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# Theory of Dilute Solutions of  $He<sup>4</sup>$  in Liquid  $He<sup>3</sup>$  at Low Temperatures

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Expressions for the  $l = 0$  forward scattering amplitudes for  $He^{3}$ -He<sup>4</sup> and He<sup>4</sup>-He<sup>4</sup> quasiparticle pairs in dilute solutions of  $He<sup>4</sup>$  in liquid  $He<sup>3</sup>$  are derived in terms of thermodynamic quantities. A Fermi-liquid theory of the solutions is developed, and various thermodynamic quantities are derived. The theory agrees well with the available experimental data.

## I. INTRODUCTION

The physics of dilute solutions of  $He<sup>4</sup>$  in liquid  $He<sup>3</sup>$  has been a subject of some interest both experimentally<sup>1,2</sup> and theoretically<sup>3-5</sup> during the past ten years or so. However, there has been little detailed theoretical work done in this area. The principle reason for this is clear: No experimental data have been available in the temperature region amenable to quantitative theoretical treatment. The He<sup>4</sup> concentration  $x_4$  is so low here (in the singlephase region) that the properties of the solutions are determined almost completely by the excitations intrinsic to the He'. Nevertheless, looking toward future improvement of experimental technique, and in view of the fact that our investigations shed some light both on the basic nature of the phase-separation transition and on the interactions between  $He<sup>3</sup>-He<sup>4</sup>$ and  $He<sup>4</sup>$ -He<sup>4</sup> quasiparticle pairs, we present our results.

Section II is devoted to a discussion of the interactions between  $\text{He}^{3}$ -He<sup>4</sup> and  $\text{He}^{4}$ -He<sup>4</sup> quasiparticle pairs. The  $l = 0$  spherically symmetric parts of the forward scattering amplitudes for these pairs are derived by phenomenological arguments (a more rigorous derivation using Green's-function techniques will be published elsehwere). In particular, we find that the  $l = 0$  forward-scattering amplitude for a He<sup>3</sup>-He<sup>4</sup> quasiparticle pair is given by

$$
a_0^{34} = (1+\alpha)/\nu(0) , \qquad (1)
$$

where  $\nu(0)$  is the density of states at the Fermi surface in pure He<sup>3</sup> and  $\alpha$  is the fractional excess volume occupied by a  $He<sup>4</sup>$  atom in liquid  $He<sup>3</sup>$ . Using



FIG. 1. Diagrams contributing to the forward scattering amplitude  $a_0^{34}$ . The external propagators are included for clarity.

the experimental result<sup>6</sup>  $\alpha \approx -0.32$  this becomes

$$
a_0^{34} = 0.68/\nu(0) , \qquad (2)
$$

a result which should be compared with the analogous quantities for parallel and antiparallel scattering in pure He<sup>3</sup>.

$$
a_0^{1*} = 2.9/\nu(0) , a_0^{1*} = -1.1/\nu(0) . \qquad (3)
$$

Equation (2) thus gives us the important result that the temperature range over which a quasiparticle description is valid for the  $He<sup>4</sup>$  atoms may be expected to be the same as that for  $He<sup>3</sup>$  quasiparticles (i.e.,  $T \leq 0.1$  K).

Another result found in Sec. II is that

$$
a_0^{44} = \left(\frac{\partial \mu_4}{\partial n_4}\right)_{\mu_3},\tag{4}
$$

in which  $\mu_{\,4}$  and  $\mu_{\,3}$  are, respectively, the He $^4$  and He<sup>3</sup> chemical potentials, and  $n_4$  is the He<sup>4</sup> number density. This equation is of the same form as that for He $^3$ -He $^3$  quasiparticle scattering in dilute solu $\cdot$ for He<sup>-</sup>-He<sup>-</sup> quasiparticle scattering in dilute s<br>tions of He<sup>3</sup> in superfluid He<sup>4</sup>.<sup>8</sup>  $a_0^{44}$  is evaluate using an assumption concerning the nature of the phase-separation transition as  $x_4$ ,  $T \rightarrow 0$ ; the result is  $a_0^{44} = 0$ , from which it follows that at small  $x_4$  and T, the transition is closely related to that of a free Bose gas.

