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COMMENTS ON THE MOLECULAR MECHANISM OF ACOUSTIC WAVE PROPAGATION IN SIMPLE LIQUIDS

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When seeking an explanation for the molecular mechanism of acoustic wave propagation in liquids it was stated that theoretical considerations applying the Lennard-Jones type expression for energy can not serve this purpose. A certain molecular interpretation of the ABL principle is proposed as a solution. On this path we obtain an expression for propagation velocity of an acoustic wave in terms of space filling a relationship between sound velocity and coefficient of viscosity and an expression for intermolecular compressibility.

Poszukując wyjaśnienia molekularnego mechanizmu propagacji fal akustycznych w cieczach stwierdzono, że takie rozważania teoretyczne, w których stosuje się wyrażenia na energię typu Lennarda-Jonesa nie mogą prowadzić do celu. Jako próbę wyjścia z tej sytuacji proponuje się pewną molekularną interpretację reguły ABL. Uzyskuje się w taki sposób wyrażenie na prędkość propagacji fali akustycznej w funkcji wypełnienia przestrzeni, zależność prędkości dźwięku od współczynnika lepkości oraz wyrażenie na ściśliwość międzymolekularną.

1. Considerations of an elementary, i.e. molecular mechanism of acoustic wave propagation in definite liquids require detailed information about the structure and internal interactions in these liquids. But even in a case of simple liquids, such informations are extremely scant. For example, data concerning free volume achieved by Kittel and Eyring differ by an order of magnitude [1]. Table 1 contains some values of free volume for several liquids at the temperature of 15°C, calculated by Kittel and Eyring and presented in the mentioned paper. Considerable differences between values of free volume determined by various authors with various methods have been also pointed out by SOCZKIEWICZ [9].

It should be further mentioned that conclusions of considerations applying an expression for intermolecular interaction potential energy of Lennard-Jones or similar (e.g in a statistical integral) type can not be applied in considerations of the elementary act of propagation of an acoustic wave. This act consists of effects taking place between molecules and of effects occurring in the molecule itself. It this case the compressibility of the molecule should be distinguished from intermolecular compressibility which is of kinetic as well as potential character anyway. The so-called compressibility of liquid – compressibility measured in macroscale – is a certain

Table 1. Values of free volume for chosen liquids at temperature of 15°C according to [1]

Type of liquid	$V_s\left[\frac{m^3}{mol}\right]\times 10^6$	
	according to EYRING	according to KITTEL
Benzene	0.217	0.0509
Toluene	0.209	0.0454
Chlorobenzene	0.181	0.0357
Bromobenzene	0.155	0.0296
Carbon tetrachloride	0.251	0.0566
Chloroform	0.271	0.0591

resultant of mentioned compressibilities. If we include molecules' own volume then we find that the potential field in which molecules move has nothing to do with the Lennard–Jones potential. Molecules as point sources of forces introduce an idealization which falsifies these details of the liquid's molecular structure which are necessary in considerations of the elementary process of acoustic wave propagation. However, sometimes certain segments on the diagram illustrating potential energy of molecule interaction in terms of distance r, described with the Lennard–Jones formula,

$$\varphi(r) = -\frac{a}{r^m} + \frac{b}{r^n} \tag{1}$$

are accepted as corresponding with the diameter of molecules; but this is logically delayed reasoning. This is so, because first of all conclusions are drawn at an assumption that molecules are point sources of forces then the diameter of molecules is defined as if the regard of the molecule's own volume did not change anything significant.

It results from the above that we should give up Lennard–Jones type expressions and models based on such expressions when seeking a relationships between propagation velocity of acoustic wave and quantities which characterize the actual structure of the liquid.

So all we can do in this situation is consider empirical data expressed in adequate principles and try to explain these principles in molecular terms. The theory requires information resulting from this procedure, not the other way round. It is natural that these informations can not be influenced by such side effects like molecule association or relaxation. Therefore, we will be concerned with simple liquids and with frequency range much below relaxation frequency in these liquids.

