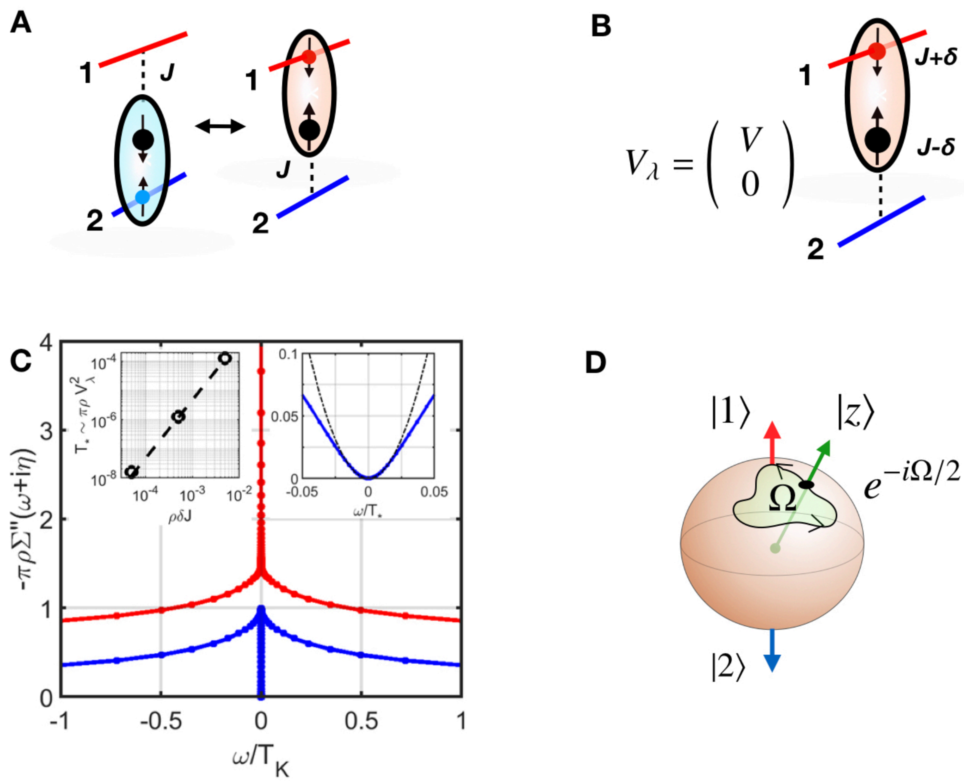

FIG. 3: **(A)** The symmetric two-channel Kondo model forms a quantum critical state where the Kondo singlets are delocalized between channels. **(B)** Application of channel asymmetry stabilizes a Kondo singlet on one channel, defined by a spinor order parameter. **(C)** A fermionic pole is induced in the strongest channel, indicating the presence of a selective hybridization $\bar{V}_\lambda = (V, 0)$ between the fractionalized moment and the two screening channels. The red curve is shifted by 0.5 unit vertically upward for clarity. (Insets) left: The Fermi temperature T_* vs $\rho\delta J$. right: the low-frequency regime of self-energy in channel 2 vs. ω showing Fermi liquid behavior (dashed black line is an ω^2 fit). **(D)** Adiabatic rotation of the channel asymmetry along a path subtending a solid angle Ω leads between channels leads to an $e^{-i\Omega/2}$ Berry phase, reflecting the half-integer nature of the induced order.

Fig. 3**C** displays the self-energy of a channel-asymmetric spin-1/2 2CK model, calculated using NRG. For $\delta J > 0$, a sharp, resolution-limited quasiparticle pole forms in the strongest $\lambda = 1$ channel, leading to a pole in the self-energy, $\Sigma_{\lambda\lambda'} \sim \bar{V}_\lambda V_{\lambda'}/\omega$. The product form of the self-energy follows from the projection into the strongest channel. By Fourier transforming this result, we confirm that at long times, the self-energy factorizes

$$\Sigma_{\lambda\lambda'}(t_2 - t_1) \xrightarrow{|t_2 - t_1| \to \infty} \bar{V}_\lambda(t_2) V_{\lambda'}(t_1) \mathrm{sgn}(t_2 - t_1)/2. \tag{7}$$

This factorization into two spinors means that the singular part of the self-energy does not transform under an irreducible representation, but instead as a *reducible* sum of a vector and scalar representation, i.e $1/2 \otimes 1/2 = 0 + 1$. If we are to preserve Landau's notion that order parameters transform under irreducible representations, then we are forced to acknowledge that the spinor V_λ is the relevant order parameter and we have order fractionalization.

