

## Observation of Dispersion in the $J = 2^+$ Collective Modes of $^3\text{He-B}$ by Nonlinear Acoustic Spectroscopy

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Parametric excitation yields new results, which cannot be obtained by conventional linear spectroscopy, on the collective modes in superfluid  $^3\text{He-B}$ . In our ultrasonic experiments, performed in a chamber with four quartz crystals, we have excited the real squashing ( $J = 2^+$ ) modes by two simultaneous sound waves which are mutually either parallel, orthogonal, or antiparallel. Qualitatively these results are in agreement with the theoretical dispersion relation of the  $J = 2^+$  modes, and quantitatively we have been able to extract the collective-mode velocities from the line splittings in zero magnetic field.

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Following the invention of powerful lasers, nonlinear optics has developed into a significant subfield of physics during the past three decades [1,2]. Two-photon processes, which are parametric nonlinearities, have proved extremely useful in the spectroscopy of solid materials and large molecules. The optical two-photon transition obeys selection rules different from those valid for the corresponding one-photon case. Thus one can excite such modes which otherwise could not be studied [3]. For the complicated spectra of molecules or impurities in crystals, this additional spectral information has been a very valuable tool.

Similar considerations are applicable to linear versus parametric excitations in ultrasonic collective-mode spectroscopy of superfluid  $^3\text{He-B}$ . Each mode can be classified according to its parity under the particle-hole transformation (+/-), its total angular momentum  $J = 0, 1, 2$ , and according to the projection of  $J$  on the quantization axis,  $m$ . The best known of these oscillations are the real squashing (RSQ,  $J = 2^+$ ) and squashing (SQ,  $J = 2^-$ ) modes, which can be excited by ultrasound [4].

In our earlier investigations [5,6] we were able to excite the RSQ modes in  $^3\text{He-B}$  parametrically by two coincident parallel zero sound pulses. In this Letter we wish to demonstrate the usefulness of two-phonon absorption (TPA) as a spectroscopic tool. In particular we have, for the first time, measured the dispersion relation of the  $J = 2^+$ ,  $m = 0$  mode, as well as the dispersion splitting of the  $m = 0$  and  $m = \pm 2$  modes in zero magnetic field.

The selection rules arising from the approximate particle-hole symmetry in  $^3\text{He}$  imply that the  $J = 2^-$  mode is strongly excited by one phonon but that the  $J = 2^+$  mode is excited only in proportion to the weak particle-hole asymmetry ( $\sim 10^{-3}$  times less). In a two-phonon process, owing to the positive net parity of the phonons involved, the  $J = 2^+$  mode is excited but the  $J = 2^-$  is not, except for the weak particle-hole asymmetry contribution. Our results are consistent with the theory: TPA was not observed for the  $J = 2^-$  mode [5,6].

In our experiments the RSQ mode was excited by two coincident zero sound pulses. The signal wave, at a frequency  $f_s$  and a wave vector  $\mathbf{q}_s$ , was monitored by a phase-sensitive spectrometer, and the high intensity pump wave, at  $f_p$  and  $\mathbf{q}_p$ , provided the parametric excitation. The dispersion relation of collective modes in zero magnetic field can be written as a series expansion, which to the lowest order in  $q$  reads

$$h^2 f^2 = a^2 [\Delta(T_m)]^2 + \hbar^2 v_m^2 q^2, \quad (1)$$

where  $\Delta(T_m)$  is the energy gap of  $^3\text{He-B}$  at the temperature  $T_m$ ; in the case of TPA,  $f = f_s + f_p$  and  $\mathbf{q} = \mathbf{q}_s + \mathbf{q}_p$  [7,8]. For the RSQ mode, the quantum number  $m$  can have values 0,  $\pm 1$ , and  $\pm 2$ , and  $a^2$  is a dimensionless coefficient weakly dependent on temperature and pressure; in the weak-coupling limit without interaction effects  $a^2 = \frac{8}{5}$ , and earlier experiments give  $a^2 \approx 1.43$  at  $p = 0$ ,  $T = 0$ , when the BCS energy gap is used [9].