In Sec. III we develop a Fermi-liquid theory for the excitations and derive formulas for various thermodynamic quantities, including the specific heat. A comparison with the available experimental data is made, and suggestions for future experimental work are given in Sec. IV.

### II. QUASIPARTICLE SCATTERING AMPLITUDES

As mentioned in the Introduction, the derivations presented here will be phenomenological ones. One of us (W. F. S.) has derived the same results using a Green's-function formalism in a paper which will be published elsewhere.

Let us consider the general diagrammatic structure of the  $He^3-He^4$  scattering amplitude. The diagrams contributing to the forward scattering amplitude (see Fig. 1) may be derived into two classes: diagrams contributing to collective -mode exchange, and all other diagrams. The only  $l = 0$  modes in He<sup>3</sup> which can be exchanged by  $He^3$  and  $He^4$  quasiparticles are sound waves (spin modes are ruled out since the  $He<sup>4</sup>$  has zero spin). The contribution since the real as zero spin). The contribution<br> $a_0^{34}$  to  $a_0^{34}$  due to collective modes must then be of the form

$$
{}^{c}a_{0}^{34} = V_{3-\text{ph}}V_{4-\text{ph}}\lim_{\vec{k}\to 0}\lim_{\omega\to 0}\chi_{33}(\vec{k},\,\omega) , \qquad (5)
$$

in which  $V_{3-\text{ph}}$  and  $V_{4-\text{ph}}$  are renormalized quasiparticle-phonon interaction vertices and  $\chi_{33}(\tilde{k}, \omega)$  is the density-density correlation function (the phonon propagator) in pure He<sup>3</sup>.  $V_{4-ph}$  just measures the change in He<sup>4</sup> chemical potential  $\mu_4$  due to a longwavelength disturbance in the He<sup>3</sup> density  $n_3$ . Hence we have

$$
V_{4-\text{ph}} = \left(\frac{\partial \mu_4}{\partial n_3}\right)_{n_4=0} = (1+\alpha)\left(\frac{\partial \mu_3}{\partial n_3}\right)_{n_4=0},\tag{6}
$$

using a thermodynamic identity in the last step. Similarly,  $V_{3-\nu h}$  measures the change in the He<sup>3</sup> quasiparticle energy  $\epsilon_{3\beta}$  due to a long-wavelength disturbance in  $n_3$ :

$$
V_{3-\text{ph}} = \left(\left.\frac{\partial \epsilon_{3\rho}}{\partial n_3}\right)_{n_4=0}\right|_{\rho=\rho_F} . \tag{7}
$$

The quasiparticle momentum  $p$  is set equal to the Fermi momentum  $p_F$  after the derivative is taken since one must exclude effects of a purely statistical nature. <sup>9</sup> The derivative in  $(7)$  may be expressed in terms of the  $l = 0$  symmetric Fermi-liquid parameter  $F_0^s$  in pure He<sup>3</sup>. We have, then,

$$
V_{3-\text{ph}} = F_0^s / \nu(0) \tag{8}
$$

Since<sup>10</sup>

$$
\lim_{\vec{k}\to 0} \lim_{\omega\to 0} \chi_{33}(\vec{k}, \omega) = -\frac{\nu(0)}{1+F_0^s} = -\left(\frac{\partial n_3}{\partial \mu_3}\right)_{n_4=0}, \quad (9)
$$

we obtain

obtain  

$$
{}^{c}a_{0}^{34} = -(1+\alpha)F_{0}^{s}/\nu(0).
$$
 (10)

