

but also from the calculation by Maris<sup>4</sup> of the magnitude and frequency dependence of our zero-pressure data and from the explanation by Jäckle of the shoulder in our data, is that all these theories require an assumption of anomalous dispersion to account for our data. This has to be interpreted as very

strong evidence for anomalous dispersion. Whether this dispersion should be described by a quadratic or a cubic term in the energy spectrum remains to be determined; more detailed theoretical calculations using both descriptions would be very desirable.

\*Based on work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup>C. J. Pethick and D. ter Haar, *Physica* **32**, 1905 (1966); see also, S. G. Eckstein, Y. Eckstein, J. B. Ketterson, and J. H. Vignos, in *Physical Acoustics*, edited by W. P. Mason and R. N. Thurston (Academic, New York, 1970), Vol. 6, and the references contained therein.

<sup>2</sup>B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and J. H. Vignos, *Phys. Rev.* **181**, 347 (1969).

<sup>3</sup>H. J. Maris and W. E. Massey, *Phys. Rev. Letters* **25**, 220 (1970); also, S. Havlin, thesis (Tel-Aviv University) (unpublished).

<sup>4</sup>H. J. Maris, *Phys. Rev. Letters* **28**, 277 (1972).

<sup>5</sup>Pat R. Roach, J. B. Ketterson, and M. Kuchnir, *Phys. Rev. Letters* **25**, 1002 (1970).

<sup>6</sup>B. M. Abraham, Y. Eckstein, J. B. Ketterson, and J. H. Vignos, *Cryogenics* **9**, 274 (1969).

<sup>7</sup>W. M. Whitney, *Phys. Rev.* **105**, 38 (1957).

<sup>8</sup>K. Dransfeld, J. A. Newell, and J. Wilks, *Proc. Roy. Soc. (London)* **A243**, 500 (1958).

<sup>9</sup>I. M. Khalatnikov and D. M. Chernikova, *Zh. Eksperim.*

*i Teor. Fiz.* **49**, 1957 (1965); **50**, 411 (1966) [*Sov. Phys. JETP* **22**, 1336 (1966); **23**, 274 (1966)].

<sup>10</sup>C. J. N. Van den Meijdenberg, K. W. Taconis, and R. De Bruyn Ouboter, *Physica* **27**, 197 (1961).

<sup>11</sup>B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and Pat R. Roach, *Phys. Rev. A* **1**, 250 (1970).

<sup>12</sup>Texas Instruments Inc., Houston, Tex.

<sup>13</sup>R. Klein and R. K. Wehner, *Phys. Rev. Letters* **23**, 1372 (1969); in *Proceedings of the Twelfth International Conference on Low-Temperature Physics, Kyoto, Japan, 1970*, edited by Eizo Kanda (Academic Press of Japan, Tokyo, 1971), p. 91.

<sup>14</sup>R. Klein and R. K. Wehner, *Helv. Phys. Acta* **44**, 550 (1971).

<sup>15</sup>N. E. Phillips, C. G. Waterfield, and J. K. Hoffer, *Phys. Rev. Letters* **25**, 1260 (1970).

<sup>16</sup>J. Jäckle and K. W. Kehr, *Phys. Rev. Letters* **27**, 654 (1971).

<sup>17</sup>E. Feenberg, *Phys. Rev. Letters* **26**, 301 (1971).

<sup>18</sup>A. Molinari and T. Regge, *Phys. Rev. Letters* **26**, 1531 (1971).

<sup>19</sup>G. Barucchi, G. Ponzano, and T. Regge (unpublished).

## Liquid-Helium Configuration around a Metastable Excited Helium Atom<sup>†</sup>

J. P. Hansen\* and E. L. Pollock

*Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14850*

(Received 22 November 1971)

The pair distribution for normal He atoms in the vicinity of a metastable  $2^3S$  He atom is calculated in the zero-temperature limit, from first principles, using a variational Jastrow wave function and the Percus-Yevick integral equations for fluid mixtures. From this pair function, the "bubble radius," coordination number, and energy per metastable atom are calculated.