The mode velocities  $v_m$  in Eq. (1) are on the order of the Fermi velocity,  $v_F = 59.0$  m/sec at  $p = 0$  [4]. They were first calculated by Vdovin [10] and later by a number of other authors [11]. In the absence of interaction effects (i.e., magnetic Fermi liquid and higher-order pairing contributions) the mode velocities can be written in the form

$$v_m^2 = c_a^2 + \frac{1}{6} (4 - m^2) c_b^2, \quad (2)$$

which indicates that out of the three  $v_m$ 's only two are independent. The coefficients  $c_a$  and  $c_b$  are given by [12]

$$c_a^2 = \frac{v_F^2}{5} \left[ 1 - \frac{2}{7} \frac{\lambda_1}{\lambda_0} \right], \quad (3)$$

$$c_b^2 = \frac{2v_F^2}{5} \left[ 1 + \frac{3}{7} \frac{\lambda_1}{\lambda_0} \right]. \quad (4)$$

The function  $\lambda_1/\lambda_0$  is weakly temperature dependent and has the value  $-0.43$  at  $T = 0$  [13]. Fishman and Sauls [14] have calculated corrections to Eqs. (3) and (4)

caused by antisymmetric Fermi-liquid effects. They find at most 10% shifts in the mode velocities at zero pressure.

There are two reasons for the superiority of parametric excitation with respect to the one-phonon process in studying the dispersion relation [Eq. (1)] of the collective modes. First, one can alter the wave number  $|\mathbf{q}_s + \mathbf{q}_p|$  of the mode while its frequency  $f_s + f_p$  stays constant. Second, the mode coupling to ultrasound depends on the relative orientation of  $\mathbf{q}_s, \mathbf{q}_p$ , and the quantization axis of the mode,  $\mathbf{Q}$  [15]. If  $\mathbf{Q} \parallel \mathbf{q}_s$  and  $\mathbf{q}_p$ , only the mode with  $m=0$  couples to ultrasound. In low magnetic fields,  $\mathbf{Q}$  is aligned with  $\mathbf{q}_s$  in the one-phonon excitation but with  $\mathbf{q}_s + \mathbf{q}_p$  in the two-phonon case. Consequently while one-phonon processes allow the observation of only the  $m=0$  mode in zero field, TPA with  $\hat{\mathbf{q}}_s \perp \hat{\mathbf{q}}_p$  permits observation of the  $m = \pm 2$  modes as well.

Using parallel ( $\uparrow\uparrow$ ), antiparallel ( $\uparrow\downarrow$ ), and orthogonal ( $\uparrow\rightarrow$ ) pulses, altogether four peaks in ultrasound attenuation, at temperatures  $T_{\uparrow\uparrow,0}$ ,  $T_{\uparrow\downarrow,0}$ ,  $T_{\uparrow\rightarrow,0}$ , and  $T_{\uparrow\rightarrow,\pm 2}$ , should be observable. It can be shown [15] that the corresponding attenuation peak heights are related by

$$\Delta\alpha_{\uparrow\uparrow,0} = \Delta\alpha_{\uparrow\downarrow,0} = 4\Delta\alpha_{\uparrow\rightarrow,0} = \frac{4}{3}\Delta\alpha_{\uparrow\rightarrow,\pm 2}, \quad (5)$$

and  $\Delta\alpha_{\uparrow\rightarrow,\pm 1} = 0$ .

In the present experiments two simultaneous zero sound pulses, either parallel, orthogonal, or antiparallel, propagated through the  $^3\text{He}$  sample. The frequencies  $f_s$  and  $f_p$  were kept constant, but the temperature and thereby  $\Delta(T)$  was varied, allowing excitations with different  $m$  or  $q$  to couple [see Eq. (1)]. Our experimental chamber consisted of two pairs of  $X$ -cut crystals. They were mounted on a cubic frame, such that the crystals of one pair faced the top and bottom of the cell, and those of the other pair were glued to two opposite vertical sides, respectively. Transmission and detection of sound pulses were thus possible both in the vertical and horizontal directions. A vertical magnetic field could be applied. The distance between the crystals of each pair was  $L=9.5$  mm, and the diameter of the central holes bored through the sides of the cube was 6 mm. Two of the vertical faces were left open to the rest of the  $^3\text{He}$  volume. The crystals operated at odd harmonic frequencies of 5.0 MHz; the pulses of ultrasound were 30 or 45  $\mu\text{sec}$  long. All measurements were made at zero pressure, using  $f_s=25.15$  MHz and  $f_p=15.10$  MHz. In these conditions, TPA could be observed at  $T/T_c \approx 0.5$ .