The remaining term  ${}^{nc}a_0^{34}$  is just the response of the energy of a He<sup>3</sup> atom when a He<sup>4</sup> atom is added,

keeping the collective-mode coordinate 
$$
n_3
$$
 constant:  
\n
$$
{}^{n}e_{a_0}^{34} = \left(\frac{\partial \mu_3}{\partial n_4}\right)_{n_3} \Big|_{n_4=0} = (1+\alpha)\left(\frac{\partial \mu_3}{\partial n_3}\right)_{n_4=0}
$$
\n
$$
= (1+\alpha)(1+F_0^8)/\nu(0) . \qquad (11)
$$

Adding the results  $(10)$  and  $(11)$ , we obtain  $(1)$ 

$$
a_0^{34} = (1+\alpha)/\nu(0) \tag{1}
$$

We note in passing that (1) is easily shown to be equivalent to

$$
a_0^{34} = \left(\frac{\partial \epsilon_{3p}}{\partial n_4}\right)_{\mu_3} \bigg|_{p=p_F, n_4=0} . \tag{1'}
$$

It is interesting to note at this point that  $a_0^{t_i}$  and  $a_0^{\dagger}$  [see Eq. (3)], given by<sup>11</sup>

$$
\nu(0)a_0^{\dagger 1} = \frac{F_0^8}{1 + F_0^8} - \frac{F_0^6}{1 + F_0^a} ;
$$
  
\n
$$
\nu(0)a_0^{\dagger 1} = \frac{F_0^8}{1 + F_0^8} + \frac{F_0^a}{1 + F_0^a} ,
$$
\n(12)

may be derived in the same fashion if one takes care to include paramagnon exchange.

The  $l = 0$  forward scattering amplitude  $a_0^{44}$  of two  $He<sup>4</sup>$  quasiparticles (in the limit of very small quasiparticle momenta) is easily derived along the same lines. The phonon exchange part is

$$
{}^{c}a_{0}^{44} = V_{4-ph}^{2} \lim_{\vec{k} \to 0} \lim_{\omega \to 0} \chi_{33}(\vec{k}, \omega) = -\left(\frac{\partial \mu_{4}}{\partial n_{3}}\right)_{n_{4}=0}^{2} \left(\frac{\partial n_{3}}{\partial \mu_{3}}\right)_{n_{4}=0},
$$
\n(13)

and the part due to other processes is

$$
{}^{nc}a_0^{44} = \left(\frac{\partial \mu_4}{\partial n_3}\right)_{n_3}\Big|_{n_4=0} . \tag{14}
$$

Adding (13) and (14) and doing some simple variable transformations yields

$$
a_0^{44} = \left(\frac{\partial \mu_4}{\partial n_4}\right)_{\mu_3}\bigg|_{n_4=0} \ . \tag{15}
$$

This result is of the same form as that used in the effective potential of Bardeen, Baym, and Pines. However, it is important to note that (15) includes exchange, so that if one were to construct an effective potential for  $He<sup>4</sup>$ -He<sup>4</sup> scattering here, then its zero-momentum transfer limit mould be

$$
V_{44}(\vec{k}=0) = \frac{1}{2} \left( \frac{\partial \mu_4}{\partial n_4} \right)_{\mu_3} \bigg|_{n_4=0} . \tag{16}
$$

With the aid of a very plausible assumption concerning the nature of the phase-separation curve, one may evaluate  $a_0^{44}$ . The assumption is that the endpoint of the phase-separation line (at  $x_4 = 0$ ,  $T=0$ ) is a consolute point [in the sense that  $(\partial \mu_4/\partial n_4)_b = 0$  there]. To understand the import of this assumption, let us examine the possible alternatives: (i) The end point is neither a  $\lambda$  point, nor a consolute point. In this case there is a metastable region at  $T = 0$  and  $x_4 \neq 0$  in which we have a nonsuperfluid gas of Bose quasiparticles. Also, in this region  $(\partial \mu_4/\partial n_4)_{P,T=0}$ , which for small  $x_4$  is equal to  $(\partial \mu_4/\partial n_4)_{\mu_3, T=0}$ , must be greater than zero.<sup>12</sup> From this it follows that the effective potential [see Eq.  $(16)$ ] is repulsive. Existing theory indicates<sup>13</sup> that a dilute Bose gas with repulsive. interactions should be superfluid at  $T=0$ . Hence, we reject alternative (i). (ii) The end point is a  $\lambda$  point and not a consolute point. In this case there would be some region for small  $x_4$  and T where

