

EXCITATION SPECTRUM OF TWO-COMPONENT LIQUIDS OR DENSE GASES CALCULATED BY DECOUPLING OF GREEN FUNCTIONS

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In the present work the method of retarded Green functions is used. The system of self-consistent equation for radial distribution functions and excitation spectrum of two-component mixture is found. The simple approximation we use is the one without damping of elementary excitations. Here for two-component liquid, the two branches of elementary excitation spectrum are obtained. For large wave vector k , they tend to the excitation energy of each compound itself. The energy spectrum is described by an expression slightly modified as compared to the expression of Parliński or earlier expression of Hubbard-Beeby or Prezhev. The properties of quantum and classical liquids are described qualitatively. For example the two branches of self energy excitation spectrum, for the mixture of Neon isotopes, are computed with the aid of an experimentally measured radial distribution function.

1. The basic problem of many body theory is to calculate properties of gases, liquids and solids from intermolecular potential energy. We shall consider the homogeneous and isotropic systems in thermal equilibrium. Spinless particles with pair intermolecular potential are assumed. Hence the many particles potential is neglected. If the phase transitions are not taken into account we may apply a method which is rather useful for one phase only. Here the system is described with the aid of the density operators in the formalism of retarded Green function. This method permits the spectrum of excitation energy and structure factors to be found. The method is applicable for dense gases. Application to one component classical quantum liquids gives satisfactory results as well. Our approximation consists in the decoupling of Green functions based on the comparison of first non zero moments of spectral densities.

The results for one component liquids were obtained by Parliński (1970 a). The correct self-energy excitation spectrum for argon and helium encourages one to apply a similar method to gaseous and liquid two-component mixtures. The generalization to more numerous component mixtures is quite simple.

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2. Let us consider a homogeneous system of N_1 particles, each of mass M_1 , and N_2 particles of mass M_2 in the volume V . Densities of two components are ρ_1, ρ_2 respectively, where $\rho = N/V$. Assume that the particle interaction potential can be described as a sum of pair potentials:

$\Phi_{11}(\mathbf{r})$ for particles of first kind

$\Phi_{22}(\mathbf{r})$ for particles of second kind

$\Phi_{12}(\mathbf{r})$ between particles of first and second kind.

The Hamiltonian of the system has the following form:

$$H = H_1 + H_2 + H_{12}$$

$$H_1 = \sum_{l|1}^{N_1} \mathbf{P}_{(1)l}^2 / 2M_1 + \frac{1}{2} \sum_l \sum_{m \neq l} \Phi_{11}(\mathbf{R}_{(1)l} - \mathbf{R}_{(1)m})$$

$$H_2 = \sum_{l|1}^{N_2} \mathbf{P}_{(2)l}^2 / 2M_2 + \frac{1}{2} \sum_l \sum_{m \neq l} \Phi_{22}(\mathbf{R}_{(2)l} - \mathbf{R}_{(2)m})$$

$$H_{12} = \sum_{l,m} \Phi_{12}(\mathbf{R}_{(1)l} - \mathbf{R}_{(2)m}) = \sum_{l,m} \Phi_{21}(\mathbf{R}_{(1)l} - \mathbf{R}_{(2)m}). \quad (1)$$

Assume also that pair potentials have Fourier transforms

$$\Phi(\mathbf{q}) = \frac{1}{V} \int_V d^3\mathbf{r} \Phi(\mathbf{r}) \exp(-i\mathbf{q}\mathbf{r}) \quad (2)$$

$$\Phi(\mathbf{r}) = \sum_{\mathbf{q}} \Phi(\mathbf{q}) \exp(i\mathbf{q}\mathbf{r}). \quad (3)$$

We may also write:

$$\sum_{\mathbf{q}} \exp(i\mathbf{q}\mathbf{r}) = V\delta(\mathbf{q}) \quad (4)$$

$$\int d^3\mathbf{r} \exp(i\mathbf{k}\mathbf{r}) = V\delta_{\mathbf{k},0}. \quad (5)$$

Our Hamiltonian does not contain external fields. Thus the coordinate system can be chosen in such a way that the centre-of-mass of the system is stationary *i. e.* $\sum \mathbf{P}_{(a)l} = 0$, (a) is 1 or 2 and shows the type of the l -th particle.

