

## Effective $^3\text{He}$ interactions in dilute $^3\text{He}$ - $^4\text{He}$ mixtures

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(Received 22 February 2012; published 27 April 2012)

The effective interaction between  $^3\text{He}$  quasiparticles in dilute liquid  $^3\text{He}$ - $^4\text{He}$  mixtures affects many of its physical properties. The interaction potential is determined here from the saturation solubility and osmotic pressure data reported recently. The interactions are examined consistently over the entire pressure range of liquid-helium mixtures at very low temperatures, that is, from 0 to 25.6 bar, where the solid phase appears. To reproduce all experimental data, it was necessary to include a concentration dependence in the potential. This dependence, however, turned out to be almost the opposite to what has been proposed earlier. The deduced potential can be used to calculate, among other things, an estimate for the superfluid transition temperature of  $^3\text{He}$ - $^4\text{He}$  mixtures. In addition, we also find values for some less well-established parameters for helium mixtures, such as the binding energy of a  $^3\text{He}$  atom in superfluid  $^4\text{He}$ .

DOI: [10.1103/PhysRevB.85.134529](https://doi.org/10.1103/PhysRevB.85.134529)

PACS number(s): 67.60.-g, 67.60.G-, 67.85.Pq

### I. INTRODUCTION

Liquid mixture of the two stable isotopes of helium,  $^3\text{He}$  and  $^4\text{He}$ , forms an interesting quantum system, the fermion density of which can be varied between zero and what is allowed by the maximum solubility of  $^3\text{He}$  in  $^4\text{He}$ . Thus, the Fermi temperature and interaction energy of the  $^3\text{He}$  system can also be changed continuously.

Certain properties of helium mixtures are governed by the mutual interactions between the fermionic  $^3\text{He}$  particles in the liquid. The most interesting, perhaps, is the possible superfluid transition of the  $^3\text{He}$  component. Since the early studies of helium mixtures, it has been obvious that the interaction among  $^3\text{He}$  is attractive. According to the BCS theory, this guarantees a superfluid state at some low enough temperature. Mixtures have been cooled down to about 100  $\mu\text{K}$  without detecting superfluidity in the  $^3\text{He}$  component. This is practically about the lowest temperature that can be achieved by external cooling due to rapidly growing thermal boundary resistance. Another experimental scheme has been proposed, in which internal cooling of mixtures is produced by adiabatic melting of a  $^4\text{He}$  crystal in superfluid  $^3\text{He}$ .<sup>1</sup> The working pressure in such an experiment is the melting pressure, about 25 bar. To calculate any properties of the system there, one needs to know about the interactions under this particular condition. None of the previous studies have been extended to this range of pressures.

No satisfactory first-principles description of the interactions exists, so that we are restricted to using a phenomenological approach. In other words, we choose a suitably parametrized functional form for the interaction and iterate the set of its variables to reproduce the measured data. The characteristics of the interaction potential can be probed through measurements of various macroscopic properties of helium mixtures. In this paper, we use recently measured solubility<sup>2</sup> and osmotic pressure<sup>3</sup> data to determine a phenomenological interaction potential between two  $^3\text{He}$  quasiparticles at any pressure of liquid mixtures. For simplicity, we use a mean-field approximation (Hartree-Fock).

Similar studies have previously been performed mostly at zero pressure.<sup>4,5</sup> Yorozu *et al.* measured the phase separation curve at low temperatures and at some pressures between 0 and 20 bar. They used those data to determine the interaction

potential at five discrete pressures.<sup>6</sup> We have used our newly retrieved interaction potential to calculate various mixture properties, some of which have already been published.<sup>3,7</sup>

### II. QUASIPARTICLE INTERACTIONS

The task of finding the proper interaction potential is challenging. There is inevitably some scatter in the experimental data required in the analysis. We must allow some of the less certain quantities to vary slightly from the measured values, increasing the number of fit parameters. Sometimes, small variation of observable quantities may introduce unacceptably large deviations to the computed potential. Many macroscopic quantities are not very sensitive to the exact shape of the potential but to some integrated values, which creates some degree of “degeneracy” in the problem: several slightly different potentials can reproduce any single measured quantity. To reach an acceptable level of confidence, we used several sets of data and required the chosen potential to be valid at any pressure with some suitably selected smoothly pressure-dependent parameters.

It is useful to consider the mutual interactions between quasiparticles in momentum space, i.e., the Fourier transform of the potential

$$V(\mathbf{k}) = \int d^3r e^{-i\mathbf{k}\cdot\mathbf{r}} V(\mathbf{r}). \quad (1)$$

Here, the potential is written in terms of the wave vector  $k = p/\hbar$ . The quasiparticle energy can now be written in the Hartree-Fock approximation as

$$\begin{aligned} \epsilon(k) = & -E_3 + \frac{(\hbar k)^2}{2m}(1 + \gamma k^2) + n_3 V(0) \\ & - \int \frac{d^3k'}{(2\pi)^3} V(\mathbf{k} - \mathbf{k}') f(k'), \end{aligned} \quad (2)$$

where  $E_3$  is the binding energy of a single  $^3\text{He}$  atom in superfluid  $^4\text{He}$ ,  $m$  is the effective mass of a  $^3\text{He}$  quasiparticle in the zero concentration limit, the  $\gamma k^2$  term is a small correction to the basic effective mass model of Landau Fermi-liquid theory, and  $n_3$  is the  $^3\text{He}$  particle density. Of the interaction terms,  $n_3 V(0)$  is the direct Hartree interaction energy. As can

be seen from the Fourier transform [Eq. (1)],  $V(0)$  represents a spatial average of the interaction. The second interaction term in Eq. (2) is the exchange energy due to interaction between particles with parallel spin. For  $\gamma$ , we used the experimental value<sup>8</sup>  $\gamma = -0.076 \text{ \AA}^2$  and for the effective mass, we used the values determined in Ref. 3. In the above dispersion relation,  $f(k)$  is the Fermi-Dirac distribution function

$$f(k) = \frac{1}{e^{\beta(\epsilon(k)-\mu)} + 1}, \quad (3)$$

where  $\beta = 1/(k_B T)$ , as usual.

