## Second wind of the Dulong-Petit Law at a quantum critical point

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Renewed interest in <sup>3</sup>He physics has been stimulated by experimental observation of non-Fermi-liquid behavior of dense <sup>3</sup>He films at low temperatures. Abnormal behavior of the specific heat C(T) of two-dimensional liquid <sup>3</sup>He is demonstrated in the occurrence of a *T*-independent  $\beta$  term in C(T). To uncover the origin of this phenomenon, we have considered the group velocity of transverse zero sound propagating in a strongly correlated Fermi liquid. For the first time, it is shown that if two-dimensional liquid <sup>3</sup>He is located in the vicinity of the quantum critical point associated with a divergent quasiparticle effective mass, the group velocity depends strongly on temperature and vanishes as *T* is lowered toward zero. The predicted vigorous dependence of the group velocity can be detected in experimental measurements on liquid <sup>3</sup>He films. We have demonstrated that the contribution to the specific heat coming from the boson part of the free energy due to the transverse zero-sound mode follows the Dulong-Petit Law. In the case of two-dimensional liquid <sup>3</sup>He, the specific heat becomes independent of temperature at some characteristic temperature of a few mK.

Almost two hundred years ago, Pierre-Louis Dulong and Alexis-Thérèse Petit [1] discovered experimentally that the specific heat C(T) of a crystal is close to constant independent of the temperature T. This behavior, attributed to lattice vibrations – i.e. phonons – is known as the Dulong-Petit Law. Later, Ludwig Boltzmann [2] reproduced the results of Dulong and Petit quantitatively in terms of the equipartition principle. However, subsequent measurements at low temperatures demonstrated that C(T) drops rapidly as the T is lowered toward zero, in sharp contrast to Boltzmann's theory. In 1912, Peter Debye [3] developed a quantum theory for evaluation of the phonon part of the specific heat of solids, correctly explaining the empirical behavior  $C(T) \sim T^3$  of the lattice component as  $T \to 0$ . In the Debye theory, the T-independence of C(T) is recovered at  $T \ge T_D$ , where  $T_D$  is a critical temperature corresponding to the saturation of the phonon spectrum. With the advent of the Landau theory of quantum liquids [4], predicting a linear-in-T dependence of C(T) for the specific heat contributed by itinerant fermions, our understanding of the low-temperature thermodynamic properties of solids and liquids thus seemed to be firmly established. However, recent measurements [5, 6] of the specific heat of two-dimensional (2D) <sup>3</sup>He as realized <sup>3</sup>He films absorbed on graphite preplated with a <sup>4</sup>He bilayer, reveal behavior strongly antithetical to established wisdom, which calls for a new understanding of the low-temperature thermodynamics of strongly correlated many-fermion systems.

Owing to its status as a fundamental exemplar of the class of strongly interacting many-fermion systems, liquid <sup>3</sup>He remains a valuable touchstone for lowtemperature condensed-matter physics. In recent years, interest in <sup>3</sup>He physics has been driven by the the observation of non-Fermi-liquid (NFL) behavior of dense <sup>3</sup>He films at the lowest temperatures  $T \simeq 1$  mK reached experimentally [5–11]. In particular, measurements of the specific heat C(T) in the 2D <sup>3</sup>He system show the presence of a term  $\beta$  tending to a finite value as  $T \rightarrow 0$ . Such behavior contrasts sharply with that of its counterpart, three-dimensional (3D) liquid <sup>3</sup>He; for this system, the lower the temperature, the better Landau Fermi-liquid (FL) theory works. (Here we shall not consider superfluid phases of <sup>3</sup>He.)

In seeking the origin of the anomalous contribution  $\beta$ remaining in C(T) at the lowest temperatures attained, it is instructive to examine the schematic low-T phase diagram of 2D liquid <sup>3</sup>He shown in Figure. The essential features of this picture are documented by the cited experiments on <sup>3</sup>He films. The effective coupling parameter is represented by  $z = \rho/\rho_{\infty}$ , where  $\rho$  is the number density of the system and  $\rho_{\infty}$  is the critical density at which a quantum critical point (QCP) occurs. This QCP is associated with a divergence of the effective mass  $M^*(z)$ , portrayed in Figure by the curve (in