A way of exposing the spinorial character of the order is to adiabatically rotate the spinor V_λ by slowly rotating the channel asymmetry, which we may write $\delta J(\hat{n}(t) \cdot \vec{\Psi})$, where $\vec{\Psi} = \psi^\dagger_\lambda \vec{\alpha}_{\lambda\lambda'} (\vec{\sigma} \cdot \vec{S}) \psi_{\lambda'}$ is the composite "channel magnetization", defined in terms of three Pauli matrices $\vec{\alpha} = (\alpha_1, \alpha_2, \alpha_3)$, and $\hat{n}(t)$ is the asymmetry field. In this adiabatic evolution of the Kondo singlet, the channel selective hybridization is then determined by $(\vec{\alpha}_{\lambda\lambda'} \cdot \hat{n}) V_{\lambda'} = +V_\lambda$. By slowly varying $\hat{n}(t)$ along a closed path, we can characterize the topology of the order parameter by examining the corresponding Berry phase factor $e^{-i\gamma} = e^{-i\Omega S}$ where Ω is the solid angle enclosed by the path and S determines the spin of the order parameter (Fig. 3**D**). To calculate γ we go to the extreme strong coupling limit, where $\delta J \gg \Lambda$, the electron band-width. In this limit the channel Kondo singlet becomes entirely local, taking the form $|z\rangle = \sum_{\lambda=1,2} z_\lambda |\lambda_s\rangle$, where $z = (z_1, z_2) \equiv V_\lambda/|V|$ is a unit spinor and $|\lambda_s\rangle = \frac{1}{\sqrt{2}}(|\lambda \uparrow, \Downarrow\rangle - |\lambda \downarrow, \Uparrow\rangle)$ denotes a singlet formed between the local moment ($\Uparrow$) and electron ($|\lambda \uparrow\rangle$) in channel λ. When $\hat{n}(t)$ is rotated through a solid angle Ω, the spinor $z_\lambda(t)$ evolves adiabatically, and the ground-state wavefunction acquires a Berry phase given by $\gamma = -i \oint dt (z^\dagger \partial_t z) = \frac{1}{2}\Omega$ (cf. Supplementary Material). The factor of 1/2 confirms the half-integer channel spin of the state.

We also note the application of the symmetry-breaking field δJ induces an expectation value of the composite order parameter $\langle \vec{\Psi} \rangle \propto V^*_\lambda \vec{\alpha}_{\lambda\lambda'} V_{\lambda'}$, indicating that the induced composite order has fractionalized into a product of channel hybridization spinors. From this exercise, we see that order fractionalization (OF) is manifested in three separate ways: (i) a splitting of the composite fermion into a spinor boson and a fermion; (ii) a fractionalization of the local moment into a product of fermion excitations and (iii) fractionalization of the composite order parameter into a product of spinorial order parameters.