If we take into account the isotropy of system, we see that in the sense of the average over a cononical ensemble we have:

$$\langle \sum_l P_{(1)l} \rangle = \langle \sum_l P_{(2)l} \rangle = 0. \quad (6)$$

The operators \mathbf{R} and \mathbf{P} follow commutation relations

$$[R_{(a)l}^\alpha; P_{(b)m}^\beta] = i\delta_{lm} \delta^{\alpha\beta} \delta_{(a),(b)}; (a), (b) = 1, 2 \quad \alpha, \beta = x, y, z. \quad (7)$$

The chosen units system is this: $\hbar = k = 1$. Here k is Boltzmann's constant and h is Planck's constant ($\hbar = h/2\pi$). Introduce the collective coordinates

$$n_{(a)}(\mathbf{k}, t) = \sum_l \exp(i\mathbf{k}\mathbf{R}_{(a)l}(t)); (a) = 1, 2$$

which are Fourier transforms of particle density operators $\sum_l \delta(\mathbf{r} - \mathbf{R}_{(a)l}(t))$. The Hamiltonian may be written in collective coordinates.

$$H_{(a)} = \sum_l \mathbf{P}_{(a)l}^2 / 2M_a + \frac{1}{2} \sum_{\mathbf{q}} \Phi_{aa}(\mathbf{q}) \{n_a(\mathbf{q})n_a(-\mathbf{q}) - N_a\} \quad (a) = 1, 2$$

$$H_{12} = \frac{1}{2} \sum_{\mathbf{q}} \Phi_{12}(\mathbf{q}) \{n_1(\mathbf{q})n_2(-\mathbf{q}) + n_2(\mathbf{q})n_1(-\mathbf{q})\}. \quad (8)$$

3. Now we will specify the radial distribution functions. They are defined as:

$$g_{11}(\mathbf{r}) = VN_1^{-2} \langle \sum_l \sum_{m \neq l} \delta(\mathbf{r} - \mathbf{R}_{(1)l} + \mathbf{R}_{(1)m}) \rangle$$

$$g_{22}(\mathbf{r}) = VN_2^{-2} \langle \sum_l \sum_{m \neq l} \delta(\mathbf{r} - \mathbf{R}_{(2)l} + \mathbf{R}_{(2)m}) \rangle$$

$$g_{12}(\mathbf{r}) = g_{21}(\mathbf{r}) = V(N_1N_2)^{-1} \langle \sum_{l,m} \delta(\mathbf{r} - \mathbf{R}_{(1)l} + \mathbf{R}_{(2)m}) \rangle. \quad (9)$$

The value $\varrho_1^2 g_{11}(\mathbf{r} - \mathbf{r}') d^3\mathbf{r} d^3\mathbf{r}'$ is the probability that one particle of type 1 will be found at \mathbf{r} in $d^3\mathbf{r}$ and another of the same type in $d^3\mathbf{r}'$ at \mathbf{r}' . The value $\varrho_2^2 g_{22}(\mathbf{r} - \mathbf{r}') d^3\mathbf{r} d^3\mathbf{r}'$ has the same meaning but for particles of the second type. The value $\varrho_1 \varrho_2 g_{12}(\mathbf{r} - \mathbf{r}') d^3\mathbf{r} d^3\mathbf{r}'$ is the probability for one particle of type 1 and other particle of type 2.

Radial distribution functions fulfill relations:

for $\mathbf{r} \rightarrow 0$, values g_{11} , g_{22} and $g_{12} \rightarrow 0$
 this region is called the hard core radius
 for $\mathbf{r} \rightarrow \infty$, values g_{11} , g_{22} and $g_{12} \rightarrow 1$.

If there is no correlation in the mixture then $g_{11} = g_{22} = g_{12} = 1$. Let us perform the Fourier transform of radial distribution functions. Using the density-density correlation function:

$$\langle n_{(a)}(\mathbf{k}, t) n_{(b)}(-\mathbf{k}, 0) \rangle = \langle \sum_{l,m} \{\exp i\mathbf{k}\mathbf{R}_{(a)l}(t)\} \{\exp i\mathbf{k}\mathbf{R}_{(b)m}(0)\} \rangle \quad (10)$$

we have:

$$\langle n_1(\mathbf{q}) n_1(-\mathbf{q}) \rangle - N_1 = \varrho_1^2 V \int g_{11}(\mathbf{r}) \exp(-i\mathbf{q}\mathbf{r}) d^3\mathbf{r}; \quad n_1(\mathbf{q}) = n_1(\mathbf{q}, 0)$$

$$\langle n_2(\mathbf{q}) n_2(-\mathbf{q}) \rangle - N_2 = \varrho_2^2 V \int g_{22}(\mathbf{r}) \exp(-i\mathbf{q}\mathbf{r}) d^3\mathbf{r}$$

$$\langle n_1(\mathbf{q}) n_2(-\mathbf{q}) \rangle = \varrho_1 \varrho_2 V \int g_{12}(\mathbf{r}) \exp(-i\mathbf{q}\mathbf{r}) d^3\mathbf{r}. \quad (11)$$

