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## Two-Phonon Absorption by the Real Squashing Mode in Superfluid $^3\text{He-B}$

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We have observed in superfluid  $^3\text{He-B}$  two-phonon absorption, which is one of the nonlinear effects recently predicted by McKenzie and Sauls. The  $J=2^+$  real squashing mode was excited with two coincident sound pulses at the temperature  $T^+$ ,  $f_{\text{signal}} + f_{\text{pump}} = f_{\text{rsq}}(T^+)$ , under 0, 2.7, and 3.5 bars pressure. The heights of the attenuation peaks scale almost linearly with the energy of the higher-intensity wave, in qualitative agreement with the theory. Furthermore, a satellite peak appears even in zero magnetic field, which originates from the reflected wave.

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Ultrasound is a very useful probe to study the properties of collective modes in the order parameter  $A_{ij}$  of superfluid  $^3\text{He}$ . In the  $B$  phase, oscillations of the order parameter about the equilibrium value  $A_{ij}^0$  can be classified by  $J$  and  $m_J$ , where  $J=0,1,2$  is the total angular momentum and  $m_J = -2, -1, \dots, 2$  is the magnetic quantum number. The  $J=2^+$  and  $J=2^-$  modes, where the plus and minus refer to the real and imaginary parts of the order parameter, are known as the real squashing (rsq) mode [ $hf = a_{\text{rsq}}\Delta(T_{\text{rsq}})$ ] and the squashing (sq) mode [ $hf = a_{\text{sq}}\Delta(T_{\text{sq}})$ ], respectively. Here  $\Delta(T)$  is the temperature-dependent BCS energy gap [ $\Delta(0) = 1.76k_B T_c$ ], and  $a_{\text{rsq}} = \sqrt{8/5}$  and  $a_{\text{sq}} = \sqrt{12/5}$  in the weak-coupling (WC) limit neglecting Fermi-liquid parameters and the higher-order pairing interactions. These modes are of particular interest because they couple to ultrasound, resulting in sharp, well-defined features in sound attenuation, phase velocity, and group velocity.<sup>1</sup>

The index  $\pm$  in  $J$  may also be considered as a parity under the particle-hole (p-h) transformation  $C$ ,<sup>2</sup> which maps a quasiparticle just above the Fermi surface into a quasiparticle just below, with the spin rotated by  $\pi$ . The real (imaginary) component of the order parameter has even (odd) parity under  $C$ ; this provides a selection rule: The rsq (sq) mode cannot (can) couple to the linear term  $\delta n$  of the density fluctuation caused by the sound under exact p-h symmetry, whereas the rsq (sq) mode can (cannot) couple to the quadratic term  $(\delta n)^2$ , even under exact p-h symmetry. However, this symmetry is weakly broken, allowing the linear rsq mode to be excited by a single zero-sound phonon.

In this Letter, we report the first detection of two-phonon absorption by the rsq mode. This is a three-wave resonance, one of the nonlinear effects in the sense that intensive  $(\delta n)^2$  allows two phonons with frequencies  $f_p, f_s$  and wave vectors  $\mathbf{q}_p, \mathbf{q}_s$  to excite a third mode with frequency  $f$  and wave vector  $\mathbf{q}$ :  $f = f_p + f_s$ ,  $\mathbf{q} = \mathbf{q}_p + \mathbf{q}_s$ . This excitation by two-phonon process is allowed by the p-h-symmetry selection rule. The effect was predicted by McKenzie and Sauls,<sup>3</sup> who derived, from microscopic

theory, dynamical equations describing the nonlinear interactions between two coincident zero sound waves and the rsq mode.

At low sound intensities, in the linear-response regime, many of the observed properties in the  $B$  phase, such as the Zeeman splitting of the modes in a magnetic field, are similar to those seen in the optical spectroscopy of atoms and molecules. Consequently, it is interesting to study whether in  $^3\text{He-B}$  there exist acoustic analogs of *nonlinear* optical effects.<sup>4</sup> Polturak *et al.*<sup>5</sup> made the first experiments in the nonlinear regime, observing effects similar to optical self-induced transparency at sound energy densities on the order of 1% of the superfluid condensation energy density. Sauls<sup>6</sup> produced a phenomenological theory on the results of these measurements, based on an optical analog. However,<sup>7</sup> his theory predicted self-induced transparency at sound energy densities *two orders* of magnitude larger than those used by Polturak *et al.* These are different types of nonlinear effects from the three-wave resonance.