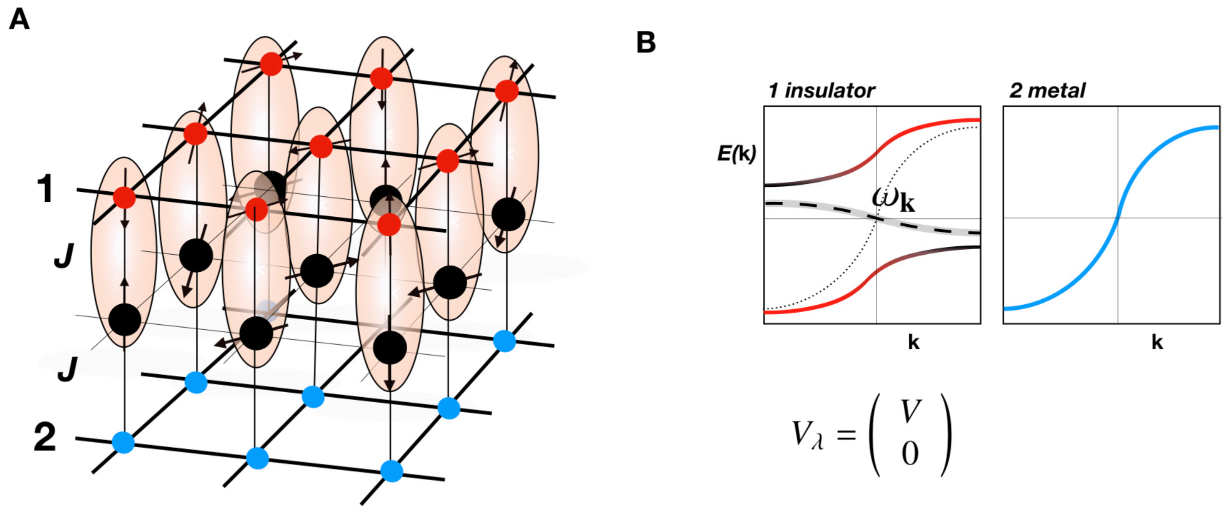

FIG. 4: (**A**) In a 2CK lattice model at $d = 2, 3$ dimensions we expect order fractionalization to lead to spontaneous channel symmetry breaking, forming Kondo singlets in one channel, with a channel-spinor defining the selective hybridization of the fractionalized local moments. (**B**) The channel spinor leads to a Kondo insulator in the hybridized channel (1) with a hybridization gap. The dashed line indicates the dispersion $\omega_{\mathbf{k}}$ of the emergent f-electron. Channel 2 remains a gapless insulator.

Discussion: Spontaneous Order Fractionalization. Induced order fractionalization in the two-channel Kondo impurity model enables us to conjecture spontaneous order fractionalization in lattice models. The key idea is that order fractionalization at one site produces a channel-symmetry breaking Weiss field at its neighbors. This then provides positive feedback that allows a fractionalized ordered state to develop spontaneously due to the fragile nature of the critical system. The NRG results confirm that the path integral gauge theory approach provides a qualitatively correct description of the fractionalization at a mean-field level, which can then be used to describe spontaneous order fractionalization in a Kondo lattice. As in conventional mean-field theories, the Gaussian order parameter fluctuations about such a mean-field theory are finite in dimensions $d \geq 2$, allowing a stable spontaneous order fractionalization [51, 52]. Thus in the two-channel Kondo lattice at half filling, spontaneous OF means that the system behaves as a Kondo insulator in one channel, remaining metallic in the other. With this reasoning, symmetry-breaking phases of this type characterized in dynamical mean-field theory calculations [32, 53–55] can be interpreted as order-fractionalized phases.

Kondo Model	3 body state	Composite Fermion	Induced Order	Asymmetry Ψ	OF
One Channel	$(\vec{\sigma}\cdot\vec{S})_{\alpha\beta}\psi_\beta$	$V\ f_\beta$	(Fermi Liquid)	—	—
		$(\vec{\sigma}\cdot\vec{\eta})_{\alpha\beta}\mathcal{V}_\beta$	Odd ω pairing [36, 56]	$\psi_\uparrow\psi_\downarrow S^+ + \mathrm{H.c}$	$\mathcal{V}_\uparrow\mathcal{V}_\downarrow + \mathrm{H.c}$
Two channel	Spin $(\vec{\sigma}\cdot\vec{S})_{\alpha\beta}\psi_{\lambda\beta}$	$V_\lambda f_\alpha$	Channel FM [34, 55]	$\psi^\dagger_\lambda(\vec{\sigma}\cdot\vec{S})\psi_{\lambda'}$	$\bar{V}_\lambda V_{\lambda'}$
		$V_\lambda f_\alpha + \Delta_\lambda\tilde{\alpha}f^\dagger_{-\alpha}$	Composite pairing [13, 37]	$\psi_1(\vec{\sigma}\cdot\vec{S})\sigma_2\psi_2$	$V_1\Delta_2 - V_2\Delta_1$
	Quadrupolar $(\vec{\gamma}\cdot\vec{S})_{\lambda\lambda'}\psi_{\lambda'\alpha}$	$V_\alpha f_\lambda$	Hastatic Order [33]	$\psi^\dagger_\alpha\vec{\sigma}(\vec{\gamma}\cdot\vec{S})\psi_\beta$	$\bar{V}_\alpha V_\beta$

TABLE I: Showing the different patterns of fractionalization in single-channel and two-channel Kondo systems, derived from both mean-field decouplings, as referenced, and the strong-coupling limit of the corresponding impurity Kondo model $H = H_I + \delta J\Psi$. In the table on the third line, $\vec{\eta} = (\eta_1, \eta_2, \eta_3)$ denotes a vector of three emergent Majorana fermions that fractionalize the local moment spin, $\vec{S} \equiv -\frac{i}{2}\vec{\eta}\times\vec{\eta}$ [36]. On the fifth line $\tilde{\alpha}$ denotes $\tilde{\alpha} \equiv \mathrm{sign}\alpha$.