there would exist a dilute solution of superfluid  $He<sup>4</sup>$ in liquid  $He<sup>3</sup>$ . Such a solution has never been observed. Finally, we ca11 attention to the calculation of Cohen and van Leeuwen<sup>5</sup> on a dilute-gas model of  $He<sup>3</sup>-He<sup>4</sup>$  solutions which verifies our assumption in a special case.

Proceeding on the assumption that the end point of the phase-separation curve is indeed a consolute point, and noting again that  $(\partial \mu_4/\partial n_4)_{P,T}$  is equal to  $(\partial \mu_4 / \partial n_4)_{\mu_3, T}$  in the limit as  $x_4 \rightarrow 0$ , we immediate see from  $(15)$  that

$$
a_0^{44} = 0 \tag{17}
$$

# III. FERMI-LIQUID THEORY

We commence the development of a Fermi-liquid theory for the mixtures in question by expanding the energy to second order in deviations of the quasiparticle distribution functions  $n_{3b}$  and  $n_{4b}$  from their values at  $T=0$  and  $x_4=0$ :

$$
E - E_0 = \sum_{p} \epsilon_{3p} \delta n_{3p} + \sum_{p} \epsilon_{4p} \delta n_{4p} + \frac{1}{2} \sum_{pp'} f_{pp'}^{33} \delta n_{3p} \delta n_{3p'}
$$
  
+
$$
\sum_{pp'} f_{pp'}^{34} \delta n_{3p} \delta n_{4p'} + \frac{1}{2} \sum_{pp'} f_{pp'}^{44} \delta n_{4p} \delta n_{4p'} + \cdots ; \qquad (18)
$$

here  $E_0$  is the ground-state energy of pure He<sup>3</sup>. This theory is, of course, valid only in the onephase region. Our object now is to evaluate the terms on the right-hand side of  $(18)$  in terms of  $T$ ,  $n_4$ , and  $\delta n_3 = n_3 - n_3^0$ ,  $n_3^0$  being the particle number density in the ground state of pure  $He<sup>3</sup>$ . We use the spectrum

$$
\epsilon_{4p} = \epsilon_{40} + p^2/2m^*_{4} \t\t(19)
$$

where  $\epsilon_{40}$  is the zero-temperature zero-concentration limit of  $\mu_4$ , and  $m_4^*$  is an effective mass. Then, denoting by  $\epsilon_B(n_4, T)$  the energy per particle of a free Bose gas, we have

$$
\sum_{p} \epsilon_{4p} \delta n_{4p} = n_4 [\epsilon_{40} + \epsilon_B(n_4, T)] \tag{20}
$$

For low temperatures (hence, small  $He<sup>4</sup>$  momenta) we take the  $l = 0$  part of  $f_{pp'}^{44}$  equal to a constant independent of  $T$ . Then we have

$$
\frac{1}{2}\sum_{\rho\rho'} f_{\rho\rho'}^{44} \delta n_{4\rho} \delta n_{4\rho'} = \frac{1}{2} f_0^{44} n_4^2 . \qquad (21)
$$