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Phase diagram of the 2D liquid <sup>3</sup>He system. The region defined by  $z = \rho/\rho_{FC} < 1$  is divided into LFL and NFL domains separated by a solid line. The dependence  $M^*(z) \propto (1-z)^{-1}$  is shown by the solid line approaching the dashed asymptote, thus depicting the divergence of the effective mass at the quantum critical point (z = 1, T = 0)indicated by the arrow. In the region  $z \gtrsim 1$ , the fermion condensate (FC) sets in and Dulong-Petit behavior of the specific heat is realized for the strongly correlated quantum many-fermion system (as represented by the dash horizontal line at T = 0)

red on line) that approaches the dashed asymptote at z = 1. The T - z phase plane is divided into regions of 2D Fermi-Liquid (FL) and non-Fermi-Liquid (NFL) behavior. The part of the diagram where z < 1 consists of a FL region at lower T and a NFL region at higher T, separated by a solid curve. The regime where  $z \gtrsim 1$  belongs to a NFL state with specific heat taking a finite value  $\beta(\rho)$  at very low temperatures. The physical source of this excess heat capacity has not been established with certainty, although it is supposed that the  $\beta$  anomaly is related to peculiarities of the substrate on which the <sup>3</sup>He film is placed.

In this letter we propose that the observed  $\beta$  term in C(T) can instead have its origin in an intrinsic mechanism analogous to that producing the classical Dulong-Petit behavior in solids. It is shown for the first time that in systems (such as 2D <sup>3</sup>He) containing a fermion condensate (FC), the group velocity of transverse zero sound depends strongly on temperature. It is this dependence that gives rise to the  $\beta$  term, granting the Dulong-Petit Law a "second wind."

As indicated above, the most challenging feature of the NFL behavior of liquid <sup>3</sup>He films involves the specific heat C(T). According to Landau theory, C(T) varies linearly with T, and at low film densities the experimental behavior of the specific heat of 2D liquid <sup>3</sup>He is in agreement with FL theory. However, for relatively dense <sup>3</sup>He films, this agreement is found to hold only at sufficiently high temperatures. If T is lowered into the millikelvin region, the function C(T) ceases to fall toward zero and becomes flat [5, 6, 11].

The common explanation [5, 6, 12] of the flattening of C(T) seen in these experiments imputes the phenomenon to disorder associated with the substrate that supports the <sup>3</sup>He film. More specifically, it is considered that there exists weak heterogeneity of the substrate (steps and edges on its surface) such that quasiparticles, being delocalized from it, give rise to the low-temperature feature  $\beta$  of the heat capacity [6]. Even if we disregard certain unjustified assumptions made in Ref. [12], there remains the disparate fact that the emergent constant term in C(T) is of comparable order for different substrates [5, 6, 11]. Furthermore, the explanation posed in Ref. [12] implies that the departure of C(T) from FL predictions shrinks as the film density increases, since effects of disorder are most pronounced in weakly interacting systems. Contrariwise, the anomaly in C(T) makes its appearance in the density region where the effective mass  $M^*$  is greatly enhanced [6, 11] and the 2D liquid <sup>3</sup>He system becomes strongly correlated. This reasoning compels us to consider that the NFL behavior of C(T) is an *intrinsic* feature of 2D liquid <sup>3</sup>He, which is associated with the divergence of the effective mass rather than with disorder.

The flattening of the curve C(T) as seen in <sup>3</sup>He films is by no means a unique phenomenon. Indeed, as expressed in the Dulong-Petit (DP) law, the specific heat C(T) of solids remains independent of T as long as Texceeds the Debye temperature  $\Omega_D$ , which is determined by the saturation of the phonon spectrum of the crystal lattice. Normally, the value of  $\Omega_D$  is sufficiently high that the DP law belongs to classical physics. However, we will argue that the DP behavior of C(T) can also make its appearance at extremely low temperatures in strongly correlated Fermi systems, with zero sound playing the role of phonons.