We have measured the attenuation and phase velocity of a low-intensity wave in the presence of a copropagating high-intensity wave. The wave of lower intensity at  $f_s, \mathbf{q}_s$ , which is being monitored, is called the *signal* wave, and the one of higher intensity at  $f_p, \mathbf{q}_p$  is called the *pump* wave. If the signal wave is of higher frequency than the pump wave ( $f_s > f_p$ ,  $E_s \ll E_p$ , where  $E_s, E_p$  are the energies per one pulse of the signal and pump waves, respectively), a two-phonon absorption (a stimulated Raman scattering) peak is expected to be seen at a temperature  $T^+$  ( $T^-$ ) such that

$$h(f_s \pm f_p) = a_{\text{rsq}}\Delta(T^\pm). \quad (1)$$

The intensity of these attenuation peaks should linearly increase with the power of the *pump* wave. The maximum sound attenuation  $\Delta\alpha$  and the change of the phase velocity  $\Delta c/c$  of the signal wave due to the nonlinear resonance with the rsq mode are given by

$$\frac{\Delta\alpha}{\alpha_s} \simeq \frac{\Delta c}{c} \simeq \frac{|\tilde{A}|^2}{(1+F\delta)^2} \frac{\Delta}{\Gamma} \frac{U_p}{U_c}. \quad (2)$$

Here  $U_c = \frac{1}{2} N(\epsilon_F) \Delta(T)^2$  is the superfluid condensation energy density,  $U_p$  is the energy density of the pump wave,  $1/\Gamma$  is the lifetime of the mode,  $\tilde{A}$  is the coupling strength which has been calculated from microscopic theory,<sup>3</sup>  $F_0^{\delta}$  is a Landau Fermi-liquid parameter, and  $N(\epsilon_F)$  is the density of states for one spin component at the Fermi surface. Since  $F_0^{\delta}$  and  $U_c$  increase monotonically with pressure,  $\Delta a$  and  $\Delta c$  are expected to be largest at low pressures. Furthermore, since the linewidth  $\Gamma$  decreases with temperature, low temperatures are favorable for experiments. The peak at  $T^-$  is difficult to observe because its location is within the temperature range where the sq mode strongly attenuates the sound signal, whereas  $T^+$  is more favorably positioned.

Our experiments were performed in the ROTA2 cryostat at Helsinki,<sup>8</sup> but measurements reported here were carried out on nonrotating liquid. The sound cell has two X-cut quartz crystals, 4 mm apart. The diameter of the cylindrical  $^3\text{He}$  chamber is 6 mm and the fluid inside is in liquid contact with the main  $^3\text{He}$  volume through several holes. We employed the pulsed sound transmission technique at frequencies  $f_p = 8.9$  MHz and  $f_s = 26.8$  or 44.7 MHz. The pump and signal waves were produced by two separate rf generators. Pulses at these two frequencies were simultaneously sent to the transmitter crystal through a directional bridge (HP8721A), so that

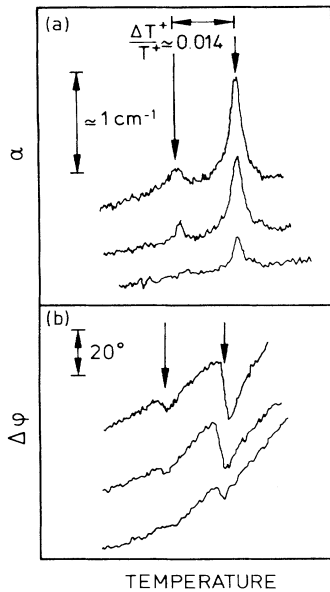


FIG. 1. Two-photon absorption by the rsq at  $T^+$ . (a) Attenuation ( $\alpha$ ) vs temperature, and (b) corresponding phase change ( $\Delta\phi$ ), obtained at zero pressure with three different energies  $E_p$  of the pump pulses.  $\Delta\phi$  is proportional to  $\Delta c$ . Frequencies of the signal and pump waves were  $f_s = 26.8$  MHz and  $f_p = 8.9$  MHz, respectively.  $E_p = 2.9, 1.9,$  and  $0.59$  nJ from top to bottom; the energy of the signal wave pulses was  $E_s = 0.11$  nJ in all cases. Magnetic field is zero.

