Discovery of an excited pair state in superfluid ^{3}He

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Collective modes are the fingerprint of a condensed phase. The spectroscopy of these modes in superfluid ^{3}He and unconventional superconductors can provide key information on the symmetry of the condensate as well as the microscopic pairing mechanism responsible for the ground state and excitation energies. Here, we use interferometry within an acoustic cavity—which is very sensitive to changes in the velocity of transverse sound—to reveal a new collective mode in the B phase of superfluid ^{3}He (^{3}He-B). We identify the mode as an excited bound state of Cooper pairs, which is weakly bound with an excitation energy within 1% of the pair-breaking edge. On the basis of the selection rules for coupling of transverse sound to a collective mode in ^{3}He-B, combined with the observation of acoustic birefringence near the collective mode frequency, we infer that the new mode is most likely a spin-triplet ($S = 1$), f-wave pair exciton (orbital momentum $L = 3$) with total angular momentum, $J = 4$. The existence of a pair exciton with $J = 4$ suggests an attractive, subdominant, f-wave pairing interaction in liquid ^{3}He.

Fifty years ago Bardeen, Cooper and Schrieffer (BCS) published their seminal paper on the theory of superconductivity in metals[1]. This theory, combined with developments over the next decade[2–5], is one of the most successful theoretical achievements of modern physics. The basic feature of BCS theory, the condensation of bound pairs of fermions, has impacted research on nuclear structure[6], our understanding of neutron star interiors, the rotational dynamics of pulsars[7], and most recently, the physics of ultracold, metastable phases of atomic gases[8,9]. In condensed matter, BCS pairing is ubiquitous. It is observed in metals, magnetic materials[10], organic conductors[11], strongly disordered films that are on the verge of becoming insulators[12] and liquid ^{3}He (refs 13,14).

Perhaps the most detailed and specific signatures of broken symmetries of the normal state are the collective modes of the pair condensate. These are the dynamical fingerprints of a multicomponent order parameter[15]. The order-parameter collective modes of superfluid ^{3}He have been extensively studied[16] using acoustic absorption spectroscopy[16–24]. There have been efforts to observe and calculate the spectra and signatures of such modes in the heavy-fermion superconductors, UPt_3 (refs 25, 26), UBe_{13} (refs 27,28), as well as for Sr_2RuO_4 (refs 29,30). Recently, observation of the Leggett mode, the interband Josephson oscillations of a two-band superconductor, was reported in MgB_2 (ref. 31). Collective modes of the amplitude of the order parameter have also been observed. In $NbSe_2$, the superconducting state develops below the onset of a charge-density-wave instability. The coupling between the amplitude mode of the charge-density wave and the amplitude mode of the superconducting order parameter produces an infrared active collective mode near the gap edge, 2Δ (refs 32,33). However, ^{3}He is the best example of complex symmetry breaking among the BCS condensates and investigation of its order-parameter collective modes holds significance for the understanding of other pairing systems.

Subdominant pairing interactions are of special interest as a manifestation of competing orders, notably in cuprate superconductors[34] and superfluid ^{3}He (ref. 22). In the latter case, the role of f-wave pairing has been identified theoretically in the temperature and magnetic-field dependence of the frequencies of order-parameter collective modes[35] and the magnetic susceptibility[36], but an experimental determination of the strength has been contradictory despite substantial effort[16]. Recent high-precision transverse sound spectroscopy measurements[17,18] have offered a new look at this problem but were found to be quantitatively inconsistent within the existing theoretical frameworks, and they indicated f-wave pairing to be repulsive. Here, we report the discovery of a new collective mode in superfluid ^{3}He. This observation alone provides compelling evidence for the existence of attractive high-angular-momentum interactions in ^{3}He, an argument based on selection rules for coupling of the order parameter to transverse sound. The most likely candidate for this new mode is the spin-triplet ($S = 1$), f-wave pair exciton ($L = 3$) with total angular momentum, $J = 4$, predicted by Sauls and Serene[35].