2. In the course of research on acoustic properties of liquids many empirical formulae have been formulated and later it became evident that they are not so accurate or not so universal as it seemed at the beginning (e.g. RAO rule, WADA principle). To us it seems that the AZIZ, BOWMAN and LIM principle [2] proved itself most accurate. Further on we will use the notation — ABL principle, for short.

According to this principle propagation velocity of an acoustic wave (w) is directly proportional to density (ϱ) independently of the fact whether density changes are caused by temperature changes at constant pressure or pressure changes at constant temperature. In other words, expressions $(\partial w/\partial \varrho)_p$ and $(\partial w/\partial \varrho)_T$ are constant quantities.

Let us consider expression $(\partial w/\partial \varrho)_p = \text{const}$, which can be expressed in the following form

$$w = \frac{A}{V} + B, \tag{2}$$

where V is the specific volume of the liquid, A and B are constants dependent on kind of liquid.

Let us extrapolate in our minds and imagine that volume V decteases to a possibly smallest value V_D when molecules touch each other. We have

$$\lim w = \lim_{V \to V_D} \left(\frac{A}{V} + B \right)$$

or (3)

$$w_D = \frac{A}{V_D} + B.$$

Quantities w_D and V_D can not be physically reached, but they are a result of formal extrapolation.

Now we can note

$$w - w_D = \frac{A}{V} - \frac{A}{V_D},$$

so the propagation velocity of an acoustic wave in liquids can be written in the following form

$$w = w_D - \frac{A}{V} \cdot \frac{V - V_D}{V_D}. \tag{4}$$

This expression shows how the wave's propagation velocity depends on "space filling". Coefficient A can be eliminated due to logarithmic differentiation of formula (4). Then we have

$$\frac{1}{w - w_D} \left(\frac{\partial w}{\partial T} \right)_p = \frac{1}{V - V_D} \left(\frac{\partial V}{\partial T} \right)_p - \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p,$$

or

$$w = w_D + \frac{\left(\frac{\partial w}{\partial T} \right)_p}{\frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p} \cdot \frac{V - V_D}{V_D}. \tag{4a}$$

Quantities A and B in expression (3) or $\left(\frac{\partial w}{\partial T}\right)_p$ and $\frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p$ in expression (4a) should be determined from experiments and in this case these expressions are equivalent. Still, volume V_D remains unknown.

We will take advantage of liquids viscosity isochores in order to determine V_D. It results that liquids viscosity depends only on volume for a definite mass; it is not effected by temperature or pressure [3]. Such behaviour is a certain analog of the ABL principle for sound velocity.

The relationship between viscosity η and specific volume V(or molar V^m) at constant atmospheric pressure is, as we know, expressed by the BACZYŃSKI principle [4]

$$\eta = \frac{C}{V - V'_D}, \tag{5}$$

where C and V'_D are constant quantities for a given liquid.

BACZYŃSKI stated good conformity of this rule with experiment for 68 liquids which do not associate. The average value V'_D was determined by him at 0.307 V_k (V_k – critical volume); it is close to the value of constant $b = \frac{1}{3} V_k$ in the van der Walls equation.

In further considerations we will call quantity $V_s = V - V'_D$ from expression (5) – free volume. We are aware of the fact that the term-free volume has different meaning in various papers. This does not cause problems when the term used in a definite paper is explicitly defined. The above can be an explanation for great differences between values of free volume achieved by various authors.

If we notice the boundary value of the rearranged ABL expression

$$w_D = \frac{A}{V_D} + B,$$

then an assumption can be made that the given V_D value is a quantity corresponding with the highest possible density of the liquid and that it corresponds with quantity V_D in BACZYŃSKI's formula, because for $V = V'_D$ we have $\eta = \infty$. Now, we will check again the rightness of this assumption. We will calculate V_D from (3) and V'_D from (5) and compare these values with each other. So we have a relationship $w(\eta)$

$$w_D = \frac{A}{V_D} + B = \frac{A}{V - \frac{C}{\eta}} + B. \tag{6}$$

While expression (4) assumes the following form

$$w = w_D - \frac{A}{V} \cdot \frac{V - V_D}{V_D} = w_D - \frac{A}{V} \cdot \frac{C}{\eta V_D},$$

what means that the propagation velocity of an acoustic wave (w) depends on the coefficient of viscosity (η) as

$$w = a + b \cdot \frac{1}{\eta V}, \tag{7}$$

where coefficients a and b can be easily determined from the ABL principle or BACZYŃSKI's principle.