each transmitter pulse was made of two frequencies. The pulse width was  $12 \mu\text{s}$  and  $\mathbf{q}_p$  and  $\mathbf{q}_s$  were parallel to each other. Particular care was taken to avoid the generation of higher harmonic frequencies. Only the signal wave was monitored by the receiver system. Temperatures were measured by a conventional  $^{195}\text{Pt}$  NMR thermometer.

Experiments on the nonlinear rsq mode are difficult to perform because the high power levels required easily cause undesirable heating in the sound cell. However, this problem can be overcome by good thermal contact between  $^3\text{He}$  and the nuclear refrigerant. In our case, a sintered-silver heat exchanger, electron-beam welded to the copper nuclear stage, provided a nominal surface area of  $100 \text{ m}^2$ .<sup>8</sup> Measurements of the thermal relaxation time between  $^3\text{He}$  and the nuclear stage yielded an effective area  $A \approx 30 \text{ m}^2$  in the heat exchanger. Using this value and the maximum averaged heating power  $E_p \approx 1.5$  nW, the calculated difference between the temperature of  $^3\text{He}$  and the electronic temperature of the nuclear stage is  $\delta T = R_K \dot{Q}/A \approx 13 \mu\text{K}$  at  $T = 1.0$  mK,<sup>9</sup> where  $R_K$  and  $\dot{Q}$  are the Kapitza thermal boundary resistance and the heat current, respectively. Therefore,  $A$  is sufficiently large for cooling to the  $T^+$  temperature of the two-phonon absorption peak, even at our high level of pump-wave intensity.

Three sets of power-dependent sound attenuation peaks, recorded during temperature sweeps at zero pressure and in zero magnetic field, are shown in Fig. 1(a), together with the corresponding phase changes, Fig. 1(b). Surprisingly, two  $T^+$  peaks were seen: The main peak was accompanied by a smaller one on the low-temperature side. We denote the maximum attenuations of these peaks by  $a_m$  and  $a_s$ , respectively. High-energy pump pulses ( $0.59 \text{ nJ} \leq E_p \leq 2.9 \text{ nJ}$ ) were used in these experiments, in contrast to our earlier measurements<sup>10</sup> during which 10-pJ pulses were typically employed to study the linear rsq mode. The measured temperatures at which the main peak appears are summarized in Table I. The result is in agreement with the theoretical prediction of Eq. (1) in the WC limit. We used the BCS energy gap and  $T_c$  determined by Greywall.<sup>11</sup>

The most important property of the two-phonon absorption is that the heights of both attenuation peaks in-

TABLE I. The temperatures  $T^+$  at which the main two-phonon absorption peak was observed. The last column gives  $a_{\text{rsq}}/\sqrt{8/5} = h(f_s + f_p)/[\sqrt{8/5}\Delta(T^+)]$ . These values should be compared to previous observations which ranged from approximately 0.91 to 0.99 for  $f_p = 0$  (Ref. 1).

$p$ (bar)	$f_s$ (MHz)	$f_p$ (MHz)	$T^+/T_c$ (observed)	$T^+/T_c$ (WC theory)	$a_{\text{rsq}}/\sqrt{8/5}$
0	26.8	8.9	$0.72 \pm 0.02$	0.70	$1.02 \pm 0.03$
2.7	44.7	8.9	$0.58 \pm 0.03$	0.59	$1.00 \pm 0.02$
3.5	44.7	8.9	$0.64 \pm 0.02$	0.66	$0.98 \pm 0.02$

crease almost linearly with  $E_p$ . In Fig. 2, this is shown for 0 and 3.5 bars pressure in zero magnetic field. In both cases, the dependence of  $\alpha_m$  and  $\alpha_s$  on  $E_p$  is not perfectly linear; some saturation is seen. Unfortunately, a sufficiently accurate measurement of the width of the peaks, to determine the contribution of line broadening, is beyond the resolution of our thermometry.

