HIERARCHIC THEORY OF CONDENSED MATTER AND ITS INTERACTION WITH LIGHT:

New Theories of Light Refraction, Brillouin Scattering and Mössbauer effect

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Materials, presented in this article are based on new quantum concept, described in:

[1]. **Book by A. Kaivarainen: Hierarchic Concept of Matter and Field. Water, biosystems and elementary particles. New York, 1995 and two of my papers:**

[2]. **"New Hierarchic Theory of Matter General for Liquids and Solids: dynamics, thermodynamics and mesoscopic structure of water and ice"** (**see URL: http://kftt.karelia.ru/~alexk**) **and:**

[3]. **Hierarchic Concept of Matter, General for Liquids and Solids: Water and ice** (**see Proceedings of the Second Annual Advanced Water Sciences Symposium, October 4-6, 1966, Dallas, Texas.**

Computerized verification of three new described here theories are presented on examples of WATER and ICE, using special computer program (**copyright, 1997, A. Kaivarainen**).

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Summary to Part I of book:

Hierarchic Theory of Matter and Field (by: A. Kaivarainen)

A basically new hierarchic quantitative theory, general for solids and liquids, has been developed.

It is assumed, that anharmonic oscillations of particles in any condensed matter lead to emergence of three-dimensional (3D) superposition of standing de Broglie waves of molecules, electromagnetic and acoustic waves. Consequently, any condensed matter could be considered as a gas of 3D standing waves of corresponding nature. Our approach unifies and develops strongly the Einstein's and Debye's models.

Collective excitations, like 3D standing de Broglie waves of molecules, representing at certain conditions the molecular Bose condensate, were analyzed, as a background of hierarchic model of condensed matter.

The most probable de Broglie wave (wave B) length is determined by the ratio of Plank constant to the most probable impulse of molecules, or by ratio of its most probable phase velocity to frequency. The waves B are related to molecular translations (tr) and librations (lb).

As the quantum dynamics of condensed matter does not follow in general case the classical Maxwell-Boltzmann distribution, the real most probable de Broglie wave length can exceed the classical thermal de Broglie wave length and the distance between centers of molecules many times.

This makes possible the atomic and molecular Bose condensation in solids and liquids at temperatures, below boiling point. It is one of the most important results of new theory, which we have confirmed by computer simulations on examples of water and ice.

Four strongly interrelated new types of quasiparticles (collective excitations) were introduced in our hierarchic model:

1. *Effectons* (*tr and lb*), existing in "acoustic" (a) and "optic" (b) states represent the coherent clusters in general case;
2. *Convertons*, corresponding to interconversions between *tr* and *lb* types of the effectons (flickering clusters);
3. *Transitons* are the intermediate $[a \rightleftharpoons b]$ transition states of the *tr* and *lb* effectons;
4. *Deformons* are the 3D superposition of IR electromagnetic or acoustic waves, activated by *transitons* and *convertons*.

Primary effectons (*tr and lb*) are formed by 3D superposition of the **most probable standing de Broglie waves** of the oscillating ions, atoms or molecules. The volume of effectons (tr and lb) may contain from less than one, to tens and even thousands of molecules. The first condition means validity of **classical** approximation in description of the subsystems of the effectons. The second one points to **quantum properties of coherent clusters due to molecular Bose condensation**.

The liquids are semiclassical systems because their primary (tr) effectons contain less than one molecule and primary (lb) effectons - more than one molecule. *The solids are quantum systems totally because both kind of their primary effectons* (*tr and lb*) *are molecular Bose condensates*. **These consequences of our theory are confirmed by computer calculations.**

The 1st order $[gas \rightarrow liquid]$ transition is accompanied by strong decreasing of rotational (librational) degrees of freedom due to emergence of primary (lb) effectons and $[liquid \rightarrow solid]$ transition - by decreasing of translational degrees of freedom due to Bose-condensation of primary (tr) effectons.

In the general case the effecton can be approximated by parallelepiped with edges corresponding to de Broglie waves length in three selected directions (1, 2, 3), related to the

symmetry of the molecular dynamics. In the case of isotropic molecular motion the effectons' shape may be approximated by cube.

The edge-length of primary effectons (tr and lb) can be considered as the "parameter of order".

The in-phase oscillations of molecules in the effectons correspond to the effecton's (a) - *acoustic* state and the counterphase oscillations correspond to their (b) - *optic* state. States (a) and (b) of the effectons differ in potential energy only, however, their kinetic energies, impulses and spatial dimensions - are the same. The *b*-state of the effectons has a common feature with **Frölich's polar mode**.

The $(a \to b)$ **or** $(b \to a)$ **transition states of the primary effectons (tr and lb), defined as primary transitons, are accompanied by a change in molecule polarizability and dipole moment without density fluctuations. At this case they lead to absorption or radiation of IR photons, respectively.**

Superposition (interception) of three internal standing IR photons of different directions (1,2,3) - forms primary electromagnetic deformons (tr and lb).

On the other hand, the [lb$\rightleftharpoons$ tr] *convertons* and *secondary transitons* are accompanied by the density fluctuations, leading to *absorption or radiation of phonons*.

Superposition resulting from interception of standing phonons in three directions (1,2,3), forms **secondary acoustic deformons (tr and lb)**.

Correlated collective excitations of primary and secondary effectons and deformons (tr and lb), localized in the volume of primary *tr* and *lb electromagnetic* deformons, lead to origination of **macroeffectons**, **macrotransitons** and **macrodeformons** (tr and lb respectively).

Correlated simultaneous excitations of tr and lb macroeffectons in the volume of superimposed *tr* and *lb* electromagnetic deformons lead to origination of **supereffectons**.

In turn, the coherent excitation of *both*: *tr* and *lb macrodeformons and macroconvertons* in the same volume means creation of **superdeformons**. Superdeformons are the biggest (cavitational) fluctuations, leading to microbubbles in liquids and to local defects in solids.

Total number of quasiparticles of condensed matter equal to 4!=24, reflects all of possible combinations of the four basic ones [1-4], introduced above. This set of collective excitations in the form of "gas" of 3D standing waves of three types: de Broglie, acoustic and electromagnetic - is shown to be able to explain virtually all the properties of all condensed matter.

The important positive feature of our hierarchic model of matter is that it does not need the semi-empiric intermolecular potentials for calculations, which are unavoidable in existing theories of many body systems. The potential energy of intermolecular interaction is involved indirectly in dimensions and stability of quasiparticles, introduced in our model.

The main formulae of theory are the same for liquids and solids and include following experimental parameters, which take into account their different properties:

[1]- **Positions of (tr) and (lb) bands in oscillatory spectra**;

[2]- **Sound velocity**;

[3]- **Density**;

[4]- **Refraction index (extrapolated to the infinitive wave length of photon)**.

The knowledge of these four basic parameters at the same temperature and pressure makes it possible using our computer program, to evaluate more than 150 *important characteristics of any condensed matter. Among them are such as: total internal energy, kinetic and potential energies, heat-capacity and thermal conductivity, surface tension, vapor pressure, viscosity,* *coefficient of self-diffusion, osmotic pressure, solvent activity, etc. Most of calculated parameters are hidden, i.e. inaccessible to direct experimental measurement.*

The new interpretation and evaluation of Brillouin light scattering and Mössbauer effect parameters may also be done on the basis of hierarchic theory. Mesoscopic scenarios of turbulence, superconductivity and superfluity are elaborated.

Some original aspects of water in organization and large-scale dynamics of biosystems - such as proteins, DNA, microtubules, membranes and regulative role of water in cytoplasm, cancer development, quantum neurodynamics, etc. have been analyzed in the framework of Hierarchic theory.

Computerized verification of our Hierarchic concept of matter on examples of water and ice is performed, using special computer program: Comprehensive Analyzer of Matter Properties (CAMP, copyright, 1997, Kaivarainen). The new opto-acoustical device (CAMP), based on this program, with possibilities much wider, than that of IR, Raman and Brillouin spectrometers, has been proposed (see URL: http://www.karelia.ru/~alexk).

This is the first theory able to predict all known experimental temperature anomalies for water and ice. The conformity between theory and experiment is very good even without any adjustable parameters.

The hierarchic concept creates a bridge between micro- and macro- phenomena, dynamics and thermodynamics, liquids and solids in terms of quantum physics.

1: New approach to theory of light refraction

1.1. Refraction in gas

If the action of photons onto electrons of molecules is considered as a force, activating a harmonic oscillator with decay, it leads to the known classical equations for a complex refraction index (Vuks, 1984).

The Lorentz-Lorenz formula obtained in such a way is convenient for practical needs. However, it does not describe the dependence of refraction index on the incident light frequency and did not take into account the intermolecular interactions. In the new theory proposed below we have tried to clear up the relationship between these parameters.

Our basic idea is that the dielectric penetrability of matter ϵ, (equal in the optical interval of frequencies to the refraction index squared n^2), is determined by the ratio of partial volume energies of photon in vacuum to similar volume energy of photon in matter:

$$\epsilon = n^2 = \frac{[\mathrm{E_p^0}]}{[\mathrm{E_p^m}]} = \frac{\mathrm{m_p c^2}}{\mathrm{m_p c_m^2}} = \frac{\mathrm{c^2}}{\mathrm{c_m^2}} \qquad 1.1$$

where $m_p = h\nu_p/c^2$ is the effective photon mass, c is the light velocity in vacuum, c_m is the effective light velocity in matter.

We introduce the notion of partial volume energy of a photon in vacuum $[E_p^0]$ and in matter $[E_p^m]$ as a product of photon energy ($E_p = h\nu_p$) and the volume (V_p) occupied by 3D standing wave of photon in vacuum and in matter, correspondingly:

$$[E_p^0] = E_p V_p^0 \qquad [E_p^m] = E_p V_p^m \qquad 1.2$$

The 3D standing photon volume as an interception volume of 3 different standing photons normal to each other was termed in our mesoscopic model as a primary electromagnetic deformon (see Introduction of [1,2,3]).

In vacuum, where the effect of an **excluded volume due to the spatial incompatibility of electron shells of molecules and photon** is absent, the volume of $3D$ photon standing wave (primary deformon) is:

$$V_p^0 = \frac{1}{\mathrm{n_p}} = \frac{3\lambda_\mathrm{p}^2}{8\pi} \qquad 1.3$$

We will consider the interaction of light with matter in this mesoscopic volume, containing a thousands of molecules of condensed matter. It is the reason why we titled this theory of light refraction as mesoscopic one.

Putting (1.3) into (1.2), we obtain the formula for the partial volume energy of a photon in vacuum:

$$[E_p^0] = E_p V_p^0 = h\nu_p \frac{9\lambda_\mathrm{p}^2}{8\pi} = \frac{9}{4}\hbar c \lambda_p^2 \qquad 1.4$$

Then we proceed from the assumption that waves B of photons can not exist with waves B of electrons, forming the shells of atoms and molecules in the same space elements. Hence, the effect of excluded volume appears during the propagation of an external electromagnetic wave through the matter. It leads to the fact that in matter the volume occupied by a photon, is equal to

$$V_p^m = V_p^0 - V_p^{\mathrm{ex}} = V_p^0 - n_M^p \bullet V_e^M \qquad 1.5$$

where $V_p^{\mathrm{ex}} = n_M^p V_e^M$ is the excluded volume which is equal to the product of the number of molecules in the volume of one photon standing wave (n_M^p) and the volume occupied by the electron shell of one molecule (V_e^M).

n_M^p is determined by the product of the volume of the photons 3D standing wave in the vacuum (1.3) and the concentration of molecules ($n_M = N_0/V_0$):

$$n_M^p = \frac{9\lambda_p^3}{8\pi}\left(\frac{N_0}{V_0}\right) \qquad 1.6$$

In the absence of the polarization by the external field and intermolecular interaction, the volume occupied by electrons of the molecule:

$$V_e^M = \frac{4}{3}\pi L_e^3 \qquad 1.7$$

where L_e is the radius of the most probable wave B $(L_e = \lambda_e/2\pi)$ of the outer electron of a molecule. As it has been shown in (7.5) that the mean molecule polarizability is:

$$\alpha = L_e^3 \qquad 1.8$$

Then taking (1.7) and (1.6) into account, the excluded volume of primary electromagnetic deformon in the matter is:

$$V_p^{\text{ex}} = \frac{9\lambda_p^3}{8\pi} n_M \frac{4}{3}\pi\alpha = \frac{3}{2}\lambda_p^3 n_M \alpha \qquad 1.9$$

Therefore, the partial volume energy of a photon in the vacuum is determined by eq.(1.4), while that in matter, according to (1.5):

$$[E_p^m] = E_p \bullet V_p^m = E_p \bullet [V_p^0 - V_p^{\text{ex}}] \qquad 1.10$$

Putting (1.4) and (1.10) into (1.1) we obtain:

$$\epsilon = n^2 = \frac{E_p V_p^0}{E_p(V_p^0 - V_p^{\text{ex}})} \qquad 1.11$$

or

$$\frac{1}{n^2} = 1 - \frac{V_p^{\text{ex}}}{V^0} \qquad 1.12$$

Then, putting eq.(1.9) and (1.3) into (1.12) we derive new equation for refraction index, leading from our mesoscopic theory:

$$\frac{1}{n^2} = 1 - \frac{4}{3}\pi n_M \alpha \qquad 1.13$$

or in another form:

$$\frac{n^2 - 1}{n^2} = \frac{4}{3}\pi n_M \alpha = \frac{4}{3}\pi \frac{N_0}{V_0}\alpha \qquad 1.14$$

where: $n_M = N_0/V_0$ **is a concentration of molecules**;

In this equation $\alpha = L_e^3$ is the average static polarizability of molecules for the case when the external electromagnetic fields as well as intermolecular interactions inducing the additional polarization are absent. This situation is realized at $E_p = h\nu_p \to 0$ and $\lambda_p \to \infty$ in the gas phase. As will be shown below the value of resulting α^* in condensed matter is bigger.

