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Characteristic features of the low-frequency region of infrared spectra and a cluster model of the structure of liquids

© G.A. Melnikov¹, N.M. Ignatenko¹, K.N. Boldyrev², O.A. Manzhos¹, A.S. Gromkov¹

¹ Southwest State University, Kursk, Russia

² Institute of Spectroscopy, Russian Academy of Sciences, Troitsk, Moscow, Russia

e-mail: melnikovga@mail.ru, inmkstu@bk.ru, kn.boldyrev@gmail.com

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The results of experimental and theoretical studies of the low-frequency region of infrared spectra and Raman scattering spectra in liquid cyclic hydrocarbons (arenes) and their halogen-substituted ones are analyzed. The results of experimental studies of 13 liquids in the range $20-700 \text{ cm}^{-1}$ using a high-resolution all-wavelength vacuum Fourier spectrometer (Bruker IFS 125 HR) are presented. It is shown that the appearance of spectral bands in the low-frequency region (the region of the "boson" peak) of the infrared spectra of liquids is due to the formation and decay of cluster formations in the structure of the liquid.

Keywords: infrared spectra, hydrocarbons (arenes), spectral bands, cluster model, "boson"peak.

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Introduction

Experimental and theoretical studies in the low frequency area of infra-red (IR) spectra by various authors have shown that in the $20-120 \text{ cm}^{-1}$ area a characteristic peak, called the "boson peak", is observed in glasses, polymers and crystals [1–4].

A number of physical models have been proposed to explain the nature of the bosonic peak. The paper [5] hypothesized the existence of resonance between acoustic vibrations and low frequency excess vibrations, with the excess low frequency vibrations arising from the rotation of coupled molecules (dimers) SiO_2 in the tetrahedron structure.

In papers [6], it is assumed that the bosonic peak arises from localized vibrational modes associated with longlived locally preferred structures, which are inherent to the liquid state and randomly distributed in the normal liquid structure. This model explains the dependence of the boson peak intensity on temperature, pressure and fragility and shows a close correlation between the boson peak and the first sharp diffraction peak. It is noted that long-lived crystalline clusters can form in the equilibrium liquid.

In [7] the role of local field, anharmonicity, and degree of disorder in boson peak formation has been studied. The Raman spectrum of small frequency light scattering of a model crystal of polymethylmethacrylate with disorder elements using the atom-atom potentials method taking into account the effect of local fields on the spectrum has been calculated.

The bosonic peak for water [8,9] has been investigated in some detail. In amorphous ice and in supercooled water, the boson peak shape was found to be universal.

In [10], the low-frequency features of phonon spectra of disordered solid solutions in which boson-type excitations

can occur are analyzed. The basic cause of such excitations is assumed to be the additional positive dispersion of sound waves due to disordering. It is shown that anomalies like the Ioffe-Regel crossover and the boson peak can arise not only in disordered systems, but also in ordered crystal structures with a complex lattice. The results obtained in [9,10] suggest that anharmonic oscillations play a significant role in clusters in which "soft" low frequency oscillators are surrounded by high frequency oscillators, with the clusters becoming mechanically unstable.

In [11,12] a structural interpretation of the librational spectrum of water and acetonitrile solutions in water is proposed. It is found that the librational band is asymmetric, with the shape of the high-frequency edge of the band well described by a Gaussian function.

A brief analysis of the publications on the physical nature of the boson peak leads to the conclusion that the properties of clusters in the structure of liquids play a decisive role on the characteristics of the boson peak.

Within the framework of the cluster model of liquid structure, it is shown that the bosonic peak formation in the low-frequency area of the IR spectrum and Raman light scattering is due to librational vibrations of dimers in the cluster structure. The proposed model allowed us to describe the mutual arrangement of the spectral bands in the IR spectrum in the area of the boson peak [13,14].