Or course relationships (4) and (7) can be applied in the range of application of mentioned above principles.

Values of the w_D quantity for certain simple liquids at the temperature of 20°C are given in Table 2; while values of w_D for the same liquids at different temperatures can be found in Table 3.

We can notice that the w_D quantity was found constant for tested liquids and independent of temperature (the difference between maximal and minimal value of w_D does not exceed $0.2 \div 0.5\%$ of the average value); while the ratio of w_D and

Table 2. Values of w_D for some simple liquids at temperature of 20°C

Type of liquids	$C \cdot 10^8$ $\left[\frac{m^2}{s}\right]$	A $\left[\frac{m^4}{kg\ s}\right]$	B $\left[\frac{m}{s}\right]$	w $\left[\frac{m}{s}\right]$	w_D $\left[\frac{m}{s}\right]$	$\frac{w_D}{w}$
n-heksane	6.484	4.950	−2163	1099.9	1605	1.459
n-heptane	6.658	5.032	−2286	1152.7	1582	1.372
n-oktane	7.145	5.107	−2392	1193.1	1561	1.308
n-nonane	7.810	5.060	−2403	1227.0	1538	1.254
benzene	5.764	4.272	−2427	1327.0	1646	1.240
toluene	5.594	4.473	−2542	1330.0	1678	1.261
carbon tetrachloride	4.752	1.591	−1597	1937.8	1153	1.229
chloroform	4.695	1.785	−1654	1001.0	1380	1.379
methyl ethyl ketone	5.989	4.086	−2069	1217.0	1642	1.348

Caution: values of w for the first four liquids come from paper [7], while for other liquids from paper [8].

Table 3. Values of w_D for some simple liquids at various temperatures

Type of liquids	t[°C]							$\frac{\Delta w_D}{w_D}$[%]
	0	10	20	30	40	50	60	
n-heksane	1605	1606	1605	1605	1606	1606	1605	0.03
n-heptane	1587	1584	1582	1581	1582	1584	1587	0.41
n-oktane	1567	1563	1561	1561	1563	1564	1571	0.40
n-nonane	1541	1538	1538	1537	1538	1538	1540	0.25
benzene	–	1648	1646	1645	1644	1645	1647	0.22
toluene	1684	1680	1678	1676	1675	1675	1675	0.53
carbon tetrachloride	1159	1155	1153	1153	1153	1154	1156	0.55
chloroform	1382	1381	1380	1380	1381	1381	1381	0.10
methyl ethyl ketone	1645	1643	1642	1642	1642	1643	1644	0.18

experimentally measured value of sound velocity w decreases from liquids with small ratio of space filling (e.g. n-hexane) to liquids with high ratio of space filling (e.g. n-octane or carbon tetrachloride).

We calculated values of sound velocity for several chosen simple liquids from formula (7) using previously found values of constants: A, C, V_D and compared them with experimental values. This confrontation is presented in Table 4.

In our calculations we used values of coefficients of viscosity for these liquids from Thorpe's and Rodger's paper [5] and specific volumes were determined from international tables of physical data [6]. Values of sound velocity in presented liquids have been taken from Boelhouwer's paper [7] and Lagemann's and Woolf's paper [8].

Data presented in Table 4 proves that expression (7) gives values of sound velocity consistent with experimental values; the standard error in the range 0 ÷ 60°C is equal to approximately 1% for all tested liquids.