1.2. Light refraction in liquids and solids

According to the Lorentz classical theory, the electric component of the outer electromagnetic field is amplified by the additional inner field (E_{ad}), related to the interaction of induced dipole moments in composition of condensed matter with each other:

$$E_{\text{ad}} = \frac{n^2 - 1}{3}E \qquad 1.15$$

The mean Lorentz *acting field* $\bar{F}$ can be expressed as:

$$\bar{F} = E + E_{\text{ad}} = \frac{n^2 + 2}{3}E \quad (\text{at } n \to 1,\ \bar{F} \to E) \qquad 1.16$$

$\bar{F}$- has a dimensions of electric field tension and tends to E in the gas phase when $n \to 1$.

In accordance with our mesoscopic model, except the Lorentz acting field, the total internal acting field, includes also two another contributions, increasing the molecules polarizability (α) in condensed matter:

1. Potential intermolecular field, including all the types of Van- der-Waals interactions in composition of coherent collective excitations, even without external electromagnetic field. Like total potential energy of matter, this contribution must be dependent on temperature and pressure;

2. Primary internal field, related with primary electromagnetic deformons (tr and lib). This component of the total acting field also exist without external fields. Its frequencies corresponds to IR range and its action is much weaker than the action of the external visible light.

Let us try to estimate the energy of the total acting field and its effective frequency (ν_f) and wavelength (λ_f), that we introduce as:

$$A_f = h\nu_f = \frac{\mathrm{hc}}{\lambda_\mathrm{f}} = A_L + A_V + A_D \qquad 1.17$$

where: $A_L,\ A_V$ and A_D are contributions, related with Lorentz field, potential field and primary deformons field correspondingly.

When the interaction energy of the molecule with a photon ($E_p = h\nu_p$) is less than the energy of the resonance absorption, then it leads to elastic polarization of the electron shell and origination of secondary photons, i.e. light scattering. We assume in our consideration that the increment of polarization of a molecule (α) under the action of the external photon ($h\nu_p$) and the total active field ($A_f = h\nu_f$) can be expressed through the increase of the most probable radius of the electron's shell ($L_e = \alpha^{1/3}$), using our (eq. 7.6 from [1]):

$$\Delta L_e = \frac{\omega_\mathrm{p}\ \mathrm{m_e}}{2\hbar}\alpha \qquad 1.18$$

where the resulting increment:

$$\Delta L^* = \Delta L_e + \Delta L_f = \frac{(\mathrm{h}\nu_\mathrm{p} + \mathrm{A_f})\mathrm{m_e}}{2\hbar^2}\alpha \qquad 1.18a$$

where: $\alpha = L_e^3$ is the average polarizability of molecule in gas phase at $\nu_f \to 0$.

For water molecule in the gas phase:

$$L_e = \alpha^{1/3} = 1.13 \bullet 10^{-10}\, m$$

is a known constant, determined experimentally [4].

The total increment of polarizability radius (ΔL^*) and resulting polarizability of molecules (α^*) in composition of condensed matter affected by the acting field

$$\alpha^* = (L^*)^3$$

can be find from the experimental refraction index (n) using our formula (1.14):

$$L^* = \left[\frac{3}{4\pi}\frac{\mathrm{V_0}}{\mathrm{N_0}} \bullet \frac{\mathrm{n}^2 - 1}{\mathrm{n}^2}\right]^{1/3} \qquad 1.19$$

$$\Delta L^* = L^* - L_e \qquad 1.20$$

where: $L^* = (\alpha^*)^{1/3}$

From (1.18) we get a formula for the increment of radius of polarizability (ΔL_f), induced by the total internal acting field:

$$\Delta L_f = \Delta L^* - \Delta L_e = \frac{\mathrm{A_f} \bullet \mathrm{m_e}}{2\hbar^2}\alpha \qquad 1.21$$

Like total internal acting field energy (1.17), this total acting increment can be presented as a sum of contributions, related to Lorentz field (ΔL_F), potential field (ΔL_V) and primary deformons field (ΔL_D):

$$\Delta L_f = \Delta L_L + \Delta L_V + \Delta L_D \qquad 1.22$$

Increment ΔL_e, induced by external photon only, can be calculated from the known frequency (ν_p) of the incident light (see 1.18a):

$$\Delta L_e = \frac{h\nu_p \bullet m_e}{2\hbar^2}\alpha \qquad 1.23$$

It means that ΔL_f can be found from (1.21) and (1.17), using (1.23). Then from (1.21) we can calculate the energy (A_f), effective frequency (ν_f) and wave length (λ_f) of the total acting field like:

$$A_f = h\nu_f = hc/\lambda_f = 2\frac{\Delta L_f \bullet \hbar^2}{m_e\alpha} \qquad 1.24$$

The computer calculations of α^*; $L^* = L_e + \Delta L^* = (\alpha^*)^{1/3}$ and A_f in the temperature range $(0 - 95^0)$ are presented on Fig.1.1.

One must keep in mind that in general case α and L are tensors. It means that all the increments, calculated on the base of eq.(1.18a) must be considered as the effective ones. Nevertheless, it is obvious that our approach to analysis of the acting field parameters can give useful additional information about the properties of transparent condensed matter.

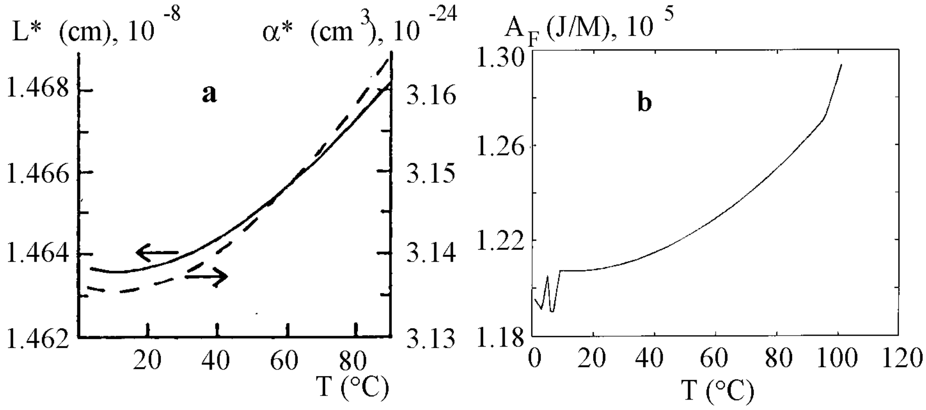

Fig. 1.1. (a)- Temperature dependencies of the most probable outer electron shell radius of H_2O (L^*) and the effective polarizability $\alpha^* = (L^*)^3$ in the total acting field; (b)- Temperature dependence of the total acting field (A_f) energy in water at the wavelength of the incident light $\lambda_p = 5.461 \bullet 10^{-5} cm^{-1}$. The experimental data for refraction index n(t) were used in calculations. The initial electron shell radius is: $L_e = \alpha_{H2O}^{1/3} = 1.13 \bullet 10^{-8} cm$ [4]. In graphical calculations in Fig.1.1a, the used experimental temperature dependence of the water refraction index were obtained by Frontas'ev and Schreiber [5].

The temperature dependencies of these parameters were computed using the known experimental data on refraction index $n(t)$ for water and presented in Fig.1.1a. The radius L^* in the range $0 - 95^0C$ increases less than by 1% at constant incident light wavelength $(\lambda = 546.1nm)$. The change of ΔL_f with temperature is determined by its potential field component change ΔL_V.

The relative change of this component: $\Delta\Delta L_V/\Delta L_f$ $(t = 0^0C)$ is about 9%. Corresponding to this change the increasing of the acting field energy A_f (eq.1.23) increases approximately by $8\,kJ/M$ (Fig 1.1 b) due to its potential field contribution.

It is important that the total potential energy of water in the same temperature range, according to our calculations, increase by the same magnitude (Fig.5b in [1] or Fig.3b in [3]). This fact points to the strong correlation between potential intermolecular interaction in

matter and the value of the acting field energy.

It was calculated that, at constant temperature (20^0) **the energy of the acting field** (A_f), $(eq.1.23)$ **in water practically does not depend on the wavelength of incident light** (λ_p). **At more than three time alterations of** λ_p: **from** $12.56 \bullet 10^{-5} cm$ **to** $3.03 \bullet 10^{-5} cm$ **and the water refraction index** (n) **from 1.320999 to 1.358100** [6] **the value of** A_f **changes less than by 1%**.

At the same conditions the electron shell radius $\mathbf{L}^*$ **and the acting polarizability** α^* **thereby increase from (1.45 to 1.5)•10**$^{-10}\, m$ **and from (3.05 to** $3.274) \bullet 10^{-30} m^3$ **respectively (Fig.1.2). These changes are due to the incident photons action only. For water molecules in the gas phase and** $\lambda_p \to \infty$ **the initial polarizability** $(\alpha = L_e^3)$ **is equal to** $1.44 \bullet 10^{-24} cm^3$ **[4], i.e. significantly less than in condensed matter under the action of external and internal fields.**

Obviously, the temperature change of energy A_f **(Fig.1.1b) is determined by the internal pressure increasing (section 11.2 of [1]), related to intermolecular interaction change, depending on mean distances between molecules and, hence, on the concentration** (N_0/V_0) **of molecules in condensed matter.**

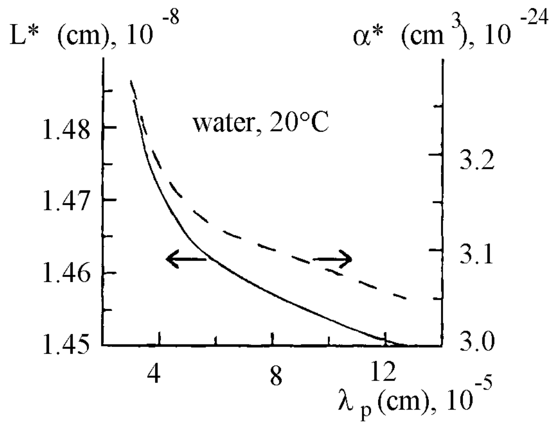

Fig. 1.2. Dependencies of the acting polarizability $\alpha^* = (L^*)^3$ and electron shell radius of water in the acting field (L^*) on incident light wavelength (λ_p), calculated from eq. (1.14) and experimental data $n(\lambda_p)$ [6]. The initial polarizability of H_2O in the gas phase at $\lambda_p \to \infty$ is equal to $\alpha = L_e^3 = 1.44 \bullet 10^{-24} cm^3$. The corresponding initial radius of the H_2O electron shell is $L_e = 1.13 \bullet 10^{-8} cm$.

On the basis of our data, changes of A_f, **calculated from (1.24) are caused mainly by the heat expansion of the matter. The photon induced increment of the polarizability** $(\alpha \to \alpha^*)$ **practically do not change** A_f.

The ability to obtain new valuable information about changes of molecule polarizability under the action of incident light and about temperature dependent molecular interaction in condensed medium markedly reinforce such a widely used method as refractometry.

The above defined relationship between the molecule polarizability and the wave length of the incident light allows to make a new endeavor to solve the light scattering problems.

2. Mesoscopic theory of Brillouin light scattering in condensed matter

2.1. Traditional approach

According to traditional concept, light scattering in liquids and crystals as well as in gases takes place due to random heat fluctuations. In condensed media the fluctuations of density, temperature and molecule orientation are possible.