To estimate the sound energy, we made calorimetric measurements. First, after stabilizing the temperature just above  $T_c$ , sound pulses were transmitted, typically with a repetition rate of 10 kHz for several tens of seconds, so that the temperature of  $^3\text{He}$  increased. The total amount of heat that was absorbed by the liquid was on the order of 100  $\mu\text{J}$ , which caused an easily measur-

able change in temperature. After 20–40 min, the temperature of the  $^3\text{He}$  sample had relaxed and a new thermal equilibrium had been established between  $^3\text{He}$  in the experimental cell and the nuclear stage (32 mol of copper in a field of typically 200 mT). This increase in temperature allowed us to estimate the heat input per pulse. The sound energies ( $E_p$ ,  $E_s$ ), obtained from these measurements, were proportional to  $V^2$  as would be expected, where  $V$  is the peak-to-peak amplitude of the transmitter pulse. Our method naturally gives upper limits for  $E_p$  and  $E_s$ . In Fig. 2, the observed attenuations at  $p=0$  and 3.5 bars are  $\approx 0.3$  and  $\approx 0.2 \text{ cm}^{-1}$ , respectively, at  $U_p/U_c=0.1$ . Theoretical predictions give 1 and 0.7  $\text{cm}^{-1}$  for these two cases.

We next discuss the possible origins of the two peaks. The temperature difference between them at zero pressure is  $(0.014 \pm 0.003)T_c$ . At first sight this doublet might be accounted for by dispersion splitting of the sq

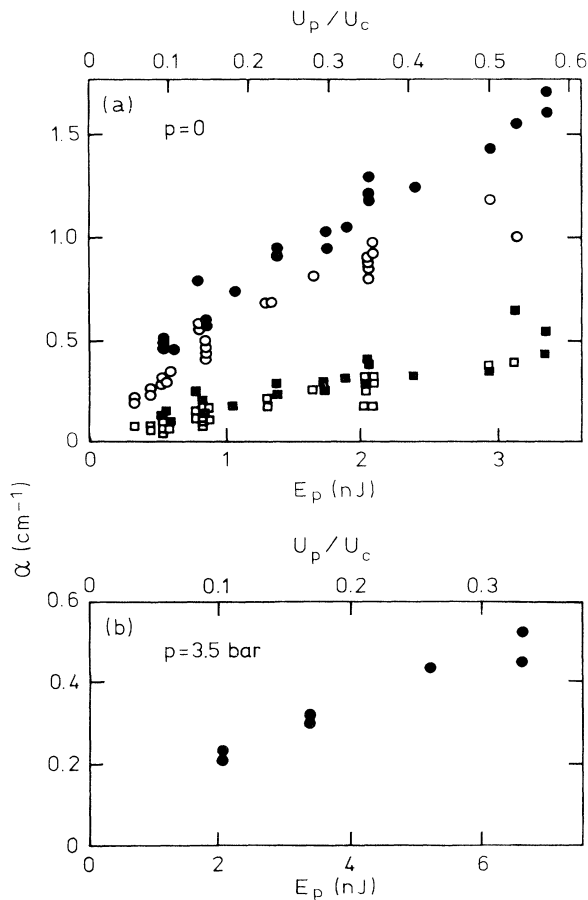


FIG. 2. Energy dependence of attenuation,  $\alpha_m$ ,  $\alpha_s$ , for the two-phonon absorption peaks in zero magnetic field. Frequencies of the signal and pump waves were (a)  $f_s=26.8$  MHz and  $f_p=8.9$  MHz at zero pressure and (b)  $f_s=44.7$  MHz and  $f_p=8.9$  MHz at 3.5 bars. The ratio  $U_p/U_c$  is also shown, where  $U_c$  is calculated using  $U_c = \frac{1}{2} N(\epsilon_F) \Delta(T^+)^2$ . At zero pressure, two different energies of the signal wave,  $E_s=0.11$  nJ (open symbols) and  $E_s=0.055$  nJ (solid symbols), were used. The circle (square) symbols refer to  $\alpha_m$  ( $\alpha_s$ ).  $E_p/E_s$  ranges from 4.5 to 27 (from 5.5 to 55) for open (solid) circle symbols.