Figure 1 shows two spectra with parallel and orthogonal pulses in zero magnetic field. The identification of the four peaks follows from the dispersion relation in Eq. (1). The two peaks for  $\mathbf{q}_s \parallel \mathbf{q}_p$  arise from parallel and antiparallel propagation, respectively. The latter one is obtained only by reflection from the receiver crystal (see Ref. [6]), hence it is much weaker. The two peaks for perpendicular waves are due to the  $m=0$  and  $m = \pm 2$  modes. Since in our experiments the overlap of the pump and signal waves is small for orthogonal pulses compared

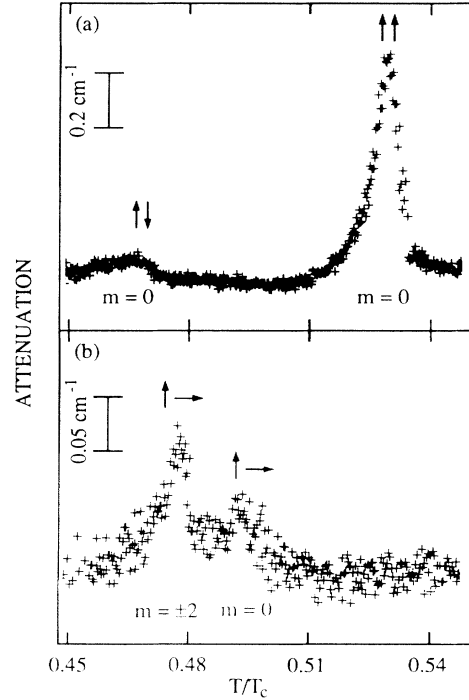


FIG. 1. The temperature dependence of the attenuation of a  $f_s=25.15$  MHz signal wave in the presence of a  $f_p=15.1$  MHz pump wave is illustrated. The pressure is zero, and no magnetic field is present. In (a) the sound pulses are parallel. The large (small) peak is due to excitation of the  $m=0$  mode at parallel (antiparallel) waves. In (b) the sound pulses are perpendicular. The  $m=0$  peak shifts from its position in (a) because of the smaller wave vector of the mode. Energy densities of the pump and signal waves,  $U_p$  and  $U_s$ , in terms of the superfluid condensation energy density  $U_c$ , are (a)  $U_p/U_c=0.11$  and  $U_s/U_c=0.0015$  and (b)  $U_p/U_c=0.16$  and  $U_s/U_c=0.0015$ . Using these values the peaks can be seen clearly but the pulses do not heat the sample excessively. The measured line splittings were used to determine the mode velocities (see text). The curves in both (a) and (b) have the same absolute position in temperature. The pulse length in each case is 45  $\mu\text{sec}$ .

to parallel ones, the peak heights for  $\mathbf{q}_s \perp \mathbf{q}_p$  are small.

Our thermometry is based on measuring the variation of the phase velocity near the peak positions, and it was calibrated against the Pt NMR thermometer in several separate runs. We can measure the absolute positions of the peaks within  $\pm 0.02T_c$ , but their relative positions can be measured much more accurately. If we fix  $T_{\uparrow\uparrow,0}/T_c=0.530$ , which is the average from several runs, the temperatures of the other peaks are  $T_{\uparrow\downarrow,0}/T_c=0.466 \pm 0.005$ ,  $T_{\uparrow\rightarrow,0}/T_c=0.493 \pm 0.005$ , and  $T_{\uparrow\rightarrow,\pm 2}/T_c=0.477 \pm 0.005$ . The difference  $(T_{\uparrow\rightarrow,0} - T_{\uparrow\rightarrow,\pm 2})/T_c = 0.0156 \pm 0.0015$  obtained from twelve experimental runs. Using these values, one can determine  $c_a^2$ ,  $c_b^2$ , and  $a^2$  from the formulas