The equilibrium phase of superfluid ^{3}He-B is a spin-triplet ($S = 1$), p-wave ($L = 1$) condensate. The order parameter has nine complex amplitudes, and correspondingly a spectrum of collective modes[16,24], the observation of which has been instrumental in establishing that the dominant pairing interaction is p-wave. For pure p-wave pairing, there are a total of 18 modes corresponding to the number of degrees of freedom of the order parameter. Theoretical predictions of order-parameter collective modes with $J = 0, 1, 2$ for a spin-triplet, p-wave superconductor were first carried out by Vdovin[37] in 1963. These modes were found independently in calculations by Maki[38] after the discovery of superfluid ^{3}He. The B phase has an isotropic gap with magnitude 2Δ, where $\Delta/\hbar$ at zero temperature varies from 34 to 97 MHz over the pressure range of the phase diagram. Acoustic techniques are well suited for these frequencies and, in general, sound can couple effectively to order-parameter modes with non-zero frequency at zero wavevector, $\Omega \sim \Delta/\hbar$. These ‘optical’ modes, shown in Fig. 1,

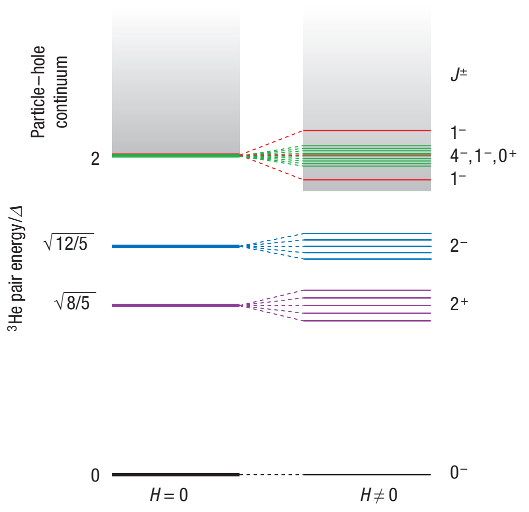

Figure 1 Energy levels of the collective modes, $J^{\pm}$, of superfluid ^{3}He-B that have been observed or predicted to couple to either longitudinal or transverse sound and their Zeeman splitting in a magnetic field in the limit of weak quasiparticle interactions. The 4^- and 0^+ modes have not been observed and evidence for the 1^- mode is not yet well established[19].