To clarify the details of this phenomenon and calculate the specific heat C(T), we evaluate a part  $F_B$  of the free energy F associated with the collective spectrum  $\omega(k) = ck$ , based on the standard formula

$$F_B = \int_0^\infty \frac{d\omega}{\pi} \frac{1}{e^{\omega/T} - 1} \int \operatorname{Im}\left[\ln D^{-1}(k,\omega)\right] d\upsilon, \quad (1)$$

where  $D(k, \omega)$  is the boson propagator, and dv is an element of momentum space. Upon integration by parts this formula is recast to

$$F_B = T \int_0^\infty \frac{d\omega}{\pi} \ln(1 - e^{-\omega/T}) \int \operatorname{Im}\left(\frac{\partial D^{-1}(k,\omega)/\partial\omega}{D^{-1}(k,\omega)}\right) dv.$$
(2)

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If damping of the collective branch is negligible (the case addressed here), then  $D^{-1}(k,\omega) \simeq (\omega - ck)$  and  $\partial D^{-1}(k,\omega)/\partial \omega \simeq 1$ , while  $\mathrm{Im} D^{-1}(k,\omega) \simeq \delta(\omega - ck)$ , and we arrive at the textbook formula

$$F_B = T \int \ln\left(1 - e^{-ck/T}\right) \theta(\Omega_0 - ck) dv, \qquad (3)$$

where  $\Omega_0$  is the characteristic frequency of zero sound. At  $T \gg \Omega_0$ , the factor  $\ln(1 - e^{-\omega/T})$  reduces to  $\ln(\omega/T)$ , yielding the result

$$F_B(T) \propto T \ln(\Omega_0/T),$$
 (4)

which, upon the double differentiation, leads to the DP law C(T) = const. At first sight, this law has nothing to do with the situation in 2D liquid <sup>3</sup>He. Its Fermi energy  $\epsilon_F^0$  is around 1 K at densities where the Sommerfeld ratio C(T)/T soars upward as  $T \to 0$ , while  $\Omega_0$  must be lower than  $T \simeq 1$  mK. Indeed, in any conventional Fermi liquid, including 3D liquid <sup>3</sup>He, there is no collective degree of freedom whose spectrum is saturated at such low ratios  $\Omega_0/\epsilon_F^0$ .

This conclusion remains valid assuming 2D liquid <sup>3</sup>He is an ordinary Fermi liquid. However, as seen from Figure, if the quantum critical point is reached at  $T \rightarrow 0$  and some critical density  $\rho_{\infty}$  where the effective mass  $M^*(\rho)$  diverges, as it does in the present case [6-8, 11, 13], the situation changes dramatically. This is demonstrated explicitly in the results of standard FL calculations of the velocity  $c_t$  of transverse zero sound, which satisfies [14, 15]

$$\frac{c_t}{2v_F}\ln\frac{c_t + v_F}{c_t - v_F} - 1 = \frac{F_1 - 6}{3F_1(c_t^2/v_F^2 - 1)},$$
 (5)

where  $v_F = p_F/M^*$  is the Fermi velocity and  $F_1 = p_F M^* f_1/\pi^2$  is a dimensionless version of the Landau first harmonic  $f_1$  [14, 16, 17]. The divergence of the effective mass  $M^*$  at the QCP implies that at the critical density determined by  $f_1 p_F M/\pi^2 = 3$  [16, 17], one has

$$c_t^2(\rho) \simeq \frac{p_F^2}{5M^*(\rho)M} \to 0, \tag{6}$$

whereas the sound velocity  $c_s$  remains finite in this limit [14, 15, 18].

We see then that in case the effective mass  $M^*$  diverges, the group velocity  $c_t$  vanishes as  $\sqrt{1/M^*}$ . Flattening of the single-particle spectrum  $\epsilon(p)$  prevails as long as  $|p - p_F|/p_F < M/M^*$ , implying that the transverse mode softens only for rather small wave numbers  $k \sim p_F M/M^*$ . Unfortunately, the associated numerical prefactor cannot be established, rendering estimation of  $\Omega_0 \sim (p_F^2/M)\sqrt{M/M^*}$  correspondingly uncertain.

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Nevertheless, one cannot exclude a significant enhancement of the Sommerfeld ratio C(T)/T at  $T \simeq 1 \,\mathrm{mK}$  due to softening of the transverse zero sound in the precritical density region.

