Study of the acoustic properties of liquids at high pressures

Umbar Esanov

Scientific – Research Institute of
Natural Gas, State Concern "Turkmengas"
744036 Turkmenistan, Ashgabat city, Archabil avenue -58.

Abstract

This paper presents the results of experimental work with liquid benzene derivatives using the Mandelstam-Brillouin spectroscopy method. It shows the results of measuring the hypersound velocity and absorption in the pressure range 0.1–100 MPa and temperatures 293K-313K in the investigated liquids. With increasing pressure, an increase in hypersound velocity and a decrease in its absorption are found.

Introduction

The study of the effect of thermodynamic conditions on molecular processes has applicational and theoretical significance for the development of the physics of the liquid state of substances. One of the little-studied processes is the effect of high external pressure on the physical mechanisms of intermolecular interactions of liquid substances. These studies contribute to the development of promising areas of molecular acoustics, such as non-equilibrium ultrafast processes with relaxation time in the high-frequency hypersonic range.

Measurement methods

The method of Mandelstam-Brillouin (MB) spectroscopy was used for the experimental work. This method is based on the registration of optical spectra obtained in the process of light diffusion by thermal phonons in liquids [1–6].
A monochromatic beam of light froma helium-neon laser with a wavelength of 6328 angstroms was used as a light source. Located in high-pressure chambers, diffused light spectra were recorded at an angle of $\theta=90°$. To calculate the velocity and absorption, formula - 1 was used.

$$V = \frac{f \cdot \lambda}{2n \cdot \sin(\theta/2)}; \qquad \alpha = \frac{\pi \cdot C \cdot (\delta v)}{v} \qquad (1)$$

where, v, α, f, are the velocity, absorption and frequency of hypersonic waves. λ is the wavelength of laser light, C is the velocity of light, θ is the angle of light diffusion, (δv) is the width (MB) of the spectrum, n is the light refractive index.

It is known that (MB) diffusion occurs due to the light diffraction by acoustic waves (phonons) caused by the thermal motion of molecules. The length of these waves varies from a few centimeters to values comparable to interatomic distances. Typically, the frequency of such sound waves is in the range of $10^4 \, Hz - 10^{12} \, Hz$.

Under thermal excitation, the phonons' motion systems correspond to a plane monochromatic sound wave. Since phonons move at the sound speed, the spectrum of light diffused due to adiabatic fluctuations has two symmetrically located components, i.e., the wavelength of diffracted light must experience a Doppler frequency shift relative to the incident light –(fig 1) [1–5].
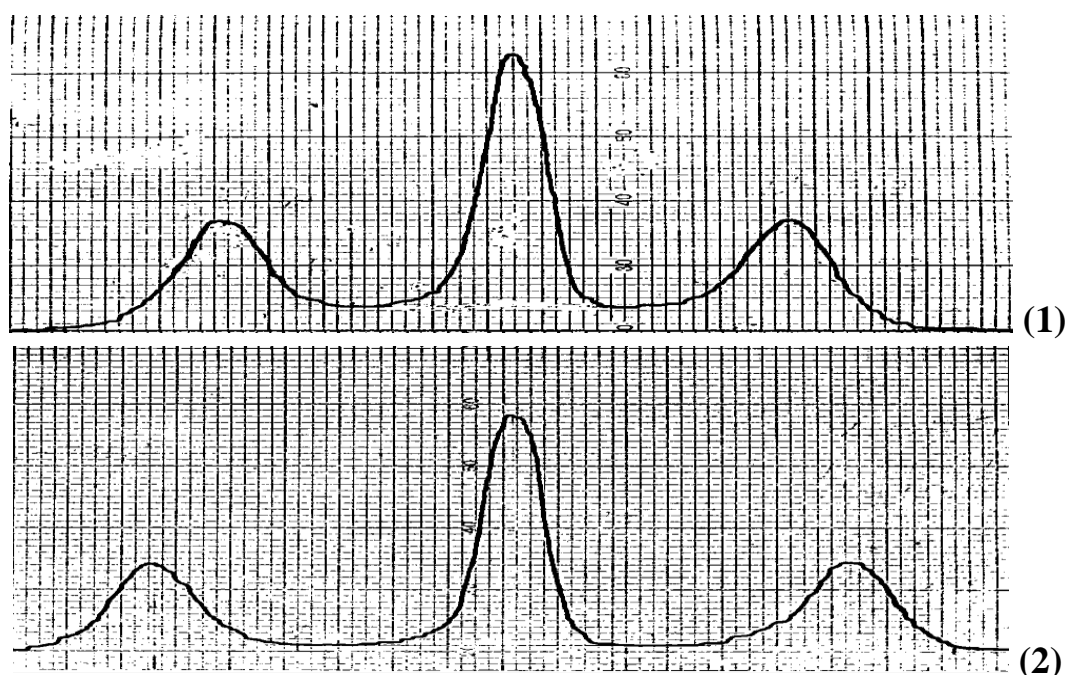

Figure 1. MB spectra of light diffusion in fluorobenzene at temperature T=303K and pressures P=0.1MPa -(1) and P=60MPa -(2).