Spectroscopic studies of the infrared emission and absorption spectra of neutral localized excitations, created in liquid helium by electron beams with an energy of the order of  $10^2$  keV, have identified these as metastable excited states of He atoms and He<sub>2</sub> molecules shifted only very slightly in energy from their free atomic or molecular values.<sup>1-3</sup> For the atomic states, Hickman and Lane, by extending the calculations of Jortner *et al.*<sup>4</sup> for an excess electron in liquid helium to include the interaction of a helium core with both the excited electron and the liquid, have shown these observa-

tions to be consistent with the existence of a cavity surrounding the excited atom.<sup>5</sup>

The purpose of this paper is to study these "bubble states" from first principles, for the special case of metastable  $2^3S$  atoms (hereafter symbolized by He\*) which have been observed at concentrations greater than  $10^{12}$  atoms cm<sup>-3</sup> by the Rice University group.<sup>1,2</sup> The calculation presented here could be extended to other excitations if the pair interaction between such excitations and normal He atoms were known.

The Hamiltonian for a system of  $N_1$  normal and

$N_2$   $2^3S$  He atoms can be written

$$H = \frac{-\hbar^2}{2m} \sum_{i=1}^{N_1+N_2} \nabla_i^2 + \sum_{i<j}^{N_1} V_{11}(r_{ij}) + \sum_{k=1}^{N_1} \sum_{i=1}^{N_2} V_{12}(r_{ki}), \quad (1)$$

where  $V_{11}(r)$  is the pair potential between two normal He atoms, which is generally taken to be the familiar Lennard-Jones 12-6 potential.  $V_{12}(r)$  is the interaction potential between a metastable and a normal He atom. This potential has an attractive well for separations of about  $2a_0$ , followed by a high repulsive "bump" around  $4a_0$  of several thousand  $^\circ K$ , through which the tunneling probability for normal He atoms, with an average kinetic energy of about  $12^\circ K$ ,<sup>6</sup> is quite negligible. Consequently for  $V_{12}$  we may use the potential given by Fitzsimmons, Lane, and Walters,<sup>7</sup> which incorporates in the long-range part adjustable parameters fitted so as to join smoothly to the theoretically determined short-range part and to give calculated diffusion coefficients in agreement with experiment. Specifically,

$$V_{12}(r) = \alpha r^2 e^{-\eta r} - \epsilon r^{-6} + \frac{1}{2}\gamma e^{-\tau r}, \quad r > 6a_0, \quad (2)$$

where  $\alpha = 0.3$ ,  $\eta = 1.4$ ,  $\gamma = 1.54$ ,  $\tau = 1.43$ , and  $\epsilon = 20$  (all in a. u.). Because of the very low He\* concentration we have omitted the potential energy term for pairs of such atoms from the Hamiltonian.

We write down a trial ground-state wave function of the Jastrow type to describe this system at  $T = 0^\circ K$ ,<sup>8</sup>

$$\psi_0 = \prod_{i<j}^{N_1} f_{11}(r_{ij}) \prod_{k=1}^{N_1} \prod_{i=1}^{N_2} f_{12}(r_{ki}). \quad (3)$$

Defining the He\* concentration  $X = N_2/N$  and the number density  $\rho = V/N$ , with  $N = N_1 + N_2$ , it is easily verified that the energy expectation value per atom is, using Eqs. (1) and (3),

$$\frac{E}{N} = \frac{\rho(1-X)^2}{2} \times \int g_{11}(r) \left[ \frac{-\hbar^2}{2m} \nabla^2 \ln f_{11}(r) + V_{11}(r) \right] d^3 r + \rho X(1-X) \int g_{12}(r) \left[ \frac{-\hbar^2}{2m} \nabla^2 \ln f_{12}(r) + V_{12}(r) \right] d^3 r, \quad (4)$$

where the radial distribution functions are

$$g_{ij}(|r-r'|) = V^2 \int \psi_0^2 dx^{3(N-2)} / \int \psi_0^2 dx^{3N} \quad (5)$$

( $r, r'$  are not integrated over in the numerator). With  $\psi_0$  from Eq. (3), Eq. (5) is identical to the expression for  $g_{ij}$  in a classical binary fluid with the identifications  $\beta U_{ij}(r) = -2 \ln f_{ij}(r)$  for the interaction potentials, and therefore (5) may be approximately calculated by any of the approximation schemes used for classical fluids.