In connection with relations (11) and (4) we may define the structure factors:

$$S_{11}(\mathbf{k}) = N_1^{-1} \langle n_1(\mathbf{k}) n_1(-\mathbf{k}) - N_1^2 \delta_{\mathbf{k},0} \rangle = 1 + \varrho_1 \int d^3\mathbf{r} \{g_{11}(\mathbf{r}) - 1\} \exp(-i\mathbf{k}\mathbf{r})$$

$$S_{22}(\mathbf{k}) = N_2^{-1} \langle n_2(\mathbf{k}) n_2(-\mathbf{k}) - N_2^2 \delta_{\mathbf{k},0} \rangle = 1 + \varrho_2 \int d^3\mathbf{r} \{g_{22}(\mathbf{r}) - 1\} \exp(-i\mathbf{k}\mathbf{r})$$

$$S_{12}(\mathbf{k}) = S_{21}(\mathbf{k}) = \{N_1 N_2\}^{-1/2} \langle n_1(\mathbf{k}) n_2(-\mathbf{k}) - N_1 N_2 \delta_{\mathbf{k},0} \rangle$$

$$= \varrho_1 \varrho_2 \int d^3\mathbf{r} \{g_{12}(\mathbf{r}) - 1\} \exp(-i\mathbf{k}\mathbf{r}). \quad (12)$$

Of course if there is no correlation, for example in dilute gases then $S_{11} = S_{22} = 1$ and $S_{12} = 0$.

4. Using the method of retarded Green functions:

$$G_{ab}(\mathbf{k}, t) = \langle\langle n_{(a)}(\mathbf{k}, t) | n_{(b)}(-\mathbf{k}) \rangle\rangle = -i\Theta(t) \langle n_{(a)}(\mathbf{k}, t); n_{(b)}(-\mathbf{k}) \rangle \quad (13)$$

the equation of motion for G_{ab} can be obtained by taking the second time-derivatives. In order to write clearly this equation we must specify the following functions:

$$T_{(a)}(\mathbf{k}, t) = \{2M_a\}^{-2} \sum_l \{ \mathbf{k}P_{(a)l}(t); \{ \mathbf{k}P_{(a)l}(t); \exp ik\mathbf{R}_{(a)l}(t) \} \}_+ \quad (14)$$

where $\{ \}_+$ denotes an anticommutator

$$\begin{aligned} V_{11}(\mathbf{k}, t) &= -(2M_1)^{-1} \sum_{\mathbf{q}} \Phi_{11}(\mathbf{q}) \mathbf{k} \mathbf{q} \{ n_1(\mathbf{k} + \mathbf{q}, t) n_1(-\mathbf{q}, t) - n_1(\mathbf{q}, t) n_1(\mathbf{k} - \mathbf{q}, t) \} \\ V_{12}(\mathbf{k}, t) &= -(2M_1)^{-1} \sum_{\mathbf{q}} \Phi_{12}(\mathbf{q}) \mathbf{k} \mathbf{q} \{ n_1(\mathbf{k} + \mathbf{q}, t) n_2(-\mathbf{q}, t) - n_2(\mathbf{q}, t) n_1(\mathbf{k} - \mathbf{q}, t) \}. \end{aligned} \quad (15)$$

We must also specify functions V_{22} and V_{21} . They may be written by replacement in (15) of 1 by 2 and of 2 by 1. Now we have:

$$\begin{aligned} \left(i \frac{d}{dt} \right)^2 G_{11}(\mathbf{k}, t) &= \delta(t) \mathbf{k}^2 N_1 / M_1 + \langle\langle T_1(\mathbf{k}, t) + V_{11}(\mathbf{k}, t) + V_{12}(\mathbf{k}, t) | n_1(-\mathbf{k}) \rangle\rangle \\ \left(i \frac{d}{dt} \right)^2 G_{12}(\mathbf{k}, t) &= \langle\langle T_1(\mathbf{k}, t) + V_{11}(\mathbf{k}, t) + V_{12}(\mathbf{k}, t) | n_2(-\mathbf{k}) \rangle\rangle \text{ etc.} \end{aligned} \quad (16)$$

In order to solve equations (16) we demand the following proportionality:

$$\begin{aligned} & \left[\langle\langle T_1 + V_{11} + V_{12} | n_1(-\mathbf{k}) \rangle\rangle; \langle\langle T_1 + V_{11} + V_{12} | n_2(-\mathbf{k}) \rangle\rangle \right] \\ & \left[\langle\langle T_2 + V_{22} + V_{21} | n_1(-\mathbf{k}) \rangle\rangle; \langle\langle T_2 + V_{22} + V_{21} | n_2(-\mathbf{k}) \rangle\rangle \right] \\ & \approx \begin{bmatrix} \Omega_{11}^2(\mathbf{k}); \Omega_{21}^2(\mathbf{k}) \\ \Omega_{12}^2(\mathbf{k}); \Omega_{22}^2(\mathbf{k}) \end{bmatrix} \times \begin{bmatrix} G_{11}; G_{12} \\ G_{21}; G_{22} \end{bmatrix}. \end{aligned} \quad (17)$$