Bardeen, Baym, and Pines (BBP) showed that in the zero-concentration limit, the form of the interaction potential at zero momentum transfer is<sup>4</sup>

$$V(0) = -\alpha^2 \frac{m_4 c_{40}^2}{n_{40}}, \quad (4)$$

where  $m_4$  is the mass of  $^4\text{He}$ ,  $c_{40}$  the speed of sound, and  $n_{40}$  the particle density in pure  $^4\text{He}$ . The factor  $\alpha$  is the BBP parameter, defined as the ‘‘excess volume occupied by  $^3\text{He}$  atoms.’’ It has weak concentration and temperature dependencies but somewhat stronger pressure dependence, being approximately 0.28 at zero pressure and 0.17 at 25 bar. The interaction presented by BBP results from the larger zero-point energy of  $^3\text{He}$  compared to  $^4\text{He}$ , which is a consequence of the smaller mass of  $^3\text{He}$ . When dissolved into liquid  $^4\text{He}$ , the  $^3\text{He}$  atoms effectively form ‘‘bubbles,’’ which tend to combine.

The result of BBP at elevated pressures was questioned by Eckstein *et al.*<sup>9</sup> They argued that  $V(0)$  should in fact become positive at high pressures. Baym responded to this by calculating a perturbation theory correction to second order in  $^3\text{He}$ - $^4\text{He}$  mass difference.<sup>10</sup> He found the correction to be surprisingly small, and the original result of BBP to be valid to good accuracy.

According to the definition of  $\alpha$ , the molar volume of helium mixture with concentration  $x$  can be written as

$$v_m = v_{40}(1 + \alpha x), \quad (5)$$

where  $v_{40}$  is the molar volume of pure  $^4\text{He}$  at the given pressure. The pure value has been determined experimentally with good accuracy. We used the data of Tanaka *et al.*<sup>11</sup> and Watson *et al.*<sup>12</sup> The temperature and concentration dependencies of  $\alpha$  were calculated by following the theory presented in Refs. 13 and 14. For  $\alpha$  itself, we used the measurements of the former. The average concentration of  $^3\text{He}$  in their experiment was 8%; the zero-concentration values were calculated on that basis.

We can now examine the pressure dependence of  $E_3$ . By starting with the Gibbs-Duhem equation and using Maxwell relations to express derivatives of chemical potentials with molar volumes, we arrive at

$$-E_3(P) = -E_0 + \frac{1}{N_A} \int_0^P v_{40}(1 + \alpha) dP'. \quad (6)$$

Here,  $\alpha$  is evaluated at zero concentration and zero temperature.

### III. CHEMICAL POTENTIAL

The chemical potential is extremely useful in connecting the interaction potential to many measurable quantities. The

value of the chemical potential is fixed by the condition

$$n_3 = 2 \int \frac{d^3 p}{(2\pi\hbar)^3} f(p). \quad (7)$$

In general, the chemical potential is obtained by a numerical solution to Eq. (7). This is, however, computationally somewhat time consuming. At temperatures well below the Fermi temperature,  $T \ll T_F$ , one may use a low-temperature series expansion of the chemical potential, which is much quicker than to determine the ‘‘exact’’ chemical potential. This makes the fitting procedure more efficient since the chemical potential is required many times at every fit cycle. The derivation of this low-temperature expansion is discussed in the Appendix. To maintain a congruent notation with previous works, we write the chemical potential in the low-temperature limit as

$$\begin{aligned} \mu_3 = & -E_3 + E_F \left[ 1 - \frac{\pi^2}{12} \left( \frac{T}{T_F} \right)^2 \right] + n_3 V(0) \\ & - \frac{1}{2} n_3 |V(0)| \left[ F + G \frac{\pi^2}{12} \left( \frac{T}{T_F} \right)^2 \right], \end{aligned} \quad (8)$$

where

$$F = \frac{3}{2} \int_0^{2k_F} \frac{k^2}{k_F^3} \left( 1 - \frac{k}{2k_F} \right) \frac{V(k)}{|V(0)|} dk \quad (9)$$

and

$$G = 3 \left( \frac{V(2k_F)}{|V(0)|} - \frac{1}{4} \int_0^{2k_F} \frac{k^3}{k_F^4} \frac{V(k)}{|V(0)|} dk \right). \quad (10)$$

Here,  $E_F$  is the Fermi energy of a noninteracting system with particle number density  $n_3$ . In our case,  $E_F = (\hbar k_F)^2 (1 + \gamma k_F^2)/(2m)$ . Similarly,  $T_F$  is the Fermi temperature of the corresponding noninteracting system, i.e.,  $T_F = E_F/k_B$ . The validity of this expansion was verified by comparing it to the numerical solution to Eq. (7).

The saturation solubility is obtained by simply setting the chemical potential of  $^3\text{He}$  in the mixture equal to that in pure  $^3\text{He}$ . That is,  $\mu_3 = \mu_{30}$ . To be exact, the comparison should be made between the dilute and rich phases of the phase-separated liquid, but at temperatures below a few hundred millikelvins, the  $^3\text{He}$ -rich phase is indeed practically pure  $^3\text{He}$ .<sup>7,15</sup> The chemical potential of pure  $^3\text{He}$  is determined from the Gibbs-Duhem equation resulting in

$$\mu_{30} = -L_{30} - \int_0^T s_{30} dT' + \frac{1}{N_A} \int_0^P v_{30} dP', \quad (11)$$

where we denote the latent heat of evaporation by  $L_{30}$ , entropy per  $^3\text{He}$  atom by  $s_{30}$ , and the molar volume of pure  $^3\text{He}$  by  $v_{30}$ . Due to the chosen path of integration,  $s_{30} = s_{30}(T, P)$  and  $v_{30} = v_{30}(T = 0, P)$  in the above equation. We calculated the entropy from the heat-capacity data of Greywall<sup>16</sup> and used the molar volume data of Greywall<sup>17</sup> and Abraham *et al.*<sup>18,19</sup> The connection between the chemical potential (interaction potential) and osmotic pressure is discussed in Ref 3.