Objects of study

Transmission spectra were recorded on a Bruker IFS 125 HR high-resolution all-wave vacuum spectrometer. A cuvette with polyethylene windows was used for the measurement. The measurements were taken at room temperature $(20^{\circ}C)$. The cuvette was filled with the liquid

Liquid and chemical formula	$\mu 10^3$, kg/mol	$ ho^{20}$, kg/m ³	$ ho_{\rm crit}$, kg/m ³	$T_{\text{boil}}, \mathbf{K}$	T _{melt} , K	n_{20}^{D}
Benzene C ₆ H ₆	78.11	879.2	304	352.75	278.66	1.5010
Toluene C ₇ H ₈	92.14	866.9	290	382.85	178.14	1.4966
o-Xylene o-C ₈ H ₁₀	106.16	875.1	287	415.95	247.97	1.5023
Ethylbenzene C ₈ H ₁₀	106.16	866.7	290	408.95	178.17	1.4959
Fluorobenzene C ₆ H ₅ F	96.10	1020.4	357	358.15	231.25	1.4662
Chlorobenzene C ₆ H ₅ Cl	112.55	1106.2	365	405.15	227.57	1.5246
Brombenzene C ₆ H ₅ Br	157.01	1494.8	485	429.35	242.33	1.5602
o-Fluorotoluene o-C ₇ H ₇ F	110.13	1004.1	326	386.65	211.15	1.473
<i>m</i> -Fluorotoluene <i>m</i> -C ₇ H ₇ F	110.13	998.6	326	388.15	186.15	1.469
<i>p</i> -Fluorotoluene <i>p</i> -C ₇ H ₇ F	110.13	1000.7	326	389.15	217.15	1.468
o-Chlorotoluene o-C7H7Cl	126.58	1082.5	357	432.15	237.15	1.525
<i>m</i> -Chlorotoluene <i>m</i> -C ₇ H ₇ Cl	126.58	1072.2	343	434.15	225.15	1.522
<i>p</i> -Chlorotoluene <i>p</i> -C ₇ H ₇ Cl	126.58	1069.7	351	435.55	280.65	1.520
2.4-dichlorotoluene C7H7Cl2	161.03	1249.8	124	473.15	259.65	1.546
2.6-dichlorotoluene C7H7Cl2	161.03	1268.6	_	469.15	275.15	1.550

Table 1. Some characteristics of the study objects

Note. The thermodynamic properties of the liquids given are the factory nameplate data for the corresponding chemical reagents, the critical density is calculated according to the Lydersen method.

to be tested, placed in the cuvette compartment where an $\sim 10^{-4}$ Torr vacuum was generated using a turbomolecular pump. The source of long-wave radiation was a low-pressure mercury lamp, a germanium-coated 6-micron lavsan film was used as a beam splitter, and a deep-cooled (4.2 K) helium bolometer with a diamond filter served as an emission detector.

The scanning step was chosen based on the current width of the observed bands and was 2 cm^{-1} for all compounds. The accuracy of the spectrometer's wave number is better than $5 \cdot 10^{-7}$ of the measured wave number.

Studies have been carried out for pure liquid arenes and their halogen containing - cyclic hydrocarbons whose molecular structure is based on a benzene ring, with one or more hydrogen atoms substituted with halogen atoms (F, Cl, Br) in the halogen substituted arenes. The selected class of liquids for research has wide practical applications in chemical technology, the perfume industry and in organic solvent processes. In theoretical terms, arenes and their halogen-containing products are used as model fluids for testing various models. Particularly due to the presence of halogen – hydrogen (H...F, H...Cl, H...Br) intermolecular interactions, the formation of cluster systems whose spectroscopic properties are observed in the far (low frequency) area of the IR spectra is possible in the structure of these liquids. One possible model of intermolecular interaction is considered in [14].

The class of liquids under study can be divided into two groups: the first group — benzene and its halogencontaining ones in which the substitution of one or more hydrogen atoms by halogen atoms leads to a lower crystallization temperature compared to the parent liquid (Table 1); the second group — toluene and its halogencontaining ones in which the substitution of one or more hydrogen atoms by halogen atoms leads to an increased crystallization temperature compared to the parent liquid. The differences in the spectra for the two groups of liquids under study are also revealed. Table 1 shows some of the characteristics of the research subjects.

Results of a pilot study

IR spectra of liquid o-xylene, ethylbenzene and halogensubstituted benzene are shown in Fig. 1.

IR spectra of liquid halogen-substituted toluene are shown in Fig. 2.

The analysis of the IR spectra of liquid arenes and their halogen-substituted products led to the following conclusions.

1. An absorption maximum was detected in the frequency range $20-120 \text{ cm}^{-1}$ for all liquids studied, which can be interpreted as a "boson" peak in the infrared spectra of the liquids. In IC spectra of the studied liquids pronounced spectral bands in the frequency range $20-700 \text{ cm}^{-1}$ are found, with the first low frequency band $(20-120 \text{ cm}^{-1})$ normally interpreted as a bosonic peak in the IC spectra (or Raman spectra) of condensed media.