The proportionality coefficients, functions Ω are found by the comparison of first non zero moments at both sides of equation (17). The n -th moment μ_n of operator O_S (in the Schrödinger picture) is defined as:

$$\langle \exp(iHt) O_S \exp(-iHt) O_S \rangle = \sum_{n|0}^{\infty} \frac{(it)^n}{n!} [H_i \dots [H_i O_S] \dots] O_S = \sum_{n|0}^{\infty} \frac{(it)^n}{n!} \mu_n. \quad (18)$$

For example the first moment of G_{11} is $-\mathbf{k}^2 N_1 / M_1$, and of G_{12} is 0. With the aid of relations (6) we obtain:

$$\begin{aligned} \Omega_{11}^2(\mathbf{k}) &= \{2\mathbf{k}^{-2} M_1\}^{-2} + \left\langle \frac{3}{N_1} \sum_l \left(\frac{\mathbf{k} P_{(1)l}}{M_1} \right)^2 \right\rangle - \frac{1}{2\mathbf{k}^2 N_1 M_1} \left\{ \sum_{\mathbf{q}} \Phi_{11}(\mathbf{q}) (\mathbf{k} \mathbf{q})^2 \times \right. \\ & \quad \times \langle 2n_1(\mathbf{q}) n_1(-\mathbf{q}) - n_1(\mathbf{k} + \mathbf{q}) n_1(-\mathbf{k} - \mathbf{q}) - n_1(\mathbf{k} - \mathbf{q}) n_1(-\mathbf{k} + \mathbf{q}) \rangle + \\ & \quad \left. + \sum_{\mathbf{q}} \Phi_{12}(\mathbf{q}) (\mathbf{k} \mathbf{q})^2 \langle n_1(\mathbf{q}) n_2(-\mathbf{q}) + n_2(\mathbf{q}) n_1(-\mathbf{q}) \rangle \right\} \\ \Omega_{12}^2(\mathbf{k}) &= \frac{1}{2\mathbf{k}^2 N_1 M_2} \sum_{\mathbf{q}} \Phi_{12}(\mathbf{q}) (\mathbf{k} \mathbf{q})^2 \langle n_1(\mathbf{q} - \mathbf{k}) n_2(\mathbf{k} - \mathbf{q}) + n_2(\mathbf{k} + \mathbf{q}) n_1(-\mathbf{k} - \mathbf{q}) \rangle \quad (19) \end{aligned}$$

etc.

We need the other approximation to calculate $\left\langle \frac{3}{N_a} \sum_l \left(\frac{\mathbf{kP}_{(a)l}}{M} \right)^2 \right\rangle$. It follows readily that:

$$\frac{3}{N_1} \left\langle \sum_l \left(\frac{\mathbf{kP}_{(1)l}}{M_1} \right)^2 \right\rangle = \frac{d^2}{dt dt'} \left\langle \sum_l \{ \mathbf{kR}_{(1)l}(t) \} \{ \mathbf{kR}_{(1)l}(t') \} \right\rangle_{t=t'} \quad (20)$$

Using the Fourier transform in ω -space and the spectral theorem we obtain:

$$\frac{3}{N_1} \left\langle \sum_l \left(\frac{\mathbf{kP}_{(1)l}}{M_1} \right)^2 \right\rangle = \frac{-3}{N_1 \pi} \int_{-\infty}^{\infty} \frac{\omega^2 \operatorname{Im} d_{11}(\omega + i\vartheta)}{\exp(\omega/\vartheta) - 1} d\omega \quad (21)$$

where ϑ is the temperature and

$$d_{11}(t) = \sum_l \langle \langle \mathbf{kR}_{(1)l}(t) | \mathbf{kR}_{(1)l}(0) \rangle \rangle; \quad d_{11}(\omega) = \int d_{11}(t) \exp(-it\omega) dt. \quad (22)$$