$$h^2(f_s + f_p)^2 = a^2[\Delta(T_{\uparrow\uparrow,0})]^2 + \hbar^2[c_a^2 + \frac{2}{3}c_b^2](q_s + q_p)^2, \quad (6)$$

$$a^2\{[\Delta(T_{\uparrow\downarrow,0})]^2 - [\Delta(T_{\uparrow\downarrow,0})]^2\} = 4\hbar^2(c_a^2 + \frac{2}{3}c_b^2)q_s q_p, \quad (7)$$

$$a^2\{[\Delta(T_{\uparrow\rightarrow, \pm 2})]^2 - [\Delta(T_{\uparrow\rightarrow, 0})]^2\} = \frac{2}{3}\hbar^2 c_b^2 (q_s^2 + q_p^2), \quad (8)$$

which follow from Eqs. (1) and (2).

Determination of  $c_a^2$ ,  $c_b^2$ , and  $a^2$  requires knowledge of the magnitude of  $\mathbf{q}_s$  and  $\mathbf{q}_p$ , and hence  $c_0$ , the phase velocity of the ultrasound at the temperature at which TPA occurs. We have calculated the latter from theory [see Eq. (129) in Ref. [8]]. The values we obtained for  $c_0$  at 15 and 25 MHz are about 5% smaller than the speed in the normal phase, which at zero pressure is 189.9 m/sec [4]. The coefficients  $c_a^2$ ,  $c_b^2$ , and  $a^2$ , deduced from our data, are presented in Table I. For comparison, we have also included their theoretical values at  $T=0$  and without interaction effects [14]. The measured value for  $c_b^2$  agrees with theory, but there is a discrepancy in the value of  $c_a^2$ , which cannot be explained by interaction effects.

The coefficient  $c_a^2$  has not been measured earlier, but two experiments exist from which  $c_b^2$  can be obtained indirectly in a nonzero magnetic field [16,17]. Using the results of Ref. [16],  $c_b^2$  can be determined in two ways which, according to Halperin and Varoquaux [4], give consistent results. The first method is based on the positions of three features in the ultrasonic signal, which were all observed in one run and which the authors interpreted as the dispersion-split modes with  $m=0, \pm 1$ , and  $\pm 2$ . From this spectrum, a value of  $(c_b/v_F)^2 = 0.29 \pm 0.06$  at  $p=4.9$  bars and  $B=7.8$  mT was found [4]. The second method is to measure the textural splitting of the  $m=0$  peak. In Ref. [17],  $c_b^2$  was determined from the nonequal spacing of the five attenuation peaks, caused by the Zeeman splitting of the RSQ mode in a nonzero magnetic field and under rotation. The values for  $(c_b/v_F)^2$  range from 0.24 to 0.38 at pressures from 3 to 12 bars.

Figure 2 is a test of the dispersion relation in the form of Eq. (1) for  $m=0$ . Here,  $[\Delta(T)/k_B T_c]^2$  has been plotted against  $(\hbar c_0/k_B T_c)^2 q^2$  for the three experimental values of  $q$ , namely,  $q_s + q_p$ ,  $(q_s^2 + q_p^2)^{1/2}$ , and  $q_s - q_p$ . We have again fixed  $T_{\uparrow\downarrow,0}/T_c = 0.530$ . The line has been plotted using the measured constants from Table I in Eq. (1). The point corresponding to  $(q_s^2 + q_p^2)^{1/2}$  lies somewhat above the line, which is not exactly consistent with Eq. (1).

TABLE I. The parameters  $a^2$ ,  $c_a^2/v_F^2$ , and  $c_b^2/v_F^2$  as derived from the theory at  $T=0$  without interaction effects [14], and from our data.