### IV. FIT PROCEDURE

As our approach is phenomenological, some decisions about the functional form of the potential and the fit parameters

have to be made. Also, some sensitive quantities, which have not been measured with good enough accuracy, are allowed to vary slightly. The fit procedure itself is straightforward. With the choice of fit parameters, a least-squares fitting is performed using the experimental data. Slightly different weights are given to different data sets.

### A. Experimental data

As mentioned, we used the saturation solubility of Ref. 2 and osmotic pressure of Ref. 3 to find the interaction potential. The saturation concentration at low temperatures is usually written as

$$x_s = x_0(1 + \beta T^2), \quad (12)$$

where both  $x_0$  and  $\beta$  depend on pressure. The data of Ref. 2 give these two quantities over the whole pressure range of liquid mixtures. Due to the scheme of measurements, the relative accuracy of  $x_0$  over pressure is very good, but the absolute value is chosen somewhat arbitrarily to match the ‘‘canonical value’’  $x_0 = 6.65\%$  at zero pressure. There is quite a lot of scatter in the data of  $\beta$ , and the values by different authors differ to some extent. While  $\beta$  is not known accurately, it is still rather sensitive to the potential. Because of this, somewhat less weight was given to  $\beta$  during the fitting process. To simulate the experimental conditions,  $\beta$  was calculated between 0 K and 40 mK (while using the low-temperature expansion for  $\mu_3$ ). Even at temperature as low as 40 mK, the quadratic approximation for the chemical potential is not completely valid anymore. A small constant correction of  $0.3 \text{ K}^{-2}$  (less than 10%) in the value of the calculated  $\beta$  was added during fitting to compensate for the quadratic approximation of  $\mu_3$ . This particular value was chosen after comparing the results of the exact calculation to those using the quadratic approximation. After the fitting, the  $\beta$  factor was calculated over the entire pressure range using the exact method, and a consistency check with the experimental values was performed. We used only the zero-temperature osmotic pressure in the fitting, as the interaction does not contribute significantly to its temperature dependence at low temperatures. This is discussed further in Ref. 3.

### B. Form of the potential

The experimental data (saturation solubility) covers the entire pressure range of liquid-helium mixtures. Therefore, we also wanted to find a potential, which is determined at any given pressure in this range. In order to keep the number of fit parameters as small as possible, we decided to treat the BBP result for  $V(0)$  as exact. For the functional form of the potential, we used three Gaussian functions added together. That is,

$$V(k) = V(0) \sum_{i=1}^3 A_i e^{-C_i [k/(2k_0)]^2}, \quad (13)$$

where the arbitrary factor  $k_0$  was chosen to be  $0.3482 \text{ \AA}^{-1}$ , which is the Fermi wave vector  $k_F$  at zero pressure and concentration  $x = 6.65\%$ . Many previous analyses of the interaction potential have used polynomial series in powers of  $k$ , which, of course, do not allow one to use the potential

at large values of momenta (high temperatures). The Gaussian functions also have the added value that they can be inverse Fourier transformed back to real space if one wishes to perform such calculations.

To see what kind of pressure dependence, in addition to the inherent pressure dependence of  $V(0)$ , is required for the parameters to account for the measured data, the fitting procedure was first performed separately at a few distinct pressures. It was noticed that a potential composed of just two Gaussians was adequate to reproduce the appropriate values at a given pressure, but would have required a rather complicated pressure dependence for its parameters. The three Gaussians, however, only required linear dependence in some of the parameters.

The pressure dependence was chosen quite arbitrarily so that  $A_1$ ,  $A_2$ ,  $C_1$ , and  $C_2$  were of the form

$$X(P) = X_0 + X_1 P. \quad (14)$$

Furthermore,  $A_3 = 1 - A_1 - A_2$  holds, while  $C_3$  is a constant.

### C. Concentration dependence

By estimating the lowest-order corrections caused by screening of the interaction by other quasiparticles, it is rather straightforward to show that the interaction should become stronger (more negative) at zero momentum transfer. Furthermore, the correction is proportional to the second power of the zero-concentration, or bare, interaction. This was indeed the result of the considerations of Fu and Pethick.<sup>20</sup> They assumed that the bare interaction remains unchanged as concentration is increased and then determined all correction terms of order  $x^{1/3}$ .

The concentration dependence due to Fu and Pethick did not yield exceptionally good agreement with measurements. One should note that already Fu and Pethick did have a problem using their concentration dependence and Baym’s result for  $V(0)$ . This may indicate a fault either in their analysis, or that of BBP.

Ignoring all concentration dependencies did not yield good results either. The most difficult quantity to reproduce was the temperature coefficient  $\beta$ , and especially its increase at higher pressures. If we forced  $\beta$  to match as well as possible by giving it extra weight in the fitting, the correspondence in osmotic pressure became poor. Even in this case, the calculated values of  $\beta$  did not fit the measured ones especially well. Relieving the BBP result for  $V(0)$  did not change these observations. Yorozu *et al.* were able to fit their measured  $x_s$  and  $\beta$  quite well using concentration-independent potentials. This is possible presumably because they fitted  $L_3 - E_0$  separately at every pressure, even though this quantity is constant by definition. Their fitted value of  $|L_3 - E_0|$  at 20 bar was over 40% smaller than at 0 bar. At 0 bar, it was  $0.2884 \text{ K}$ , which is yet 10% smaller than what is suggested by measurements. They also did not need to concern themselves so much with the apparent increase of  $\beta$  at the highest pressures.

