Three-Body Bound States and the Development of Odd-Frequency Pairing

P. Coleman,[1] E. Miranda,[1] and A. Tsvelik[2]
[1]Serin Physics Laboratory, Rutgers University, P.O. Box 849, Piscataway, New Jersey 08855
[2]Department of Physics, Oxford University, 1 Keble Road, Oxford OX1 3NP, United Kingdom
(Received 9 June 1994)

We propose that the development of odd-frequency superconductivity is driven by the growth of an anomalous three-body scattering amplitude. Using this as an ansatz we develop a mean-field theory for odd-frequency pairing within the Kondo lattice model. The three-body bound-state formation leads to the formation of a gapless band of strongly paired quasiparticles whose spin and charge coherence factors vanish linearly with energy. Possible links with heavy fermion superconductors are discussed.

PACS numbers: 74.70.Tx, 74.25.Ha, 75.30.Mb

Although three-body bound states are ubiquitous in many branches of physics, little is known about their role in collective condensed matter behavior. According to current wisdom, quantum phase transitions are an exclusive response to the growth of anomalous two-body scattering amplitudes. Here we discuss the possibility of phase transitions driven by an instability in a three-fermion channel [1]. We are led to propose this as a mechanism for the development of odd-frequency superconductivity, where the gap function is an odd function of frequency $\underline{\Delta}(\omega, k) = -\underline{\Delta}(-\omega, k)$ [2,3].

Our interest in this topic is physically motivated by heavy fermion superconductors (HFSC) [4,5], where superconductivity is intimately associated with the compensation of local moments. In these dense local moment systems, huge amounts of spin entropy are liberated by the condensation process: In UBe_{13}, for example, the condensation entropy $\Delta S = C_V(T_{c^+}) = 1$ J/K mol is of order $0.2R\ln 2$. To microscopically explain how the order parameter involves the local moment degrees of freedom is a major challenge.

A striking feature of these compounds if the failure of a single quasiparticle density of states to reconcile their thermodynamics and nuclear relaxation rates. Heavy fermion (HF) superconductors exhibit a universal T^3 dependence in their NMR and NQR relaxation rates $1/T_1 \propto T^3$, but exhibit no corresponding universality in their specific heat C_V. In superconducting UPt_3, for example, $C_v = \gamma_s T + BT^2$, [6] yet in UPd_2Al_3, $C_v = \gamma_s T + BT^3$ [7] despite a T^3 NMR or NQR relaxation rate over three to four decades of the relaxation rate [8,9]. This dichotomy appears to rule out the energy dependence of the density of states $N(\omega)$ as a root cause of the universal relaxation rates. Qualitatively, the NMR or NQR relaxation rate at temperature T scales as

$$(T_1)^{-1} \sim T[N(\omega)|\langle\omega|S_\pm|\omega\rangle|]^2_{\omega\sim k_BT}\,, \tag{1}$$

where

$$|\langle\omega|S_\pm|\omega\rangle|^2 = \overline{|\langle\mathbf{k}|S_\pm|\mathbf{k}'\rangle|^2\delta(\omega - E_\mathbf{k})\delta(\omega - E_{\mathbf{k}'})} \tag{2}$$

is a momentum average of quasiparticle spin-matrix elements. A T^3 NMR or NQR relaxation rate in the presence of a finite quasiparticle density of states $N(0) \sim \gamma_s$ leads us to speculate that in a HFSC, spin coherence factors must scale linearly with energy

$$|\langle\omega|S_\pm|\omega\rangle| \sim \omega\,. \tag{3}$$

In a BCS superconductor, the quasiparticles take the form $a_\mathbf{k} = \underline{u}_\mathbf{k}c_\mathbf{k} + \underline{v}_\mathbf{k}c^\dagger{}_{-\mathbf{k}}$, where

$$\begin{pmatrix}|\underline{u}_\mathbf{k}|^2\\|\underline{v}_\mathbf{k}|^2\end{pmatrix} = \frac{1}{2}\left[1 \pm \frac{1}{\sqrt{1 + (\Delta_\mathbf{k}/\epsilon_\mathbf{k})^2}}\right]. \tag{4}$$