Density (ρ) fluctuations leading to dielectric penetrability (ϵ) fluctuations are of major importance. This contribution is estimated by means of Einstein formula for scattering coefficient of liquids [7]:

$$R = \frac{\mathrm{Ir}^2}{\mathrm{I_0V}} = \frac{\pi}{2\lambda^4} kT\beta_T \left(\rho \frac{\partial\epsilon}{\partial\rho}\right)_T \qquad 2.1$$

where β_T is isothermal compressibility.

Many authors made attempts to find a correct expression for the variable $(\rho \frac{\partial\epsilon}{\partial\rho})$.

The formula derived by M.F.Vuks [8, 9] is most consistent with experimental data:

$$\rho \frac{\partial\epsilon}{\partial\rho} = (n^2 - 1)\frac{3\mathrm{n}^2}{2\mathrm{n}^2 - 1} \qquad 2.2$$

2.2. Fine structure of scattering

The fine structure - spectrum of the scattering in liquids is represented by two Brillouin components with frequencies shifted relatively from the incident light frequency: $\nu_\pm = \nu_0 \pm \Delta\nu$ and one unshifted band like in gases (ν_0).

The shift of the Brillouin components is caused by the Doppler effect resulting from a fraction of photons scattering on phonons moving at sound speed in two opposite directions [8].

This shift can be explained in different way as well. If in the antinodes of the standing wave the density oscillation occurs at frequency (Ω):

$$\rho = \rho_0 \cos\Omega t, \qquad 2.3$$

then the scattered wave amplitude will change at the same frequency. Such a wave can be represented as a superposition of two monochromatic waves having the frequencies:$(\omega + \Omega)$ and $(\omega - \Omega)$, where

$$\Omega = 2\pi f \qquad 2.4$$

is the elastic wave frequency at which scattering occurs when the Wolf-Bragg condition is satisfied:

$$2\Lambda \sin\varphi = 2\Lambda \sin\frac{\theta}{2} = \lambda' \qquad 2.5$$

or

$$\Lambda = \lambda'/(2\sin\frac{\theta}{2}) = \frac{\mathrm{c}}{\mathrm{n}\nu}(2\sin\frac{\theta}{2}) = v_{ph}/f \qquad 2.6$$

where Λ is the elastic wave length corresponding to the frequency f; $\lambda' = \lambda/n = c/n\nu$ (λ' and λ are the incident light wavelength in matter and vacuum, respectively); φ is the angle of sliding; θ is the angle of scattering; n is the refraction index of matter; c is the light speed.

The value of Brillouin splitting is represented as:

$$\pm\Delta\nu_{M-B} = f = \frac{\mathrm{V_{ph}}}{\Lambda} = 2\nu\frac{\mathrm{V_{ph}}}{\mathrm{c}} n \sin\frac{\theta}{2} \qquad 2.7$$

where: $\nu n/c = 1/\lambda$; n is the refraction index of matter; ν is incident light frequency;

$$v_{ph} = v_S \qquad 2.8$$

is the phase velocity of a scattering wave equal to hypersonic velocity.

The formula (2.7) is identical to that obtained from the analysis of the Doppler effect:

$$\frac{\Delta\nu}{\nu} = \pm 2\frac{V_S}{c} n \sin\frac{\theta}{2} \qquad 2.9$$

According to the classical theory, the central line, which is analogous to that observed in gases, is caused by entropy fluctuations in liquids, without any changes of pressure [8]. On the basis of Frenkel theory of liquid state, the central line can be explained by fluctuations of ”hole” number - cavitational fluctuations [10].

The thermodynamic approach of Landau and Plachek leads to the formula, which relates the intensities of the central (I) and two lateral (I_{M-B}) lines of the scattering spectrum with compressibility and heat capacities:

$$\frac{I}{2I_{M-B}} = \frac{I_p}{I_{ad}} = \frac{\beta_T - \beta_S}{\beta_S} = \frac{C_p - C_v}{C_v} \qquad 2.10$$

where: β_T and β_S are isothermal and adiabatic compressibilities; C_p and C_v are isobaric and isohoric heat capacities.

In crystals, quartz for example, the central line in the fine structure of light scattering is usually absent or very small. However, instead of one pair of shifted components, observed in liquids, there appear *three* Brillouin components in crystals. One of them used to be explained by scattering on the longitudinal phonons, and two - by scattering on the transversal phonons.

2.3. New mesoscopic approach to problem

In our hierarchic theory the thermal ”random” fluctuations are ”organized” by different types of superimposed quantum excitations.

According to our Hierarchic model, **including microscopic**, **mesoscopic and macroscopic scales of matter** (**see Introduction of** [**1**,**2**,**3**]), **the most probable** (**primary**) **and mean** (**secondary**) **effectons**, **translational and librational are capable of quantum transitions between two discreet states**: $(a \Leftrightarrow b)_{tr,lb}$ **and** ($\Leftrightarrow \bar{b})_{tr,lb}$ **respectively**. **These transitions lead to origination/annihilation of photons and phonons**, **forming primary and secondary deformons**.

The mean heat energy of molecules is determined by the value of 3kT, **which as our calculations show**, **has the intermediate value between the discreet energies of a and b quantum states of primary effectons** (**Fig**.**19 of** [**1**]), **making**, **consequently**, **the non equilibrium conditions in condensed matter**. **Such kind of instability is a result of ”competition” between classical and quantum distributions of energy** .

The maximum deviations from thermal equilibrium and that of the dielectric properties of matter occur when the same states of primary and secondary quasiparticles, e.g. a, and $b,\bar{b}$ occur simultaneously. Such a situation corresponds to the A and B states of macroeffectons. The $(A \Leftrightarrow B)_{tr,lb}$ transitions represent thermal fluctuations. The big density fluctuations are related to ”flickering clusters” (macroconvertions between librational and translational primary effectons) and the maximum fluctuations correspond to Superdeformons.

Only in the case of spatially independent fluctuations the interference of secondary scattered photons does not lead to their total compensation.

The probability of the event that two spatially uncorrelated events coincide in time is equal to the product of their independent probabilities [10].

Thus, the probabilities of the coherent (a,) and ($b,\bar{b}$) states of primary and secondary effectons, corresponding to A and B states of the macroeffectons (tr and lib), independent on each other, are equal to:

$$\left(P_M^A \right)^{ind}_{tr,lb} = \left(P_{ef}^a \bar{P}_{ef}^a \right)^S_{tr,lb} \bullet \left(\frac{1}{Z^2} \right) = \left(\frac{P_M^A}{Z^2} \right)_{tr,lb} \qquad 2.11$$

$$(P_M^B)_{tr,lb}^{\text{ind}} = \left(P_{ef}^b \bar{P}_{ef}^b \right)_{tr,lb}^S \bullet \left(\frac{1}{Z^2}\right) = \left(\frac{P_M^B}{Z^2}\right)_{tr,lb} \qquad 2.12$$

where

$$\frac{1}{Z}(P_{ef}^a)_{tr,lb} \quad \text{and} \quad \frac{1}{Z}\left(\bar{P}_{ef}^a\right)_{tr,lb} \qquad 2.13$$

are the independent probabilities of *a* and states determined according to formulae (4.10 and 4.18 of [2,3]), while probabilities $(P_{ef}^b/Z)_{tr,lb}$ and $\left(\bar{P}_{ef}^b/Z\right)_{tr,lb}$ are determined according to formulae (4.11 and 4.19 of [2,3]);

Z is the sum of probabilities of all types of quasiparticles states - eq.(4.2 of [2, 3]).

The probabilities of molecules being involved in the spatially independent translational and librational macrodeformons are expressed as the products (2.11) and (2.12):

$$\left(P_D^M \right)_{tr,lb}^{\text{ind}} = \left[\left(P_M^A \right)^{\text{ind}} \bullet \left(P_M^B \right)^{\text{ind}} \right]_{tr,lb} = \frac{P_D^M}{Z^4} \qquad 2.14$$

Formulae (2.11) and (2.12) may be considered as the probabilities of space-independent but coherent macroeffectons in A and B states, respectively.

For probabilities of space-independent supereffectons in A^* and B^* states we have:

$$\left(P_S^{A^*} \right)^{\text{ind}} = \left(P_M^A \right)_{tr}^{\text{ind}} \bullet \left(P_M^A \right)_{lb}^{\text{ind}} = \frac{P_S^{A^*}}{Z^4} \qquad 2.15$$

$$\left(P_S^{B^*} \right)^{\text{ind}} = \left(P_M^B \right)_{tr}^{\text{ind}} \bullet \left(P_M^B \right)_{tr}^{\text{ind}} = \frac{P_S^{b^*}}{Z^4} \qquad 2.15a$$

In a similar way we get from (2.14) the probabilities of spatially independent superdeformons:

$$\left(P_S^{D^*} \right)^{\text{ind}} = \left(P_M^D \right)_{tr} \bullet \left(P_M^D \right)_{lb} = \frac{P_S^{D^*}}{Z^4} \qquad 2.16$$

The concentrations of molecules, the states of which markedly differ from the equilibrium one and which cause light scattering in composition of spatially independent macroeffectons and macrodeformons, are equal to:

$$\left[N_M^A = \frac{N_0}{Z^2 V_0}\left(P_M^A \right) \right]_{tr,lb}; \qquad \left[N_M^B = \frac{N_0}{Z^2 V_0}\left(P_M^B \right) \right]_{tr,lb} \qquad 2.17$$

$$\left[N_M^D = \frac{N_0}{Z^4 V_0}\left(P_M^D \right) \right]_{tr,lb}$$

The concentrations of molecules, involved in a-convertons, b- convertons and Macroconvertons or c-Macrotransitons (see Introduction) are correspondingly:

$$N_M^{ac} = \frac{N_0}{Z^2 V_0} P_{ac}; \qquad N_M^{bc} = \frac{N_0}{Z^2 V_0} P_{bc}; \qquad N_M^C = \frac{N_0}{Z^4 V_0} P_{\text{cMt}} \qquad 2.18$$

The probabilities of convertons-related excitations are the same as used in Chapter 4 of book [1].

The concentration of molecules, participating in the independent supereffectons and superdeformons:

$$N_M^{A^*} = \frac{N_0}{Z^4 V_0} P_s^{A^*}; \qquad N_M^{B^*} = \frac{N_0}{Z^4 V_0} P_S^{B^*} \qquad 2.19$$

$$N_M^{D^*} = \frac{N_0}{Z^8 V_0} P_S^{D^*} \qquad 2.20$$

where N_0 and V_0 are the Avogadro number and the molar volume of the matter.

Substituting (2.17 - 2.20) into well known **Raleigh formula for scattering coefficient**, **measured at the right angle between incident and scattered beams**:

$$R = \frac{I}{I_0}\frac{r^2}{V} = \frac{8\pi^4}{\lambda^4}\alpha^2 n_{\text{M}} \ (cm^{-1}) \qquad 2.20a$$

we obtain the values of contributions from different states of quasiparticles to the resulting scattering coefficient:

$$(R_A^M)_{tr,lb} = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^2}\frac{N_0}{V_0}\left(P_M^A \right)_{tr,lb}; \quad R_A^s = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^4}\frac{N_0}{V_0}P_s^{A*} \qquad 2.21$$

$$(R_B^M)_{tr,lb} = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^2}\frac{N_0}{V_0}\left(P_M^B \right)_{tr,lb}; \quad R_B^s = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^4}\frac{N_0}{V_0}P_s^{B*} \qquad 2.22$$

$$(R_D^M)_{tr,lb} = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^2}\frac{N_0}{V_0}\left(P_M^D \right)_{tr,lb}; \quad R_D^s = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^4}\frac{N_0}{V_0}P_s^{D*} \qquad 2.23$$

The contributions of excitations, related to [*tr*/*lb*] convertons are:

$$R_{ac} = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^2}\frac{N_0}{V_0}R_{bc} = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^2}\frac{N_0}{V_0}P_{bc}$$

$$R_{\mathrm{abc}} = \frac{8\pi^4}{\lambda^4}\frac{(\alpha^*)^2}{Z^4}\frac{N_0}{V_0}P_{\mathrm{cMt}}$$

where: α^* is the acting polarizability determined by eq.(1.24) and (1.25).