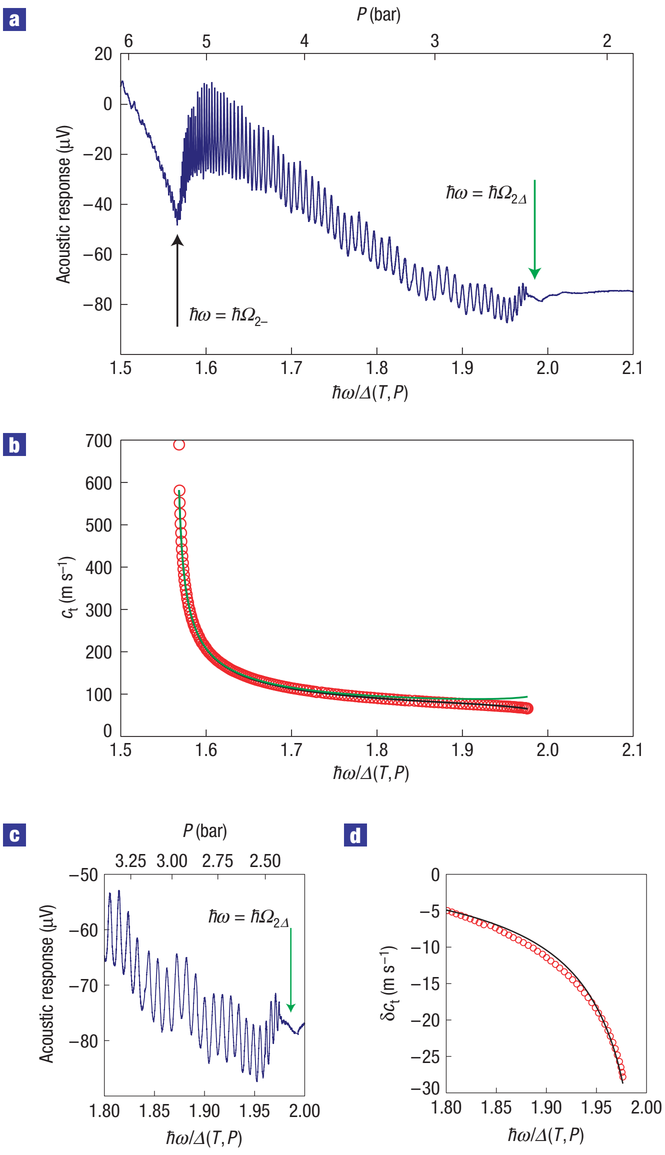

Figure 2 Acoustic cavity response to pressure as a function of energy normalized to the gap energy, at 88 MHz and ≈550 μK in zero magnetic field. **a**, Interference oscillations obtained from a pressure sweep, where the arrows mark the well-established $J = 2^-$ mode (black) and a new mode at the gap edge (green). **b**, Transverse sound velocities (red circles) from **a** compared with the theory, equation (1) (green curve). The black curve is calculated by including an extra mode in equation (1). **c**, Detail of **a** near 2Δ. The green arrow marks where the period of the oscillations goes to zero, indicating the 2Δ mode, $\hbar\Omega_{2\Delta}$. **d**, The difference between the measured and theoretical (equation (1)) transverse sound velocities (red circles) and the difference between the phenomenological model and equation (1) (black curve).

correspond to excited pair states with total angular momentum quantum numbers[38], $J = 0, 1, 2$, and so on, each with $2J + 1$ substates, labelled by $-J \le m_J \le J$. In addition, the order-parameter modes are classified by their parity under particle–hole symmetry, $J^{\pm}$, where plus and minus distinguish between modes with even (+) or odd (−) parity under particle–hole transformation, that is, particle ↔ hole conversion. Figure 1 shows the energy level diagram for the excited pair states in superfluid ^{3}He-B that have been observed, or are predicted, to couple to sound[16,24].

Acoustic waves have a linear dispersion, $\omega = ck$, where c is the sound velocity. The sound frequency, ω, can be chosen to match an order-parameter collective mode, $\Omega_{J^{\pm},m_J}(T, P)$, which is only weakly dependent on wavevector k, but can be tuned by sweeping the pressure or temperature. If there is coupling between sound and the modes they can be identified by significant changes in the attenuation and velocity of the propagating sound wave at the crossing point[16,17]. In addition, we use a unique spectroscopy based on magneto-acoustics that has high spectral resolution[18] and well-defined selection rules for coupling to the order parameter. We constructed a cavity defined by an a.c.-cut quartz piezoelectric transducer and a polished quartz reflector that are separated by $D = 31.6 \pm 0.1$ μm; details are given in Supplementary Information, Section I. Transverse sound is both generated and detected by our transducer. We sweep the ^{3}He pressure, holding the temperature near $T \approx 550$ μK $\ll T_c$. The resulting changes in the phase velocity alter the number of half-wavelengths in the cavity and are manifested as acoustic interference producing an oscillatory electrical response, shown in Fig. 2a. The acoustic signal can be represented as $A = A_0 + A_1 \cos\theta \sin(2D\omega/c + \phi)$, where A_0 is a smoothly varying background in the absence of cavity wave interference and is determined by acoustic impedance[39]; A_1 is the amplitude of the oscillatory signal modulation from wave interference; ϕ is a phase angle; θ is the angle between the polarization of the sound wave at the surface of the transducer and the direction of linear polarization that the transducer can generate and detect. We apply magnetic fields, H, up to 305 G, parallel to the propagation direction. These acoustic techniques offer a precise means to investigate the order-parameter structure of superfluid ^{3}He.

Transverse sound does not ordinarily propagate in fluids. However, it was predicted by Moores and Sauls[40] to exist in superfluid ^{3}He-B as a consequence of coupling to the $J = 2^-$, $m_J = \pm 1$ modes. The experiments of Lee *et al.*[41] confirmed this theory, which was later exploited by Davis *et al.*[17,18] to investigate the $J = 2^-$ mode with much higher precision than is possible with longitudinal sound[21]. Furthermore, Moores and Sauls predicted that ^{3}He-B becomes circularly birefringent in a magnetic field, where the $J = 2^-$, $m_J = \pm 1$ modes couple differently to right and left circularly polarized transverse sound waves[40], giving them different velocities. Circular birefringence leads to rotation of the plane of polarization of linearly polarized transverse sound by an angle proportional to the component of magnetic field in the propagation direction[18,40,41], the acoustic analogue of optical Faraday rotation. Selection rules govern the coupling of the collective modes with quantum numbers $J^{\pm}, m_J$ to right and left circularly polarized transverse sound. Circularly polarized transverse waves in ^{3}He-B, propagating in the direction of the magnetic field, preserve axial symmetry. As a result, transverse sound couples only to $m_J = \pm 1$ modes. Application of a magnetic field lifts the degeneracy of the $m_J = \pm 1$ states through the nuclear Zeeman energy, producing circular birefringence. Therefore, an order-parameter collective mode, $\Omega_{J^{\pm}, m_J}$, that induces acoustic circular birefringence requires $J \geq 1$ with $m_J = \pm 1$. In addition, Moores and Sauls have shown that in zero field only even angular momentum modes with $m_J = \pm 1$ couple to transverse currents[40]. These selection rules follow from the invariance of the B-phase ground state under joint spin and orbital rotations (a $J = 0$ ground state) and are applicable for the geometry of the acoustic experiments described here.

