Massless Leggett Mode in Three-Band Superconductors with Time-Reversal-Symmetry Breaking

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The Leggett mode associated with out-of-phase oscillations of the superconducting phase in multiband superconductors usually is heavy due to interband coupling, which makes its excitation and detection difficult. We report on the existence of a massless Leggett mode in three-band superconductors with time-reversal-symmetry breaking. The mass of this Leggett mode is small close to the time-reversal-symmetry-breaking transition and vanishes at the transition point, and thus locates within the smallest superconducting energy gap, which makes it stable and detectable, e.g., by means of the Raman spectroscopy. The thermodynamic consequences of this massless mode and possible realization in iron-based superconductors are also discussed.

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Introduction.—Spontaneous breaking of a continuous symmetry and the associated low-energy collective excitation govern the physical properties in many systems ranging from condensed matter physics to particle physics. Superconductivity emerging as the spontaneous breaking of the $U(1)$ gauge symmetry is associated with a massless excitation known as Bogoliubov-Anderson-Goldstone (BAG) boson [1,2]. Coupled with an electromagnetic field, the BAG boson becomes the massive plasma mode due to the Anderson-Higgs mechanism.

Because of the discoveries of MgB_2 [3] and iron pnictides [4], it is now accepted that multicomponent superconductors are ubiquitous. Multiband superconductors are not always straightforward extensions of the single-band counterpart, and novel features may arise [5–10]. A famous example is the Leggett mode (LM) associated with the relative phase oscillation between different bands in a multicomponent superconductor (see Fig. 1), with the mass proportional to the interband coupling. [5] The LM was observed in MgB_2 with the Raman spectroscopy [9], and in the point-contact transport measurements [11]. The mass of the mode lies between the two superconducting energy gaps, which is consistent with the theoretical calculations [12]. The LM in MgB_2 therefore decays into a quasiparticle continuum associated with the band of a smaller energy gap.

For iron-based superconductors, many studies have revealed the sign-reversal pairing symmetry between different bands [13–16]. The system of more than three bands is somehow frustrated, and under appropriate conditions there may exist time-reversal-symmetry-breaking (TRSB) states even with conventional s-wave pairing symmetry, which involve nontrivial phase differences (i.e., $\delta\varphi \neq 0$ or π) among superconducting gaps [6,17–19]. With the new TRSB transition below T_c, the spectrum of collective excitations, and thus low-energy physical properties of the superconductors, should be modified significantly. It was reported that the LM may exist below the two-particle continuum in iron-based superconductors under appropriate conditions, [20] and that the mass of the LM may be reduced in some dynamical classes of multiple interband Josephson coupling in three-band superconductors. [21] We note that in other TRSB superconducting systems with mixed-symmetry order parameters with nodes such as $d + is$, a massive LM in the TRSB state was found in Ref. [22]. Having these recent progresses in mind, a question of fundamental interest arises: What are the effects of frustration on the LM and is it possible to have a massless LM?

In the present work, we demonstrate that the mass of the LM can be reduced significantly and even vanishes at the

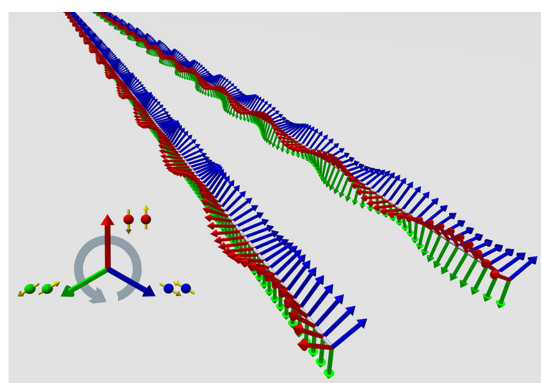

FIG. 1 (color online). Frustrated interband scatterings force Cooper pairs in different bands to carry different phases, which results in interband Josephson currents. Two dynamical modes associated with superconducting phases in three-band superconductors: the LM, where two phases oscillate out-of-phase while the third one stays unchanged, becomes massless at the TRSB transition (left), and the BAG mode, where all the three phases rotate in the same direction during the propagation of plasma wave in space (right).

TRSB transition upon tuning interband coupling or density of states in multiband superconductors. It is shown that the LM of the vanishing mass can be detected by Raman scattering, which also serves as smoking gun evidence for the TRSB transition. The appearance of massless excitations modifies superconducting properties qualitatively, such as changing an exponential temperature dependence of the electronic specific heat C_v predicted for fully gapped systems to a power-law one. Finally, we discuss how several recent experiments on iron-based superconductors can be explained by the existence of massless LM.