	Theoretical	Measured
$a^2$	1.60	$1.47 \pm 0.03$
$(c_a/v_F)^2$	0.224	$0.30 \pm 0.07$
$(c_b/v_F)^2$	0.327	$0.30 \pm 0.06$

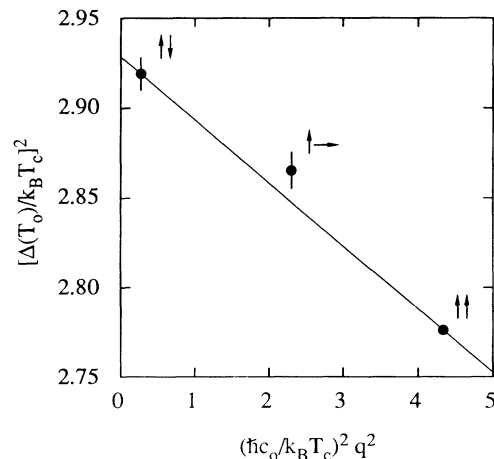


FIG. 2. Measurement of the dispersion of the  $J=2^+$ ,  $m=0$  mode under the experimental conditions of Fig. 1. The square of the BCS gap function,  $[\Delta(T)]^2$ , at the temperature of TPA for  $m=0$ , scaled by  $(k_B T_c)^2$ , is shown as a function of  $q^2 = |\mathbf{q}_s + \mathbf{q}_p|^2$ , multiplied by a constant  $(\hbar c_0/k_B T_c)^2$ . Here  $q$  is the magnitude of the wave vector at which the mode is excited. Equation (1) predicts a linear relationship between  $[\Delta(T_0)]^2$  and  $q^2$ .

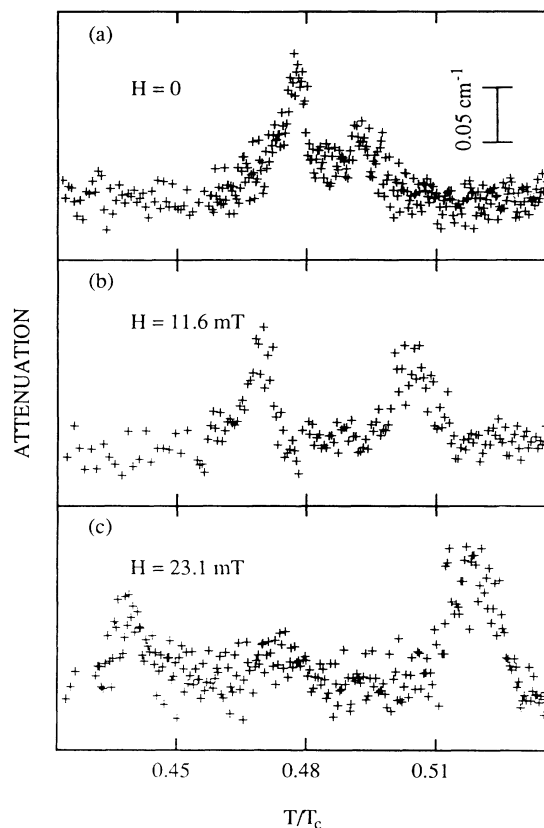


FIG. 3. Magnetic field dependence of two-phonon absorption for perpendicular pulses. Values of experimental parameters are the same as in Figs. 1 and 2. The magnetic field is parallel to the direction of the pump wave.

Figure 3 shows three attenuation spectra of crossed pulses ( $\mathbf{q}_s \perp \mathbf{q}_p$ ) when a magnetic field is applied in the direction of the pump wave propagation. The temperature splitting between the farthestmost peaks is almost twice as large as in the case of  $\hat{\mathbf{q}}_s \uparrow \hat{\mathbf{q}}_p \perp \mathbf{H}$  in an equal magnetic field [6]. This is due to the flattening of  $\Delta(T)$  towards lower temperatures of the present measurement. It is obvious that when  $H > 20$  mT, the high-temperature ( $m = +2$ ) peak already dominates like in the case of parallel propagation [6]. We do not, however, understand why it happens in this case.

In summary, using the three-wave resonance as a spectroscopic tool in zero sound experiments, we have directly tested the dispersion relation of the  $J = 2^+$  collective mode in  $^3\text{He-B}$ . Such a study is not possible by linear spectroscopy, with just one propagating sound wave, because of the vanishing coupling strengths of all except one of the  $m$  modes in zero magnetic field and because of the fixed value of the wave number of excitation,  $q = 2\pi f_s/c_0$ . In the quantitative part of our discussion we have extracted the  $J = 2^+$  mode velocities at  $p = 0$  and at a low temperature ( $T/T_c \approx 0.5$ ).

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