Vanishing coherence factors occur when the magnitudes $|\underline{u}_\mathbf{k}|^2$ and $|\underline{v}_\mathbf{k}|^2$ are equal, i.e., when $\Delta_\mathbf{k}/\epsilon_\mathbf{k} \to \infty$. At a gap node $\Delta_\mathbf{k} = 0$, $\underline{u}_\mathbf{k}$ and $\underline{v}_\mathbf{k}$ are either unity or zero, so quasiparticles are unpaired and coherence factors are unity. Coherence factors that vanish at low energies thus require a fundamentally new type of theory where gapless quasiparticles are strongly paired. In this Letter we show how the development of an anomalous pole in a three-fermion channel leads to a singular gap function $\Delta_\mathbf{k}(\omega) \propto 1/\omega$, whose divergence at low energies enforces a linear energy dependence of coherence factors.

Our discussion hinges on a generalization of the concept of field contractions to three-body bound states. To illustrate this idea, consider the example of a ^{3}He atom: a bound state between a spin-$\frac{1}{2}$ nucleus and two electrons. In a many-body description, the ^{3}He atom is a bound-state pole in a three-fermion channel. Low-energy correlation functions of the bound fermions are determined by their factorization into three-body contractions

$$\hat{\psi}_\uparrow(\mathbf{1})\hat{\psi}_\downarrow(\mathbf{2})\mathcal{N}_\sigma(\mathbf{3}) = \int \Lambda(\mathbf{1},\mathbf{2},\mathbf{3};x)\hat{\Phi}_\sigma(x)dx\,, \tag{5}$$

where $\Phi^\dagger(x)$ creates the ^{3}He fermion at center of mass x, ψ represents the electron fields, $\mathcal{N}$ the nucleus, and Λ is the atomic wave function. The key observation is that a new Fermi field $\Phi_\sigma(x)$ is dynamically generated by the development of a bound-state pole. The atomic wave function Λ is a *three-body amplitude* that scatters incoming fermions into the bound state.

With this picture in mind, we are led to generalize the concept of three-body contractions to embrace the

0031-9007/95/74(9)/1653(4)$06.00

possibility of symmetry-breaking three-body amplitudes that act as collective order parameters. Consider a hypothetical bound triad of two electrons and a hole on a lattice [10]:

$$\psi_\alpha^\dagger(1)\psi_\beta(2)\psi_\gamma(3) = \sum_j \Lambda_{\alpha\beta\gamma}(1,2,3;j)\hat{\phi}_j\,. \tag{6}$$

Suppose the three-body amplitude $\Lambda_{\alpha\beta\gamma}$ is *complex* and carries the charge and spin of the bound state, transforming like the electron field ψ under gauge transformations. In this case, the residual fermionic pole at site j carries no phase and must be represented by a "real" fermion

$$\phi_j = \phi_j^\dagger\,. \tag{7}$$

A fermion of this type is a "Majorana fermion." Unlike a conventional fermion, its square is a pure number $\phi_j^2 = \frac{1}{2}\{\phi_j, \phi_j\} = \frac{1}{2}$; its bare Feynman propagator is proportional to the inverse frequency

$$\langle\phi_j(\omega)\phi_{j'}(-\omega)\rangle = \delta_{jj'}\frac{1}{\omega}\,, \tag{8}$$

and it is represented by a line without an arrow. A simple consequence is that electrons scattering into the three-body channel acquire an anomalous self-energy with a singular, odd-frequency pole $\Delta_{\mathbf{k}}(\omega) \propto 1/\omega$ (Fig. 1).