We shall calculate "d" using the same approximation as Parliński (1970a):

$$\left(i \frac{d}{dt} \right)^2 d_{11}(\mathbf{k}, t) \approx \delta(t) \mathbf{k}^2 N_1 / M_1 + \alpha_{(1)}^2(\mathbf{k}) d_{11}(\mathbf{k}, t); \quad d_{11}(\mathbf{k}, t) = d_{11}(t)$$

$$\left(i \frac{d}{dt} \right)^2 d_{22}(\mathbf{k}, t) \approx \delta(t) \mathbf{k}^2 N_2 / M_2 + \alpha_{(1)}^2(\mathbf{k}) d_{22}(\mathbf{k}, t)$$

where

$$d_{22}(t) = d_{22}(\mathbf{k}, t) = \sum_l \langle \langle \mathbf{kR}_{(2)l}(t) | \mathbf{kR}_{(2)l}(0) \rangle \rangle. \quad (23)$$

The value α is obtained by the following comparison of the first moments:

$$\sum_l \alpha_{(1)}^2(\mathbf{k}) \langle [H, \mathbf{kR}_{(1)l}] | \mathbf{kR}_{(1)l} \rangle = \sum_l \langle [H; [[\mathbf{kR}_{(1)l}; H]; H]] | \mathbf{kR}_{(1)l} \rangle. \quad (24)$$

Converting (24) into \mathbf{r} -space:

$$\alpha_{(1)}^2(\mathbf{k}) = (M_1 \mathbf{k}^2)^{-1} \{ \varrho_1 \int g_{11}(\mathbf{r}) (\mathbf{k} \nabla_{\mathbf{r}})^2 \Phi_{11}(\mathbf{r}) d^3 \mathbf{r} + \varrho_2 \int g_{12}(\mathbf{r}) (\mathbf{k} \nabla_{\mathbf{r}})^2 \Phi(\mathbf{r}) d^3 \mathbf{r} \}$$

$$\alpha_{(2)}^2(\mathbf{k}) = (M_2 \mathbf{k}^2)^{-1} \{ \varrho_2 \int g_{22}(\mathbf{r}) (\mathbf{k} \nabla_{\mathbf{r}})^2 \Phi_{22}(\mathbf{r}) d^3 \mathbf{r} + \varrho_1 \int g_{12}(\mathbf{r}) (\mathbf{k} \nabla_{\mathbf{r}})^2 \Phi_{12}(\mathbf{r}) d^3 \mathbf{r} \}. \quad (25)$$

Green functions "d" are:

$$d_{aa}(\mathbf{k}, \omega) = \mathbf{k}^2 N_a / \{ M_a \times (\omega^2 - \alpha_{(a)}^2(\mathbf{k})) \} \quad a = 1, 2. \quad (26)$$

The expressions, we have looked for, are:

$$\frac{3}{N_a} \left\langle \sum_l \left(\frac{\mathbf{kP}_{(a)l}}{M_a} \right)^2 \right\rangle = \frac{\mathbf{k}^2}{2M_a} 3\alpha_{(a)}(\mathbf{k}) \operatorname{cth} \left\{ \frac{\alpha_{(a)}(\mathbf{k})}{2\vartheta} \right\} = \frac{\mathbf{k}^2}{2M_a} 6K_a. \quad (27)$$

It is easily seen that for $\vartheta \rightarrow \infty$ we get: $K_1 = K_2 = \vartheta$. For $\vartheta \rightarrow 0$, $K_1 = 0.5 \times \alpha_{(1)}(\mathbf{k})$ and $K_2 = 0.5 \times \alpha_{(2)}(\mathbf{k})$. To write clearly our expressions we ought to convert (19) and (20) in \mathbf{r} -space. Knowing that:

$$-V \times (\mathbf{k}\mathbf{q})^2 \Phi(\mathbf{q}) = \int d^3r \exp(-i\mathbf{q}\mathbf{r}) (\mathbf{k}\nabla_r)^2 \Phi(\mathbf{r}) \quad (28)$$

and using (2), (4), (11) we obtain our expressions in shorter form:

$$\begin{aligned} \Omega_{11}^2(\mathbf{k}) &= \frac{\mathbf{k}^4}{4M_1^2} + \frac{\mathbf{k}^2}{2M_1} 6K_1 + \frac{\varrho_1}{\mathbf{k}^2 M_1} \int g_{11}(\mathbf{r}) \{1 - \cos \mathbf{k}\mathbf{r}\} [\mathbf{k}\nabla_r]^2 \Phi_{11}(\mathbf{r}) d^3r + \\ &\quad + \frac{\varrho_2}{\mathbf{k}^2 M_1} \int g_{12}(\mathbf{r}) (\mathbf{k}\nabla_r)^2 \Phi_{12}(\mathbf{r}) d^3r \\ \Omega_{12}^2(\mathbf{k}) &= \frac{-\varrho_2}{\mathbf{k}^2 M_2} \int g_{12}(\mathbf{r}) \cos(\mathbf{k}\mathbf{r}) (\mathbf{k}\nabla_r)^2 \Phi_{12}(\mathbf{r}) d^3r = \frac{\varrho_2 M_1}{\varrho_1 M_2} \times \Omega_{21}^2(\mathbf{k}) \\ \Omega_{22}^2(\mathbf{k}) &= \frac{\mathbf{k}^4}{4M_2^2} + \frac{\mathbf{k}^2}{2M_2} 6K_2 + \frac{\varrho_2}{\mathbf{k}^2 M_2} \int g_{22}(\mathbf{r}) \{1 - \cos \mathbf{k}\mathbf{r}\} (\mathbf{k}\nabla_r)^2 \Phi_{22}(\mathbf{r}) d^3r + \\ &\quad + \frac{\varrho_1}{\mathbf{k}^2 M_2} \int g_{12}(\mathbf{r}) (\mathbf{k}\nabla_r)^2 \Phi_{12}(\mathbf{r}) d^3r. \end{aligned} \quad (29)$$