In our NRG studies we have highlighted just one example of how broken channel symmetry induces order fractionalization. By varying the asymmetry field Ψ we can induce a variety of different kinds of fractionalized order (Table I). In principle one could repeat an NRG calculation for each of these cases; however we can determine the pattern of fractionalization by using a fractionalized mean-field theory or by analyzing the structure of the extreme strong-coupling asymmetry limit. A simple generalization is to the quadrupolar Kondo model, where the roles of channel and spin are reversed. Here the fractionalized excitations carry the channel index $\left[(\vec{S}\cdot\vec{\sigma})\psi\right]_{\lambda\alpha} \to V_\alpha f_\lambda$ while the order parameter V_α is a spinor that breaks time reversal symmetry, an example of the "hastatic" hidden order proposed for phases of URu_2Si_2 [33] and $PrTi_2Al_{20}$ [34]. The rich symmetry structure of the 2CK [57], allows one to explore a wide class of symmetry breaking operators Ψ, with many possible fractionalization patterns. If one employs a Nambu formalism, writing $\tilde{\psi}_{\lambda\sigma+} = \psi_\sigma$ and $\tilde{\psi}_{\lambda\sigma-} = \mathrm{sgn}\sigma\psi^\dagger_{\lambda-\sigma}$, the conduction electron $\tilde{\psi}_{\lambda\sigma\tau}$ now carries three indices, channel, spin and isospin (λ, σ, τ). Adding a composite pair asymmetry term to the Kondo model induces Andreev reflection off the Kondo impurity, and the composite fermion is now a mixture of particle and hole; this results in composite two-channel pairing [37], a form of order hypothesized for the heavy fermion superconductor $NpPd_2Al_5$ [38]. We also note that by adding a composite triplet pairing term to the Hamiltonian $\Psi = \psi_\uparrow\psi_\downarrow S^+ + \mathrm{H.c}$ or more generally, by replacing the spin density operator $\psi^\dagger\vec{\sigma}\psi$ of the conduction sea (at the origin) by a combination of conduction spin and isospin operators $\tilde{\psi}^\dagger\vec{\sigma}\tilde{\psi} \to \tilde{\psi}^\dagger(\vec{\sigma}+\lambda\vec{\tau})\tilde{\psi}$ [58], we can induce an odd-frequency paired state in which the local moment fractionalizes into Majorana fermions [36, 59]. Such a state has been recently proposed as a candidate for the strange insulating state in SmB_6 [60].

We can also envisage spontaneous order fractionalization in broader contexts beyond Kondo lattices; for example in the Hubbard model where the combination of spin and fermion is now replaced by a three fermion bound-state. In this more general situation, the OF will involve the fractionalization of a three-fermion bound-state into two components: a bosonic "*corona*" surrounding a *"dark fermion"* located at the center-of-mass. This process has the effect of partitioning the quantum numbers $\Lambda = (\{\lambda\}, \{\alpha\})$ of three-body composite, into two parts, the λ variables reside exclusively in the bosonic corona, while the α variables reside in the dark fermion. The order fractionalization conjecture for this general case takes the form

$$\left(\psi\psi\psi\right)_\Lambda(x) = V^\lambda_{\alpha\alpha'}(x) f_{\alpha'}(x). \tag{8}$$

Here $(\psi\psi\psi)_\Lambda(x)$ corresponds to a combination of creation or annihilation operators with center of mass x, that transform under fundamental representations of the Λ. $V^\lambda_{\alpha\alpha'}(x)$ and $f_\alpha(x)$ are the order parameter corona and the dark fermion respectively. In the simplest cases, $V^\lambda_{\alpha\beta} = V^\lambda\delta_{\alpha\beta}$ is diagonal, and the corona and dark fermion share a common $U(1)$ gauge symmetry. The general matrix structure of the order fractionalization allows for a non-abelian partition of the quantum numbers, with an internal SU(N) gauge symmetry associated with the quantum numbers α. This more general form is required to understand the example of composite pairing shown in Table. I.