 $\delta n_{3p}$  will be nonzero only very near the Fermi surface. Hence we find

$$
\sum_{\rho\rho'} f_{\rho\rho}^{34} \delta n_{3\rho} \delta n_{4\rho'} = f_0^{34} n_4 \delta n_3 , \qquad (22)
$$

$$
\frac{1}{2}\sum_{\rho\rho'} f_{\rho\rho}^{33} \cdot \delta n_{3\rho} \delta n_{3\rho'} = \frac{1}{2} \frac{F_0^s}{\nu(0)} (\delta n_3)^2 \,, \tag{23}
$$

where  $f_0^{34}$  is the  $l = 0$  part of  $f_{pp'}^{34}$  in the limit of small He<sup>4</sup> momentum p'. The sum  $\sum_{p} \epsilon_{p} \delta n_{3p}$  must be carefully evaluated, taking into account both the effect due to nonzero  $T$  and that due to the fact that addition of He<sup>4</sup>'s changes the equilibrium He<sup>3</sup> density. The latter effect is

$$
\left(\sum_{p} \epsilon_{p} \delta n_{3p}\right)^{(1)} = \mu_{3}^{0} \delta n_{3} + [2\nu(0)]^{-1} (\delta n_{3})^{2} , \qquad (24)
$$

the second term accounting for the change in the Fermi momentum  $p_F$ . The former effect is the usual<sup>14</sup> change in  $\delta n_{3p}$  due to an increase in T, evaluated at the new density  $n_3=n_3^0+\delta n_3$ :

$$
\left(\sum_{\rho} \epsilon_{\rho} \delta n_{3\rho}\right)^{(2)} = \frac{1}{6} \pi^{2} \nu(0) \Big|_{n_{3}^{0} + \delta n_{3}} (K_{B}T)^{2}
$$

$$
= \frac{\pi^{2}}{6} \nu(0) (K_{B}T)^{2} \Big(1 + \frac{1}{\nu(0)} \frac{\partial \nu(0)}{\partial n_{3}} \delta n_{3}\Big).
$$
(25)

Thus, we find

$$
\sum_{p} \epsilon_{p} \delta n_{3p} = \mu_{3}^{0} \delta n_{3} + \frac{1}{2\nu(0)} (\delta n_{3})^{2} + \frac{\pi^{2}}{6} \nu(0)
$$

$$
\times (K_{B}T)^{2} \left(1 + \frac{1}{\nu(0)} \frac{\partial \nu(0)}{\partial n_{3}} \delta n_{3}\right). \tag{26}
$$

Collecting our results, we have

$$
E - E_0 = \mu_3^0 \delta n_3 + [\epsilon_{04} + \epsilon_B(n_4, T)] n_4 + f_0^{34} \delta n_3 n_4
$$
  
+ 
$$
\frac{1}{2\nu(0)} (1 + F_0^s) (\delta n_3)^2 + \frac{1}{2} f_0^{44} n_4^2
$$
  
+ 
$$
\frac{1}{6} \pi^2 (K_B T)^2 \nu(0) \left(1 + \frac{1}{\nu(0)} \frac{\partial \nu(0)}{\partial n_3} \delta n_3\right). (27)
$$

From (27) we may calculate various thermodynamic quantities of interest. The specific heat is given by (we use the classical limit  $\epsilon_B = \frac{3}{2}K_B T$  here; the validity of this use is shown below)

$$
C_V = \frac{3}{2} n_4 K_B + \frac{\nu(0)\pi^2}{3} K_B^2 T \left( 1 + \frac{1}{\nu(0)} \frac{\partial \nu(0)}{\partial n_3} \delta n_3 \right). \quad (28)
$$

Since the He' is added at constant pressure (which is equivalent to constant  $He<sup>3</sup>$  chemical potential for small  $x_4$ , we have

$$
\delta n_3 = - (1+\alpha) n_4 \tag{29}
$$

In addition, we have

$$
\frac{n_3}{\nu(0)} \frac{\partial \nu(0)}{\partial n_3} = \frac{1}{3} + \frac{n_3}{m_3^*} \frac{\partial m_3^*}{\partial n_3}
$$
 (30)

and<sup>15</sup>

$$
\frac{n_3}{m_3^*} \frac{\partial m_3^*}{\partial n_3} \approx 2.2 , \qquad (31)
$$

so that  $(28)$  becomes<sup>16</sup>

$$
C_{v} = \frac{3}{2} n_{4} K_{B} + \frac{1}{3} v(0) \pi^{2} K_{B}^{2} T[1 - 2.5(1 + \alpha)x_{4}]. \quad (32)
$$