It is sometimes more convenient to determine characteristic quantities V_D and $V_s = V - V_D$, omitting the constant parameter C in Baczyński's formula. Logarithmic differentiation immediately changes Baczyński's expression into

$$\frac{1}{V_s}\left(\frac{\partial V}{\partial T}\right)_p = -\frac{1}{\eta}\left(\frac{\partial \eta}{\partial T}\right)_p. \tag{8}$$

Hence

$$V_s = -V\cdot\frac{\frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p}{\frac{1}{\eta}\left(\frac{\partial \eta}{\partial T}\right)_p}, \tag{9a}$$

and

$$V_D = V - V_s = V\left[1+\frac{\frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p}{\frac{1}{\eta}\left(\frac{\partial \eta}{\partial T}\right)_p}\right], \tag{9b}$$

or from expression (4a)

$$w_D = w\left[1+\frac{\frac{1}{w}\left(\frac{\partial w}{\partial T}\right)_p}{\frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p+\frac{1}{\eta}\left(\frac{\partial \eta}{\partial T}\right)_p}\right]. \tag{10}$$

Table 4. Values of sound velocity in some simple liquids, calculated from expression [7]

Type of liquids	$w\left[\frac{m}{s}\right]$	t[°C]							Standard error [m/s]
		0	10	20	30	40	50	60	
n-heksane	w_{cal}	1188.2	114.58	1102.2	1057.3	1022.4	964.6	917.1	0.9
	w_{exp}	1191.9	1145.7	1099.9	1054.5	1009.5	964.7	920.2	
	Δw	−3.7	+0.1	+2.3	+1.8	+1.9	−0.1	3.1	
n-heptane	w_{cal}	1235.5	1196.5	1155.8	1113.5	1069.8	1024.7	978.2	1.3
	w_{exp}	1240.7	1196.5	1152.9	1109.7	1066.9	1024.6	982.8	
	Δw	−5.2	−0.1	+2.9	+2.8	+2.9	+0.1	−4.6	
n-oktane	w_{cal}	1274.3	1236.3	1196.4	1154.8	1111.5	1066.8	1025.2	1.2
	w_{exp}	1278.0	1235.6	1193.5	1151.9	1110.8	1070.1	1029.9	
	Δw	−3.7	+0.7	+2.9	+2.9	+0.7	−3.3	−4.7	
carbon tetrachloride	w_{cal}	996.5	969.1	940.3	910.4	879.4	874.4	814.6	1.3
	w_{exp}	1001.9	969.8	938.1	906.8	875.9	845.4	815.3	
	Δw	−5.4	−0.7	+2.2	+3.6	+4.5	+2.0	−0.7	
chloroform	w_{cal}	1069.6	1037.2	1004.0	970.0	935.4	900.1	864.5	0.9
	w_{exp}	1072.9	1037.2	1002.0	967.4	933.5	900.2	867.6	
	Δw	−3.3	0.0	+2.0	+2.6	+1.9	−0.1	−3.1	
benzene	w_{cal}	–	1369.3	1326.4	1282.0	1236.2	1189.1	1141.1	1.1
	w_{exp}	–	1374.0	1326.9	1280.2	1233.9	1188.1	1143.0	
	Δw	–	−4.3	−0.5	+1.8	+3.3	+1.0	−1.9	
toluene	w_{cal}	1406.1	1368.5	1329.5	1289.5	1248.3	1206.3	1163.5	1.3
	w_{exp}	1413.1	1371.0	1329.1	1287.4	1245.7	1204.1	1162.4	
	Δw	−7.0	−2.5	+0.4	+2.1	+2.6	+2.2	+1.1	

A decision should be made in every separate case whether it is more convenient to use parameters A, B, C in discussed principles, or relative thermal coefficients in accordance with formulae (9a), (9b) and (10). These coefficients not always can be determined with desirable accuracy for example. This is in case when there is few measuring points. Let us also notice that the relative temperature coefficient of the relative thermal coefficient can be considerable. Because for an arbitrary physical quantity x we have

$$\frac{\dfrac{\partial}{\partial T}\left(\dfrac{1}{x}\cdot\dfrac{\partial x}{\partial T}\right)}{\dfrac{1}{x}\left(\dfrac{\partial x}{\partial T}\right)} = -\frac{1}{x}\cdot\frac{\partial x}{\partial T} + \frac{\dfrac{\partial^2 x}{\partial T^2}}{\dfrac{\partial x}{\partial T}}. \tag{11}$$

Also for this reason the possible error of determination of coefficient $\frac{1}{x}\left(\frac{\partial x}{\partial t}\right)$ can lead to a not too accurate temperature dependence.