The OFC also implies that the corresponding self-energy factorizes as follows

$$\Sigma_{\Lambda\Lambda'}(2,1) \xrightarrow{|2-1|\to\infty} \bar{V}_\lambda(2) g(2-1) V_{\lambda'}(1). \tag{9}$$

where $g(2-1)$ is the one-particle propagator of the dark fermion. In a lattice, the dark fermions will generically delocalize with dispersion $\omega_{\mathbf{k}}$, forming a Fermi surface $\mathbf{k} \in \{\mathbf{k}^*_F\}$ where $\omega_{\mathbf{k}^*_F} = 0$ vanishes. In space time, the asymptotic Green's function of the dark Fermions,

$$g(\vec{x}, t) \sim \delta_{\alpha\alpha'} \frac{e^{i\vec{k}^*_F \cdot \vec{x}}}{x - v_F(\hat{x})t}, \tag{10}$$

where $\vec{k}^*_F(\hat{x}) = \mathbf{k}^*_F$ is the Fermi wavevector at the extremal point on the Fermi surface where the group velocity $\vec{v}_F = v_F \hat{x}$ is parallel to $\vec{x}$. This defines a kind of "light cone" on which g is arbitrarily large. The factorization of the self-energy into a product of spinors is the conjectured outcome of order fractionalization in a fermionic system, and constitutes a generalization of the concept of off-diagonal long range order into the time domain. We also note that the singularity $\Sigma_{\lambda,\lambda'}(\omega, \mathbf{k}^*_F) \sim \bar{V}_\lambda V_{\lambda'}/(\omega - \omega_{\mathbf{k}})$ in the self-energy at the dark Fermi surface leading to zeroes in the electronic Green's function $G(\omega, \mathbf{k}^*_F) = 0$ [61, 62]. There is an interesting possible link with singularities in the electron self-energy observed in cluster dynamical mean-field studies of the Hubbard model [63] and also proposed as a phenomenological explanation of Fermi arcs in under-doped cuprate superconductors [64, 65].

Conventional and fractionalized order can be delineated in various ways. There are a number of quantum materials, including $NpPd_5Al_2$, $CeCoIn_5$, UBe_{13}, PrV_2Al_{20} and $PrTi_2Al_{20}$ where spin or quadrupolar Kondo effects coincide with phase transitions into broken symmetry states. An important "fingerprint" of fractionalization is the appearance of dark fermionic bound-states, that may be detected using spectroscopies such as angle resolved photoemission (ARPES) or scanning tunneling microscopy (STM). For example if in $CeCoIn_5$ the Kondo effect coincides with the development of superconductivity, then STM should detect an expansion of the Fermi surface at the superconducting transition; in neutral cases the thermal conductivity would be an ideal probe for this Fermi surface change.

An intriguing question is whether the different topologies of fractionalized order can be detected experimentally. For example in UBe_{13} and Pr $(V,Ti)_2Al_{20}$ where the channel index is spin, it may be possible to externally manipulate the channel-symmetry breaking Kondo-effect: rotating the order spatially through 360° to create a π phase shift may be detected in a channel interferometer; rotating the order in time using optical methods may lead to a breathing Fermi surface which might be measured using a channel-selective conductivity.

We end by noting that despite the spin-statistics theorem, relativistic versions of order fractionalization are possible since Lorentz invariance does not prohibit bosons that carry half-integer isospin. A classic example is the Higgs boson, a spinor which carries half-integer weak isospin and could conceivably emerge as a fractionalized order parameter of more fundamental Fermi fields.

In conclusion we have presented a mechanism for the fractionalization of order parameters through the formation of fermionic poles in the self-energy; this enables long-time factorization of the self-energy into products of order parameters transforming under the fundamental representation of the symmetry group. We have provided crucial substantiating examples in the single and two-channel impurity Kondo models. These results have led us to conjecture that such phenomena may appear spontaneously in a lattice as suggested by several experimental, mean-field and computational results.

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I. SUPPLEMENTARY MATERIAL

This section contains additional details and proofs for key statements in the paper. Section A contains details of the Numerical Renormalization Group (NRG) calculation. Section B contains a derivation of the fermionic pole in the self-energy of the Kondo problem using Fermi-liquid theory, in the single-channel and channel-asymmetric two-channel Kondo models. Section C contains a proof of the Berry phase accumulated by the ground state under an adiabatic time-evolution, establishing the spinorial character of the order parameter.