Leggett mode.—The Hamiltonian for three separate pieces of the isotropic Fermi surface can be written as

$$H = \sum_{l,\sigma} \int d^3 r \psi^\dagger_{l\sigma}(\mathbf{r})(\varepsilon_l - \mu)\psi_{l\sigma}(\mathbf{r}) - \sum_{j,l} \int d^3 r \psi^\dagger_{j\sigma}(\mathbf{r})\psi^\dagger_{j\bar{\sigma}}(\mathbf{r}) V_{jl} \psi_{l\bar{\sigma}}(\mathbf{r})\psi_{l\sigma}(\mathbf{r}), \quad (1)$$

where $\psi^\dagger_{l\sigma}(\psi_{l\sigma})$ is the electron creation (annihilation) operator in the lth band with the dispersion $\varepsilon_l(\mathbf{k})$ and the chemical potential μ and spin index σ. V_{jl} is the intraband for $l = j$ and interband for $l \neq j$ scattering, respectively, which can be either repulsive or attractive depending, for instance, on the strength of the Coulomb and electron-phonon interaction. The interband repulsion may cause frustration of the superconductivity in different bands and results in TRSB [6,17]. Introducing the Nambu spinor operator $\Psi_j = (\psi_{j\uparrow}, \psi^\dagger_{j\downarrow})^T$ and the energy gap Δ_j through the Hubbard-Stratonovich transform, we arrive at the following action in the imaginary time representation after integrating out the fermionic fields [23]:

$$S = \int d\tau d^3 r \sum_{j,l}^{3} \Delta_j g_{jl} \Delta_l^* - \sum_j \mathrm{Tr}\ln \mathcal{G}_j^{-1}, \quad (2)$$

with $\hat{g} = \hat{V}^{-1}$ and the Gor'kov green function

$$\mathcal{G}_j^{-1} = -\begin{pmatrix} \partial_\tau + (\varepsilon_j - \mu) & -\Delta_j \\ -\Delta_j^* & \partial_\tau - (\varepsilon_j - \mu) \end{pmatrix}. \quad (3)$$

The superconducting energy gaps at $T = 0$ are given by

$$\sum_{l=1}^{3} \Delta_l g_{lj} = N_j(0)\Delta_j \sinh^{-1}\left(\frac{\hbar\omega_{cj}}{|\Delta_j|}\right), \quad (4)$$

with $N_j(0)$ the density of states (DOS) at the Fermi surface in the normal state. Here ω_{cj} is a cutoff frequency and depends on the pairing mechanism. For electron-phonon coupling, ω_{cj} is the Debye frequency.

For a demonstration of our basic idea, we take a set of simplified interband couplings [24],

$$\hat{g} = \frac{1}{V}\begin{pmatrix} \alpha & 1 & 1 \\ 1 & \alpha & \eta \\ 1 & \eta & \alpha \end{pmatrix}, \quad (5)$$

and assume that the DOS and the cutoff frequencies are identical for three bands [$N_j(0) = N$, and $\omega_{cj} = \omega_c$] [17]. The prediction of a massless LM, however, is not restricted to the specific choice of $\hat{g}$ as discussed later. Here $g_{ij} > 0$ corresponds to a repulsive interaction. We take Δ_1 as positive real, and $\Delta_2 = \Delta e^{i\varphi}$, $\Delta_3 = \Delta e^{-i\varphi}$ because they are symmetric under the condition of Eq. (5). Hereafter, we take $\hbar\omega_c$ as the unit for Δ_l.

For a small η, the interband repulsion g_{12} and g_{13} dominates and the system takes $\varphi = \pi$. For a large η, a state with a finite phase difference between Δ_2 and Δ_3 appears, corresponding to a state of TRSB where $(\Delta_1, \Delta_2, \Delta_3) \neq (\Delta_1, \Delta_2, \Delta_3)^*$, even apart from the common phase factor. In the TRSB state, the energy gaps are given by

$$\Delta_1 = 1/\sinh\left(\frac{\alpha\eta - 1}{\eta}\frac{1}{NV}\right) \quad \text{and} \quad \Delta = 1/\sinh\left(\frac{\alpha - \eta}{NV}\right), \quad (6)$$

and $\cos\varphi = -\Delta_1/(2\eta\Delta)$. The system undergoes a second-order TRSB transition at η_c given by $\eta_c = \Delta_1(\eta_c)/[2\Delta(\eta_c)]$, as shown in Figs. 2(a) and 2(b).