To make a link with heavy fermions consider the case where the three-body amplitude is symmetric in positions 2 and 3, so that $\Lambda_{\alpha\beta\gamma} = -\Lambda_{\alpha\gamma\beta}$. By contracting the spin indices, we find

$$[\mathbf{S}(1)\cdot\boldsymbol{\sigma}_{\alpha\beta}]\Psi_\beta(2) = \sum_j A_\alpha(1,1,2;j)\hat{\phi}_j\,, \tag{9}$$

where $\mathbf{S} = \frac{1}{2}\psi^\dagger\boldsymbol{\sigma}\psi$ is the spin density and $A_\alpha = \frac{1}{3}\epsilon_{\alpha\eta}\epsilon_{\beta\gamma}\Lambda_{\eta\beta\gamma}$ is a two-component spinor. This type of three-body bound state thus describes a collective binding of spins to electrons that is of particular interest in the contest of HFSC.

Though a three-fermion state cannot condense [11], the development of an anomalous three-body amplitude does imply off-diagonal long-range order. The square of Eq. (9) is a complex number, thanks to the Majorana character of the pole. When we square the left-hand side of this expression we may cast it as the expectation value of a *composite* operator

$$\langle\mathbf{S}(1)[\sigma\psi_{-\sigma}(2)\psi_\sigma(3)]\rangle = \sum_j \tilde{A}_j^T(1,2)i\sigma_2\boldsymbol{\sigma}\tilde{A}_j(1,3)\,, \tag{10}$$

where $\tilde{A}_j(1,2) \equiv A(1,1,2;j)$. Composite off-diagonal order of this type between spin and singlet pair density has been discussed in connection with odd-frequency pairing [3,12,13].

We illustrate how this idea of three-body bound states leads naturally to odd-frequency pairing within the Kondo lattice Hamiltonian,

$$H = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}}\psi_{\mathbf{k}}^\dagger\psi_{\mathbf{k}} + \sum_j H_{\rm int}[j]\,, \tag{11}$$

where $\psi_{\mathbf{k}}^\dagger$ is a conduction electron spinor, coupled to an array of $S = \frac{1}{2}$ local moments $\mathbf{S}_j$ via an antiferromagnetic exchange interaction

$$H_{\rm int}[j] = J(\psi_j^\dagger\boldsymbol{\sigma}\psi_j)\cdot\mathbf{S}_j\,. \tag{12}$$

Here ψ_j denotes the conduction electron in a tight-binding representation. This Hamiltonian provides a toy model for heavy fermion metals.

An electron scattering at site j couples directly to the three-body spinor $\xi_{j\alpha} = (\mathbf{S}_j\cdot\boldsymbol{\sigma}_{\alpha\beta})\psi_{j\beta}$. To examine the possibility of anomalous bound-state formation in this channel, we use the result $(\mathbf{S}\cdot\boldsymbol{\sigma})^2 = \frac{3}{4} - \mathbf{S}\cdot\boldsymbol{\sigma}$ to cast the interaction in the form

$$H_{\rm int}[j] = -J(\xi_j^\dagger\xi_j)\,. \tag{13}$$

We apply our bound-state ansatz to ξ_j by writing

$$-J\xi_j(t) = 2V_j\hat{\phi}_j(t) - J\delta\xi_j(t)\,, \tag{14}$$

where V_j is a two component spinor representing the anomalous three-body amplitude, and $\delta\xi_j$ represents fluctuations that are neglected in the mean-field theory. Next we substitute (14) into (13) so that $\sum_j H_{\rm int}[j] \rightarrow \tilde{H} + O(\delta\xi^\dagger\delta\xi)$, where

$$\tilde{H} = \sum_j 2\left[\left\{\psi_j^\dagger(\boldsymbol{\sigma}\cdot\mathbf{S}_j)\hat{\phi}_jV_j + \text{H.c.}\right\} + \frac{V_j^\dagger V_j}{J}\right]. \tag{15}$$