A. Details of the NRG calculations

NRG calculations were performed using the density-matrix NRG code [66] with a flat density of states, which produces the imaginary part of the local Green's functions (e.g. $G(z) = \langle\langle\psi_\sigma; \psi_\sigma^\dagger\rangle\rangle_z$) at discrete frequencies determined by the Wilson discretization as follows

$$-G''(\omega + i\eta) = \frac{1}{Z}\sum_{n,m} |\left\langle m|\psi_\sigma^\dagger|n\right\rangle|^2 \pi\delta(\omega + E_n - E_m)(e^{-\beta E_m} + e^{-\beta E_n}), \tag{11}$$

where $|m\rangle$ and $|n\rangle$ are many-body eigenstates with energies E_m and E_n, respectively, and $Z = \sum_n e^{-\beta E_n}$ is the partition function. Our methodology takes account of the $\mathrm{SU}_{charge}(2)\times\mathrm{SU}_{spin}(2)$ and $\mathrm{SU}_{spin}(2)\times\mathrm{SU}_{charge1}(2)\times\mathrm{SU}_{charge2}(2)$ symmetries of the one and the two-channel models, respectively to simplify calculation of the matrix elements. The calculations employed 750 multiplets and a Wilson parameter $\Lambda = 1.8$, with a Wilson chain length $L = 120$. The full-density matrix calculation ensures that the discrete results (11) satisfy the sum-rules enforced by the commutation relations. The code uses an interpolative log-Gaussian broadening, with the Kernel defined as follows [66, 67]

$$\begin{aligned} K_{int}(\omega, \omega_i) &= \frac{1}{b\sqrt{\pi}} e^{-[x(\omega) - x(\omega_i)]^2/b^2} \frac{dx}{d\omega_i}, \\ x(\omega) &= \frac{1}{2}\tanh(\omega/T_Q)\log[(\omega/T_Q)^2 + e^\gamma] \end{aligned} \tag{12}$$

where the 'quantum temperature' T_Q is chosen to be 10^{-15} and $\gamma \approx T_Q$ for our zero temperature calculations. We have taken the logarithmic broadening parameter b to be $b = 0.6$. This leads to

$$-G''(\omega + i\eta) = \frac{1}{Z}\sum_{n,m} |\left\langle m|\psi_\sigma^\dagger|n\right\rangle|^2 \pi K(\omega, E_m - E_n)(e^{-\beta E_m} + e^{-\beta E_n}). \tag{13}$$

The normalization

$$\int d\omega K_{int}(\omega, \omega_i) = 1 \tag{14}$$

ensures that the sum-rules are satisfied. Following the standard approach, the Hilbert transform is applied to the broadened data to obtain both real and imaginary parts of the Green's function.

We compute the irreducible self-energy for the Kondo problem from the conduction electron Green's function $G(z)$. In principle, the self-energy can be calculated directly from the relation

$$\Sigma(z) = g^{-1}(z) - G^{-1}(z) \tag{15}$$

where $g(z)$ is the bare conduction electron propagator in the absence of the impurity ($J = 0$). However, at the Fermi energy $G^{-1}(z)$ is singular and so this expression requires careful regularization.

In practice, we found it easier to divide the calculation into two parts. First we calculated the electron T-matrix $T(z)$, defined by the relation

$$G(z) = g(z) + g(z)T(z)g(z) \tag{16}$$

from which we obtain

$$T(z) = \frac{G(z) - g(z)}{g(z)^2}. \tag{17}$$

The regularized self- energy was then calculated from the T-matrix, using the relationship

$$\Sigma(z) = \lim_{\epsilon\to 0} \frac{T(z)}{1 + (1-\epsilon)g(z)T(z)}, \qquad g(z) = \langle\langle\psi_\sigma;\psi_\sigma^\dagger\rangle\rangle_z\Big|_{J=0}. \tag{18}$$

This limiting procedure was used to ensure that the $\Sigma(z)$ has the correct analytical properties. A finite ϵ leads to some residual broadening of the peak. In our calculations, we used $\epsilon = 10^{-8}$.

B. Fermionic pole in the Kondo self energy: relationship to Fermi-liquid theory

We can gain analytic insight into our results using Fermi-liquid theory. From Fermi-liquid theory [49, 68] we know that the phase shifted quasi-particles have a Fermi-liquid interaction with a single scale: the Kondo temperature. This gives rise to a low energy scattering T-matrix of the form

$$-\pi\rho T(\omega + i\eta) = i - \frac{\omega}{T'_K} - i\gamma\frac{\omega^2}{T'^2_K} + O(\omega^3), \tag{19}$$

where T'_K is proportional to the weak-coupling Kondo temperature $T_K \sim D\sqrt{J\rho}e^{-1/\rho J}$. Inserting this expression, together with a flat density of states $g(\omega + i\eta) = -i\pi\rho$ into (18), the corresponding irreducible self-energy is then

$$\pi\rho\Sigma(\omega + i\eta) = \left[\frac{1}{\pi\rho T(\omega + i\eta)} - i\right]^{-1} = \frac{T'_K}{\omega + i(\gamma - 1)\omega^2/T'_K + i\eta}. \tag{20}$$