If we Fourier transform G_{ab} into ω -space then $\left(i \frac{d}{dt}\right)^2 \rightarrow \omega^2$ and (16) may be written in following form:

$$\begin{bmatrix} \omega^2 - \Omega_{11}^2(\mathbf{k}); & -\Omega_{21}^2(\mathbf{k}) \\ -\Omega_{12}^2(\mathbf{k}); & \omega^2 - \Omega_{22}^2(\mathbf{k}) \end{bmatrix} \times \begin{bmatrix} G_{11}(\mathbf{k}, \omega); & G_{12}(\mathbf{k}, \omega) \\ G_{21}(\mathbf{k}, \omega); & G_{22}(\mathbf{k}, \omega) \end{bmatrix} \begin{bmatrix} \frac{\mathbf{k}^2 N_1}{M_1}; & 0 \\ 0; & \frac{\mathbf{k}^2 N_2}{M_2} \end{bmatrix}. \quad (30)$$

The self-energy of the system can be obtained by solving the equation:

$$\det \left\{ \begin{bmatrix} \omega^2 - \Omega_{11}^2; & -\Omega_{21}^2 \\ -\Omega_{12}^2; & \omega^2 - \Omega_{22}^2 \end{bmatrix} - \lambda \begin{bmatrix} 1; & 0 \\ 0; & 1 \end{bmatrix} \right\} = 0$$

and from the condition $\lambda_1 \times \lambda_2 = 0$.

Finally we have:

$$\omega^2(\mathbf{k}) = \frac{1}{2} \{ \Omega_{11}^2(\mathbf{k}) + \Omega_{22}^2(\mathbf{k}) \mp \sqrt{ \{ \Omega_{11}^2(\mathbf{k}) - \Omega_{22}^2(\mathbf{k}) \}^2 + 4 \Omega_{12}^2(\mathbf{k}) \Omega_{21}^2(\mathbf{k}) } \}. \quad (31)$$

We see that here two solutions exist — we call them ω^+ and ω^- in correspondence to the choice of the sign in the expression (31). Conventionally we will call one branch of excitation spectrum “acoustic” and other “optical”. From the analysis of (31) we can see that for $\mathbf{k} \rightarrow \infty$ the potential part of (31) is negligible, only the kinetic part is large. Both branches at large \mathbf{k} are almost equal Ω_{11}^2 or Ω_{22}^2 . For illustration (31) a computation is given for the mixture of 9% ^{22}Ne and 91% ^{20}Ne (see Figs 1 and 2). This composition of Neon isotopes is almost like the composition of natural existing Neon. The only difference is that: in natural Neon 0.257% admixture of the ^{21}Ne isotope is present.

Experimental data for computations are taken from the article of Jong (1967 a). The potential is (6,12):

$$\Phi_{11} = \Phi_{22} = \Phi_{12} = 4\varepsilon \times (x^{12} - x^6) \text{ here } x = \sigma/r$$

$\varepsilon = 35.8^\circ\text{K}$, $\sigma = 2.75 \text{ \AA}$ (Rowlison's data).