The resulting coefficient of the isotropic scattering (R_{iso}) is defined as the sum of contributions (2.21-2.23) and is subdivided into three kinds of scattering: caused by translational quasiparticles, caused by librational quasiparticles and by the mixed type of quasiparticles:

$$R_{\mathrm{iso}} = [R_A^M + R_B^M + R_D^M] + [R_A^M + R_B^M + R_D^M] + [R_{ac} + R_{bc} + R_{\mathrm{abc}}] + [R_A^s + R_B^s + R_D^s] \qquad 2.24$$

Total contributions, related to convertons and superexcitations are correspondingly:

$$R_C = R_{ac} + R_{bc} + R_{\mathrm{abc}} \quad \text{and} \quad R_S = R_A^s + R_B^s + R_D^s$$

The polarizability of anisotropic molecules having no cubic symmetry is a tensor. In this case, total scattering (R) consists of scattering at density fluctuations (R_{iso}) and scattering at fluctuations of the anisotropy $\left(R_{\mathrm{an}} = \frac{13\Delta}{6-7\Delta}R_{\mathrm{iso}}\right)$:

$$R = R_{\mathrm{iso}} + \frac{13\Delta}{6-7\Delta}R_{\mathrm{iso}} = R_{\mathrm{iso}}\frac{6+6\Delta}{6-7\Delta} = R_{\mathrm{iso}}K \qquad 2.25$$

where R_{iso} corresponds to *eq*.(2.24); Δ is the depolarization coefficient.

The factor: $\left(\frac{6+6\Delta}{6-7\Delta}\right) = K$ was obtained by Cabanne and is called after him. In the case of isotropic molecules when $\Delta = 0$, the Cabanne factor is equal to 1.

The depolarization coefficient (Δ) could be determined experimentally as the ratio:

$$\Delta = I_x/I_z, \qquad 2.26$$

where I_x and I_z are two polarized components of the beam scattered at right angle with respect to each other in which the electric vector is directed parallel and perpendicular to the incident beam, respectively. For example, in water $\Delta = 0.09$ (Vuks, 1977).

According to the proposed theory of light scattering in liquids the central unshifted (like in gases) component of the Brillouin scattering spectrum, is caused by fluctuations of concentration and self-diffusion of molecules, participating in the convertons, macrodeformons (tr and lib) and superdeformons. The scattering coefficients of the central line (R_{centr}) and side lines ($2R_{\mathrm{side}}$) in transparent condensed matter, as follows from (2.24) and (2.25), are equal correspondingly to:

$$R_{\mathrm{cent}} = K\left[\left(R_D^M \right)_{tr} + \left(R_D^M \right)_{lb}\right] + K(R_C + R_S) \qquad 2.27$$

and

$$2R_{\mathrm{side}} = \left(R_A^M + R_B^M \right)_{tr} + \left(R_A^M + R_A^M \right)_{lb} \qquad 2.27a$$

where K is the Cabanne factor.

The total coefficient of light scattering is:

$$R_t = R_{\mathrm{cent}} + 2R_{\mathrm{side}} \qquad 2.28$$

In accordance with our model the fluctuations of anisotropy (Cabanne factor) should be taken into account for calculations of the central component only. The orientations of molecules in composition of A and B states of Macroeffectons are correlated and their coherent oscillations are not accompanied by fluctuations of anisotropy of polarizability (see Fig.2.1).

The probabilities of the convertons, macrodeformons and superdeformons excitations (eqs.2.14, 4.16, 4.27 in [1]) are much lower in crystals than in liquids and hence, the central line in the Brillouin spectra of crystals is not usually observed.

The lateral lines in Brillouin spectra are caused by the scattering on the molecules forming (A) and (B) states of spatially independent macroeffectons, as it was mentioned above.

The polarizabilities of the molecules forming the independent macroeffectons, synchronized in $(A)_{tr,lb}$ and $(B)_{tr,lb}$ states and dielectric properties of these states, differ from each other and from that of transition states (macrodeformons). Such short-living states should be considered as the non equilibrium ones.

In fact we must keep in mind, that static polarizabilities in the more stable ground A state of the macroeffectons are higher than in B state, because the energy of long-term Van der Waals interaction between molecules of the A state is bigger than that of B-state.

If this difference may be attributed mainly to the difference in the long-therm dispersion interaction, then from (1.33) we obtain:

$$E_B - E_A = V_B - V_A = -\frac{3}{2}\frac{E_0}{r^6}\left(\alpha_B^2 - \alpha_A^2 \right) \qquad 2.29$$

where polarizability of molecules in A-state is higher, than that in B-state:

$$\alpha_A^2 > \left[\left(\alpha^* \right)^2 \approx \alpha_D^2 \right] > \alpha_B^2$$

The kinetic energy and dimensions of ”acoustic” and ”optic” states of macroeffectons are the same: $T_{\text{kin}}^A = T_{\text{kin}}^B$.

In our present calculations of light scattering we ignore this difference (2.29) between polarizabilities of molecules in A and B states.

But it can be taken into account if we assume, that polarizabilities in (A) and (a), (B) and (b) states of primary effectons are like:

$$\alpha_A \simeq \alpha_a \simeq \alpha^*; \quad \alpha_B \simeq \alpha_b$$

and the difference between the potential energy of (a) and (b) states is determined mainly by dispersion interaction (eq.2.28).

Experimental resulting polarizability ($\alpha^* \simeq \alpha_a$) can be expressed as:

$$\alpha_a = f_a\alpha_a + f_b\alpha_b + f_t\alpha \qquad 2.29a$$

where $\alpha_t \simeq \alpha$ is polarizability of molecules in the gas state (or transition state);

$$f_a = \frac{P_a}{P_a + P_b + P_t}; \quad f_b = \frac{P_b}{P_a + P_b + P_t};$$

$$\text{and} \quad f_t = f_d = \frac{P_t}{P_a + P_b + P_t}$$

are the fractions of (a), (b) and transition (t) states (equal to 2.66) as far $P_t = P_d = P_a \bullet P_b$.

On the other hand from (1.33) at $r = const$ we have:

$$\Delta V_{\text{dis}}^{b\to a} = -\frac{3}{4}\frac{(2\alpha\Delta\alpha)}{r^6} \bullet I_0 \qquad (r_a = r_b; \quad I_0^a \simeq I_0^b) \text{ and}$$

$$\frac{\Delta V_{\text{dis}}^{b\to a}}{V^b} = \frac{h\nu_p}{h\nu_b} = \frac{\Delta\alpha_a}{\alpha} \ \text{ or } \ \Delta\alpha_a = \alpha_a\frac{\nu_p}{\nu_b} \qquad 2.29b$$

$$\alpha_b = \alpha_a - \Delta\alpha_a = \alpha_a(1 - \nu_p/\nu_b)$$

where: $\Delta\alpha_a$ is a change of each molecule polarizability as a result of the primary effecton energy changing: $E_b \to E_a + h\nu_p$ with photon radiation; ν_b is a frequency of primary effecton in (b)- state (eq.2.28).

Combining (2.29) and (2.29b) we derive for α_a and α_b of the molecules composing primary translational or librational effectons:

$$\alpha_a = \frac{\mathrm{f_t}\alpha}{1 - \left(\mathrm{f_a} + \mathrm{f_b} + \mathrm{f_b} \bullet \frac{\mathrm{v_p}}{\mathrm{v_b}}\right)} \qquad 2.30$$

$$\alpha_b = \alpha_a\left(1 - \frac{\mathrm{v_p}}{\mathrm{v_b}}\right) \qquad 2.30a$$

The calculations by means of (2.30) are approximate in the framework of our assumptions mentioned above. But they correctly reflect the tendencies of α_a and α_b changes with temperature.

The ratio of intensities or scattering coefficients for the central component to the lateral ones previously was described by Landau- Plachek formula (2.10). According to our mesoscopic theory this ratio can be calculated in another way leading from (2.27) and (2.28):

$$\frac{\mathrm{I_{centr}}}{2\mathrm{I_{M-B}}} = \frac{\mathrm{R_{cent}}}{2\mathrm{R_{side}}} \qquad 2.30b$$

Combining (2.30) and Landau- Plachek formula (2.10) it is possible to calculate the ratio (β_T/β_S) and (C_P/C_V) using our mesoscopic theory of light scattering.

2.4. Factors that determine the Brillouin line width

The known equation for Brillouin shift is (see 2.7):

$$\Delta\nu_{M-B} = \nu_0 = 2\frac{\mathrm{v_s}}{\lambda}n\sin(\theta/2) \qquad 2.31$$

where: v_s is the hypersonic velocity; λ is the wavelength of incident light, n is the refraction index of matter, and θ - scattering angle.

The deviation from ν_0 that determines the Brillouin side line half width *may be expressed as the result of fluctuations of sound velocity v_s and n* related to A and B states of tr and lib macroeffectons:

$$\frac{\Delta\nu_0}{\nu_0} = \left(\frac{\Delta\mathrm{v_s}}{\mathrm{v_s}} + \frac{\Delta\mathrm{n}}{\mathrm{n}}\right) \qquad 2.32$$

$\Delta\nu_0$ is the most probable side line width, i.e. the true half width of Brillouin line. It can be expressed as:

$$\Delta\nu_0 = \Delta\nu_{\mathrm{exp}} - F\Delta\nu_{\mathrm{inc}}$$

where $\Delta\nu_{\mathrm{exp}}$ is the half width of the experimental line, $\Delta\nu_{\mathrm{inc}}$ - the half width of the incident line, F - the coefficient that takes into account apparatus effects.

Let us analyze the first and the second terms in the right part of (2.32) separately.

The v_s squared is equal to the ratio of the compressibility modulus (K) and density (ρ):

$$v_s^2 = K^2/\rho \qquad 2.33$$

Consequently, from (2.33) we have:

$$\frac{\Delta\mathrm{v_s}}{\mathrm{v_s}} = \frac{1}{2}\left(\frac{\Delta\mathrm{K}}{\mathrm{K}} - \frac{\Delta\rho}{\rho}\right) \qquad 2.34$$

In the case of independent fluctuations of K and ρ ::

$$\frac{\Delta\mathrm{v_s}}{\mathrm{v_s}} = \frac{1}{2}\left(\left|\frac{\Delta\mathrm{K}}{\mathrm{K}}\right| - \left|\frac{\Delta\rho}{\rho}\right|\right) \qquad 2.35$$

From our equation (1.14) we obtain for refraction index:

$$n^2 = \left(1 - \frac{4}{3}N\alpha^*\right)^{-1}, \qquad 2.36$$

where $N = N_0/V_0$ is the concentration of molecules.

From (2.36) we can derive:

$$\frac{\Delta\mathrm{n}}{\mathrm{n}} = \frac{1}{2}(n^2 - 1)\left(\frac{\Delta\alpha^*}{\alpha^*} + \frac{\Delta\mathrm{N}}{\mathrm{N}}\right) \qquad 2.37$$

where:

$$(\Delta N/N) = (\Delta\rho/\rho) \qquad 2.38$$

and

$$\left(\frac{\Delta\alpha^*}{\alpha^*}\right) \simeq \left(\frac{\Delta K}{K}\right) \qquad 2.39$$

we can assume eq.(2.39) as far both parameters: polarizability (α^*) and compressibility models (K) are related with the potential energy of intermolecular interaction.

For the other hand one can suppose that the following relation is true:

$$\frac{\Delta\alpha^*}{\alpha^*} \simeq \frac{|\quad {}^{a}_{ef} - 3kT\ |}{3kT} = \frac{\Delta K}{K} \qquad 2.40$$

where: $\ {}^{a}_{ef}$ is the energy of the secondary effectons in ( ) state; $E_0 = 3kT$ is the energy of an "ideal" quasiparticle as a superposition of 3D standing waves.

The density fluctuations can be estimated as a result of the free volume (v_f) fluctuations (see 2.45):

$$\left(\frac{\Delta v_f}{v_f}\right)_{tr,lb} = \frac{1}{Z}(P_D^M)_{tr,lb} \simeq (\Delta N/N)_{tr,lb} \qquad 2.41$$

Now, putting (2.40) and (2.41) into (2.37) and (2.34) and then into (2.32), we obtain the semiempirical formulae for the Brillouin line half width calculation:

$$\frac{\Delta v_f}{v_f} \simeq \frac{n^2}{2}\left[\frac{|\quad {}^{a}_{ef} - 3kT\ |}{3kT} + \frac{1}{Z}\left(\ P_D^M\ \right)\right]_{tr,lb} \qquad 9.42$$

Brillouin line intensity depends on the half-width $\Delta\nu$ of the line in following ways:

for a Gaussian line shape:

$$I(\nu) = I_0^{\max}\exp\left[-0.693\left(\frac{\nu - \nu_0}{\frac{1}{2}\Delta\nu_0}\right)^2\right]; \qquad 2.43$$

for a Lorenzian line shape:

$$I(\nu) = \frac{I_0^{\max}}{1 + \left[\ (\nu - \nu_0)/\frac{1}{2}\Delta\nu_0\ \right]^2} \qquad 2.44$$

The traditional theory of Brillouin line shape gives a possibility for calculation of $\Delta\nu_0$ taking into account the elastic (acoustic) wave dissipation.