A remarkable result permits us to solve this mean-field theory, despite the trilinear combination of field operators. Consider the combination

$$\boldsymbol{\eta}_j = 2\mathbf{S}_j\phi_j\,. \tag{16}$$

Since ϕ_j commutes with the spin operator, it follows that these operators are real $\boldsymbol{\eta}_j = \boldsymbol{\eta}_j^\dagger$ and satisfy a canonical anticommutation algebra, $\{\eta_j^a, \eta_k^b\} = \delta^{ab}\delta_{jk}$. In other words, the fusion of ϕ_j with each spin-$\frac{1}{2}$ operator transmutes it into a fermion [14]. We can thus rewrite $\tilde{H}$ as a bilinear Hamiltonian

$$\tilde{H} = \sum_j\left[\psi_j^\dagger(\boldsymbol{\sigma}\cdot\boldsymbol{\eta}_j)V_j + \text{H.c.}\right] + \frac{2V_j^\dagger V_j}{J}\,. \tag{17}$$

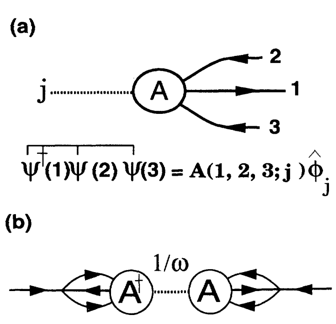

FIG. 1. Odd frequency pairing. Illustrating (a) anomalous amplitude for three-particle bound state and (b) anomalous pairing amplitude that scales as $1/\omega$ in this picture [12].

This type of Hamiltonian was previously derived by starting from a Majorana spin representation [12]. The current discussion enables us to link the appearance of fermionic spin modes and vanishing coherence factors with the development of bound states. For convenience, consider a cubic lattice. Here the mean-field free energy is minimized in staggered configurations, where $V_j = e^{i\mathbf{Q}\cdot\mathbf{R}_j/2}V$, $\mathbf{Q} = (\pi,\pi,\pi)$ [12]. The staggered phase may be absorbed by a gauge transformation of the electrons $\psi_{\mathbf{k}} \rightarrow \psi_{\mathbf{k}+\mathbf{Q}/2}$, which leads to the following mean-field Hamiltonian:

$$H_{\mathrm{MFT}} = \sum_{\mathbf{k}} \tilde{\epsilon}_{\mathbf{k}} \psi^{\dagger}_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\mathbf{k}} \Big[\psi^{\dagger}_{\mathbf{k}} (\boldsymbol{\sigma} \cdot \boldsymbol{\eta}_{\mathbf{k}}) V + \mathrm{H.c.} \Big] + N_s \frac{2V^{\dagger}V}{J},$$

where N_s is the number of sites and $\tilde{\epsilon}_{\mathbf{k}} = \epsilon_{\mathbf{k}-\mathbf{Q}/2}$. If we represent the propagator of the Majorana fermions by a dashed line without an arrow then the effect of the anomalous three-body scattering amplitude is to introduce vertices of the form

$$\overset{\beta}{\rightarrow}\!\!\triangle\!\!\overset{a}{- - -} = [V^{\dagger} \cdot \boldsymbol{\sigma}^{\alpha}]_{\beta}$$
$$\overset{a}{- - -}\!\!\triangle\!\!\overset{\alpha}{\rightarrow} = [\boldsymbol{\sigma}^{a} \cdot V]_{\alpha} . \qquad (18)$$

The main effect of these vertices is to introduce a singular pairing self-energy into the electron propagators

$$\overset{\beta}{\leftarrow}\!\!\triangle\!\!\overset{1/\omega}{- - -}\!\!\triangle\!\!\overset{\alpha}{\rightarrow} = [\underline{\Delta}(\omega)]_{\alpha\beta} .$$