To obtain a candidate for the proper concentration dependence, consistent with the experimental results, we first fitted the data at 25.3 bar pressure, where we had the most data, and used a polynomial form for the concentration dependence. It was noticed that the  $x$  dependence fitted in this way was

surprisingly close to being proportional to the second power of the potential. In order to deal with the pressure dependence of the  $x$  dependence, and limiting the number of fit parameters, we decided to use the following form:

$$V(k,x) = V(0) \left[ \frac{V(k)}{V(0)} + ax^b \left( \frac{V(k)}{V(0)} \right)^2 \right], \quad (15)$$

where  $a$  and  $b$  were to be found by fitting. Since we wanted to incorporate all concentration dependence in this one term, we used the BBP result at zero concentration. That is, the parameters  $\alpha$ ,  $c_4$ , and  $n_4$  appearing in  $V(0)$  were all evaluated at  $x = 0$ .

Some of the concentration correction terms, as discussed by Fu and Pethick, also depend on the angle between the quasi-particle momenta. Since these complicate all calculations to some extent, and are hard to account for in a phenomenological approach, such effects are not included here.

#### D. Additional fit parameters

Connecting the interaction potential to the measured data requires knowledge of some other quantities. One of the central factors is the BBP parameter  $\alpha$ , which appears both directly in the interaction potential and also in the molar volume (and thus, for example, in the Fermi momentum) of mixtures. There are some conflicting data about the pressure dependence of  $\alpha$ . As stated above, we decided to use the data of Watson *et al.* as the starting point. Hatakeyama *et al.* measured  $\alpha$  as a function of temperature, pressure, and concentration.<sup>14</sup> Unfortunately, their data only extend to 10 bar. At this pressure, their value is 3% larger than that of Watson *et al.* Unpublished data of Abraham, Brandt, and Eckstein suggest that  $\alpha$  might in fact be smaller than Watson's by approximately the same amount.<sup>21</sup> Boghosian and Meyer measured the BBP parameter between 0 and 20 bar, and at 20 bar obtained a value, which is 15% smaller than that of Watson *et al.*<sup>22</sup> Considering the spread in the measured data, it is clear that some degree of freedom for  $\alpha$  is justified. We decided to adjust the data of Watson *et al.*,  $\alpha_0$ , in the following manner:

$$\alpha = (a_1 + a_2 P)\alpha_0, \quad (16)$$

which allows the tuning of the zero-pressure value as well as the slope. Another quantity, which is not perfectly well known, is the binding energy of  $^3\text{He}$ . We fitted  $(L_3 - E_0)$  as a constant over the entire pressure range.

### V. RESULTS

The obtained values for all fit parameters of the potential are shown in Table I.

The fitted "binding energy difference" is  $(L_3 - E_0)/k_B = -0.311$  K. This is in excellent agreement with the experimental value of  $-0.312 \pm 0.007$  K.<sup>23</sup> The adjustment parameters of  $\alpha$ , as defined in Eq. (16), are  $a_1 = 0.971$  and  $a_2 = 0.038$   $1/25 \text{ bar}^{-1}$ . Thus, the corrections to the experimental values (Watson *et al.*) of the BBP parameter are at most 3%.

The zero-concentration interaction potentials at various pressures are plotted in Fig. 1. The shape of the potential seems reasonable. The zero- $k$  value remains negative at all pressures, as is expected due to treating the BBP result for  $V(0)$  as

TABLE I. Fitted parameters for the interaction potential. The various parameters are defined through Eqs. (13), (14), and (15). The pressure factors are in units  $1/(25 \text{ bar})$ .

X	$X_0$	$X_1$ ( $1/25 \text{ bar}^{-1}$ )
$A_1$	-5.8553	-1.9882
$A_2$	-9.9140	-4.6585
$C_1$	0.395	-0.301
$C_2$	0.149	0.270
$C_3$	0.383	
$a$	-2.6064	
$b$	1.0071	

exact. At low momenta, the potential increases approximately quadratically, as was already reasoned by BBP.<sup>4</sup> Our  $V(k)$  becomes positive at around  $k = 1.4k_0$ , about the same value as the potentials obtained by other authors. The increase of  $V(k)$  then continues almost linearly, which differs from some previous considerations. If we inverse Fourier transform the potential to real space, the form of  $V(r)$  resembles that of the Lennard-Jones potential, as one might expect. The exact form of  $V(r)$ , however, can not be deduced from these considerations alone, as we have not pinpointed the behavior of the potential at high momentum transfers  $p \gtrsim 2p_F$ .

The concentration dependence of the potential is illustrated in Fig. 2, where the  $P = 0$  potential is plotted at concentrations  $x = 0$  and  $x = 6.65\%$ . The potential of Yorozu *et al.* at  $P = 0$  is shown for comparison. Since the concentration dependence is proportional to the second power of the potential and the correction term is positive, the value of  $V(k, x \neq 0)$  is larger than  $V(k, 0)$  at small and large momenta. We see that the potential of Yorozu *et al.* has a clearly smaller slope at high momentum transfers. Even our best-fitting concentration-independent potential, which is also shown in Fig. 2, has a clearly larger slope. This seems to be required by  $\beta$ , as it was not well fitted for a potential shaped like that of Yorozu *et al.* Again, we believe they were able to obtain good consistency

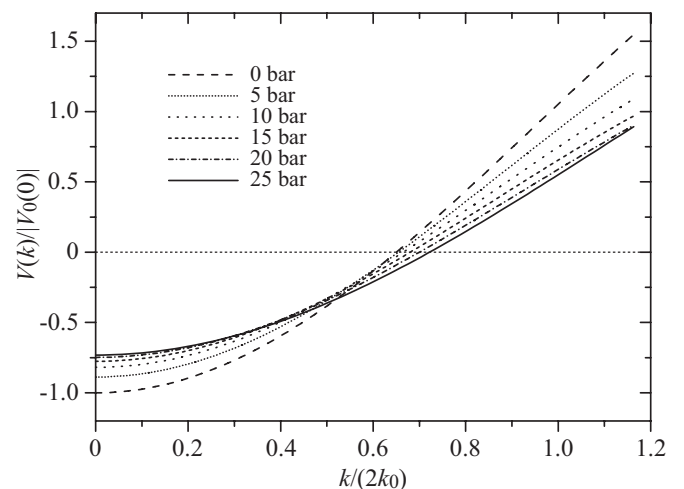


FIG. 1. Fitted interaction potential  $V(k)$  at zero concentration and six different pressures. The potentials are normalized by the zero-pressure value of  $V(0)$ .