In the superfluid state, transverse sound propagates according to the dispersion relation[40],

$$\left(\frac{\omega}{q v_{\mathrm{F}}}\right)^2_{m_J} = \Lambda_0 + \Lambda_{2^-} \frac{\omega^2}{\omega^2 - \Omega_{2^-, m_J}(H)^2 - \frac{2}{5} q^2 v_{\mathrm{F}}^2}. \quad (1)$$

This equation can be solved for the phase velocity as shown by the green curve in Fig. 2b, where v_{F} is the Fermi velocity, Λ_0 is the quasiparticle restoring force and the 2^- mode frequency is $\Omega_{2^-, m_J}(H) = \Omega_{2^-} + m_J g_{2^-} \gamma_{\mathrm{eff}} H$, to first order in magnetic field. The Landé g factor, g_{2^-}, gives the Zeeman splitting[18] of the mode, γ_{eff} is the effective gyromagnetic ratio[42] of ^{3}He and the complex wavevector is $q = k + i\alpha$, where α is the attenuation. For substates $m_J = \pm 1$, there is a non-zero magneto-acoustic coupling, Λ_{2^-}, between the 2^- mode and transverse sound. The lifetime of the 2^- mode due to thermal quasiparticle damping is omitted in equation (1) and is negligible at low temperatures but should become apparent at higher temperatures than in the work we present here. Further details for Ω_{2^-, m_J}, Λ_0 and Λ_{2^-} are given in Supplementary Information, Section II.

From equation (1) it is apparent that the sound velocity diverges for acoustic frequencies approaching the 2^- mode, resulting in faster oscillations in the acoustic response as is evident in Fig. 2a. Quantitative comparison with theory can be made by converting the oscillations into the transverse sound phase velocity. To obtain absolute values for the velocity, we fix one adjustable parameter by comparison of our data with the velocity calculated from equation (1) near the 2^- mode, as shown in Fig. 2b. The agreement between the calculation, green curve, and the experiment, red circles, is excellent for energies below $\sim 1.8\Delta$. However, above this energy we observe a downturn in the transverse sound velocity. This effect is quite clear in the raw data shown in Fig. 2c, indicated by a decreasing period of the oscillations with increasing energy. This is a classic signature of the approach to an order-parameter collective mode[16]. Figure 2d shows, as red circles, the difference between the measured sound velocity and the value calculated from equation (1), based on coupling only to the 2^- mode. The downturn in velocity is quite evident here. By extrapolating the period of the oscillations in Fig. 2c to zero, to the point where the velocity apparently diverges as indicated by the green arrow, we determine the frequency (excitation energy) of the new collective mode. This procedure is described in detail in Supplementary Information, Section III. We find the excitation energy of the mode, $\hbar\Omega_{2\Delta}$, to vary systematically with pressure, but to remain within 1% of 2Δ for pressures from 1 to 20 bar, as shown in Fig. 3a. The precision of the extrapolation is given by the error bars; the accuracy of 1% is determined by the absolute temperature scale[43] in the framework of the weak-coupling-plus model for the energy gap[44].

To model the sound velocity near the gap edge, we amend equation (1) by adding a new term to represent the coupling of transverse sound to the 2Δ collective mode with a form similar to that of the 2^- mode, $\omega^2 \Lambda_{2\Delta}/(\omega^2 - \Omega_{2\Delta}^2)$. The velocity calculated from this model is given by the black curve in Fig. 2b,d, describing our data quite well with a coupling strength, $\Lambda_{2\Delta} = 0.18$. Furthermore, we note that the amplitude, A_1, of the interference oscillations near the gap edge decreases in a manner similar to the period. As the amplitude is proportional to the inverse of the sound attenuation, this is a consistent identification of a collective mode, where it can be shown from equation (1) that the attenuation diverges at the mode location, as does the velocity.