The chemical potentials are found to be

$$
\mu_3 = \mu_3^0(n_3^0) - \frac{\pi^2}{12} \frac{(K_B T)^2}{\epsilon_F^0} \left( 1 + \frac{1}{\epsilon_F^0} \frac{\partial \epsilon_F^0}{\partial n_3} \delta n_3 \right), \quad (33)
$$

$$
\mu_4 = \epsilon_{04}(n_3^0) + \left(\frac{\partial \mu_4}{\partial n_4}\right)_{\mu_3} n_4 + \mu_B(n_4, T) , \qquad (34)
$$

where  $\mu_B(n_4, T)$  is the chemical potential of a free Bose gas composed of particles of mass  $m_4^*$  and characterized by number density  $n_4$ . Note that  $\mu_3$ changes from its value at  $x_4=0$ ,  $T=0$  only by the usual temperature factor, corrected to the new He<sup>3</sup> number density. Using (15) and (17), (34) reduces to

$$
\mu_4 = \epsilon_{04}(n_3^0) + \mu_B(n_4, T) \tag{35}
$$

Since we have

$$
\frac{n_3}{\epsilon_p^0} \frac{\partial \epsilon_p^0}{\partial n_3} = \frac{2}{3} - \frac{n_3}{m_3^*} \frac{\partial m_3^*}{\partial n_3} = -1.9 , \qquad (36)
$$

 $Eq. (33) becomes$ 

(26) 
$$
\mu_3 = \mu_3^0 (n_4^0) - \frac{\pi^2}{12} \frac{(K_B T)^2}{\epsilon_F^0} [1 + 1.9(1 + \alpha)x_4].
$$
 (37)

The result (35) is important since it states that, to order  $n_4$ , there are no corrections to the free-Bose-gas result for  $\mu_4$ . In addition, (35) is consistent with the assumption of Sec. II that the end point of the phase-separation curve at  $x_4 = 0$ ,  $T = 0$  is a consolute point. This follows from the fact that the  $\lambda$  line determined by  $\mu_B(n_4, T)$  ends at  $n_4=0$ ,  $T=0$ , and that on this line  $(\partial \mu_B / \partial n_4)_T = 0$ .<sup>13</sup> However, since this  $\lambda$  line is actually inside the phase-transition curve (it in some sense "bounds" the metastable region), our argument is not a rigorous one.

# IV. DISCUSSION OF EXPERIMENTS

To date there exist no experimental data in the temperature range ( $T \leq 0.1$  K) where we expect our theory to have quantitative validity. The most complete experimental data on the determination of the phase-separation curve do, however, extend down to 0.17 K.<sup>2</sup> It is thus worthwhile to apply our theory since a good determination of the phase-separation curve would provide an experimental test of the basic relation (17).

The phase-separation curve is found by equating the He<sup>4</sup> chemical potential in the upper (small  $x_4$ ) and lower (large  $x_4$ ) phases. Denoting the latter by  $\mu_4^L$  and using (35) together with the formula

$$
\mu_B(n_4, T) = K_B T [\ln(n_4 \lambda_T^3) - n_4 \lambda_T^3 / 2^{3/2} + \cdots],
$$
 (38)

we obtain

$$
x_4 = (1/n\lambda_T^3)(1 + e^{-\epsilon/K_BT}/2^{3/2})e^{-\epsilon/K_BT} \t\t(39)
$$