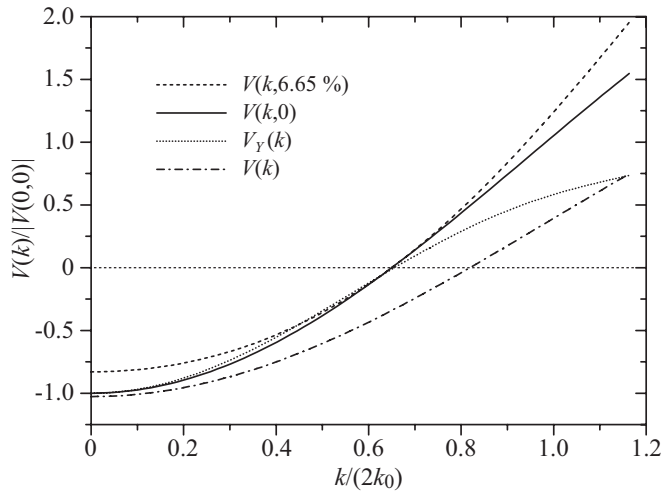


FIG. 2. Fitted interaction potential  $V(k,x)$  at zero pressure and concentrations  $x = 0$  and  $x = 6.65\%$ . The zero-pressure potential of Yorozu *et al.* (Ref. 6)  $V_Y(k)$  is shown for comparison. Also shown is the potential we obtained when no concentration dependence is assumed. The potentials are normalized by the ( $P = 0$ ,  $x = 0$ ) value of  $V(0)$ .

with experimental data because they used a smaller value of  $|L_3 - E_0|$ .

All  $\beta$  data could be fitted quite well without a concentration dependence if the potential was allowed to make an unrealistic turn to even larger slope at large  $k$  (some of the exponents becoming positive). The shape of this kind of potential was suspicious and increased exponentially at large  $k$ . Therefore, we neglected such potentials, and limited the exponents to negative values.

The measured and calculated saturation solubility are shown in Fig. 3, illustrating the excellent correspondence between the two. Throughout the fitting process, it was clear that the solubility was “the easiest” to fit. By this we mean that the solubility was almost always well reproduced early on during the fitting process, regardless of the concentration

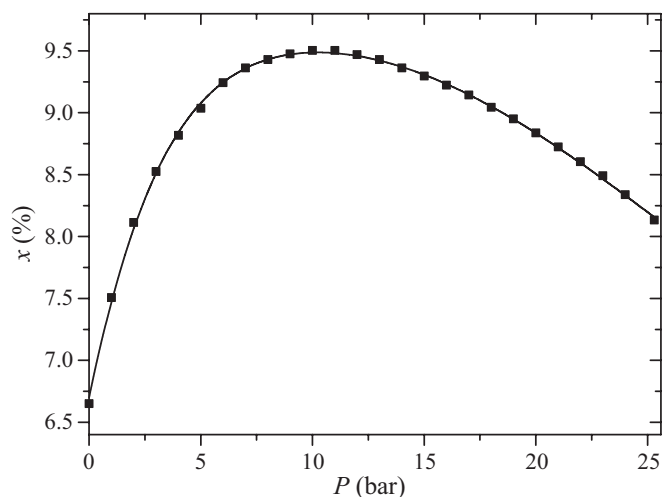


FIG. 3. Measured and calculated zero-temperature saturation solubility over the entire pressure range of liquid mixtures. The experimental data are from Ref. 2.

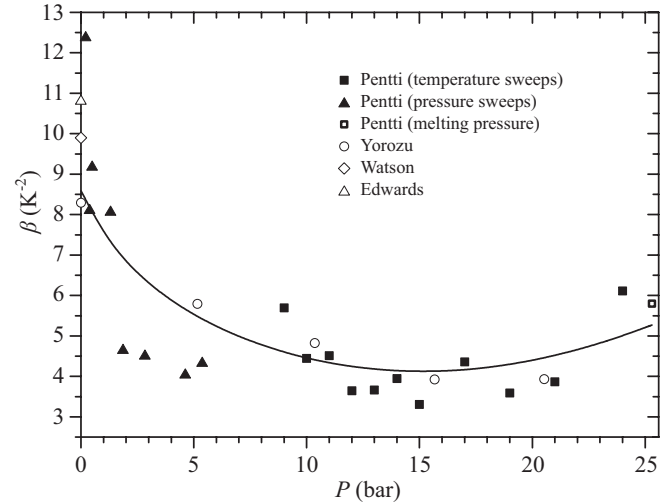


FIG. 4. Measured and calculated temperature coefficient  $\beta$  of saturation solubility over the entire pressure range of liquid mixtures. It has been calculated using the “exact” chemical potential and is evaluated at temperatures between 20 and 40 mK. The experimental data are from Pentti *et al.* (Ref. 2) (the melting pressure data point is different from that given in Ref. 2, as this is due to reanalysis performed after the publication of Ref. 2), Yorozu *et al.* (Ref. 6), Edwards *et al.* (Ref. 24), and Watson *et al.* (Ref. 12).

dependence, etc. This was possibly due to the ability of  $L_3 - E_0$  to adjust the solubility accordingly. This was not so with  $\beta$ , which is shown in Fig. 4. The general features of the calculated  $\beta$  correspond to experimental values fairly well, especially considering the apparent uncertainty in the data. One should note that Edwards *et al.* and Watson *et al.* found their values of  $\beta$  at temperatures somewhat high, between 25 and 150 mK. This causes  $\beta$  to appear larger than in the zero-temperature limit, as the quadratic temperature expansion of the solubility is no longer valid. A comparison between the measured and calculated osmotic pressure is shown and discussed in Ref. 3.