The fading out of acoustic wave amplitude may be expressed as:

$$A = A_0 e^{-\alpha x} \quad \text{or} \quad A = A_0 e^{-\alpha v_s} \qquad 2.45$$

where α is the extinction coefficient; $x = v_s t$ - the distance from the source of waves; v_s and t - sound velocity and time, correspondingly.

The hydrodynamic theory of sound propagation in liquids leads to the following expression for the extinction coefficient:

$$\alpha = \alpha_s + \alpha_b = \frac{\Omega^2}{2\rho v_s^3}\left(\frac{4}{3}\eta_s + \eta_b\right) \qquad 2.46$$

where: α_s and α_b are contributions to α, related to share viscosity (η_s) and bulk viscosity (η_b), respectively; $\Omega = 2\pi f$ is the angular frequency of acoustic waves.

When the side lines in Brillouin spectra broaden slightly, the following relation between their intensity (I) and shift ($\Delta\omega = |\ \omega - \omega_0\ |$) from frequency ω_0, corresponding to maximum intensity ($I = I_0$) of side line is correct:

$$I = \frac{I_0}{1 + (\frac{\omega - \omega_0}{a})}, \qquad 2.47$$

where:

$$a = \alpha v_s.$$

One can see from (2.46) that at $I(\omega) = I_0/2$, the half width:

$$\Delta\omega_{1/2} = 2\pi\Delta\nu_{1/2} = \alpha v_s \quad \text{and} \quad \Delta\nu_{1/2} = \frac{1}{2}\pi\alpha v_s \qquad 2.48$$

It will be shown in Chapter 12 how one can calculate the values of η_s and consequently α_s on the basis of the mesoscopic theory of viscosity.

2.5. Quantitative verification of mesoscopic theory of Brillouin scattering

The calculations made according to the formula (2.21 - 2.27) are presented in Fig.2.1-2.7. The proposed theory of scattering in liquids, based on our hierarchic concept, is more adequate than the traditional Einstein, Mandelschtamm-Brillouin, Landau-Plachek theories based on classical thermodynamics. It describes experimental temperature dependencies and the $I_{\text{centr}}/2I_{M-B}$ ratio for water very well (Fig.2.3).

The calculations are made for the wavelength of incident light: $\lambda_{ph} = 546.1nm = 5.461 \bullet 10^{-5}cm$. The experimental temperature dependence for the refraction index (n) at this wavelength was taken from the Frontas'ev and Schreiber paper (1965). The rest of data for calculating of various light scattering parameters of water (density the location of translational and librational bands in the oscillatory spectra) are identical to those used above in Chapter 6.

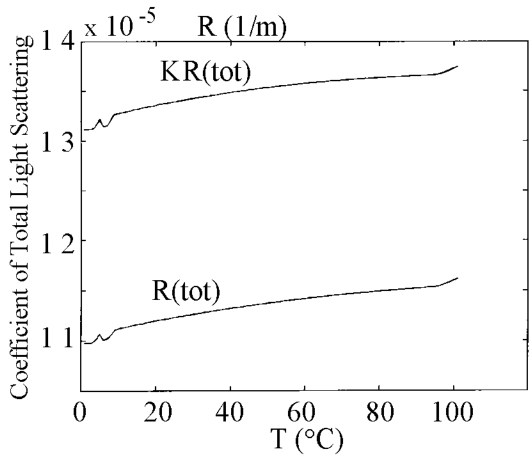

Fig. 2.1. Theoretical temperature dependencies of the total scattering coefficient for water without taking into account the anisotropy of water molecules polarizability fluctuations in the volume of macroeffectons, responsible for side lines: $[R(tot)]$ - eq.(2.27a; 2.28) and taking them into account: $[KR(tot)]$, where K is the Cabanne factor (eq.2.25).

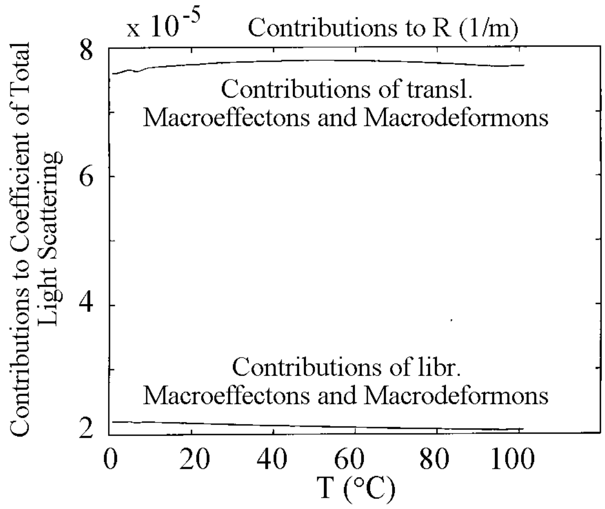

Fig. 2.2. Theoretical temperature dependencies of contributions to the total coefficient of total light scattering (R) caused by translational and librational macroeffectons and macrodeformons (without taking into account fluctuations of anisotropy).

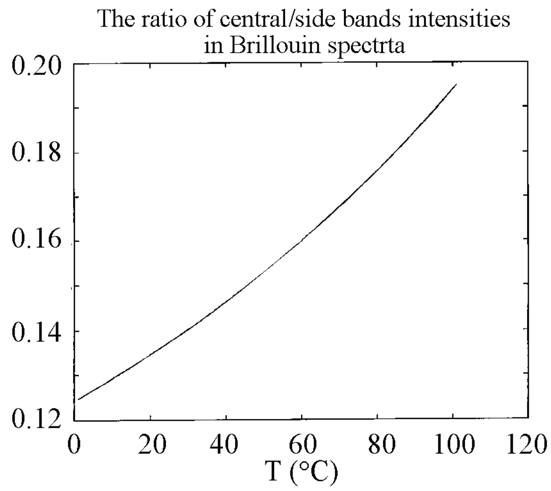

Fig. 2.3. Theoretical temperature dependencies of central to side bands intensities ratio in Brillouin spectra (eq.2.30).

Mesoscopic theory of light scattering can be used to verify the correctness of our formula for refraction index of condensed matter we got from our theory (eq. 1.14):

$$\frac{n^2-1}{n^2} = \frac{4}{3}\pi\frac{N_0}{V_0}\alpha^* \qquad 2.48a$$

and to compare the results of its using with that of the Lorentz-Lorenz formula:

$$\frac{n^2-1}{n^2+1} = \frac{4}{3}\pi\frac{N_0}{V_0}\alpha \qquad 2.49$$

From formula (2.48a) the resulting or effective molecular polarizability squared $(\alpha^*)^2$ used in eq.(2.21-2.23) is:

$$(\alpha^*)^2 = \left[\frac{(n^2-1)/n^2}{(4/3)\pi(N_0/V_0)}\right]^2 \qquad 2.50$$

On the other hand, from the Lorentz-Lorenz formula (2.49) we have another value of polarizability:

$$\alpha^2 = \left[\frac{(n^2-1)/(n^2+2)}{(4/3)\pi(N_0/V_0)} \right]^2 \qquad 2.51$$

It is evident that the light scattering coefficients (eq.2.28), calculated using (2.50) and (2.51) taking refraction index: $n = 1.33$ should differ more than four times as far:

$$\frac{R(\alpha^*)}{R(\alpha)} = \frac{(\alpha^*)^2}{(\alpha)^2} = \frac{(n^2-1)/n^2}{(n^2-1)/(n^2+2)} = \left(\frac{n^2+2}{n^2}\right)^2 = 4.56 \qquad 2.52$$

At 25^0 and $\lambda_{ph} = 546nm$ the theoretical magnitude of the scattering coefficient for water, calculated from our formulae (2.28) is equal (see Fig.2.1) to:

$$R = 11.2 \bullet 10^{-5}m^{-1} \qquad 2.53$$

This result of our theory coincides well with the most reliable experimental value (Vuks, 1977):

$$R_{\mathrm{exp}} = 10.8 \bullet 10^{-5}m^{-1}$$

Multiplication of the side bands contribution ($2R_{\mathrm{side}}$) to Cabanne factor increases the calculated total scattering to about 25% and makes the correspondence with experiment worse. This fact confirms our assumption that fluctuations of anisotropy of polarizability in composition of A and B states of macroeffectons should be ignored in light scattering evaluation due to correlation of molecular dynamics in these states, in contrast to that of macrodeformons.

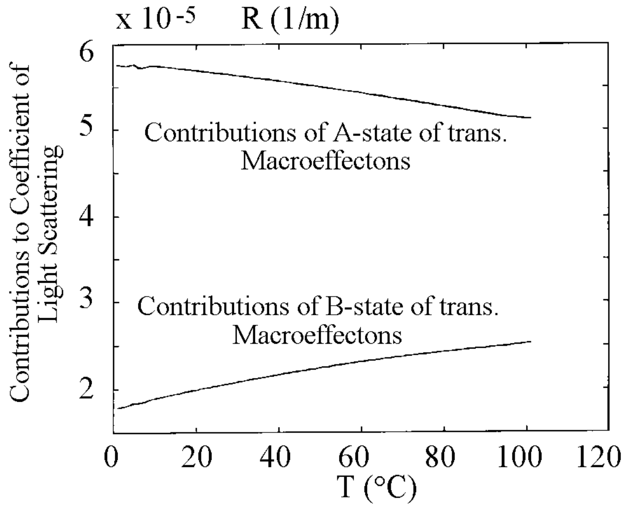

Fig. 2.4. Theoretical temperature dependencies of the contributions of A and B states of translational Macroeffectons to the total scattering coefficient of water (see also Fig.2.2);

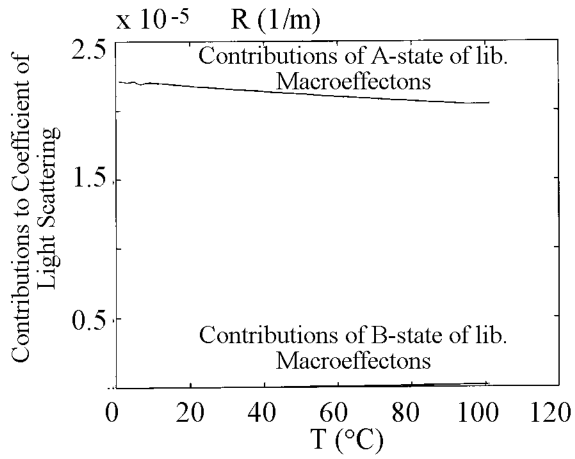

Fig. 2.5. Theoretical temperature dependencies of the contributions of the A and B states of librational Macroeffectons to the coefficient of light scattering (R).

It follows from the Fig.2.4 and 2.5 that the light scattering depends on ($A \Leftrightarrow B$) equilibrium of macroeffectons because $(R_A) > (R_B)$, i.e. scattering on A states is bigger than that on B states.

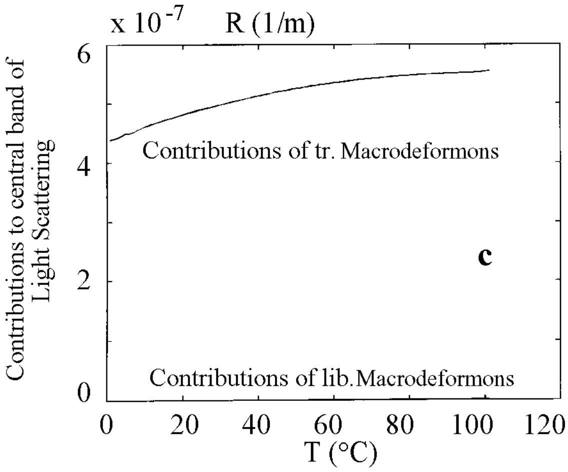

Fig. 2.6. Theoretical temperature dependencies of the contributions to light scattering (central component), related to translational $(R_D)_{tr}$ and librational $(R_D)_{lb}$ macrodeformons.

Comparing Figs. 2.1; 2.3, and 2.6 one can see that the main contribution to central component of light scattering is determined by [lb/tr] convertons R_c (see eq.2.27).