This result is plotted in Fig. 5. The background beneath the delta-function pole is thus understood as a result of the ω^2 Fermi liquid scattering rate. The parameter γ determines

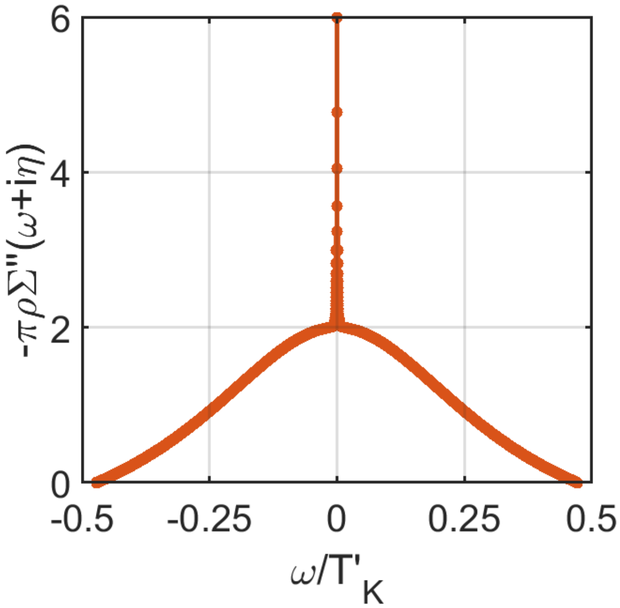

FIG. 5: The irreducible self-energy extracted from Fermi-liquid theory. The background is a result of the inelastic scattering of the composite quasiparticle at finite energy as shown in Eq. (20).

the value of background offset $-\pi\Sigma''(\omega \to 0) = \gamma - 1$, and does not affect any qualitative features of the discussion. In order to recover our numerical results $-\pi\Sigma''(\omega \to 0) = 2$, one must set $\gamma = 3$ here. This is a factor of two larger than the value of γ derived in [49]. The origin of this discrepancy is unclear to us at this point.

For the two-channel Kondo impurity model with channel symmetry, the T-matrix at $T = 0$ is given by [49]

$$-\pi\rho T(\omega + i\eta) = i/2 + O(\sqrt{\omega}) \tag{21}$$

which is responsible for the $-\pi\rho\Sigma''(0 + i\eta) = 1$ at the 2CK fixed point. In the presence of channel asymmetry, the system flows to a local Fermi liquid fixed point with resonant scattering in the strongest channel. The Fermi liquid temperature is given by [69]

$$T_* = T_K\kappa^2, \qquad \kappa^2 = 4\frac{(\rho J_1 - \rho J_2)^2}{(\rho J_1 + \rho J_2)^4}, \tag{22}$$

in the scaling limit $D \to \infty$ and $\rho J_i \to 0$. The latter limit has to be taken such that κ^2 remains finite, so that T_*/T_K remains finite. Our extracted T_* vs. $\rho\delta J$ is shown in the left inset of Fig. 3**B** and it agrees with this formula (shown as a broken line). At $T = 0$ and $|\omega| \ll T_*$ the self-energy in the two-channels are distinctively different. The self-energy of the stronger channel contains a sharp pole on top of the Fermi liquid background, whereas the weaker channel only contains a Fermi-liquid ω^2 contribution. The result can be fit with the following formula:

$$-\pi\rho\Sigma(\omega+i\eta) = \begin{pmatrix} -[(\omega+i\eta)/T^* + 2i\omega^2/T_*^2]^{-1} & 0 \\ 0 & 2i\omega^2/T_*^2 \end{pmatrix}, \tag{23}$$