The fit parameters obtained, when no concentration dependence is assumed, are listed in Table II. In this case again, the binding-energy difference comes close to the expected value  $(L_3 - E_0)/k_B = -0.309$  K. The  $\alpha$  parameters were  $a_1 = 0.985$  and  $a_2 = 0.002$ . The concentration-independent potential at zero pressure is also plotted in Fig. 2. Generally, concentration-independent potential seemed to require  $V(k)$  to have its zero at larger values of  $k$  than the

TABLE II. Fitted parameters for the interaction potential when no concentration dependence is assumed. The various parameters are defined through Eqs. (13) and (14). The pressure factors are in units  $1/(25 \text{ bar})$ .

$X$	$X_0$	$X_1$ ( $1/25 \text{ bar}^{-1}$ )
$A_1$	-9.5533	0.3575
$A_2$	-6.5724	-3.1747
$C_1$	0.268	-0.163
$C_2$	0.158	0.242
$C_3$	0.312	

concentration-dependent potentials or those by other authors. The zero-temperature solubility and osmotic pressure data were fitted well also with the concentration-independent potential. The values of  $\beta$ , on the other hand, were considerably smaller than the experimental data. At 25 bar, the calculated value in this case was  $\beta \approx 3$ .

### VI. SUPERFLUID TRANSITION

One of the most interesting predictions, which can be obtained from the potential, is the superfluid transition temperature of the  $^3\text{He}$  component. Unfortunately, this is also perhaps the most unreliable result as it has an exponential dependence on the potential. According to the BCS theory, any fermion system with an attractive interaction between the particles will form Cooper pairs and condense into its ground state at sufficiently low temperature. As the interaction between  $^3\text{He}$  atoms has turned out to be attractive, a dual superfluid phase of helium mixtures must exist. This phase has not yet been detected experimentally. It would be an interesting intermingled liquid Fermi-Bose superfluid, the first of its kind to be observed. Preparations for an experiment to find this state are on the way.

The transition temperature depends, of course, on the details of the interaction between the fermions. Using the generalized BCS theory, the transition temperature can be calculated approximately from

$$T_c \lesssim T_F e^{N(0)V_l}, \quad (17)$$

where  $N(0)$  is the density of states at the Fermi level and  $V_l$  is the interaction component with angular momentum  $l$ . In the above equation,  $T_F$  is the true Fermi temperature, i.e., with the concentration-dependent effective mass. In  $^3\text{He}$ - $^4\text{He}$  mixtures, two ways of pairing are possible, namely,  $l = 0$  and 1, called the  $s$ -wave and  $p$ -wave pairing, respectively. Pairing states of larger values of angular momentum have vanishingly small  $T_c$ . In general, the  $s$ -wave pairing is favorable at low concentrations, while the  $p$ -wave pairing overcomes this when the concentration increases, which is illustrated in Fig. 5. One should note that, due to the exponential dependence on the

interaction strength, the quantitative result is quite sensitive to the precise form of the potential. Since the solubility and osmotic pressure are not that sensitive to the detailed form of  $V(k)$ , we must admit at least an order of magnitude uncertainty in the calculated  $T_c$ . The numerical values are deduced as follows.

The exponents in Eq. (17) can be determined from the Landau parameters  $F_l^{a,s}$  as

$$N(0)V_0 = -1/F_0^a, \quad (18)$$

$$N(0)V_1 = -3/F_1^s. \quad (19)$$

To preserve antisymmetry of the total wave function of Cooper pairs, we must choose the antisymmetric Landau parameter  $F_l^a$  for  $l = 0$  and the symmetric  $F_l^s$  for  $l = 1$ . Thus, the critical temperatures for the  $s$ -wave ( $s$ ) and  $p$ -wave ( $p$ ) are

$$T_c^s \approx T_F e^{-1/F_0^a}, \quad (20)$$

$$T_c^p \approx T_F e^{-3/F_1^s}, \quad (21)$$

respectively. The Landau parameters are related to the potential in the Hartree-Fock approximation as

$$F_0^a = -\frac{N(0)}{2k_F^2} \int_0^{2k_F} kV(k)dk, \quad (22)$$

$$F_1^s = -\frac{3N(0)}{2k_F^2} \int_0^{2k_F} k \left(1 - \frac{k^2}{2k_F^2}\right) V(k)dk, \quad (23)$$

where the density of states at the Fermi level is

$$N(0) = \frac{m^* p_F}{2\pi^2 \hbar^3}. \quad (24)$$

The calculated transition temperatures at several pressures as a function of  $^3\text{He}$  concentration are presented in Fig. 5. The general shapes of the curves are similar to many previous estimates. The  $s$ -wave pairing dominates at low concentrations, but is heavily suppressed beyond  $x \approx 3\%$ . At higher concentrations, the  $p$ -wave pairing becomes preferable, and yields the maximum  $T_c$ . The highest transition temperature obtained with our potential is almost  $200 \mu\text{K}$  and is such high at pressures between 5 and 10 bar at the saturation solubility. Mixtures have been cooled to temperatures in this range. Oh *et al.* cooled a helium mixture at 10 bar pressure down to  $100 \mu\text{K}$  without observing the superfluid transition.<sup>25</sup> The concentration in their experiment, however, was not quite the maximum at this pressure. Rather, it was 9% instead of 9.5%. Our calculated estimate for the  $T_c$  under these conditions is approximately the same  $100 \mu\text{K}$ . The predicted maximum  $T_c$  for the  $s$ -wave pairing is a few microkelvins, and seems to be out of reach by experiments. The highest  $T_c$  at 25.3 bar, the ‘‘natural pressure’’ of the upcoming adiabatic-melting experiment,<sup>26</sup> is  $40 \mu\text{K}$ . This is within the range of plausible experimental temperatures.