At  $T \rightarrow 0$  and densities exceeding  $\rho_{\infty}$ , the system undergoes a cascade of topological phase transitions in which the Fermi surface acquires additional sheets [19-21]. As indicated in Figure, FL theory continues to hold with quasiparticle momentum distribution n(p)satisfying  $n^2 = n$ , until a greater critical density  $\rho_{FC}$  is reached where a new phase transition, known as fermion condensation, takes place [21-27]. Beyond the point of fermion condensation, the single-particle spectrum  $\epsilon(p)$ acquires a flat portion. The range L of momentum space adjacent to the Fermi surface where the FC resides depends on the difference between the effective coupling constant and its critical value. As will be seen, L is a new dimensional parameter that serves to determine the key quantity  $\Omega_0$ .

At finite T, the dispersion of the FC spectrum  $\epsilon(p)$  existing at  $\rho > \rho_{FC}$  acquires a nonzero value proportional to temperature [21,24-26]:

$$\epsilon(p,T) = T \ln \frac{1 - n_*(p)}{n_*(p)}, \quad p_i (7)$$

where  $0 < n_*(p) < 1$  is the FC momentum distribution and  $p_i$  and  $p_f$  are the lower and upper boundaries of the FC domain in momentum space. Consequently, in the whole FC region, the FC group velocity, given by

$$v(p,T) = \frac{\partial \epsilon(p)}{\partial p} = -T \frac{\partial n_*(p)/\partial p}{n_*(p)(1-n_*(p))}, \quad p_i 
(8)$$

is proportional to T. Significantly, in the density interval  $\rho_{\infty} < \rho < \rho_{FC}$  the formula (7) describes correctly the single-particle spectrum  $\epsilon(p,T)$  in case the temperature T exceeds a very low transition temperature [21]. The FC itself contributes a T-independent term to the entropy S; hence its contribution to the specific heat C(T) = TdS/dT is zero. Accordingly, we focus on the zero-sound contribution to C(T) in systems having a FC.

Due to the fundamental difference between the FC single-particle spectrum and that of the remainder of the Fermi liquid, a system having FC is, in fact, a twocomponent system. Remarkably, the FC subsystem possesses its own set of zero-sound modes, whose wave numbers are relatively small, not exceeding  $L = (p_f - p_i) > 0$ . The mode of prime interest for our analysis is that of transverse zero sound. As may be seen by comparison of formulas (6) and (8), its velocity  $c_t$  depends on temperature so as to vanish like  $\sqrt{T}$  as  $T \to 0$ . To verify the latter property explicitly, we observe first that for systems with a rather small proportion of FC, evaluation of the spectrum of collective excitations may be performed by employing the familiar FL kinetic equation [14, 28]

$$(\omega - \mathbf{k}\mathbf{v})\,\delta n(\mathbf{p}) = -\mathbf{k}\mathbf{n}\frac{\partial n(p)}{\partial p}\int \mathcal{F}(\mathbf{p},\mathbf{p}_1)\delta n(\mathbf{p}_1)dv_1.$$
(9)

Focusing on transverse zero sound in 2D liquid <sup>3</sup>He, one need only retain the term in the Landau amplitude  $\mathcal{F}$ proportional to the first harmonic  $f_1$ . To proceed further we make the usual identification  $(c_t - \cos \theta) \delta n(\mathbf{p}) =$  $(\partial n(p)/\partial p) \phi(\mathbf{n})$ , where  $\cos \theta = \mathbf{kv}/kv$ . Equation (9) then becomes

$$\phi(\theta) = -f_1 p_F \cos \theta \int \cos \chi \frac{\partial n(p_1)/\partial p_1}{c_t - v(T) \cos \theta_1} \phi(\theta_1) \frac{dp_1 d\theta_1}{(2\pi)^2},$$
(10)

where  $\cos \chi = \cos \theta \cos \theta_1 + \sin \theta \sin \theta_1$ , while v(T) is given by equation (8). The solution describing transverse zero sound is  $\phi(\mathbf{n}) \sim \sin \theta \cos \theta$ .

We see immediately that  $c_t \gg v(T) \sim T$ ; therefore the transverse sound in question does not suffer Landau damping. In this situation, we are led to the simple result

$$c_t^2 = -\frac{p_F}{5M} \int \frac{\partial n(p)}{\partial p} v(p,T) dp \qquad (11)$$

upon keeping just the leading relevant term  $v(T)\cos\theta/c_t^2$  of the expansion of  $(c_t - v(T)\cos\theta)^{-1}$  and executing straightforward manipulations. Factoring out an average value of the group velocity  $v(p,T) \propto T/p_F$ , we arrive at the stated behavior

$$c_t(k) \simeq \sqrt{T/M} \tag{12}$$

for wave numbers k not exceeding the FC range L. Transverse sound can of course propagate in the other, noncondensed subsystem of 2D liquid <sup>3</sup>He consisting of quasiparticles with normal dispersion [14, 15, 28]. However, its group velocity is *T*-independent, so the corresponding contribution to the free energy is irrelevant.