In the diffused light spectrum in liquids, in addition to components (MB), an unshifted (central) component is observed. The interpretation of this experimental fact was first given by L. Landau and G. Placzek [1– 4]. In contrast to crystals, there is also an isobaric density fluctuation in liquidsin addition to the adiabatic fluctuation. The rates of these two types of fluctuations are different. The adiabatic density fluctuation is due to the propagation of thermoelastic Debye waves and is associated with pressure fluctuations. A very fast rate of this type of fluctuation leads to a change in the wavelength of the diffused light, resulting in the observation of (MB) side components. Isobaric density fluctuations caused by entropy fluctuations do not mix during the time and leadto the appearance of an unshifted (central) line component in the spectrum of diffused light in pure liquids.

Measurement results

Liquid fluorobenzene (C_6H_5F), chlorobenzene (C_6H_5Cl), bromobenzene (C_6H_5Br) and iodobenzene (C_6H_5I) were selected for the study. The experiments were carried out in the pressure range 0.1 MPa – 100 MPa and temperatures 293K– 313K. The results of the experiment are presented in Figure-2 and Table-1.
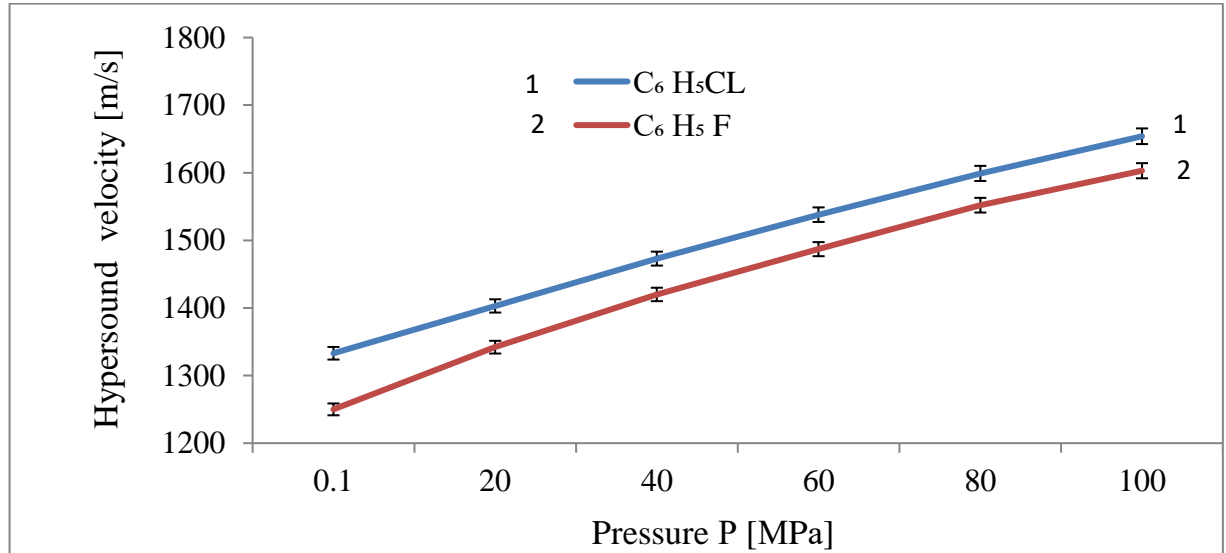

Fig.2. The hypersonic velocity behavior of liquid Chlorobenzene and Fluorobenzene as a function of pressure at temperature T=293K.

Table 1. The hypersonic velocity value of liquid Bromobenzene and Iodobenzene at high pressures

T	P	Bromobenzene		Iodobenzene	
		V	(α/f^2)	V	(α/f^2)
K	MPa	m/s	$10^{-15}\cdot m^{-1}\cdot Hz^{-2}$	m/s	$10^{-15}\cdot m^{-1}\cdot Hz^{-2}$
293	0,1	1194	85	1157	86
	20	1254	79	1200	80
	40	1308	68	1242	73
	60	1358	57	1282	64
	80	1405	48	1319	53
	100	1443	43	1354	48
313	0,1	1127	122	1097	109
	20	1182	99	1144	101
	40	1240	85	1193	96
	60	1295	71	1239	87
	80	1342	60	1286	76
	100	1383	53	1329	65

The discussion of the results

In the experiments performed in chlorobenzene, fluorobenzene, bromobenzene and iodobenzene the baric dependence of the hypersonic velocity is observed. It has been established that the hypersound velocity increases monotonically with increasing pressure. As the temperature rises, a decrease in the hypersonic velocity is observed. A similar process is observed in the dependence of the hypersound absorption coefficient on high pressure. Variations in the hypersonic waves frequency in the pressure range of 0.1 – 100 MPa and a temperature of 293K were 4.4 – 5.54 GHz for fluorobenzene, 4.4 – 5.78 GHz for chlorobenzene, 4.11 – 5.15 GHz for bromobenzene and 4.0 – 5.0 GHz for iodobenzene. There is an increase in the frequency of hypersonic waves with increasing pressure. With increasing pressure, the physical state of the liquid charges toward the state of a solid body, where the loss of sound energy intensity is less than in a liquid. As a result, the absorption of the hypersound decreases with increasing pressure. An increase in the hypersonic velocity with an increase in external pressure is associated with a decrease in the intermolecular distance and the appearance of a more densely packed state of liquid molecules.

Thus, studies of liquid systems under various thermodynamic conditions are promising for solving problems related to the control of intermolecular interaction processes.

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