In particular we have used the PY1 mixture equations<sup>9</sup> to determine  $g_{12}(r)$ . In the limit  $N_1 \ll N$  these equations decouple and

$$Y_{12}(r) = \rho \int h_{12}(|r-s|) [h_{11}(s) - Y_{11}(s)] d^3 s, \quad (6)$$

where  $Y_{ij} = g_{ij} e^{\beta V_{ij}} - 1$  and  $h_{ij} = g_{ij} - 1$ , with  $i, j = 1, 2$ .  $h_{11} - Y_{11}$ , corresponding to the pure liquid in this low-He\* concentration limit, was taken from Francis's work<sup>10</sup> based on Schiff and Verlet's wave function<sup>6</sup> in conjunction with the more accurate PY2XS integral equation. For  $f_{12}(r)$  we have chosen a simple one-parameter form which corresponds to the WKB solution at zero energy for tunneling into the dominant (repulsive) part of the potential (2):

$$f_{12}(r) = e^{-U_{12}(r)/2}, \quad U_{12}(r) = C(\frac{1}{2}\eta r + 1)e^{-\eta r/2}. \quad (7)$$

Equation (6) was solved by iteration for various values of the variational parameter  $C$  and then the second term in (4) was minimized to determine the optimum  $C$ . The corresponding  $g_{12}(r)$  allowed us to determine the "bubble radius"  $r_b$  as the distance from the origin to the first peak of  $g_{12}$  and the coordination number  $\nu$  around a He\* impurity,

$$\nu = 4\pi\rho \int_0^{r_{\min}} r^2 g_{12}(r) dr,$$

where  $r_{\min}$  is the position of the first minimum of  $g_{12}(r)$ . Also shown in Table I is the energy per He\* atom in the liquid for several values of  $C$  around the optimum value, as given by the second term in (4). The calculations were performed for two densities,  $\rho = 2.2 \times 10^{22}$  atoms  $\text{cm}^{-3}$ , the normal density of liquid He at  $0^\circ K$ , and  $\rho = 2.41 \times 10^{22}$  atoms  $\text{cm}^{-3}$ .

In Fig. 1 we compare the  $g_{12}(r)$  calculated here for  $\rho = 2.2 \times 10^{22}$  atoms  $\text{cm}^{-3}$  with  $g_{11}(r)$ <sup>6</sup> and with

TABLE I. Energy per He\* atom, coordination number, and "bubble radius" from  $g_{12}(r)$  corresponding to several values of the variational parameter  $C$  at two densities.

$C$	$E/N_2$ ( $^\circ K$ )	$\nu$	$r_b$ ( $a_0$ )
$\rho = 2.2 \times 10^{22}$ atoms $\text{cm}^{-3}$			
250	32.35	31	11.3
300	27.96	34	11.6
340	27.30	34	11.8
365	27.61	35	11.8
400	28.65	35	12.1
500	33.52	38	12.6
$\rho = 2.41 \times 10^{22}$ atoms $\text{cm}^{-3}$			
250	43.17	33	11.1
300	37.00	34	11.6
350	35.64	35	11.8
380	36.03	37	11.8
410	37.00	37	12.1