### VII. CONCLUSIONS

We estimated the effective interaction potential between  $^3\text{He}$  quasiparticles in helium mixtures by fitting simultaneously the experimental solubility and osmotic pressure

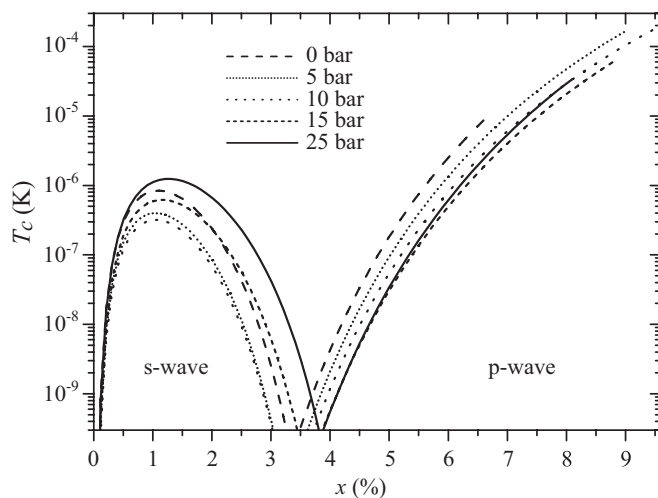


FIG. 5. Predicted superfluid transition temperature at five different pressures.

data covering wide ranges in concentration, pressure, and temperature. The simultaneous fitting was done in order to reduce “the degeneracy” in the problem. Indeed, it was found that any single experimental value could be reproduced well in more than one way, if considered separately, emphasizing the need for fitting several quantities simultaneously. Demanding consistent variation of the parameters over concentration and pressure supports reliability of the method by effectively averaging the data.

The interaction potential may be even more sensitive to transport properties, such as spin diffusion and viscosity, but we did not have means to retrieve such data in our experiments. Measurements by other groups at various pressures and concentrations are available, but it is difficult to ensure full consistency between data sets. Also, in the light of the upcoming melting pressure experiment, we are particularly interested in the interaction potential at the melting pressure, where other measurements have not been performed.

We were able to fit all data satisfactorily only by assuming a concentration dependence in the interaction potential. The unexpected sign of this contribution does raise some concern over its validity. This result to be correct requires the existence of an effect, which has a stronger concentration dependence than the screening by other quasiparticles and becomes significant only at larger concentrations. Here, we can only speculate the origin of such an effect, one possibility being the interactions giving rise to superfluidity in pure  $^3\text{He}$ .

It is plausible, of course, that the measured data are still too inaccurate to yield reliable detailed solutions, especially relating to the temperature coefficient  $\beta$  of solubility. One may also question if the Hartree-Fock approximation is a sufficient description for this system. It is difficult to judge whether the suggested concentration dependence is realistic or not because no valid microscopic theory exists. The proposed concentration dependence may only be a crude approximation to the full complexity of the actual situation.

Our estimate for the maximum superfluid transition temperature  $T_c \approx 200 \mu\text{K}$  is optimistic in the sense that it is around the upper limit of the  $T_c$  set by experiments. Estimates by some authors have yielded values many orders of magnitude lower. If the calculated  $T_c$  is on the right scale, it is in the realm of the planned experiments on helium mixtures.

#### ACKNOWLEDGMENTS

The authors wish to thank E. Pentti and A. Sebedash for useful discussions. The work was supported by the Finnish Cultural Foundation, the National Graduate School for Materials Physics, the Academy of Finland, and the European Commission.

#### APPENDIX: LOW TEMPERATURE CHEMICAL POTENTIAL

We outline here the derivation of the low-temperature series expansion of the chemical potential [Eq. (8)] as we have not come across it in the literature. The expression for  $G$  was only mentioned by Yorozu *et al.* This expansion is applicable also

more generally to systems of weakly interacting fermions at low temperatures in the Hartree-Fock approximation. Let us first evaluate the exchange term at zero temperature. That is, we want to calculate

$$- \int \frac{d^3k'}{(2\pi)^3} V(\mathbf{k} - \mathbf{k}') f(k') \quad (\text{A1})$$

in the case where the Fermi function is a step function, and thus the integration should be performed in the range  $k' \leq k_F$ . It is convenient to make a change of variables  $\mathbf{q} = \mathbf{k} - \mathbf{k}'$ . We perform the integration in spherical coordinates and divide the integral into two intervals:  $q \in (0, k_F - k)$  and  $q \in (k_F - k, k_F + k)$ . For the former, the angular integrations are trivial, but for the latter, we must take some care of the integration limits as the azimuthal angle depends on the momenta. After the integration, the exchange term becomes

$$- \frac{1}{4\pi^2} \left( 2 \int_0^{k_F - k} q^2 V(q) dq + \int_{k_F - k}^{k_F + k} q^2 \left[ 1 + \frac{k_F^2 - q^2 - k^2}{2kq} \right] V(q) dq \right). \quad (\text{A2})$$

From this, we readily obtain the zero-temperature term  $-1/2n_3|V(0)|F$  in Eq. (8) by setting  $k = k_F$ .