We have previously established[18,41] that an applied magnetic field rotates the plane of polarization of propagating transverse sound in the near vicinity of the $J = 2^-$ mode, $\omega \geq \Omega_{2^-}$. Increasing the frequency above the $J = 2^-$ mode decreases the Faraday rotation rate, which eventually becomes immeasurably small as the coupling to the mode decreases. However, at even higher frequencies near the pair-breaking edge, we find that the Faraday rotation reappears (Fig. 3b) in the same frequency region where we observe the downturn in the velocity of transverse sound. The magnetic field modulates the interference amplitude, A_1, by a factor $\cos\theta$, from which we extract the Faraday rotation angle, θ. In magneto-optics, Faraday rotation is parameterized by the Verdet constant, $V = \theta/2DH$. For our magneto-acoustic data, we find V to be a monotonically increasing function of frequency, consistent with a divergence near $\Omega_{2\Delta}$, shown as a green arrow in the inset of Fig. 3c. Apparently the birefringence originates from the 2Δ collective mode. The existence of acoustic Faraday rotation (circular birefringence) requires that the 2Δ mode corresponds to an excitation with total angular momentum $J \geq 1$ with substates $m_J = \pm 1$ coupling to transverse currents. It is particularly noteworthy that the changes in velocity and attenuation, which we associate with the new mode, occur in both zero and non-zero applied fields at the excitation energy shown in Fig. 3a.

Order-parameter modes predicted to lie close in energy to the pair-breaking edge include the $J = 0^+, 1^-, 4^-$ modes (Fig. 1). Observations of the 0^+ and 4^- modes have not previously been reported. The observation of circular birefringence enables us to rule out the 0^+ mode. A peak in the longitudinal sound attenuation that appears for non-zero magnetic field[20,23] was attributed[19,45] to the coupling of the $J = 1^-, m_J = 0$ mode to the longitudinal current (see Supplementary Information, Section IV). However, in zero field the $J = 1^-$ modes do not contribute to the stress tensor and cannot couple to either the longitudinal or transverse currents. Consequently, neither the $J = 1^-$ nor the $J = 0^+$ modes can account for our observations in zero field. Furthermore, in a non-zero magnetic field the predicted Zeeman splitting[46] of the $J = 1^-$ modes should decrease the energy of the $m_J = -1$ level as shown by the blue dashed line in Fig. 3d, overlapping a region of energy where we have observed acoustic interference oscillations. As the velocity is not perturbed in this region, it seems that the Zeeman splitting of the 2Δ mode is much smaller than that predicted for the 1^- mode and so the 1^- mode cannot explain our data in a magnetic field. Finally, pair breaking is absent for frequencies below a threshold 2Δ in zero magnetic field, and thus cannot account for