Values of molar volumes V_s^m and V_D^m calculated from formulae (9a) and (9b), and w_D determined according to formula (10) for a group of chosen simple liquids at the temperature of 20°C are given in Table 5. Table 6 presents the V_D/V_s ratio which is a certain measure of space filling. Calculation results presented in Table 5 and 6 prove that very similar w_D values are achieved from formulae (6) and (10), and that the factor of space filling defined by the V_D/V_s ratio rapidly decreases with a temperature increase.

Table 5. Values of V_s^m, V_D^m and w_D for some simple liquids at temperature of 20°C

Type of liquids	$V_s^m \cdot 10^6$ $\left[\frac{\mathrm{m}^3}{\mathrm{mol}}\right]$	$V_D^m \cdot 10^6$ $\left[\frac{\mathrm{m}^3}{\mathrm{mol}}\right]$	$\frac{V_D^m}{V_s^m}$	w_D $\left[\frac{\mathrm{m}}{\mathrm{s}}\right]$	w_D/w Eq. (10)	w_D/w Eq. (6)
n-heksane	17.40	113.26	6.51	1618	1.471	1.459
n-heptane	16.60	129.99	7.83	1601	1.389	1.372
n-oktane	15.35	147.33	9.60	1570	1.316	1.308
n-nonane	14.03	164.69	11.74	1554	1.258	1.253
benzene	7.22	81.65	11.31	1670	1.258	1.240
toluene	9.16	97.27	10.62	1197	1.277	1.262
carbon tetrachloride	7.91	88.57	11.20	1170	1.247	1.230
chloroform	9.91	70.27	7.03	1393	1.390	1.378
methyl ethyl ketone	8.59	66.08	7.69	1652	1.356	1.348

Table 6. Values of V_D/V_s ratio in terms of temperature for some simple liquids

Type of liquids	t[°C]						
	0	10	20	30	40	50	60
n-heksane	7.95	7.19	6.51	5.90	5.36	4.87	4.43
n-heptane	9.04	8.42	7.83	7.32	6.73	6.21	5.72
n-oktane	11.16	10.35	9.60	8.89	8.22	7.89	6.99
n-nonane	14.59	13.28	11.74	10.34	9.51	8.51	7.69
benzene	—	12.74	11.31	10.09	9.05	8.14	7.34
toluene	13.01	11.74	10.62	9.63	8.75	7.95	7.22
carbon tetrachloride	16.23	12.83	11.20	9.71	8.64	7.89	7.39
chloroform	8.41	7.69	7.04	6.43	5.87	5.34	4.86
methyl ethyl ketone	9.37	8.47	7.67	7.00	6.39	5.84	5.36

3. We can also determine the compressibility of intermolecular space β_s. If we neglect changes of molecule's volume with respect to changes of intermolecular distances during deformation of liquid, then, because $V = V_D + V_s$ and $V_D = \text{const}$, we have

$$\left(\frac{\partial V}{\partial p}\right)_T = \left(\frac{\partial V_s}{\partial p}\right)_T$$

or

$$-\frac{1}{V}\left(\frac{\partial V}{\partial p}\right)_T = -\frac{1}{V}\left(\frac{\partial V_s}{\partial p}\right)_T = -\frac{V_s}{V}\cdot\frac{1}{V_s}\left(\frac{\partial V_s}{\partial p}\right)_T.$$

Hence, we have

$$\beta = \beta_s \cdot \frac{V_s}{V}. \tag{12}$$

Taking advantage of expression (9a) we achieve a formula for intermolecular compressibility

$$\beta_s = \beta\frac{V}{V_s} = -\beta\frac{\dfrac{1}{\eta}\left(\dfrac{\partial \eta}{\partial T}\right)_p}{\dfrac{1}{V}\left(\dfrac{\partial V}{\partial T}\right)_p}, \tag{13}$$

β denotes effective compressibility of liquid determined from acoustic measurements here.