If we write $V = (V_0/\sqrt{2})z_0$, where z_0 is a unit spinor, then the anomalous self-energy takes the form

$$\underline{\Delta}(\omega) = -\Delta(\omega)[\underline{z}_0 \otimes \underline{z}_0^T], \qquad (19)$$

where $\Delta(\omega) = V_0^2/2\omega$. By decomposing $\psi_{\mathbf{k}}$ into four Majorana components $\psi_{\mathbf{k}} = (1/\sqrt{2})[\psi^0_{\mathbf{k}} + i\boldsymbol{\psi}_{\mathbf{k}} \cdot \boldsymbol{\sigma}]\underline{z}$ we find that only the vector components hybridize with the spin fermions: $\tilde{H} = (V/2)\sum_{\mathbf{k}}[-i\boldsymbol{\psi}_{\mathbf{k}}{}^{\dagger} \cdot \boldsymbol{\eta}_{\mathbf{k}} + \mathrm{H.c.}]$. These components develop a gap $\Delta_g \sim V_0^2/D$, while the scalars remain gapless. Diagonalizing H_{MFT}, choosing $z_0^{\dagger} = (1,0)$, the explicit form of the gapless quasiparticles is

$$a_{\mathbf{k}} = \sqrt{Z_{\mathbf{k}}}[u_{\mathbf{k}}\psi_{\mathbf{k}\uparrow} + v_{\mathbf{k}}\psi^{\dagger}_{-\mathbf{k}\uparrow}] + \sqrt{1 - Z_{\mathbf{k}}}\,\eta^3_{\mathbf{k}} . \qquad (20)$$

Here

$$\begin{pmatrix} u^2_{\mathbf{k}} \\ v^2_{\mathbf{k}} \end{pmatrix} = \frac{1}{2}\left[1 \pm \frac{1}{\sqrt{1 + [\Delta(\omega)/\mu_{\mathbf{k}}]^2}} \right]_{\omega = E_k} , \qquad (21)$$

where $\mu_{\mathbf{k}}$ is the symmetric part of $\tilde{\epsilon}_{\mathbf{k}}$, $Z_{\mathbf{k}} = [1 + \mu^2_{\mathbf{k}}/V^2]^{-1}$ and $E_{\mathbf{k}}$ is the quasiparticle energy. Unlike a Cooper-paired superconductor, the divergence of the gap function at low frequencies leads to an equal weight of particle and hole at the Fermi energy. The coherence factor $\langle \omega | S_{\pm} | \omega \rangle \sim u^2(\omega) - v^2(\omega)$ grows like $\Delta(\omega)^{-1}$ and is hence linear in energy.

In a conventional superconductor, the spectral weight of the electrons that develop a gap is transferred to the condensate. Here, the consistency of this model requires that the gapped electrons combine to produce a low-energy three-body fermion. The original model is invariant under sign changes of the Majorana fermions $\phi_j \rightarrow -\phi_j$ ($\boldsymbol{\eta}_j \rightarrow -\boldsymbol{\eta}_j$). This symmetry is broken by the mean-field theory, which then allows for the possibility of "kinks" in time, when the V_j changes sign (Fig. 2). These topological objects give rise to gapless fermionic zero modes which we are able to identify as the three-body bound states [15].

The operator $\mathcal{P}_j$ that effects the Z_2 transformation $\mathcal{P}_j \boldsymbol{\eta}_j \mathcal{P}_j^{\dagger} = -\boldsymbol{\eta}_j$ is

$$\mathcal{P}_j = -4\Phi_j \eta^1_j \eta^2_j \eta^3_j , \qquad (22)$$

where Φ_j is an independent Majorana fermion introduced to make $\mathcal{P}_j$ a bosonic rather than fermionic operator. We can sum over all tunneling processes by assuming that the duration of a kink is extremely brief in comparison with the delay between kinks, permitting us to associate a fixed tunneling amplitude Γ with a kink. A kink at site k, time τ_{jk} ($j = 1, n_k$), may be introduced into the partition function by applying the operator $\mathcal{P}_j$ to the time evolution operator. The contribution to the partition function associated with a set of kinks is