We proceed to investigate phase fluctuations at the TRSB point, where the amplitudes of the superconducting gap can be considered as rigid [25]. For this purpose, we perform the following gauge transformation which separates the phase and amplitude of the gap [12,27]:

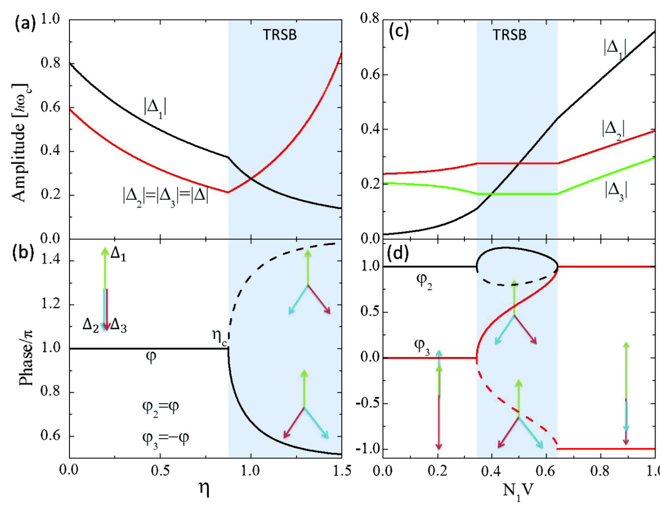

FIG. 2 (color online). Amplitudes and phases of order parameters at the TRSB phase transition, in (a) and (b) as a function of η, and in (c) and (d) as a function of DOS N_1V of the first component. Δ_1 is taken as real and positive. In (a) and (b), an identical DOS $NV = 0.5$ is taken for the three bands and $\alpha = 2$ in Eq. (5). In (c) and (d), $N_2V = 0.5$ and $N_3V = 0.4$, $\alpha = 2$ and $\eta = 1$ [see also Eq. (17)]. In the TRSB regime, there are two degenerate ground states $(\Delta_1, \Delta_2, \Delta_3)$ (solid lines) and $(\Delta_1^*, \Delta_2^*, \Delta_3^*)$ (dashed lines). The two solid lines for $N_1V > 0.64$ in (d) refer to the same state without TRSB.

$$\Delta_j \to |\Delta_j| e^{i\theta_j} \text{ and } \Psi_j(\tau, r) \to \begin{pmatrix} e^{i\theta_j/2} & 0 \\ 0 & e^{-i\theta_j/2} \end{pmatrix} \Psi_j(\tau, r), \tag{7}$$

and derive the action for the phase fluctuation

$$S = \int d\tau d^3 r \sum_{j,l} |\Delta_l| g_{lj} |\Delta_j| e^{i(\theta_l - \theta_j)} - \sum_j \mathrm{Tr}[\ln(\mathcal{G}_j^{-1} - \Sigma_j)], \tag{8}$$

where $\Sigma_j = -\frac{\hbar^2}{2m_j}(\frac{i}{2}\nabla^2\theta_j + i\nabla\theta_j\nabla)\sigma_0 + [i\frac{\partial_\tau\theta_j}{2} + \frac{\hbar^2}{8m_j} \times (\nabla\theta_j)^2]\sigma_3$ with σ_j being the Pauli matrices, σ_0 the unit matrix and m_j the electron mass [28,29]. From this action, one can obtain the time-dependent nonlinear Schrödinger Lagrangian for the phase fluctuations [30,31]. Considering small phase fluctuations around the saddle point $\phi_j = \theta_j - \varphi_j$ and expanding S up to the second order in ϕ_j, we have [24]

$$S_\phi[\phi_j] = \frac{1}{8} \sum_l \int d^3 q \hat{\phi}(-\Omega_l, -q)^T \mathbf{M} \hat{\phi}(\Omega_l, q), \tag{9}$$

with $\hat{\phi}(\Omega_l, q) \equiv [\phi_1(\Omega_l, q), \phi_2(\Omega_l, q), \phi_3(\Omega_l, q)]^T$ and

$$\mathbf{M} = \begin{pmatrix} P_1 - 2D_1 & D_1 & D_1 \\ D_1 & P_2 - D_1 - D_2 & D_2 \\ D_1 & D_2 & P_3 - D_1 - D_2 \end{pmatrix}, \tag{10}$$