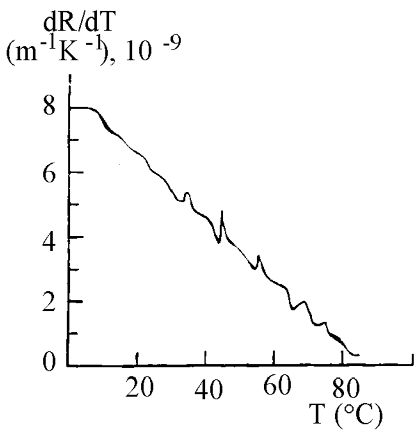

Fig. 2.7. Theoretical temperature dependences for temperature derivative (dR/dT) of the total coefficient of light scattering of water.

Nonmonotonic deviations of the dependencies dR/dT (Fig.2.7) reflect the nonmonotonic changes of the refraction index for water $n_{H_2O}(T)$, as indicated by available experimental data (Frontas'ev and Schreiber, 1965). The deviations of dependence $n_{H_2O}(t)$ from the monotonic way in accordance with hierarchic theory, are a consequence of the nonmonotonic change in the stability of water structure, i.e. nonlinear change of $(A \Leftrightarrow B)_{tr,lb}$ equilibrium. Some possible reasons of such equilibrium change were discussed in Chapter 6.

It is clear from (2.52) that the calculations based on the Lorentz-Lorentz formula (2.51) give scattering coefficient values of about 4.5 times smaller than experimental ones. It

means that the true α^* value can be calculated just on the basis of our mesoscopic theory of light refraction (eq.2.50).

The traditional Smolukhovsky-Einstein theory, valid for the integral light scattering only (eq. 2.1), yield values in the range of $R = 8.85 \bullet 10^{-5} m^{-1}$ to $R = 10.5 \bullet 10^{-5} m^{-1}$ [4, 8].

All the results, discussed above, mean that our mesoscopic theory of light scattering works better and is much more informative than the conventional one.

2.6. Light scattering in solutions

If the guest molecules are dissolved in a liquid and their sizes are much less than incident light wavelength, they do not radically alter the solvent properties. For this case the described above mechanism of light scattering of pure liquids does not changed qualitatively.

For such dilute solutions the scattering on the fluctuations of concentration of dissolved molecules (R_c) is simply added to the scattering on the density fluctuations of molecules of the host solvent (eq.2.28). Taking into account the fluctuations of molecule polarizability anisotropy (see 2.25) the total scattering coefficient of the solution (R_S) is:

$$R_S = R_t + R_c \qquad 2.54$$

Eqs. (2.21 - 2.28) could be used for calculating R_t until critical concentrations (C_{cr}) of dissolved substance when it start to destroy the solvent structure, so that the latter is no longer able to form primary librational effectons. Perturbations of solvent structure will induce low-frequency shift of librational bands in the oscillatory spectrum of the solution until these bands totally disappear.

If the experiment is made with a two-component solution of liquids, soluble in each other, e.g. water-alcohol, benzol-methanol etc., and the positions of translational and librational bands of solution components are different, then at the concentration of the dissolved substance: $C > C_{cr}$, the dissolved substance and the solvent (the guest and host) can switch their roles. Then the translational and librational bands pertinent to the guest subsystem start to dominate. In this case, R_t is to be calculated from the positions of the new bands corresponding to the ”new” host-solvent. The total ”melting” of the primary librational ”host effectons” and the appearance of the dissolved substance ”guest effectons” is *like the second order phase transition* and should be accompanied by a heat capacity jump. The like experimental effects take place indeed [8].

According to our concept, the coefficient R_c in eq.(2.54) is caused by the fluctuations of concentration of dissolved molecules in the volume of translational and librational macro- and superdeformons of the solvent. If the destabilization of the solvent is expressed in the **low frequency** shift of librational bands, then the coefficients $(R_A$ and $R_B)_{lb}$ increase (eq.2.21 and 2.22) with the probability of macro-excitations.. The probabilities of convertons and macro- and superdeformons and the central component of Brillouin spectra will increase also. Therefore, the intensity of the total light scattering increases correspondingly.

The fluctuations of concentration of the solute molecules, in accordance with our model, occur in the volumes of macrodeformons and superdeformons. Consequently, the contribution of solute molecules in scattering (R_c value in eq.2.54) can be expressed by formula, similar to (2.23), but containing the molecule polarizability of the dissolved substance (”guest”), equal to $(\alpha_g^*)^2$ instead of the molecule polarizability (α^*) of the solvent (”host”), and the molecular concentration of the ”guest” substance in the solution (n_g) instead of the solvent molecule concentration ($n_M = N_0/V_0$). For this case R_c could be presented as a sum of the following contributions:

$$(R_c)_{tr,lb} = \frac{8\pi^4}{\lambda^4}(\alpha_g^*)^2 n_g \bullet [(P_M^D)_{tr,lb} + P_S^{D^*}] \qquad 2.55$$

$$R_c^{D^*} = \frac{8\pi^4}{\lambda^4}(\alpha_g^*)^2 n_g \bullet (P_S^{D^*}) \qquad 2.55a$$

The resulting scattering coefficient (R_e) on fluctuations of concentration in (2.54) is equal to:

$$R_c = (R_c)_{tr} + (R_c)_{lb} + R_c^{D^*} \qquad 2.56$$

If **several** substances are dissolved with concentrations lower than (C_{cr}), then their R_c are summed up additively.

Formulae (2.55) and (2.56) are valid also for the dilute solutions.

Eqs.(2.21-2.28) and (2.54-2.56) should, therefore, be used for calculating the resulting coefficient of light scattering in solutions (R_S).

The traditional theory represents the scattering coefficient at fluctuations of concentration as (Vuks, 1977):

$$R_c = \frac{\pi^2}{2\lambda^4}\left(\frac{\partial\epsilon}{\partial x}\right)^2 \Delta x^2 v \qquad 2.57$$

where $(\partial\epsilon/\partial x)$ is the dielectric penetrability derivative with respect to one of the components: $\Delta\bar{x}^2$ is the fluctuations of concentration of guest molecules squared in the volume element v.

The transformation of (2.57) on the basis of classical thermodynamics [8] leads to the formula:

$$R_c = \frac{\pi^2}{2\lambda^4 N_0}\left(2n\frac{\partial n}{\partial x}\right)\left(\frac{9n^2}{(2n^2+1)(n^2+2)}\right)^2 x_1 x_2 V_{12} f, \qquad 2.58$$

where N_0 is the Avogadro number, x_1 and x_2 are the molar fractions of the first and second components in the solution, V_{12} is the molar volume of the solution, f is the function of fluctuations of concentration determined experimentally from the partial vapor pressures of the first (P_1) and second (P_2) solution components [8]:

$$\frac{1}{f} = \frac{x_1}{P_1}\frac{\partial P_1}{\partial x_1} = \frac{x_2}{P_2}\frac{\partial P_2}{\partial x_2} \qquad 2.59$$

In the case of ideal solutions

$$\frac{\partial P_1}{\partial x_1} = \frac{P_1}{x_1}; \quad \frac{\partial P_2}{\partial x_2} = \frac{P_2}{x_2}\ ; \ \text{and} \ \ f = 1.$$

For application the mesoscopic theory of light scattering to study of crystals, liquids and solutions, the following information is needed:

1. **Positions of translational and librational band maxima in oscillatory spectra**;
2. **Concentration of all types of molecules in solutions**;
3. **Refraction index or polarizability in the acting field of each component of solution at given temperature**.

Application of our theory to quantitative analysis of transparent liquids and solids yields much more information about properties of matter, its mesoscopic and hierarchic dynamic structure than the traditional one.

3. Mesoscopic theory of Mössbauer effect

3.1. General background

When the atomic nucleus with mass (M) in the gas phase irradiates γ-quantum with energy of

$$E_0 = h\nu_0 = m_p c^2 \qquad 3.1$$

where: m_p is the effective photon mass, then according to the law of impulse conservation, the nuclear acquires *additional* velocity in the opposite direction:

$$v = -\frac{E_0}{Mc} \qquad 3.2$$

The corresponding additional kinetic energy

$$E_R = \frac{Mv^2}{2} = \frac{E_0^2}{2Mc^2} \qquad 3.3$$

is termed **recoil energy**.

When an atom which irradiates γ-quantum is in composition of the solid body, then **three situations** are possible:

1. The recoil energy of the atom is higher than the energy of atom - lattice interaction. In this case, the atom irradiating γ-quantum would be knocked out from its position in the lattice. That leads to defects origination;

2. Recoil energy is insufficient for the appreciable displacement of an atom in the structure of the lattice, but is higher than the energy of phonon, equal to energy of secondary transitons and phonons excitation. In this case, recoil energy is spent for heating the lattice;

3. Recoil energy is lower than the energy of primary transitons, related to [emission/absorption] of IR translational and librational photons $(h\nu_p)_{tr,lb}$ **and phonons** $(h\nu_{ph})_{tr,lb}$. **In that case, the probability (f) of γ-quantum irradiation without any the losses of energy appears, termed the probability (fraction) of a recoilless processes.**

For example, when $E_R << h\nu_{ph}$ (ν_{ph} - the mean frequency of phonons), then the mean energy of recoil:

$$E_R = (1-f)h\nu_{ph} \qquad 3.4$$

Hence, the probability of recoilless effect is

$$f = 1 - \frac{E_R}{h\nu_{ph}} \qquad 3.5$$

According to eq.(3.3) the decrease of the recoil energy E_R of an atom in the structure of the lattice is related to increase of its effective mass (M). In our model M corresponds to the mass of the effecton.

The effect of γ-quantum irradiation without recoil was discovered by Mössbauer in 1957 and named after him.

The value of Mössbauer effect is determined by the value of $f \leq 1$.

The big recoil energy may be transferred to the lattice by portions that are **resonant** to the frequency of IR photons (tr and lb) and phonons. **The possibility of superradiation of IR quanta stimulation as a result of such recoil process is a consequence of our mesoscopic model**.

The scattering of γ-quanta without lattice excitation, when $E_R << h\nu_{ph}$, is termed the *elastic* one. The general expression [11, 12] for the probability of such phononless elastic γ-quantum radiation acts is equal to:

$$f = \exp\left(-\frac{4\pi < x^2 >}{\lambda_0^2}\right) \qquad 3.6$$

where $\lambda_0 = c/\nu_0$ is the real wavelength of γ-quantum; $<x^2>$ - the nucleus oscillations mean amplitude squared in the direction of γ-quantum irradiation.

The γ-quanta wavelength parameter may be introduced like:

$$L_0 = \lambda_0/2\pi, \qquad 3.7$$

where: $L_0 = 1.37 \bullet 10^{-5} cm$ for Fe^{57}, then eq.(3.6) could be written as follows:

$$f = \exp\left(-\frac{< x^2 >}{L_0^2}\right) \qquad 3.8$$

It may be shown [12], proceeding from the model of crystal as a system of 3N identical quantum oscillators, that when temperature (T) is much lower than the Debye one (θ_D) then:

$$< x^2 >= \frac{9\hbar^2}{4Mk\theta_D}\left\{1 + \frac{2\hbar^2 T^2}{3\theta_D^2}\right\}, \qquad 3.9$$

where $\theta_D = h\nu_D/k$ and ν_D is the Debye frequency.

From (3.1), (3.3) and (3.7) we have:

$$\frac{1}{L} = \frac{E_0}{\hbar c} \qquad 3.10$$

where: $E_0 = h\nu = c(2ME_R)^{1/2}$ is the energy of γ-quantum

Substituting eqs.(3.9 and 3.10) into eq.(3.8), we obtain the Debye-Valler formula:

$$f = \exp\left[-\frac{E_R}{k\theta_D}\left\{\frac{3}{2} + \frac{\pi^2 T^2}{\theta_D}\right\}\right] \qquad 3.11$$

when $T \to 0$, then

$$f \to \exp\left(-\frac{3E_R}{2k\theta_D}\right) \qquad 3.12$$

3.2. Probability of elastic effects

Mean square displacements $<x^2>$ of an atoms or molecules in condensed matter (eq. 3.8) is not related to excitation of thermal photons or phonons (i.e. primary or secondary transitons). According to our concept, $< x^2 >$ is caused by the mobility of the atoms forming effectons and differs for primary and secondary translational and librational effectons in $(a, \quad)_{tr,lb}$ and $(b, b)_{tr,lb}$ states.