The temperature dependence β_s can be determined through logarithmic differentiation

$$\frac{1}{\beta_s}\left(\frac{\partial \beta_s}{\partial T}\right)_p = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p + \frac{1}{\beta}\left(\frac{\partial \beta}{\partial T}\right)_p - \frac{1}{V_s}\left(\frac{\partial V}{\partial T}\right)_p. \tag{14}$$

Taking into consideration that

$$\beta_s = \frac{V^2}{Mw^2}, \quad \text{so } \beta_s = \frac{V^2}{Mw^2}\frac{1}{V_s}$$

and applying expression (8) we reach

$$\frac{1}{\beta_s}\left(\frac{\partial \beta_s}{\partial T}\right)_p = 2\left[\frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p - \frac{1}{w}\left(\frac{\partial w}{\partial T}\right)_p\right] + \frac{1}{\eta}\left(\frac{\partial \eta}{\partial T}\right)_p. \tag{15}$$

Absolute values of β_s and temperature dependence of β_s for chosen simple liquids are given in Table 7.

Table 7. Values of intermolecular compressibility for some liquids at temperature of 20°C

Type of liquids	$\beta \times 10^{10}$ $\left[\frac{\text{m}^2}{n}\right]$	$\beta_s \times 10^{10}$ $\left[\frac{\text{m}^2}{n}\right]$	$\frac{\beta_s}{\beta}$	$\frac{\partial \beta_s}{\partial T} \times 10^{12}$ $\left[\frac{\text{m}^2}{n \cdot k}\right]$	$\frac{1}{\beta_s}\left(\frac{\partial \beta_s}{\partial T}\right) \times$ $\times 10^4 \left[\frac{1}{k}\right]$
n-pentane	14.98	89.72	5.99	+31.45	+35.06
n-heksane	12.53	94.11	7.51	+7.89	+8.38
n-heptane	11.01	97.20	8.83	−8.96	−9.22
n-oktane	10.00	105.94	10.60	−31.14	−29.39
n-nonane	9.25	117.86	12.74	−60.23	−51.10
benzene	6.46	79.54	12.31	−43.02	−54.09
toluene	5.54	75.99	11.62	−30.36	−39.95
carbon tetrachloride	7.13	86.92	12.20	−49.35	−56.78
chloroform	6.69	53.76	8.04	−3.71	−6.90
methyl ethyl ketone	8.38	72.78	8.69	−11.39	−15.65

Among 20 investigated simple liquids only lighter aliphatic hydrocarbons (*n*-pentane, *n*-hexane) had a positive temperature coefficient of intermolecular compressibility. The compressibility of all other liquids decreases with temperature at constant pressure. To us it seems that this can be explained by the fact that the lightest hydrocarbons have much smaller space filling than higher homologues compare with results obtained by SOCZKIEWICZ [9]. It this case intermolecular compressibility will be mainly of kinetic character, because the actual potential well is much more flat than in a case ,of high degree of space filling, when the compressibility will have potential nature. Such a conclusion finds confirmation also in KRZAK'S research [10]. Yet, it will be possible to draw more detailed conclusions when the temperature and pressure dependence of intermolecular compressibility will be investigated for a much greater number of simple liquids. We plan to continue our research.

We consider investigations of above mentioned dependencies particularily important in understanding thermal motions of liquids molecules and of the method of transmission of an acoustic pulse in liquids. Also, we consider the application of a model of an ideal solid body (sphere-spring type) or perfect gas model to these problems groundless, because we accept that the liquids intermolecular compressibility has kinetic character as well as potential even in such a case when we accept the bottom of the potential of intermolecular interactions is flat.

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