Figure 1. IR spectra of liquid *o*-xylene, ethylbenzene and halogenated benzene: a - o-xylene; b - ethylbenzene; c - fluorobenzene; <math>d - chlorobenzene; e - bromobenzene.

2. The presence of a boson peak in the low-frequency area of the IR spectrum in condensed media can be explained by the processes of cluster systems formation and disintegration in their structure. In liquids and dense gases with chaotic particle motion, the formation of small clusters containing up to 1000 particles is characteristic, and under dynamic equilibrium conditions the distribution of clusters over the number of particles contained in their composition is established.

The numerical distribution of clusters can be described by a two-parameter Erlang probability density function [13,14]

$$f(Z) = \frac{\lambda^m}{(m-1)!} Z^{m-1} e^{-\lambda Z},\tag{1}$$

which made it possible to calculate the average number of particles in a cluster in a medium by its density and critical

density

$$\bar{Z} = \int_{0}^{\infty} Zf(Z)dZ = \frac{m!}{(m-1)!} \frac{1}{\lambda} = \frac{m}{\lambda} = m\theta = \exp\frac{\rho}{\rho_{\text{crit}}}.$$
(2)

The distribution function (1) allows we to find the frequency distribution function in the IR spectrum of the liquid [14]

$$f(\omega_*) = \frac{\lambda^4}{3} \,\omega_*^7 e^{-\lambda \omega_*^2}.\tag{3}$$

Here, $\omega_* = \omega/\omega_{\min}$ — the frequency applied.

From the spectral width at $1/2f_{max}(\omega_*)$ the lifetime of the most likely cluster at the selected fluid state parameters is estimated

$$\tau = \frac{1}{\Delta\omega_*} = \frac{1}{\omega_*'' - \omega_*'}.$$
(4)



Figure 2. IR spectra of liquid halogen-substituted toluene: a - o-fluorotoluene; b - m-fluorotoluene; c - p-fluorotoluene; d - o-chlorotoluene; e - m-chlorotoluene; f - p-chlorotoluene; g - 2.4-dichlorotoluene; h - 2.6-dichlorotoluene.

The boson peak has a fine structure comprising a series of low-energy spectral lines whose mutual arrangement is related to a series of Fibonacci numbers. Mathematically, the spectral series in the boson peak structure can be described by the simple relation [14,15]

$$\frac{\omega_i}{\omega_{\min}} = \omega_* = \sqrt{F_i}.$$
 (5)

The low-frequency boundary of the bosonic peak ω_{\min} is defined with the Fibonacci number F = 1 in formula (5) and is defined by the expression [14,15]

$$\omega_{\rm min} = \sqrt{\frac{2\Delta H_{\rm dim}}{J_{\rm dim}}},\tag{6}$$

Calculation/experiment	ω , cm ⁻¹ , at F_i									
	1	2	3	5	8	13	21	34	55	89
Benzene										
Calculation (5), (6)	23.5	33.2	40.7	52.5	66.5	84.7	108	138	174	222
Experiment [26]	_	34.0	_	-	_	87	_	131	169	228
Experiment [27]	_	_	_	_	_	_	96	130	198	255
Experiment [28,29]	_	31.4	_	_	61	87	112	128	179	214
Chlorobenzene										
Calculation (5), (6)	_	30	36	47	59	76	96	122	156	198
Experiment [30]	21	33	37	47	56	81	92	118	155	202
Brombenzene										
Calculation (5), (6)	25	35	43	56	71	90	115	146	185	236
Experiment [26]	29	34	_	_	_	90	_	130	198	230
Experiment [28,29]	-	-	_	_	-	89	_	-	182	245
	<i>m</i> -Chlorotoluene									
Calculation (5), (6)	30.4	43	53	68	86	110	139	177	225	287
Experiment [26]	-	43	50	_	_	125	_	228	247	—
	<i>m</i> -Bromotoluene									
Calculation (5), (6)	33.0	47	57	74	93	119	151	192	245	311
Experiment [26]	33	_	-	_	Ι	120	_	_	242	314
Experiment [28,29]	-	_	_	_	Ι	_	171	197	225	306
				o-Cresol						
Calculation (5), (6)	28.3	40	49	63	80	102	130	165	211	269
Experiment [26]	-	40	-	68	85	90	-	-	190	280
Experiment [28,29]		-	-	-	Ι	-	-	—	190	274
m-Cresol										
Calculation (5), (6)	26.3	37	46	59	74	95	121	153	195	248
Experiment [26]	-	_	-	_	Ι	95	_	_	219	249
Experiment [28,29]	-	_	_	_	Ι	_	-	_	214	239
Anisol										
Calculation (5), (6)	26.8	38	46	60	76	97	123	156	199	253
Experiment [26]	_	_	_	60	75	97	126	_	_	226
Experiment [28,29]	-	_	_	_	_	_	_	_	210	264