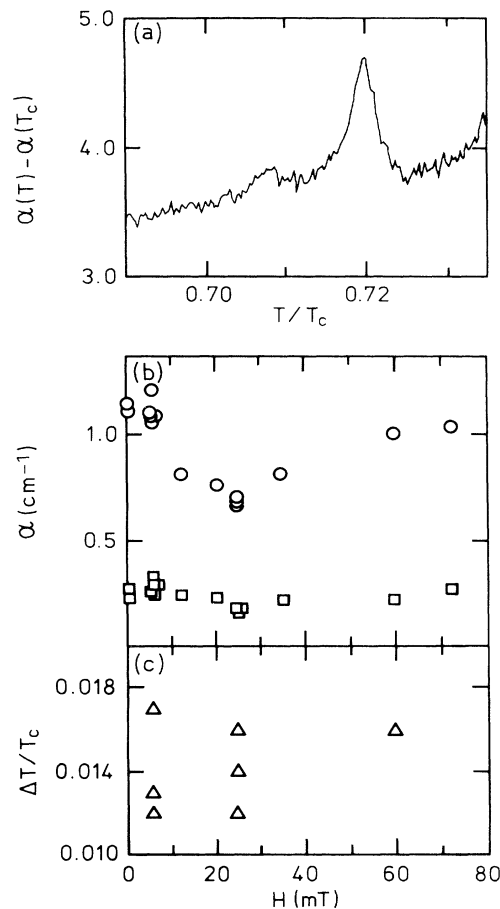


FIG. 3. (a) Two-phonon absorption peak at  $H=59.2$  mT. (b) Magnetic-field dependence of  $\alpha_m$  (circles) and  $\alpha_s$  (squares) at zero pressure. Frequencies  $f_s=26.8$  MHz and  $f_p=8.9$  MHz were employed. The magnetic field and the propagation of both sound waves were parallel. (c) Magnetic-field dependence of the temperature interval between the two peaks.

mode which, in zero magnetic field, has three temperatures  $T_{|m_J|}$ ,<sup>12,13</sup>

$$(\hbar f)^2 = a_{\text{rsq}}^2 \Delta(T_{|m_J|})^2 + b_{|m_J|} (\hbar q v_F)^2, \quad (3)$$

corresponding to  $|m_J|=0, 1$ , and  $2$ , respectively;  $v_F$  is the Fermi velocity, and  $b_0 = \frac{7}{15}$ ,  $b_1 = \frac{2}{5}$ , and  $b_2 = \frac{1}{5}$ . The splitting  $\Delta T$  between  $T_0$  and  $T_2$  is  $\Delta T/T_c \approx 0.013$ ,<sup>14</sup> which is approximately what we observe. If the two peaks really correspond to  $|m_J|=0$  and  $2$ , their distance should increase with the field when we apply a magnetic field up to 80 mT parallel to the axis of sound propagation. In Fig. 3, the field dependences of the peak heights, 3(b), and their splitting, 3(c), are shown for the fixed energy  $E_p = 2.0$  nJ. Two peaks still appeared but the temperature interval between them did not change. Hence it seems unlikely that dispersion splitting could give rise to the observed two peaks. Their field evolution does not seem to follow the conventional crossover from dispersion to Zeeman splitting.<sup>13,15</sup>

The texture splitting of the  $m_J=0$  mode,<sup>10,16</sup> in which the interval between the two peaks is independent of the field, is improbable because it should occur only in high magnetic fields.<sup>17</sup> The possibility of superflow splitting of the collective mode can also be ruled out by two observations: (i) Even when the liquid was rotated up to 1.0 rad/s in zero field, the doublet structure did not change, and (ii) the temperature drift rate in the measurement was varied with no observable changes in the spectrum.

Recently Volovik<sup>18</sup> and Shen<sup>19</sup> have suggested independently that the smaller peak is the two-phonon absorption excited by the signal wave and the *reflected* pump wave. The magnitude of the wave vector is different in the two cases;  $q = q_s \pm q_p$ , respectively. Therefore the mode splits, and the temperature interval which is independent of the field, is estimated to be  $0.017T_c$  [see Eq. (3)]. This agrees with the experiment.

In summary, we conclude that both peaks in Fig. 1 must be identified with the two-phonon absorption at  $T^+$  because the temperatures at which they appear agree with the theoretically predicted values (cf. Table I), and especially because their heights are almost proportional to the input energy.

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