with $D_1 = 8\Delta_1 \Delta \cos\bar{\varphi}/V$ and $D_2 = 8\eta\Delta^2 \cos(2\bar{\varphi})/V$ with $\bar{\varphi} \equiv \varphi_2 - \varphi_1 = \varphi_1 - \varphi_3$. $\Omega_l = 2l\pi k_B T$ with k_B the Boltzmann constant, and the excitations are bosons. In the hydrodynamic limit at $T = 0$, the dissipation is absent and $P_j = 2N(-\Omega^2 + 1/3 v_j^2 q^2)$ after the analytical continuation $i\Omega_l \leftarrow \Omega + i0^+$, where v_j is the Fermi velocity. From $\mathrm{Det}\mathbf{M} = 0$, we obtain the dispersion relations

$$\Omega_{\mathrm{BAG}}^2 = \tfrac{1}{3} q^2 v_j^2, \tag{11}$$

$$\Omega_{L-}^2 = -\frac{D_1 + 2D_2}{2N} + \frac{1}{3} q^2 v_j^2, \tag{12}$$

$$\Omega_{L+}^2 = -\frac{3D_1}{2N} + \frac{1}{3} q^2 v_j^2. \tag{13}$$

The first mode is the massless BAG mode corresponding to the uniform rotation of phases. The second and third are the LM Ω_{L-} and Ω_{L+} in the three-band system considered. Especially, the mode Ω_{L-} corresponds to the dynamics of the relative phase φ_{23} between the gaps of Δ_2 and Δ_3, and becomes massless at the TRSB transition depicted in Fig. 3. One may regard φ_{23} as the order parameter for the TRSB transition: It increases continuously from 0 at the transition, and therefore, the associated fluctuations become massless at the TRSB transition.

A magnetic field can be introduced into S_ϕ through the standard replacement $\nabla\phi_l \to \nabla\phi_l - 2\pi\mathbf{A}/\Phi_0$ with Φ_0 the flux quantum and $\mathbf{A}$ the vector potential. In this case, it is more convenient to rewrite the phase fluctuations in terms of ϕ_1, $\phi_{12} \equiv \phi_1 - \phi_2$, and $\phi_{13} \equiv \phi_1 - \phi_3$. ϕ_1 describes the BAG mode, and ϕ_{12} and ϕ_{13} correspond to the LMs. The gauge field couples with ϕ_1 in the form $(\nabla\phi_1 - 2\pi\mathbf{A}/\Phi_0)$. One may integrate out ϕ_1, resulting in the massive plasma mode due to the Anderson-Higgs mechanism. In contrast to the BAG mode, the LMs remain massless at the TRSB transition since ϕ_{12} and ϕ_{13} are decoupled from the gauge field $\mathbf{A}$.

In stark contrast to conventional symmetry-broken systems, there exist the stable massive LMs both before and after TRSB transition as shown in Fig. 3, because the relative phase between different condensates is fixed in both the states with and without TRSB.

Raman scattering.—Interband scatterings do not involve the gauge field; thus, the LMs do not respond to a magnetic field. However, the LMs are coupled indirectly with the electric field through the charge density, which renders them detectable by the Raman spectroscopy through the inelastic scattering of the photon with the charge density [32–35]. The interaction between the incident photon and the charge can be modeled as $\tilde{\rho}(\tau, q) = \sum_{j=1}^{3} \sum_{k,\sigma} \gamma_j(k) \psi_{j\sigma}^\dagger(\tau, k + \frac{q}{2}) \psi_{j\sigma}(\tau, k - \frac{q}{2})$, where $\gamma_j(k)$ is the scattering coefficient determined by the polarization of the incident and scattered photon. In the following, we derive the experimentally measurable Raman response function $\chi_{\tilde{\rho}\tilde{\rho}}(\tau - \tau', q) = -\langle T_\tau \tilde{\rho}(\tau, q) \tilde{\rho}(\tau', -q) \rangle$ with T_τ being a time-ordering operator. We introduce a source term coupled with $\tilde{\rho}$, $H_J(\tau) = -\sum_q \tilde{\rho}(\tau, q) J(\tau, -q)$ because $\chi_{\tilde{\rho}\tilde{\rho}}$ can be computed by the linear response theory with respect to J. The effective action in the presence of the incident photon reads [24]

$$S = \int d\tau d^3 r \sum_{l,j} \Delta_l g_{lj} \Delta_j^* - \sum_l \mathrm{Tr} \ln(\mathcal{G}_{J,l}^{-1} + \mathcal{G}_l^{-1}), \tag{14}$$

with $\mathcal{G}_{J,l}^{-1} = -\gamma_l(k) J(\tau, -q) \sigma_3$. For a weak incident wave, we may neglect the fluctuations of the amplitude of the