The radial distribution function is $g(\mathbf{r}) = \{V/(N_1 + N_2)^2\} \langle \sum_{l,m,a,b} \delta(\mathbf{r} - \mathbf{R}_{(a)l} + \mathbf{R}_{(b)m}) \rangle$. It is certainly a rather good assumption that $g = g_{11} = g_{22} = g_{12}$. The proof is that the sole not very large difference between the atoms of ^{22}Ne and ^{20}Ne , is their mass. Admixture of

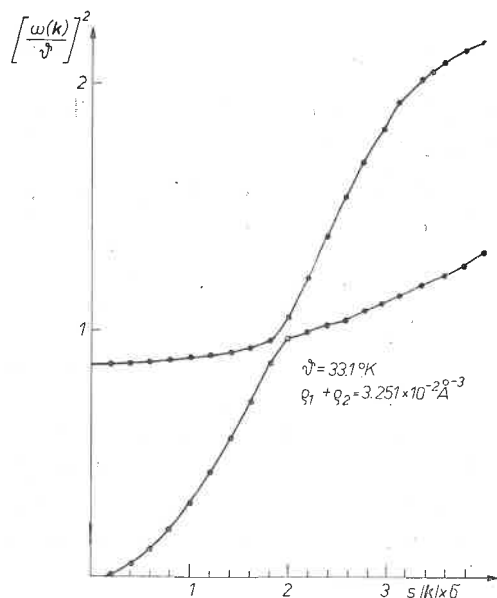


Fig. 1

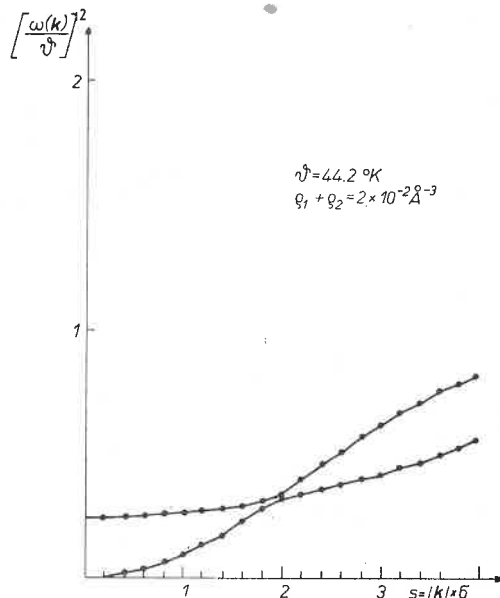


Fig. 2

Fig. 1 and Fig. 2. Self-energy excitation spectrum of the mixture of 9% ^{22}Ne and 91% ^{20}Ne

0.257% of ^{21}Ne seems not to be important. We will use, experimentally measured $g(\mathbf{r})$ for our computations (Stripe and Tomson's data). For each \mathbf{k} there exist two vibrations, one acoustic and other "optical" in disagreement with the results of Prezzhev (1970 a) who used the quasi-crystalline approximation. In the case of our results the energy of the acoustic branch is zero for $\mathbf{k} = 0$. Next with \mathbf{k} the energy increases. Very small oscillations appear on both curves. Their period is smaller than the value $|\mathbf{k}| = 2\pi/\sigma$.

5. Equation (30) helps us to calculate the structure factors. The first step is to obtain the G_{ab} functions.

$$\begin{aligned} G_{11}(\mathbf{k}, \omega) &= \mathbf{k}^2 N_1 \{\omega^2 - \Omega_{22}^2(\mathbf{k})\} \times \{M_1 \times D(\mathbf{k})\}^{-1} \\ G_{22}(\mathbf{k}, \omega) &= \mathbf{k}^2 N_2 \{\omega^2 - \Omega_{11}^2(\mathbf{k})\} \times \{M_2 \times D(\mathbf{k})\}^{-1} \\ G_{12}(\mathbf{k}, \omega) &= G_{21}(\mathbf{k}, \omega) = \mathbf{k}^2 N_1 \Omega_{12}^2(\mathbf{k}) \times \{M_1 \times D(\mathbf{k})\}^{-1} \end{aligned} \quad (33)$$

where

$$D(\mathbf{k}) = \{\omega^2 - \Omega_{11}^2(\mathbf{k})\}\{\omega^2 - \Omega_{22}^2(\mathbf{k})\} - \Omega_{12}^2(\mathbf{k})\Omega_{21}^2(\mathbf{k}).$$

The spectral theorem gives the result we need. For example:

$$N_1 S_{11}(\mathbf{k}) = -\frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\text{Im } G_{11}(\mathbf{k}, \omega + i\delta)}{\exp(\omega/\vartheta) - 1} d\omega; \quad \delta \rightarrow 0^+. \quad (34)$$