As noted above, the characteristic wave number of the soft transverse zero-sound mode is given by the FC range  $L(\rho) = p_f - p_i$ , treated here as an input parameter. The key quantity  $\Omega_0$  is therefore estimated as  $\Omega_0 \simeq k_{\max}c_i$ , where  $k_{\max}$  is the maximum value of the zero sound momentum at which the zero sound still exists. In our case the zero sound is associated directly with the FC; hence  $k_{\max} \simeq \sqrt{Lp_F}$  and we have

$$\Omega_0 \simeq k_{\max} c_t \simeq \sqrt{\frac{T L p_F}{M}}.$$
(13)

As long as the inequality  $Lp_F/M < T$  is met (or equivalently,  $T/\epsilon_F^0 > L/p_F$  holds), the ratio  $\Omega_0/T$  is small, and we are led to the DP result C(T) = const. Then, in spite of the low temperature, C behaves as if the system were situated in the classical limit rather than at the QCP. Such a behavior is ensured by the fact that the system contains a macroscopic subsystem with heavy quasiparticles. As the temperature ultimately goes down to zero at the fixed density  $\rho$ , the inequality  $Lp_F/M < T$ eventually fails, the quantum regime is restored and the dominant contribution to C comes from the "normal" fermions. In other words, there exists an extremely low temperature  $T_0$  below which the usual FL behavior of zero sound is recovered.

Interestingly, the value of the constant term in C(T) can be evaluated in closed form in terms of the FC range L. Upon inserting  $\omega_t(k) = c_t k$  into Eq. (3) and integrating, the *T*-independent term in the specific heat is found to be

$$C/N \simeq Lp_F/8\pi\rho,\tag{14}$$

where N is the number of atoms in the film. The FC range parameter L also enters the result derived analogously for the spin susceptibility  $\chi$ . The FC component of  $\chi$  is given by [21, 29]

$$\chi_*(T) \simeq \chi_C(T) \frac{L}{p_F},\tag{15}$$

where  $\chi_C(T) = \mu_B^2 \rho / T$ .

The results (14) and (15) jointly establish an unambiguous relation within our model between the Tindependent term in the specific heat C(T) and the Curie component of the spin susceptibility  $\chi(T)$  (which has also been observed experimentally [7, 8]). This relation can be tested using existing experimental data [6]. The T-independent specific heat C/N exists in the density region around  $\rho = 9.5 \text{ nm}^{-2}$ . Being referred to one particle, it is readily evaluated from  $\beta \simeq 0.25 \text{ mJ/K}$ . One finds  $C/N \simeq 0.01$ , yielding  $L/p_F \simeq 0.05$ . On the other hand, the data for the spin susceptibility given in Fig. 2(B) of Ref. [6] supports a Curie-like component at  $\rho = 9.25$  nm<sup>-2</sup>. The value of the corresponding numerical factor extracted from the data, which according to Eq. (15) is to be identified with the ratio  $L/p_F$ , is approximately 0.07. Given the uncertainties involved, our model is consistent with the experimental data of Ref. [6].

In summary, we have analyzed the group velocity of transverse zero sound propagating in a strongly correlated Fermi liquid. We have shown for the first time that if two-dimensional liquid <sup>3</sup>He is located in the vicinity of the quantum critical point associated with a divergent quasiparticle effective mass, the group velocity depends strongly on temperature and vanishes at diminishing temperatures. Such a vivid dependence of the group velocity can be detected in experimental measurements on the liquid. We have demonstrated that the contribution to the specific heat coming from the boson part of the free energy contributed by the transverse zerosound mode follows the Dulong-Petit law. Accordingly, the specific heat becomes independent of temperature at some characteristic temperature. In the case of twodimensional liquid <sup>3</sup>He, this temperature can be a few mK. At sufficiently lower temperature the usual FL behavior of zero sound is recovered. The model developed from the analysis is found to be in reasonable agreement with experimental measurements.

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