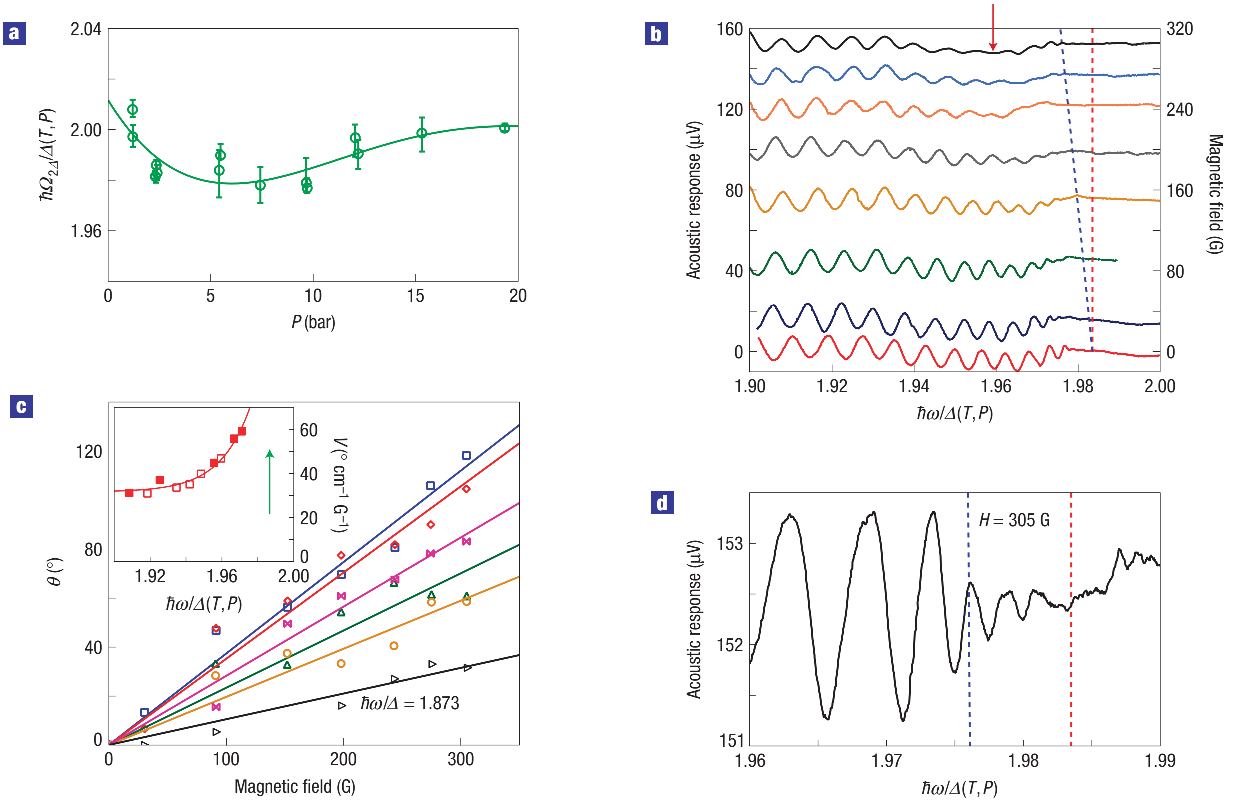

Figure 3 Pressure and magnetic-field dependence of the 2Δ mode. a, $\hbar\Omega_{2\Delta}$ as a function of pressure in zero magnetic field. The curve is a guide to the eye and error bars were determined from an extrapolation procedure as described in Supplementary Information, Section III. **b**, The acoustic response at 88 MHz and $\approx$550 μK as a function of energy, offset by magnetic field (right axis), indicated by the crossing of the data traces with the red dashed line which also marks the zero-field energy of the 2Δ mode. Acoustic birefringence rotates the plane of polarization by an angle θ, producing a minimum signal amplitude at $\theta = \pi/2$, indicated by the red arrow for 305 G and $\approx$1.96Δ. The blue dashed line represents the predicted field dependence ($g = 0.4$) of the $J = 1^-$, $m_J = -1$ mode. **c**, The Faraday rotation angle, θ, is proportional to magnetic field at energies given by filled squares in the inset. Inset: Verdet constant as a function of energy, diverging at $\hbar\Omega_{2\Delta}$, marked by the green arrow. **d**, Detail of the 305 G trace from **b**.

the downturn in velocity below 2Δ. Higher angular momentum modes, in particular modes with $J = 4^-$, $m_J = \pm 1$, do couple to transverse sound even in zero field. On the basis of the experimental geometry and the selection rules for transverse sound and acoustic birefringence, the only known theoretical candidate that might account for our observations are the $J = 4^-$ modes. The two $J = 4^-$ modes with $m_J = \pm 1$, that couple to transverse current, have a weaker Zeeman interaction than the $J = 2^-$ modes owing to the smaller relative strength of f-wave pairing interactions compared with p-wave pairing interactions. Quantitative predictions for the coupling strength and the magnitude of the Zeeman splitting are needed for a conclusive identification.

Predictions for the $J = 4^-$ mode frequency[35] depend on existence of an attractive f-wave pairing interaction[22], subdominant with respect to p-wave pairing, as well as the unknown Fermi liquid parameter, F^s_4. Several experiments, including magnetic susceptibility and $J = 2^\pm$ collective mode spectroscopy[16–18,42,47,48], were analysed to try and determine the f-wave pairing interaction, as well as Fermi liquid interactions, in an effort to predict the $J = 4^-$ mode frequency. However, the results of these different analyses are ambiguous owing to imprecision of the Fermi liquid parameters, $F^{a,s}_2$, as well as non-trivial strong coupling effects[18]. Our observation of a new order-parameter collective mode, identified on the basis of selection rules as the $J = 4^-$ mode, provides direct evidence for an attractive f-wave pairing interaction. Precise measurements of the mode frequency in this work, combined with a future measurement of the Zeeman splitting, should provide constraints on the microscopic pairing mechanism in ^{3}He. In addition, our results may lead to realization of predictions of mixed symmetry pairing near impurities, surfaces and interfaces; surface phases with broken time inversion; and novel vortex phases[34,49].

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