Next, we consider the lowest-order temperature term  $G$ . We could make a low-temperature series expansion in Eq. (7) and invert the series to obtain an expression for the chemical potential. Instead of this rather involved calculation, we present perhaps a slightly more transparent way of obtaining  $G$ . We consider the lowest-order temperature correction to the free energy and obtain the chemical potential from  $\mu_3 = (\partial F / \partial N_3)_{T, V}$ . Most of this calculation has already been done, for example, by Baym and Pethick<sup>27</sup> and need not be repeated here. They calculated a low-temperature correction of the form

$$\mu(T) = \mu(0) - \frac{\pi^2}{2} \left( \frac{m^*}{3} + n \frac{\partial m^*}{\partial n} \right) \frac{(k_B T)^2}{(\hbar k_F)^2}, \quad (\text{A3})$$

where  $n$  is the particle density and the effective mass is now defined as

$$m^* \equiv p_F \left( \frac{\partial \epsilon}{\partial p} \right)_{|p=p_F}^{-1}. \quad (\text{A4})$$

By using Eq. (A2) in Eq. (2), taking the derivative, expanding the denominator in powers of the interaction potential, and retaining only first-order terms in  $V$ , we obtain

$$m^* = m - \frac{m^2}{4\hbar^2 \pi^2 k_F} \int_0^{2k_F} k \left( 1 - \frac{k^2}{2k_F^2} \right) V(k) dk. \quad (\text{A5})$$

Here, we have set  $\gamma = 0$  in the dispersion relation. Equation (A5) is the well-known relation between the interaction potential and the concentration-dependent effective mass. The expression for  $G$  is obtained by simply substituting this into Eq. (A3) and collecting all terms involving  $V$ .

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- <sup>1</sup>J. Tuoriniemi, J. Martikainen, E. Pentti, A. Sebedash, S. Boldarev, and G. Pickett, *J. Low Temp. Phys.* **129**, 531 (2002).
- <sup>2</sup>E. M. Pentti, J. T. Tuoriniemi, A. J. Salmela, and A. P. Sebedash, *Phys. Rev. B* **78**, 064509 (2008).
- <sup>3</sup>A. Salmela, A. Sebedash, J. Rysti, E. Pentti, and J. Tuoriniemi, *Phys. Rev. B* **83**, 134510 (2011).
- <sup>4</sup>J. Bardeen, G. Baym, and D. Pines, *Phys. Rev.* **156**, 207 (1967).
- <sup>5</sup>C. Ebner, *Phys. Rev.* **156**, 222 (1967).
- <sup>6</sup>S. Yorozu, M. Hiroi, H. Fukuyama, H. Akimoto, H. Ishimoto, and S. Ogawa, *Phys. Rev. B* **45**, 12942 (1992).
- <sup>7</sup>J. Rysti, J. Tuoriniemi, A. Salmela, and A. Sebedash (accepted in *J. Phys.: Conf. Ser.*).
- <sup>8</sup>J. R. Owers-Bradley, P. C. Main, and R. M. Bowley, *J. Low Temp. Phys.* **72**, 201 (1988).
- <sup>9</sup>S. G. Eckstein, Y. Eckstein, C. G. Kuper, and A. Ron, *Phys. Rev. Lett.* **25**, 97 (1970).
- <sup>10</sup>G. Baym, *J. Low Temp. Phys.* **18**, 335 (1975).
- <sup>11</sup>E. Tanaka, K. Hatakeyama, S. Noma, and T. Satoh, *Cryogenics* **40**, 365 (2000).
- <sup>12</sup>G. E. Watson, J. D. Reppy, and R. C. Richardson, *Phys. Rev.* **188**, 384 (1969).
- <sup>13</sup>B. M. Abraham, O. G. Brandt, Y. Eckstein, J. Munarin, and G. Baym, *Phys. Rev.* **188**, 309 (1969).
- <sup>14</sup>K. Hatakeyama, S. Noma, E. Tanaka, S. N. Burmistrov, and T. Satoh, *Phys. Rev. B* **67**, 094503 (2003).
- <sup>15</sup>M. Nakamura, Y. Fujii, M. Yamamoto, M. Kinoshita, and M. Yamaguchi, *J. Low Temp. Phys.* **129**, 43 (2002).
- <sup>16</sup>D. S. Greywall, *Phys. Rev. B* **27**, 2747 (1983).
- <sup>17</sup>D. S. Greywall, *Phys. Rev. B* **33**, 7520 (1986).
- <sup>18</sup>B. M. Abraham and D. W. Osborne, *J. Low Temp. Phys.* **5**, 335 (1971).
- <sup>19</sup>B. M. Abraham, D. Chung, Y. Eckstein, J. B. Ketterson, and P. R. Roach, *J. Low Temp. Phys.* **6**, 521 (1972).
- <sup>20</sup>H. H. Fu and C. J. Pethick, *Phys. Rev. B* **14**, 3837 (1976).
- <sup>21</sup>C. Ebner and D. O. Edwards, *Phys. Rep.* **1**, 77 (1971).
- <sup>22</sup>C. Boghosian and H. Meyer, *Phys. Lett. A* **25**, 352 (1967).
- <sup>23</sup>P. Seligmann, D. O. Edwards, R. E. Sarwinski, and J. T. Tough, *Phys. Rev.* **181**, 415 (1969).
- <sup>24</sup>D. O. Edwards, E. M. Ifft, and R. E. Sarwinski, *Phys. Rev.* **177**, 380 (1969).
- <sup>25</sup>G.-H. Oh, Y. Ishimoto, T. Kawae, M. Nakagawa, O. Ishikawa, T. Hata, T. Kodama, and S. Ikehata, *J. Low Temp. Phys.* **95**, 525 (1994).
- <sup>26</sup>E. Pentti, J. Rysti, A. Salmela, A. Sebedash, and J. Tuoriniemi, *J. Low Temp. Phys.* **165**, 132 (2011).
- <sup>27</sup>G. Baym and C. Pethick, *Landau Fermi-Liquid Theory: Concepts and Applications* (Wiley, New York, 1991).