Here  $n = n_3 + n_4$  and

$$
\lambda_T = \left(\frac{2\pi\hbar^2}{m\frac{\pi}{4}K_BT}\right)^{1/2} \tag{40}
$$

is the thermal wavelength for the  $He<sup>4</sup>$  quasiparticle

gas

$$
\epsilon \equiv \epsilon_{40}(n_3^0) - \mu_4^L \tag{41}
$$

The second term in brackets in (39) results from the quantum correction [second term in brackets in (38)] to the chemical potential of a noninteracting Boltzmann gas. This correction is negligibly small at very low temperatures, but with the figures we obtain is alrealy  $\approx 2.2\%$  at  $T=0.2$  K. According to our theory it is the only correction when  $a_0^{44} = 0$ . It is possible to write (39) as

$$
\ln \frac{T^{3/2}}{x_4} = \frac{\epsilon}{K_B T} - \ln \left( 1 + \frac{\exp(-\epsilon/K_B T)}{2^{3/2}} \right)
$$

$$
+ \ln \left[ \left( \frac{2\pi\hbar^2}{m_4^*K_B} \right)^{3/2} \right]. \tag{42}
$$

At very low temperatures a plot of the left-hand side of (42) versus  $1/T$  should give a straight line from which the parameters  $\epsilon_{40}$  and  $m_{4}^{*}$  could be deduced. Such a plot is given in Fig. 2 for the range of temperature 0.17 K  $\leq$  T  $\leq$  0.5 K using the data of Ref. 2 on phase separation in dilute  $He<sup>4</sup>$  mixtures. The best fit with the right-hand side of (42), using for  $\mu_{4}^{L}$  the computed values of Radebaugh,  $^{17}$  is obtained for  $\epsilon_{40}$  = -6.61 K and  $m_4^* \cong 4.5 m_4$  ( $m_4$  is the He<sup>4</sup> bare mass). <sup>18</sup> It is remarkable that even at a temperature as high as 0. 5 K the theory is in very good agreement with the experimental results. This seems to indicate that concentration and temperature-dependent corrections to  $a_0^{44}$  are very small.

The values of  $\epsilon_{40}$  and  $m_4^*$  differ from those obthat the values of  $\epsilon_{40}$  and  $m_4$  allocated that the tained earlier.<sup>19</sup> But it should be noticed that the previous value of  $\epsilon_{40}$  was obtained from specificheat measurements on a 4. 6% mixture at temperatures higher than 0. <sup>5</sup> K and using for the excitation spectrum one different from  $(19)$ . Also the extra-

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- J. R. Dillinger (Wisconsin U.P., Madison, 1958), p. 158.  $^7$ See, e.g., D. Pines and P. Nozieres, The Theory of



FIG. 2.  $\ln(T^{3/2}/x_4)$  vs  $1/T$ . The heavy dots denote the experimental points and the line denotes our theoretical curve  $(\epsilon_{40} = -6.61 \text{ K}, m_4^* = 4.5 m_4)$ .

polation of the phase-separation curve was at that time more difficult because no experimental data were available below 0.3 K.<sup>18</sup>

The phase-separation curve in the zero-temperature limit now becomes

$$
x_4 \cong 0.85 T^{3/2} e^{-0.56/T} . \tag{43}
$$

In conclusion it appears that the theory developed here is in very good agreement with experiment. The parameters  $\epsilon_{40}$  and  $m_{4}^{*}$  could be determined with better accuracy (especially in the case of  $m_4^*$ ) if phase-separation measurements were extended to lower temperatures.

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(1969).  $^9$ One does not want to take into account the density dependence of the  $p_F^2$  factor in  $\epsilon_{3p_F}$ .

 $10$ This is just the well-known compressibility sum rule. See, e.g. , Ref. 7, Chap. 2.

 $^{11}$ Ref. 7, Chap. 1.

- $12$ See, e.g., I. Prigogine and R. Defay, Chemical
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- 1963), Chap. 19. Note that we assume  $m<sub>4</sub><sup>*</sup>$  independent of  $n_4$  for small  $x_4$ .