Finally:

$$S_{11}(\mathbf{k}) = \frac{\Omega_{12}^2 \Omega_{21}^2 A(\mathbf{k})}{M_1} \left\{ \frac{\text{cth}(\omega_+/2\vartheta)}{\omega_+(\omega_+^2 - \Omega_{11}^2)} + \frac{\text{cth}(\omega_-/2\vartheta)}{\omega_-(\omega_-^2 - \Omega_{11}^2)} \right\} \quad (35)$$

$$S_{22}(\mathbf{k}) = \frac{\Omega_{12}^2 \Omega_{21}^2 A(\mathbf{k})}{M_2} \left\{ \frac{\text{cth}(\omega_+/2\vartheta)}{\omega_+(\omega_+^2 - \Omega_{22}^2)} + \frac{\text{cth}(\omega_-/2\vartheta)}{\omega_-(\omega_-^2 - \Omega_{22}^2)} \right\} \quad (36)$$

$$S_{12}(\mathbf{k}) = S_{21}(\mathbf{k}) = \frac{2|\Omega_{12}\Omega_{21}|}{\sqrt{M_1 M_2}} A(\mathbf{k}) \left\{ \frac{\text{cth}(\omega_+/2\vartheta)}{\omega_+} + \frac{\text{cth}(\omega_-/2\vartheta)}{\omega_-} \right\} \quad (37)$$

where

$$A(\mathbf{k}) = 0.5 \times \mathbf{k}^2 / \{(\Omega_{11}^2 - \Omega_{22}^2)^2 + 4\Omega_{12}^2 \Omega_{21}^2\}^{1/2}.$$

It is easy to see from (35)–(37) that if $\mathbf{k} \rightarrow \infty$ and $\omega_+ \rightarrow \Omega_{11}$ then only the first term in (35) is large. Function “cth” tends to 1, and having ω_- from (31) we can prove that $S_{11} \rightarrow 1$. The same follows in the case of S_{22} . However S_{12} tends to zero if $\mathbf{k} \rightarrow \infty$. A remark should be made that there is no agreement between $S(\mathbf{k})$ in (34)–(37) and experiment (but only for \mathbf{k} not too large). For large \mathbf{k} there is good agreement. The computations for one component liquids by Parliński (1970 a) showed that the computed value of $S(\mathbf{k})$ is smaller than the experiment.

The reason is the used approximation of the decoupling of Green functions. Taking into account the life time of excitation one may improve the theoretical results.

6. It is possible to get the equation of state.

From the condition:

$$\left\langle i \frac{d}{dt} \right\rangle \left\langle \sum_{l,a} \mathbf{P}_{(a)l} \mathbf{R}_{(a)l} \right\rangle = 0; \quad a = 1, 2$$

we have:

$$\left\langle \sum_{l,a} [\mathbf{P}_{(a)l}; H] \times \mathbf{R}_{(a)l} \right\rangle + \left\langle i \sum_{l,a} \mathbf{F}_{(a)l} \mathbf{R}_{(a)l} \right\rangle + \left\langle \sum_{l,a} \frac{\mathbf{P}_{(a)l}^2}{M_a} \right\rangle = 0 \quad (38)$$

over surface

the second term is equal $-3ipV$, where “ p ” is the pressure. Using the commutation relation we obtain:

$$3pV = \sum_{a=1,2} \left\{ \left\langle 2 \sum_l \mathbf{P}_{(a)l}^2 / 2M_a \right\rangle - \frac{1}{2} \sum_{b=1,2} \sum_{\mathbf{q}} \Phi_{ab}(\mathbf{q}) \mathbf{q} \frac{\partial}{\partial \mathbf{q}} \langle n_a(\mathbf{q}) n_b(-\mathbf{q}) \rangle \right\}. \quad (39)$$

This is the equation of state. Converting into \mathbf{r} -space we obtain a neater expression:

$$p = \varrho_1 K_1 + \varrho_2 K_2 - \frac{1}{6} \{ \varrho_1^2 \int d^3\mathbf{r} g_{11}(\mathbf{r})(\mathbf{r}\nabla_{\mathbf{r}})\Phi_{11}(\mathbf{r}) + \varrho_2^2 \int d^3\mathbf{r} g_{22}(\mathbf{r})(\mathbf{r}\nabla_{\mathbf{r}})\Phi_{22}(\mathbf{r}) + 2\varrho_1\varrho_2 \int d^3\mathbf{r} g_{12}(\mathbf{r})(\mathbf{r}\nabla_{\mathbf{r}})\Phi_{12}(\mathbf{r}) \}. \quad (40)$$

7. *Conclusion.* The method of approximation — the decoupling of Green functions leads to a self-consistent equations for structure factors and self-energy excitation spectrum. Two branches of excitation spectrum are obtained. They suggest that for each value of \mathbf{k} there are two vibrations in the mixture. The situation is almost identical to that in crystals which contain two atoms in the elementary cell, of course without taking into account the transversal phonons in crystals. In the case of the mixture for $\mathbf{k} \rightarrow \infty$ potential effects are small. Expressions for structure factors approach the correct value for $\mathbf{k} \rightarrow \infty$ but are doubtful for not too large \mathbf{k} . This is the reason why no large profit from the self-consistent equations systems may be obtained. One must hence work with experimental data.

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