Table 2. Low-frequency spectra for hydrocarbons and their halogen-containing different structures

where $J_{\rm dim}$ — moment of inertia of the dimer with respect to the principal axes for different dimer configurations in the cluster structure, $\Delta H_{\rm dim}$ — the enthalpy of formation of the dimer configuration. In disordered condensed media (dense gases, liquids) with chaotic motion of particles the processes of self-organization of particles into cluster systems based on the Efimov effect are possible [16-21]. As a result of the

Calculation/experiment	ω , cm ⁻¹ , at F_i										
	1	2	3	5	8	13	21	34	55	89	
o-Xylene											
Calculation (5)	_	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	35	50	58	79	94	127	-	_	255	
Ethylbenzene											
Calculation (5)	_	35	43	56	71	90	115	146	185	236	
Authors' experiment	25	35	42	58	71	87	112	158	_	245	
Fluorobenzene											
Calculation (5)	—	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	39	—	_	_	—	-	158	197	241	
Chlorobenzene											
Calculation (5)	—	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	33	48	_	_	—	125	158	195	258	
				Bromben	zene						
Calculation (5)	—	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	33	_	_	_	94	127	—	-	_	
			_	o-Fluoroto	luene	-					
Calculation (5)	—	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	35	48	56	_	_	125	_	_	_	
			1	<i>m</i> -Fluoroto	oluene						
Calculation (5)	_	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	40	48	-	-	—	125	158	212	245	
				p-Fluoroto	oluene						
Calculation (5)	-	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	40	48	-	-	—	125	158	204	258	
	1	T		o-Chloroto	oluene	ſ		1		1	
Calculation (5)	-	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	40	48	56	_	—	125	166	204	255	
		1	1	<i>n</i> -Chloroto	oluene						
Calculation (5)	-	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	40	48	—	_	—	127	—	—	—	
<i>p</i> -Chlorotoluene											
Calculation (5)		38	47	60	76	97	124	157	200	255	
Authors' experiment	27	40	48	56	—	_	133	—	204	255	
	1	T	2,	4-Dichloro	toluene			1			
Calculation (5)	_	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	42	50	64	-	_	133	—	202	272	
	1	1	2.	6-Dichloro	toluene			1			
Calculation (5)	_	38	47	60	76	97	124	157	200	255	
Authors' experiment	27	42	50	64	79	—	118	166	—	255	

Table 3. Comparisons of low-frequency IR spectra for hydrocarbons and their different structure halogens obtained by the authors, withfrequency calculations by Fibonacci numbers



Figure 3. Pattern of cluster disintegration (F = 8).

interaction of the formed cluster with single particles in the medium, the number of particles in the cluster may become equal to one of the Fibonacci numbers. In this case, the cluster begins to divide according to the "gold" proportion, forming a chain fission before breaking up into free, chaotically moving particles.

Fig. 3 shows a diagram of Fibonacci cluster decay with a number of particles 8, with a quantum of electromagnetic radiation being re-radiated at each stage of decay. These processes explain the appearance of spectral bands in the low-frequency area of the IR spectra of organic liquids.

3. Irrespective of the internal structure and isometry of the liquid molecules, their spectra in the low-frequency area are universal, and the mutual arrangement of the reduced frequencies for each liquid is described by the formula (5) using a set of Fibonacci numbers.

The low-frequency boundary of the boson peak is an individual spectroscopic characteristic of the liquid and is defined by formula (6). Table 2 gives results of calculations of the low frequency limit of the boson peak (these frequencies are in bold) and the frequencies in the boson peak structure with Fibonacci numbers, compared to experimental data for liquids of different molecular structures.