C. Berry phase calculation

In this section, we calculate the Berry phase associated with a slow time-dependent change in the channel asymmetry of the two channel Kondo model. Since the Berry phase is a topological quantity, it is independent of coupling strength, and we can carry out this calculation in the strong-coupling limit of the model, given by

$$H[\hat{n}] = H_I + \Delta J \hat{n} \cdot \vec{\Psi}$$

where $H_I = J \sum_\lambda \psi_\lambda^\dagger \vec{\sigma} \psi_\lambda \cdot \vec{S}$ is the symmetric Kondo interaction and

$$\vec{\Psi} = \psi_\lambda^\dagger \vec{\alpha}_{\lambda\lambda'} (\vec{\sigma} \cdot \vec{S}) \psi_{\lambda'} \tag{24}$$

is the "channel magnetization", where $\vec{\alpha}$ are a set of Pauli matrices in channel space and

$$\vec{n}(t) = (\sin\theta_t \cos\phi_t, \sin\theta_t \sin\phi_t, \cos\theta_t). \tag{25}$$

is the time-dependent asymmetry field. We have replaced $\delta J \to \Delta J$ to denote a large finite value of the channel asymmetry. Provided ΔJ and J are much larger than the electron band-width, we can ignore everything except the one-site Hamiltonian. At $\Delta J = 0$, the ground-state is an over-screened local moment with ground-state energy $E = -2J$. Beyond a critical asymmetry, a channel asymmetric singlet state with energy $-\frac{3}{2}(J+\Delta J)$ is stabilized. This requires that $\Delta J > J/3$.

We can parameterize the asymmetry field using a CP^1 representation $\hat{n} = \bar{z}\vec{\alpha}z$, where

$$z[\hat{n}] = \begin{pmatrix} z_1 \\ z_2 \end{pmatrix} = \begin{pmatrix} \cos\theta/2 \\ \sin\theta/2 e^{i\phi} \end{pmatrix}. \tag{26}$$

Then the ground-state for a particular fixed value of $\hat{n}$ can be written as

$$
\begin{aligned}
|z\rangle &= \sum_{\lambda=1,2} |\lambda_s\rangle\, z_\lambda, \\
|\lambda_s\rangle &= \frac{1}{\sqrt{2}} \left(|\lambda \uparrow, \Downarrow\rangle - |\lambda \downarrow, \Uparrow\rangle \right)
\end{aligned}
\tag{27}
$$

Here the $\Uparrow, \Downarrow$ refer to the spin state of the local moment and $|\lambda \uparrow\rangle, |\lambda \downarrow\rangle$ refer to the spin state of the electron in channel λ. At the site of the moment, the weaker channel is either empty or doubly occupied, so that its spin is shut down. This charge state in the weaker channel $\alpha\, |0_{\bar{\lambda}}\rangle + \beta\, |2_{\bar{\lambda}}\rangle$ forms an isospin variable in the charge sector which, since it commutes with the Hamiltonian, is not relevant for this discussion and we do not include it in $|\lambda_s\rangle$.

When evolved adiabatically, the spin singlet follows the direction of the channel asymmetry. If the applied field $\hat{n}(t = T) = \hat{n}(t = 0)$ returns to its original direction, the state returns to its original ground-state, up to a finite phase. The state at time t can be written as

$$
|\psi(t)\rangle = e^{-i\alpha(t)}\, |z(t)\rangle \tag{28}
$$

where

$$
\alpha(t) = \int_0^t dt' E(t') + \gamma(t). \tag{29}
$$

Here the first term is the Schrödinger phase accumulation associated with the energy, which in our case is constant $E(t') = E$. The second term $\gamma(t)$ is the Berry phase. Inserting this into the time-dependent Schroedinger equation

$$
i\hbar\partial_t\, |\psi(t)\rangle = H[\vec{n}(t)]\, |\psi(t)\rangle \tag{30}
$$

we find

$$
\partial_t\, |z(t)\rangle - i\dot{\gamma}\, |z(t)\rangle = 0 \tag{31}
$$

Multiplying from left by $\langle \bar{z}|$ we find

$$
\dot{\gamma} = -i\, \langle \bar{z}|\partial_t|z\rangle \tag{32}
$$

From Eq. (26)

$$
-i\, \langle \bar{z}|\partial_t|z\rangle = -i\bar{z}(t)\partial_t z(t) = \dot{\phi}\frac{1-\cos\theta}{2} = \frac{1}{2}\dot{\Omega} \tag{33}
$$

where $\dot{\Omega}$ is the rate at which the vector $\hat{n}$ sweeps out solid angle in channel space. The total accumulated Berry phase associated with a closed path in channel space is then

$$
\gamma = \frac{1}{2}\int dt\dot{\Omega} = \frac{1}{2}\Omega \tag{34}
$$

where Ω is the total solid angle subtended by the path. This has the general form of $\gamma = \Omega S$. The fact that we find the pre-factor $S = 1/2$ shows the spinorial character of the ground state and the underlying order parameter.

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