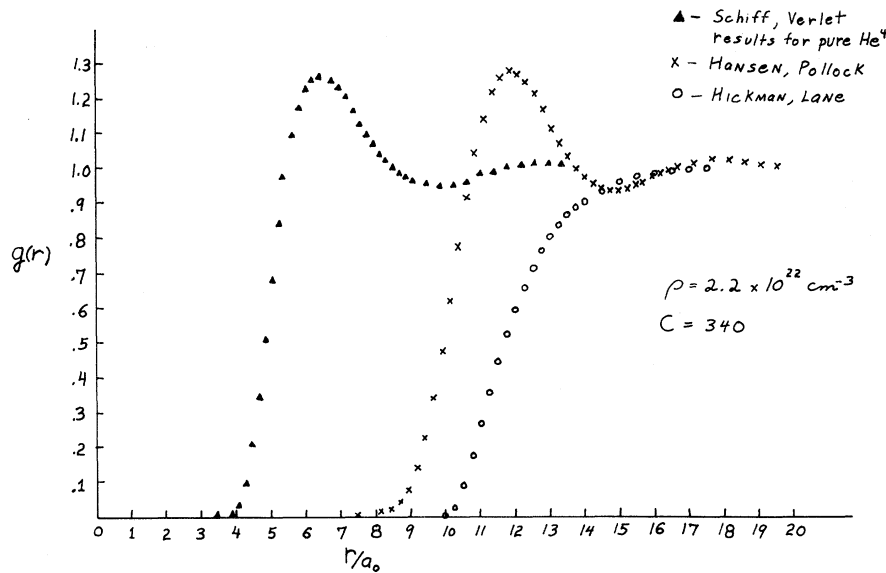


FIG. 1. Comparison of  $g_{12}(r)$  at  $\rho = 2.2 \times 10^{22}$  atoms  $\text{cm}^{-3}$  for the optimum  $C = 340$  with  $\rho(r)/\rho_0$  as given by Hickman and Lane (Ref. 5) and with  $g_{11}(r)$  for the pure liquid as calculated by Schiff and Verlet (Ref. 6).

$\rho(r)/\rho_0$  as given by Hickman and Lane.<sup>11</sup> Hickman and Lane predict a slightly larger bubble radius and we feel this may be due to their use of the strongly repulsive free-electron helium pseudopotential rather than the nonlocal pseudopotential.<sup>4</sup> Experimental verification for either  $g_{12}(r)$  from line-shift measurements would require a  $2^3P-1^1S$  interac-

tion potential valid at large  $r$ .

The authors are grateful to H. A. Bethe, D. M. Lee, N. F. Lane, and A. P. Hickman for useful discussions of this problem. We are indebted to W. P. Francis for providing his liquid helium results obtained with the PY2XS equation and to J. A. Krumhansl for a critical reading of the manuscript.

<sup>†</sup>Work supported by the U. S. Atomic Energy Commission under Contract No. AT (30-1)-3699, Technical Report No. NYO-3699-63.

\*Present address: Laboratoire de Physique Théorique et Hautes Energies, Université de Paris, Faculté des Sciences - Orsay, Batiment 211, 91 - Orsay, France.

<sup>1</sup>W. S. Dennis, E. Durbin, Jr., W. A. Fitzsimmons, O. Heybey, and G. K. Walters, Phys. Rev. Letters **23**, 1083 (1969).

<sup>2</sup>J. C. Hill, O. Heybey, and G. K. Walters, Phys. Rev. Letters **26**, 1213 (1971).

<sup>3</sup>C. M. Surko and F. Reif, Phys. Rev. **175**, 229 (1968).

<sup>4</sup>J. Jortner, N. R. Kestner, S. A. Rice, and M. H.

Cohen, J. Chem. Phys. **43**, 2614 (1965).

<sup>5</sup>A. P. Hickman and N. F. Lane, Phys. Rev. Letters **26**, 1216 (1971).

<sup>6</sup>D. Schiff and L. Verlet, Phys. Rev. **160**, 208 (1967).

<sup>7</sup>W. A. Fitzsimmons, N. F. Lane, and G. K. Walters, Phys. Rev. **174**, 193 (1968).

<sup>8</sup>J. P. Hansen and D. Schiff, Phys. Rev. Letters **23**, 1488 (1969).

<sup>9</sup>J. L. Lebowitz, Phys. Rev. **133**, A895 (1964).

<sup>10</sup>W. P. Francis, G. V. Chester, and L. Reatto, Phys. Rev. A **1**, 86 (1970).

<sup>11</sup>Hickman and Lane have informed us that the abscissa in Fig. 1 of their paper is  $R_0$  rather than  $R_b$  as shown.