We will ignore below the contributions of macro- and supereffectons in Mössbauer effect as very small. Then the resulting probability of elastic effects at γ-quantum radiation is determined by the sum of the following contributions (see *eqs*.4.2 – 4.4 of [1, 2]):

$$f = \frac{1}{Z}\sum_{tr,lb}\left[\left(P^a_{ef} f^a_{ef} + P^b_{ef} f^b_{ef}\right) + \left(\bar{P}^a_{ef}\bar{f}^a_{ef} + \bar{P}^b_{ef}\bar{f}^b_{ef}\right)\right]_{tr,lb} \qquad 3.13$$

where: $P^a_{ef},\ P^b_{ef},\ \bar{P}^a_{ef},\ \bar{P}^b_{ef}$ are the relative probabilities of the *acoustic and optic* states for primary and secondary effectons; Z is the total partition function.

These parameters are calculated as described in Chapter 4 of book [1] and in papers cited in the Summary of this article. Each of contributions to resulting probability of the elastic effect can be calculated separately as:

$$\left(f^a_{ef}\right)_{tr,lb} = \exp\left[-\frac{<\left(x^a\right)^2_{tr,lb}>}{L_0^2}\right] \qquad 3.14$$

$\left(f^a_{ef}\right)_{tr,lb}$ is the probability of elastic effect, related to dynamics of primary translational and librational effectons in *a*-state;

$$\left(f^b_{ef}\right)_{tr,lb} = \exp\left[-\frac{<\left(x^b\right)^2_{tr,lb}>}{L_0^2}\right] \qquad 3.15$$

$\left(f^b_{ef}\right)_{tr,lb}$ is the probability of elastic effect in primary translational and librational effectons in *b*-state;

$$\left(\bar{f}^a_{ef}\right)_{tr,lb} = \exp\left[-\frac{<\left(\bar{x}^a\right)^2_{tr,lb}>}{L_0^2}\right] \qquad 3.16$$

$\left(\bar{f}^a_{ef}\right)_{tr,lb}$ is the probability for secondary effectons in -state;

$$\left(\bar{f}_{ef}^{b}\right)_{tr,lb} = \exp\left[-\frac{<\left(\bar{x}^{b}\right)^{2}_{tr,lb}>}{L_0^2}\right] \qquad 3.17$$

$\left(\bar{f}_{ef}^{b}\right)_{tr,lb}$ is the probability of elastic effect, related to secondary effectons in $\bar{b}$-state.

Mean square displacements within different types of effectons in eqs.(3.14-3.17) are related to their phase and group velocities. At first we express the displacements using group velocities of the waves $B(v_{gr})$ and periods of corresponding oscillations (T) as:

$$<\left(x^{a}\right)^{2}_{tr,lb}> = \frac{<(v^{a}_{gr})^{2}_{tr,lb}>}{<\nu_a^2>_{tr,lb}} = <\left(v^{a}_{gr}T^{a}\right)^{2}_{tr,lb}> \qquad 3.18$$

where $(T^a)_{tr,lb} = (1/\nu_a)_{tr,lb}$ is a relation between the period and the frequency of primary translational and librational effectons in *a*-state;

$(v^{a}_{gr} = v^{b}_{gr})_{tr,lb}$ are the group velocities of atoms forming these effectons equal in (a) and (b) states.

In a similar way we can express the displacements of atoms forming (b) state of primary effectons (tr and lib):

$$<\left(x^{b}\right)^{2}_{tr,lb}> = \frac{<(v^{b}_{gr})^{2}_{tr,lb}>}{<\nu_b^2>_{tr,lb}} \qquad 3.19$$

where ν_b is the frequency of *primary* translational and librational effectons in *b*-state.

The mean square displacements of atoms forming *secondary* translational and librational effectons in and $\bar{b}$ states:

$$<\left(\bar{x}^{a}\right)^{2}_{tr,lb}> = \frac{<(\bar{v}^{a}_{gr})^{2}_{tr,lb}>}{<\bar{\nu}_a^2>_{tr,lb}} \qquad 3.20$$

$$<\left(\bar{x}^{b}\right)^{2}_{tr,lb}> = \frac{<(\bar{v}^{b}_{gr})^{2}_{tr,lb}>}{<\bar{\nu}_b^2>_{tr,lb}} \qquad 3.21$$

where: $(\bar{v}^{a}_{gr} = \bar{v}^{b}_{gr})_{tr,lb}$

Group velocities of atoms in primary and secondary effectons may be expressed using corresponding phase velocities (v_{ph}) and formulae for waves B length as follows:

$$\left(\lambda_a\right)_{tr,lb} = \frac{h}{m<v_{gr}>_{tr,lb}} = \left(\frac{v^{a}_{ph}}{\nu_a}\right)_{tr,lb} = \qquad 3.22$$

$$= \left(\lambda_b\right)_{tr,lb} = \left(\frac{v^{b}_{ph}}{\nu_b}\right)_{tr,lb}$$

hence for the group velocities of the atoms or molecules forming primary effectons (*tr and lb*) squared we have:

$$\left(v^{a,b}_{gr}\right)^{2}_{tr,lb} = \frac{h^2}{m^2}\left(\frac{\nu_{a,b}}{v^{a,b}_{ph}}\right)^{2}_{tr,lb} \qquad 3.23$$

In accordance with mesoscopic theory, the wave B length, impulses and group velocities in *a* and *b* states of the effectons are equal. Similarly to (3.23), we obtain the group velocities of particles, composing secondary effectons:

$$\left(\bar{v}^{a,b}_{gr}\right)^{2}_{tr,lb} = \frac{h^2}{m^2}\left(\frac{\bar{\nu}_{a,b}}{\bar{v}^{a,b}_{ph}}\right)^{2}_{tr,lb} \qquad 3.24$$

Substituting eqs.(3.23) and (3.24) into (3.18-3.21), we find the important expressions for the average coherent displacements of particles squared as a result of their oscillations in the volume of the effectons (*tr*, *lib*) in both discreet states (acoustic and optic):

$$< (x^a)^2_{tr,lb} >= (h/mv^a_{ph})^2_{tr,lb} \qquad 3.25$$

$$< (x^b)^2_{tr,lb} >= (h/mv^b_{ph})^2_{tr,lb} \qquad 3.26$$

$$< (\bar{x}^a)^2_{tr,lb} >= (h/m\bar{v}^a_{ph})^2_{tr,lb} \qquad 3.27$$

$$< (\bar{x}^b)^2_{tr,lb} >= (h/m\bar{v}^b_{ph})^2_{tr,lb} \qquad 3.28$$

Then, substituting these values into eqs.(3.14-3.17) we obtain a set of different contributions to the resulting probability of effects without recoil:

$$\left.\begin{aligned} \left(f_f^a \right)_{tr.lb} &= \exp\left[-\left(\frac{h}{mL_0 v^a_{ph}} \right)^2 \right]_{tr,lb}; \\ \left(f_f^b \right)_{tr.lb} &= \exp\left[-\left(\frac{h}{mL_0 v^b_{ph}} \right)^2 \right]_{tr,lb}; \end{aligned}\right\} \qquad 3.29$$

$$\left.\begin{aligned} \left(\bar{f}_f^a \right)_{tr.lb} &= \exp\left[-\left(\frac{h}{mL_0 \bar{v}^a_{ph}} \right)^2 \right]_{tr,lb}; \\ \left(\bar{f}_f^b \right)_{tr.lb} &= \exp\left[-\left(\frac{h}{mL_0 \bar{v}^b_{ph}} \right)^2 \right]_{tr,lb}; \end{aligned}\right\} \qquad 3.30$$

where the phase velocities $(v^a_{ph},\ v^b_{ph},\ \bar{v}^a_{ph},\ \bar{v}^b_{ph})_{tr,lb}$ are calculated from the resulting sound velocity and the positions of translational and librational bands in the oscillatory spectra of matter at given temperature using eqs.2.69-2.75. The wavelength parameter:

$$L_0 = \frac{c}{2\pi\nu_0} = \frac{hc}{2\pi E_0} = 1.375 \bullet 10^{-11}\, m$$

for gamma-quanta, radiated by nuclear of Fe^{57}, with energy:

$$E_0 = 14.4125 \text{ kev} = 2.30167 \bullet 10^{-8} \text{ erg}$$

Substituting eqs.(3.29) and (3.30) into (3.13), we find the total probability of recoilless effects (f_{tot}) in the given substance. Corresponding computer calculations for ice and water are presented on Figs.3.1 and 3.2.

As far the second order phase transitions in general case are accompanied by the alterations of the sound velocity and the positions of translational and librational bands, they should also be accompanied by alterations of f_{tot} and its components.

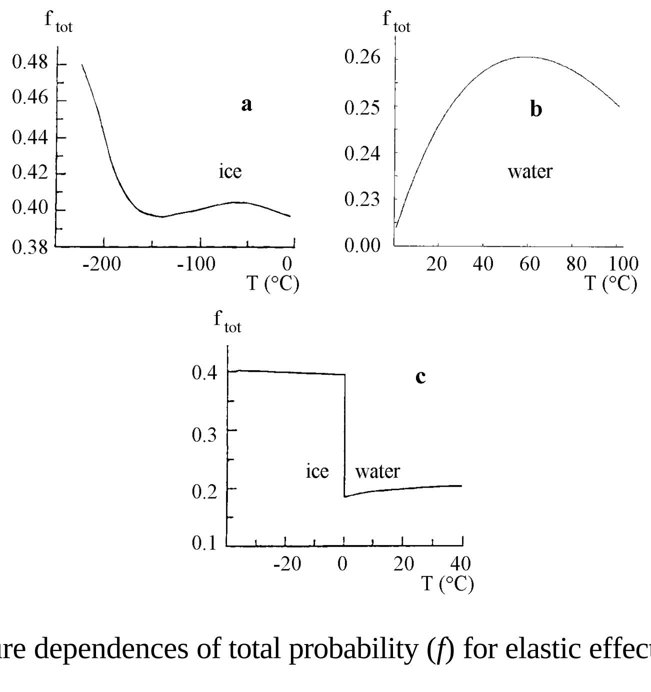

Fig. 3.1. Temperature dependences of total probability (f) for elastic effect without recoil and

phonon excitation: (a) in ice; (b) in water; (c)-during phase transition. The calculations were performed using eq.(3.13).

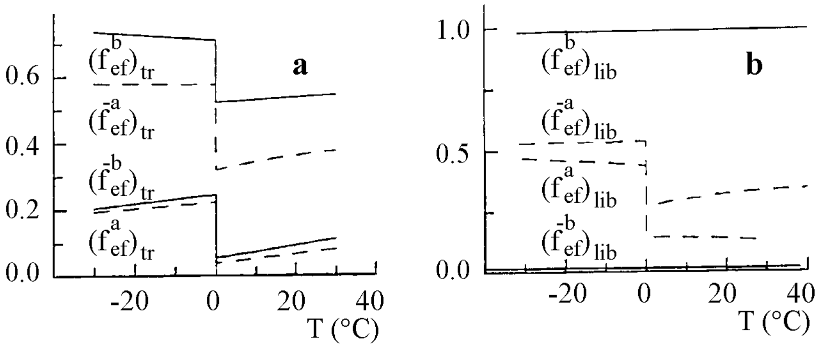

Fig. 3.2. (a) - The contributions to probability of elastic effect (f) (see Fig.3.1) for primary $(f_{ef}^{a,b})_{tr}$ and secondary $(\bar{f})_{tr}$ translational effectons and (**b**) and those of librational effectons $(f_{ef}^{a,b})_{lb}$ and $(\bar{f})_{lb}$ near the temperature of [ice ⇔ water] phase transition.

The total probability (f) and its components, caused by primary and secondary quasiparticles were calculated according to formula (3.13). The value of (f) determines the magnitude of the Mössbauer effect registered by γ-resonance spectroscopy.

The band width caused by recoilless effects is determined by the uncertainty principle and expressed as follows:

$$\Gamma = \frac{h}{\tau} \approx \frac{10^{-27}}{1.4 \bullet 10^{-7}} = 7.14 \bullet 10^{-21}\ \text{erg} = 4.4 \bullet 10^{-9} eV \qquad 3.31$$

where τ is the lifetime of nucleus in excited state (for Fe^{57} $\tau = 1.4 \bullet 10^{-7} s$).

The position of the band depends on the mean square velocity of atoms, i.e. on second order Doppler effect. In the experiment, such an effect is compensated by the *velocity of γ-quanta source* motion relative to absorbent. In the framework of our model this velocity is interrelated with the mean velocity of the secondary effectons diffusion in condensed matter.

3.3. Doppler broadening in spectra of nuclear gamma-resonance (NGR)

Mössbauer effect is characterized by the nonbroadened component of NGR spectra only, with probability of observation determined by eq.(3.13).