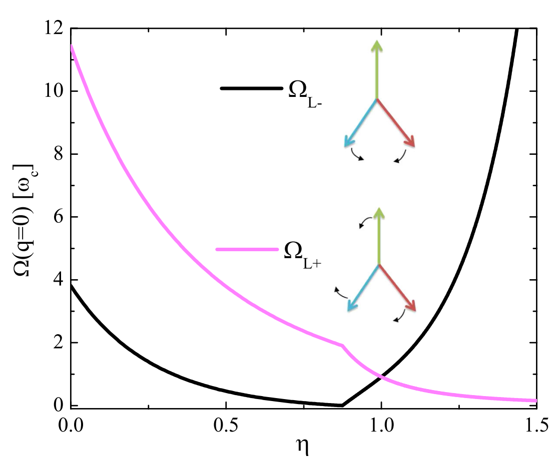

FIG. 3 (color online). Dependence of the masses of LMs on the interband coupling η. Here $NV = 0.5$ and $\alpha = 2$, and the masses are in units of ω_c.

order parameters, and the fluctuations for the superconducting phase acquires a form $S = S_\phi + S_J$, with S_ϕ defined in Eq. (9) and

$$S_J = \frac{1}{2}\sum_{j,q}[J(q)Z_j(q)\phi_j^T(-q) + J(-q)\tilde{Z}_j(-q)\phi_j(q) + J(q)J(-q)\Pi_{j,33}^{\gamma\gamma}], \tag{15}$$

where $Z_j(q) = \Delta_j[-\sin\varphi_j\Pi_{j,31}^{\gamma}(q) - \cos\varphi_j\Pi_{j,32}^{\gamma}(q)]$ and $\tilde{Z}_j(q) = \Delta_j[-\sin\varphi_j\Pi_{j,13}^{\gamma}(q) - \cos\varphi_j\Pi_{j,23}^{\gamma}(q)]$. The polarization functions are defined as $[\Pi_{j,ml}^{\gamma\gamma}, \Pi_{j,ml}^{\gamma}] \equiv 1/(L^3\beta)\sum_n \int d^3k \Upsilon_{j,ml}[\gamma_j(k+\frac{q}{2})\gamma_j(k-\frac{q}{2}), \gamma_j(k+\frac{q}{2})]$.

Integrating out the fluctuations ϕ_j, we then obtain the correlation function

$$\chi_{\tilde{\rho}\tilde{\rho}}(i\Omega, q=0) = \sum_j\{\Pi_{j,33}^{\gamma\gamma} - Z_j[\mathbf{M}^{-1}]_{jj}\tilde{Z}_j^T\}. \tag{16}$$

The first term gives the resonant scattering at $\Omega = 2\Delta_j$ and the second term accounts for the resonance with the LMs, as depicted in Fig. 4. When the energy difference between the incident and scattered photon matches the energy of the LMs, $\mathbf{M}^{-1}$ becomes singular and gives δ peaks in the spectroscopy. In reality, the delta-function peaks are rounded by both the damping effect and interactions between Leggett bosons when the oscillations of the LM become strong, which are neglected in Eq. (16). Although the response of a genuinely massless LM is hidden into the elastic scatterings, it can be traced out clearly if one changes η systematically and generates a LM of small mass, which can be achieved by electron or hole doping because the interband scattering is renormalized by the DOS as in Eq. (4).

Discussions.—At $T > 0$, the Landau damping by quasiparticles sets in and the lifetime of the LM decreases. A local time-dependent equation for the phase fluctuations does not exist due to the singularity of the DOS in the superconducting state [36]. In the vicinity of T_c, the dynamics of superconductivity can be described by the standard time-dependent Ginzburg-Landau equation [36]. In this region, the lifetime of the LM is much smaller than the inverse of its energy due to severe damping by quasiparticles; therefore, there are no well-defined Leggett excitations. Nevertheless, in the static case, the massless feature manifests as the divergence of the characteristic length for the relative phase variation in the vicinity of the TRSB [19].