 $^{14}$ See, e.g., Ref. 7.

 $15J.$  Wilks, The Properties of Liquid and Solid Helium (Oxford U. P. , London, 1967), p. 677, Table A14.

 $16$ Note that this differs significantly from the simple interpolation formula used by De Bruyn Ouboter  $et \ al.$  , in Ref. 1.

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<sup>18</sup>The value of  $m<sub>4</sub><sup>*</sup>$  is, however, very sensitive to the way in which the theory is fitted to the data. For example, the best fit to the six lowest-temperature points in Fig. 2 gives  $\epsilon_{40}$ =-6.67K and  $m_4^* \approx 3.7 m_4$ .

<sup>19</sup> De Bruyn Ouboter et al. (Ref. 1) found  $\epsilon_{40}$  = -6.45 K, and Edwards and Daunt (Ref. 3) found  $m_4^*$  = 5.3  $m_4$ .

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# Relaxation Oscillations in Stimulated Raman and Brillouin Scattering'

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The equations describing the transfer of intensity from a pump beam simultaneously to forward and backward stimulated scattered beams in an infinite medium are shown to lead to pulsations similar to relaxation oscillations in the scattered intensities. The period of the pulsations is simply re1ated to the gain length for the forward-scattered beam, and may be much longer than the ordinary transient time (related to the damping of material excitations) usually associated with stimulated scattering. Similar oscillations occur for backward scattering alone (stimulated Brillouin scattering) if the scattering medium is finite. In this case the period equals the photon round-trip time in the medium if the latter exceeds the backward gain length. Both phenomena should be observable in scattering from gases such as  $N_2$  and  $H_2$  at high pressure, and may also play a role in determining the temporal structure of 1ight scattered from the region of a self-focus in liquids.

## I. INTRODUCTION

The transient growth of light scattered inelastically from thermalor quantum fluctuations in a medium has been examined theoretically by previous authors in a variety of special cases. A typical analysis begins with the set of coupled equations describing the motion of a limited number of modes of the optical radiation field and of the medium. These are then simplified using various plausible assumptions about the mechanisms responsible for the particular phenomenon under investigation. Thus Kroll' has analyzed the growth of stimulated Brillouin scattering in the low conversion regime where linearized equations are valid. Similar studies of stimulated Raman, thermal, and Rayleigh wing scattering have appeared more recently. $2$ Tang' has extended the Brillouin scattering analysis to the nonlinear high conversion regime. Recently Maier, Kaiser, and Giordmaine<sup>4</sup> have investigated transient solutions of nonlinear equations describing the interaction of backward-Raman-scattered light with the driving beam. In this paper we report the results of a similar theoretical study in which it was found that the backward-scattered beam experiences transient behavior resembling relaxation oscillations, the duration of which may vastly exceed the transient periods studied in the linear regime.

This behavior has nothing to do with self-focusing, but occurs under very general conditions.

There are two related kinds of "relaxation" oscillations in stimulated scattering situations involving backward-scattered waves. Both are most conveniently observed in the time dependence of the backscattered intensity at the entrance to the scattering medium.

The simplest case is the  $finite-cell$  oscillation which can occur whenever a forward-traveling pump beam drives a stimulated scattered beam in the backward direction in a finite medium. The period is proportional to the sample length  $L$ , and therefore no oscillations of this type are observed if the sample length is infinite.

In contrast, three-wave oscillations may be observed even in a medium of infinite length, but stimulated scattering must occur in the forward as well as in the backward direction. These are similar to the finite-cell oscillations in that the depletion of the incident pump beam intensity by the forward wave effectively truncates the region in which the backward wave experiences gain. Thus the period is proportional to the distance  $L_f$  over which the forward wave travels before growing to an appreciable fraction of the pump intensity. (We are assuming for simplicity that the pump intensity remains constant at the cell entrance, after the