Table 3 compares the low-frequency IR spectra for hydrocarbons and their halogen-containing compounds of different structures obtained by the authors with the calculated frequencies from the Fibonacci numbers. Studies of the properties of systems of several particles in the framework of quantum scattering theory have given a system of equations of the Faddeev [16–18] type, the fundamental solution of which was done by Efimov for the three-particle system by using Jacobi [19–21] relations for the particle coordinates.

In hyperspherical coordinates, the Schrödinger equation is written as [19–21]

$$\left(-\frac{\partial^2}{\partial R^2} + V_n(R) - k^2\right)\sqrt{R} F_n(R) = 0, \qquad (7)$$

$$V_n(R) = \frac{s_n^2 - 1/4}{R^2},$$
 (8)

where $V_n(R)$ — hyper radial Efimov potential, R — hyper radius.

The solutions of equation (7) with potential (8) are valid, but calculations have shown that the solution becomes imaginary at the value of the parameter $s_n = s_0 \approx \pm 1.00624i$. In this case, the potential (8) becomes repulsive and is interpreted as a centrifugal barrier to the free motion of the three-body system, while resulting in an effective attraction of the three bodies [21].

The presence of "gold"in the cluster structure suggests a certain law of interaction between particles [22]. Let's use the power potential of G. Mie [23,24]

$$p(r_{ij}) = \frac{\varepsilon_0}{n-m} \left(\frac{n^n}{m^m}\right)^{\frac{1}{n-m}} \left[\left(\frac{\sigma}{r_{ij}}\right)^n - \left(\frac{\sigma}{r_{ij}}\right)^m \right], \quad (9)$$

$$C_{ij} = \frac{n}{(n-m)} \left(\frac{n}{m}\right)^{\frac{m}{n-m}}, \quad \sigma_0 = \left(\frac{m}{n}\right)^{\frac{1}{n-m}} R_1.$$
(10)

At m = 2 and n = 3, the potential (9) takes the form

$$\varphi(r_{ij}) = \frac{9}{4} \varepsilon_0 \left[\left(\frac{\sigma_0}{r_{ij}} \right)^3 - \left(\frac{\sigma_0}{r_{ij}} \right)^2 \right].$$
(11)

The peculiarity of the potentials (8) and (11) is that in the three-particle Efimov system the arising weak, "cost-grqq interaction is accompanied by the appearance of a stronger interaction proportional to R^{-2} . In this case, a hierarchy of interactions arises in a multi-particle system, where weak interactions lead to stronger ones [25]. This is the reason for the formation of a bound state of three particles with the successive formation of a disc-shaped cluster system [31].

Conclusions

Experimental studies of the IR spectra of liquid hydrocarbons (arenes) and their halogenated substituents have shown that in the low frequency area of the spectra $(20-120 \text{ cm}^{-1})$ a transmittance band is observed which in crystals, glasses and polymers is called the "boson peak". On the small frequency side the transmittance band is limited to a frequency limit (low frequency boson peak limit — $20-30 \text{ cm}^{-1}$), which is an unambiguous characteristic of the fluid.

The proposed cluster model of liquid structure predicts a bosonic peak in the low-frequency area of IR and Raman spectra in condensed matter, whose presence is explained by the processes of formation and decay of cluster systems in the structure of matter. In liquids and dense gases are characterized by the formation of small clusters containing up to 1000 particles, and dynamic equilibrium conditions establish the distribution of clusters according to the number of particles contained in their composition. Mathematically, the distribution of clusters by numerical composition can be described by a two-parameter Erlang function.

The low-frequency boundary of the boson peak within the proposed model is determined by the characteristics of dimer formation (energy of formation of a particular dimer configuration ΔH_{dim} , moment of inertia of the configuration J_{dim}) in the cluster structure according to the relation (6).

Calculations of the low-frequency boson peak limit showed that their values lie within the range $15-40 \text{ cm}^{-1}$, depending on the composition and structure of the condensed media. For simple organic liquids and their solutions the low frequency limit of the boson peak is $20-30 \text{ cm}^{-1}$.

The mutual arrangement of the spectral bands with the given most probable frequencies in the boson peak area is a universal function for all liquids with different molecular structures and atomic compositions of the molecules and can be described with an error of 1-2% by a number of Fibonacci numbers using the formula (5).

Conflict of interest

The authors declare that they have no conflict of interest.

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