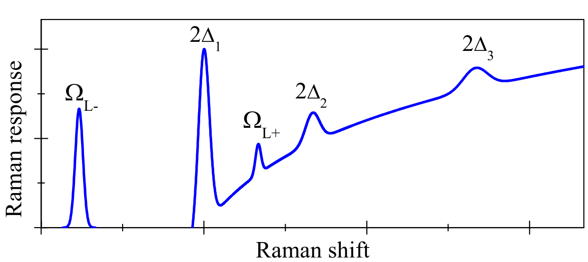

FIG. 4 (color online). Schematic view of the Raman response in three-band superconductors with TRSB. The finite linewidth of peaks is due to the damping and interaction between Leggett bosons. The background at an energy larger than $2\Delta_1$ is due to the quasiparticle excitations.

Let us discuss the applicability of our results to the iron-based superconductors. In order to demonstrate the mass reduction of the LM by TRSB, we adopt a simple and general BCS-like Hamiltonian in Eq. (1). An implicit expectation behind this treatment is that more realistic models would merely lead to quantitative corrections. It seems that this simplification is not far from the situation in some iron-based superconductors, since the s wave with sign-reversal ($s\pm$) pairing symmetry is favored by many experiments [15,16]. It also became clear recently that the interband hopping in single particle channels using a more realistic tight-binding model gives an additional contribution to the interband Josephson coupling, and that the Hund's interaction only gives a higher order correction to the LM [20]. It was also shown [37] that the $s\pm$ pairing can result from the moderate electronic correlations [38–42] in iron-based superconductors; thus, the electronic correlations presumably do not hamper the massless LM much.

The reason that no direct experimental observation on the TRSB state in iron-pnictide superconductors has been reported to date may come from its requirement for sufficiently strong frustration interactions among different bands. Here we observe that the TRSB transition can be induced not only by interband coupling but also by DOS $N_j(0)$. In order to demonstrate this we derive the TRSB solution to Eq. (4) under a general coupling matrix $\hat{g}$. Complex gap functions as the solution to Eq. (4) appear when there is only one independent vector in the matrix $\hat{g} - \hat{g}'$, with $g'_{jj} = N_j(0)\sinh^{-1}(\hbar\omega_{cj}/|\Delta_j|)$ and 0 otherwise. From this constraint we obtain [24]

$$\frac{|\Delta_j|}{\hbar\omega_{cj}} = \frac{1}{\sinh[(g_{jj}g_{kl} - g_{jk}g_{jl})/N_j(0)g_{kl}]}, \tag{17}$$

with $j \neq k \neq l$. It is easy to see that to find further the phases of the gap functions is equivalent to forming a triangle with the three segments $|\Delta_j|/g_{kl}$, which is possible when and only when $|\Delta_j|/g_{kl} + |\Delta_k|/g_{jl} > |\Delta_l|/g_{jk}$ for all the three combinations. The phase transition from a TRSB state to a state without TRSB takes place when one of the above inequalities is broken, for example $|\Delta_1|/g_{23} = |\Delta_2|/g_{13} + |\Delta_3|/g_{12}$. The results for the DOS-driven TRSB transition are displayed in Figs. 2(c) and 2(d). There are two TRSB transitions and the TRSB state is realized in a finite region of DOS. Therefore, experimentally one can tune $N_j(0)$ by a careful chemical doping, which hopefully drives the system to the TRSB transition.

Although the massless LM does not change the magnetic properties of the system, it affects qualitatively several thermodynamic behaviors of s-wave superconductivity. For fully gapped superconductors, the electronic contribution to the specific heat C_v at $T \ll T_c$ depends exponentially on the temperature $(\Delta/k_BT)^{3/2}\exp(-\Delta/k_BT)$. The contribution of the massless Leggett excitations can be obtained analytically by treating the Leggett bosons as free quantum gas. The contribution is of a power-law temperature dependence T^3, which can be detected experimentally.

It is worth noting that a T^3 dependence of the electronic specific heat C_v in iron-base superconductors after subtracting the residue electronic contribution (linear in T) and phonon contribution (also T^3 dependence) has been reported in several experiments, [43–45]; fully gapped order parameters are inferred from measurements for the dependence of electronic C_v on the magnetic field, which excluded the possibility of the gap function of the line node. Actually, in Ref. [44], the authors suggested that the additional T^3 contribution might be due to some bosonic modes. These experimental observations can be naturally explained by the existence of massless LM. Additional measurements such as the Raman spectroscopy on similar samples [43–45] are much anticipated, which may well be in the vicinity of the TRSB transition.

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