$$A\{\tau_{jk}\} = \mathrm{Tr}\Big[T \prod_{j,k} \Gamma \mathcal{P}_j(\tau_{jk}) e^{-\int_0^{\beta} H_{\mathrm{MFT}}\, d\tau} \Big],$$

where the trace contains the trace over the Φ field. Contributions from paths with odd n_k are zero. The complete partition function is obtained by summing over all such paths

$$Z = \sum_{\{n_k\}} \prod_k \left(\frac{1}{n_k!} \prod_{j=1,n_k} \int_0^{\beta} d\tau_{jk} \right) A\{\tau_{jk}\} .$$

This expression is recognized as the expansion of a simple exponential, $Z = \mathrm{Tr}[e^{-\beta(H_{\mathrm{MFT}} + H_{\mathrm{int}})}]$, where

$$H_{\mathrm{int}} = -2\Gamma \sum_j \Phi_j \eta^1_j \eta^2_j \eta^3_j . \qquad (23)$$

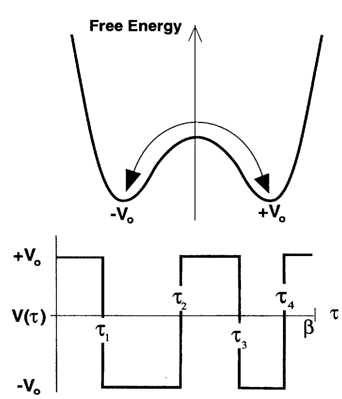

FIG. 2. Kink configuration. Configuration with four kinks in the order parameter V at a given site.

Thus by taking account of kinks, we reveal the three-body bound state, represented here by the Majorana fermion Φ_j. This particle couples to the triplet of gapped "vector" quasiparticles. Below the three-fermion threshold $3\Delta_g$, Φ_j propagates via the virtual excitation of quasiparticles above the gap, forming a sharp three-body band of width $\sim\Gamma^2/\Delta_g$.

In essence, we have followed the consequences of linear coherence factors to their logical conclusion. Should these ideas prove relevant to HFSC, then there are several interesting consequences. Vanishing coherence factors should lead to the development of a quadractic temperature or frequency dependence in a wide variety of response functions [transverse ultrasound attenuation, the depletion of the superfluid density $\Delta\rho_s$, and the quasiparticle conductivity $\sigma(\omega)$], despite a linear specific heat and an essentially isotropic thermal conductivity. In addition, a superconductor with vanishing coherence factors will exhibit a much larger Andreev reflection current than a gapless BCS superconductor. Furthermore, the absorption of an incoming electron into a three-body bound state within the condensate should result in the reflection of a particle *and* hole, creating a *diffuse Andreev scattering background* below T_c. Finally, it is worth noting that recent measurements on UPt_3 have observed a very large low-temperature specific heat anomaly [7]. Conservatively, this anomaly is a Pt nuclear ordering transition, however, it could conceivably be a signature of a narrow band of three-body bound states. In this speculative scenario, the large specific heat anomaly would occur without an NMR signature, but would coincide with a corresponding anomaly in the thermal conductivity.

We have attempted to elucidate the physics of odd-frequency superconductivity with the proposal that it is driven by the development of an anomalous three-body amplitude. Unlike a Cooper-paired superconductor, this type of superconductor involves the cooperative pairing of electrons and spins, and leads to the unique feature of gapless paired quasiparticles with vanishing coherence factors. We have speculated that this picture may prove useful in developing our understanding of heavy fermion superconductors.

We wish to particularly thank N. Andrei and L. Ioffe for their insights on the link between kinks in the semi-classical approach and three-body bound states [15]. Discussions with E. Abrahams and A. Ruckenstein are also gratefully acknowledged. This work was supported by NSF Grant No. DMR-93-12138, Grant No. GR/H-26680 from the EPSRC, UK, and CNPq, Brazil.

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