When the energy of absorbed γ-quanta exceeds the energy of thermal IR *photons* (*tr*,*lib*) or *phonons excitation*, the absorbance band broadens as a result of Doppler effect. Within the framework of our mesoscopic concept the Doppler broadening is caused by thermal displacements of the particles during $[a \Leftrightarrow b$ and $\Leftrightarrow \bar{b}]_{tr,lb}$ transitions of primary and secondary effectons, leading to origination/annihilation of the corresponding type of deformons (electromagnetic and acoustic).

The *flickering clusters*: [*lb*/*tr*] convertons (*a* and *b*), can contribute in the NGR line broadening also.

In that case, the value of Doppler broadening ($\Delta\Gamma$) of the band in the NGR spectrum could be estimated from corresponding kinetic energies of these excitations, related to their group velocities (see eq. 4.31). In our consideration we take into account the **reduced to one molecule** kinetic energies of primary and secondary translational and librational transitons, *a*-convertons and *b*-convertons. The contributions of macroconvertons, macro- and superdeformons are much smaller due to their small probability and concentration:

$$\Delta\Gamma = \frac{V_0}{N_0 Z} \sum_{tr,lb} \left(n_t P_t T_t + \bar{n}_t \bar{P}_t \bar{T}_t \right)_{tr,lb} + \qquad 3.32$$

$$+ \frac{V_0}{N_0 Z} (n_{ef})_{lb} [P_{ac} T_{ac} + P_{bc} T_{bc}]$$

where: N_0 and V_0 are the Avogadro number and molar volume;

Z is the total partition function (*eq*.4.2); n_t and $\bar{n}_t$ are the concentrations of primary and secondary transitons (eqs.3.5 and 3.7);

$(n_{ef})_{lb} = n_{\mathrm{con}}$ is a concentration of primary librational effectons, equal to that of the convertons; P_t and $\bar{P}_t$ are the relative probabilities of primary and secondary transitons (eqs. 4.26 and 4.27); P_{ac} and P_{bc} are relative probabilities of (*a* and *b*) -convertons (see Chapter 4 of [1]);

T_t and $\bar{T}_t$ are the kinetic energies of primary and secondary transitons, related to the corresponding total energies of these excitations (E_t and $_t$), their masses (M_t and $\overline{M}_t$) and the resulting sound velocity (v_s, see eq.2.40) in the following form:

$$(T_t)_{tr,lb} = \frac{\sum_1^3 \left(E_t^{1,2,3}\right)_{tr,lb}}{2M_t (v_s^{res})^2} \qquad 3.33$$

$$(T_t)_{tr,lb} = \frac{\sum_1^3 \left(\ _t^{1,2,3}\right)_{tr,lb}}{2\bar{M}_t (v_s^{res})^2} \qquad 3.34$$

The kinetic energies of (a and b) convertons are expressed in a similar way:

$$(T_{ac}) = \frac{\sum_1^3 (E_{ac}^{1,2,3})_{tr,lb}}{2M_c (v_s^{res})^2}$$

$$(T_{bc}) = \frac{\sum_1^3 \left(E_{bc}^{1,2,3}\right)_{tr,lb}}{2M_c (v_s^{res})^2}$$

where: $E_{ac}^{1,2,3}$ and $E_{bc}^{1,2,3}$ are the energies of selected states of corresponding convertons; M_c is the mass of convertons, equal to that of primary librational effectons.

The broadening of NGR spectral lines by Doppler effect in liquids is generally expressed using the diffusion coefficient (D) at the assumption that the motion of Mössbauer atom has the character of unlimited diffusion [13]:

$$\Delta\Gamma = \frac{2E_0^2}{\hbar c^2} D \qquad 3.35$$

where: $E_0 = h\nu_0$ is the energy of gamma quanta; c is light velocity and

$$D = \frac{kT}{6\pi\eta a} \qquad 3.36$$

where: η is viscosity, (a) is the effective Stokes radius of the atom Fe^{57}

The probability of recoilless γ-quantum absorption by the matter containing for example Fe^{57}, decreases due to diffusion and corresponding Doppler broadening of band ($\Delta\Gamma$):

$$f_D = \frac{\Gamma}{\Gamma + \Delta\Gamma} \qquad 3.37$$

where $\Delta\Gamma$ corresponds to eq.(3.32).The formulae obtained here make it possible to experimentally verify a set of consequences of our mesoscopic theory using the gamma- resonance method. A more detailed interpretation of the data obtained by this method also becomes possible.

The magnitude of ($\Delta\Gamma$) was calculated according to formula (3.32). It corresponds well to experimentally determined Doppler widening in the nuclear gamma resonance (NGR) spectra of ice.

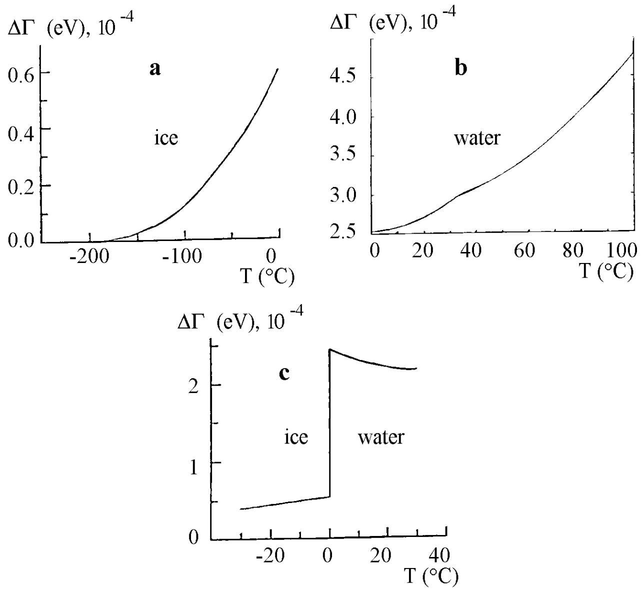

Fig. **3**.**3**. The temperature dependences of the parameter ΔΓ, characterizing the nonelastic effects and related to the excitation of thermal phonons and IR photons: **a**) in ice; **b**) in water; **c**) near phase transition.

3.4. Acceleration and forces, related to thermal dynamics of molecules and ions. Hypothesis of Vibro-gravitational interaction

During the period of particles thermal oscillations (tr and lb), their instant velocity, acceleration and corresponding forces alternatively and strongly change.

The change of wave B instant group velocity, averaged during the molecule oscillation period in composition of the (a) and (b) states of the effectons, determines the average *acceleration*:

$$\left[a_{gr}^{a,b} = \frac{\mathrm{dv}_{\mathrm{gr}}^{\mathrm{a,b}}}{\mathrm{dt}} = \frac{\mathrm{v}_{\mathrm{gr}}^{\mathrm{a,b}}}{\mathrm{T}} = v_{gr}\nu^{a,b}\right]_{tr,lb}^{1,2,3} \qquad 3.38$$

We keep in mind that group velocities, impulses and wave B length in (a) and (b) states of the effectons are equal, in accordance with our model.

Corresponding to (3.38) forces:

$$[F^{a,b} = ma_{gr}^{a,b}]_{tr,lb}^{1,2,3} \qquad 3.39$$

The energies of molecules in (a) and (b) states of the effectons also can be expressed via accelerations:

$$\left[E^{a,b} = h\nu^{a,b} = F^{a,b}\lambda = ma^{a,b} \bullet \lambda = ma^{a,b}(v_{ph}^{a,b}/\nu^{a,b})\right]_{tr,lb}^{1,2,3} \qquad 3.40$$

From (3.40) one can express the accelerations of particles in the primary effectons of condensed matter, using their phase velocities as a waves B:

$$\left[a_{gr}^{a,b} = \frac{\mathrm{h}(\nu^{\mathrm{a,b}})^2}{\mathrm{mv}_{\mathrm{ph}}^{\mathrm{a,b}}}\right]_{tr,lb}^{1,2,3} \qquad 3.41$$

The accelerations of particles in composition of secondary effectons have a similar form:

$$\left[\ {}_{gr}^{a,b} = \frac{\mathrm{h}(\bar{\nu}^{\mathrm{a,b}})^2}{\mathrm{m}\bar{\mathrm{v}}_{\mathrm{ph}}^{\mathrm{a,b}}}\right]_{tr,lb}^{1,2,3} \qquad 3.42$$

These parameters are important for understanding the dynamic properties of condensed systems.

The accelerations of the atoms, forming primary and secondary effectons can be calculated, using eqs.(2.74-2.75 of [1]) to determine phase velocities and eqs. (2.27, 2.28, 2.54, 2.55 [1]) to find a frequencies.

Multiplying (3.41) and (3.42) by the atomic mass m, we derive the most probable and mean forces acting upon the particles in both states of primary and secondary effectons in condensed matter:

$$\left[F_{gr}^{a,b} = \frac{\mathrm{h}(\nu^{a,b})^2}{\mathrm{v}_{\mathrm{ph}}^{a,b}} \right]_{tr,lb}^{1,2,3} \qquad \left[\bar{F}_{gr}^{a,b} = \frac{\mathrm{h}(\bar{\nu}^{a,b})^2}{\bar{\mathrm{v}}_{\mathrm{ph}}^{a,b}} \right] \qquad 3.43$$

The comparison of calculated accelerations with empirical data of the Mössbauer effect - supports the correctness of our approach.

According to eq.(2.54) in the low temperature range, when $h\nu_a << kT$, the frequency of *secondary tr and lb* effectons in the (a) state can be estimated as:

$$\nu^a = \frac{\nu_a}{\exp\left(\frac{\mathrm{h}\nu_a}{\mathrm{kT}}\right) - 1} \approx \frac{\mathrm{kT}}{\mathrm{h}} \qquad 3.44$$

For example, at $T = 200K$ we have $\bar{\nu}^a \approx 4 \bullet 10^{12} s^{-1}$.

If the phase speed in eq.(3.42) is taken equal to $\bar{v}_{ph}^a = 2.1 \bullet 10^5 cm/s$ (see Fig.2a) and the mass of water molecule:

$$m = 18 \bullet 1.66 \bullet 10^{-24} g = 2.98 \bullet 10^{-23} g,$$

then from (3.42) we get the acceleration of molecules in composition of secondary effectons of ice in (a) state:

$$_{gr}^{a} = \frac{\mathrm{h}(\bar{\nu}^a)^2}{\mathrm{m}\bar{\mathrm{v}}_{\mathrm{ph}}^{\mathrm{a}}} = 1.6 \bullet 10^{16} cm/s^2$$

This value is about 10^{13} times more than that of free fall acceleration ($g = 9.8 \bullet 10^2 cm/s^2$), which agrees well with experimental data, obtained for solid bodies [11].

Accelerations of H_2O molecules in composition of *primary librational* effectons (a_{gr}^a) in the ice at 200K and in water at 300K are equal to: $0.6 \bullet 10^{13} cm/s^2$ and $2 \bullet 10^{15} cm/s^2$, correspondingly. **They also exceed to many orders the free fall acceleration**.

It was shown experimentally (Sherwin, 1960), that heating of solid body leads to decreasing of gamma-quanta frequency (red Doppler shift) i.e. increasing of corresponding quantum transitions period. This can be explained as the relativist time-pace decreasing due to elevation of average thermal velocity of atoms [11].

The thermal vibrations of particles (atoms, molecules) in composition of primary effectons as a partial Bose-condensate are coherent. The increasing of such clusters dimensions, determined by most probable wave B length, as a result of cooling, pressure elevation or under magnetic field action (see section 14.6 of [1]) leads to enhancement of coherent regions.

Each coherently vibrating cluster of particles with big alternating accelerations, **like librational and translational effectons is a source of coherent gravitational waves**.

The frequency of these vibro-gravitational waves (VGW) is equal to frequency of particles vibrations (i.e. frequency of the effectons in *a* or *b* states). The amplitude of VGW (A_G) is proportional to the number of vibrating coherently particles (N_G) in composition of primary effectons:

$$A_G \sim N_G \sim V_{ef}/(V_0/N_0) = (1/n_{ef})/(V_0/N_0)$$

The resonant long-distance gravitational interaction between coherent clusters of the same body or between that of different bodies is possible. **The formal description of this vibro-gravitational interaction (VGI) could be like that of distant macroscopic Van der Waals interaction**.

Different patterns of nonlocal Bose-condensate of standing gravitational waves in vacuum represent the field-informational copy of local Bose- condensate of the effectons of condensed matter.

Very important role of proposed here distant resonant VIBRO-GRAVITATIONAL INTERACTION (VGI) in elementary acts of perception and memory can be contributed by coherent primary librational water effectons in microtubules of the nerve cells (see paper "Hierarchic Model of Consciousness" in URL: http://www.karelia.ru/~alexk).

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