## L. I. Mandel'shtam and the propagation<sup>1)</sup> of sound in microscopically inhomogeneous media

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In 1936, L. I. Mandel'shtam, together with M. A. Leontovich, extended the concept of Kneser-type relaxation to liquid media, and at the same time introduced the notion of microscopically inhomogeneous acoustical media and of another type of relaxation appearing in such media ("nonlocal relaxation"). These ideas were used (first at the suggestion of L. I. Mandel'shtam) for studying the acoustical behavior of such media as suspensions, emulsions, polycrystals, liquids with gaseous bubbles, etc. At the present time the same picture is used for constructing a theory of the sound propagation in pure high-viscosity liquids (the behavior of which does not fit into the scheme of Kneser-type relaxations) and for clarifying their structure. It has been established that the existing experimental material concerning the propagation of different types of waves in a number of high-viscosity liquids agrees well with the notion of a high-viscosity liquid represented by a microscopically inhomogeneous medium with structure: a disordered liquid with ordered regions ("clusters"). The assumed applicability of the picture of a microscopically inhomogeneous medium to high-viscosity liquids and nonlocal relaxation is likewise supported by observed changes in the values of a number of parameters for such liquids as a result of distillation and the gradual return of these parameters to their initial values.

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The propagation of waves in microscopically inhomogeneous media interested L. I. Mandel'shtam almost from the very beginning of his scientific career. The present paper is dedicated to a single relatively little known idea of L. I. Mandel'shtam in this field.

In the 1930's acoustics was overtaken by an ultrasonic catastrophe: on the one hand, it was observed that in most liquids, in agreement with classical theory, the absorption coefficient of sound increased quadratically with frequency over the entire known range of ultrasonic frequencies (up to  $10^8$  Hz); but, the magnitude of the absorption coefficient greatly exceeded the value calculated according to the classical (Newtonian) viscosity<sup>2)</sup> of a liquid.

On the other hand, in the same liquids, there was observed sharp lines of the Mandel'shtam-Brillouin doublet, predicted by Mandel'shtam as the result of the scattering of light by hypersonic Debye waves  $(10^{10} \text{ Hz})$  in a manner similar to scattering by diffraction grat-ings.

But, with a quadratic increase in the absorption coefficient, calculation shows that high-frequency Debye waves are damped by a factor of hundreds and thousands over a single wavelength, which means that a doublet should not be visible.

In 1936, L. I. Mandel'shtam, in a paper written with M. A. Leontovich,<sup>1</sup> pointed out that this contradiction is easily resolved if it is assumed that the anomalously large value of the absorption coefficient of ultrasound is caused by a second viscosity that, however, has a relaxation character, similar to Kneser-type phenomena in polvatomic gases (delayed exchange of energy between the translational and internal degrees of freedom in a molecule) as noted earlier by M. A. Leontovich.<sup>2</sup> The first absorption coefficient, even though it continues to increase according to a quadratic law, is small even at high frequencies: as for the relaxing second viscosity, a quadratic increase in the absorption coefficient is retained only in the low-frequency range, while as the characteristic relaxation frequency is approached, it rapidly decreases as a result of the fact that the internal molecular degrees of freedom are "frozen." If it is assumed that this frequency lies somewhere in the range of  $10^8-10^9$  Hz, then the apparent inconsistency between the acoustic and optical data is eliminated. The subsequent development of acoustics and optics graphically confirmed this assumption.

(The expression "ultrasonic catastrophe" is used by analogy with the term "ultraviolet catastrophe": in both cases rescue comes from the fact that not all degrees of freedom are excited.)

But, in the same work, it is further mentioned that besides Kneser-type phenomena, a second relaxation mechanism is possible. In particular, the following is stated:

"Such a generalization of the Kneser theory can be carried out. This will be done elsewhere. Here we shall note only that the coefficient of the absorption of sound in the liquid is given by an expression similar

<sup>&</sup>lt;sup>1)</sup>Paper presented at the joint scientific session of the Division of General Physics and Astronomy and the Division of Nuclear Physics of the Academy of Sciences USSR, dedicated to the centennial of the birth of the Academician L. I. Mandel'shtam.

<sup>&</sup>lt;sup>2)</sup>Translator's note. The author uses the terminology "Newtonian viscosity" here and elsewhere in the original Russian text; however, the terminology "classical viscosity" appears in the American literature in the same context so that the latter usage is applied in the translated text.

to Eq. (2)," i.e., the formula of the Kneser theory presented in the paper under certain assumptions concerning the existence of two components in the liquid. Note that the expression used is "similar to", and not "the same as."

The generalization mentioned here (generalization of the Kneser theory liquids to) was given by Mandel'shtam in his next work with Leontovich.<sup>3</sup> This work laid the foundation for theoretical treatments of the anomalous absorption and dispersion of the velocity of sound, as well as the appearance of fine structure in the scattering of light in pure liquids.

The second relaxation mechanism, which is the main subject of the present paper, began its path in acoustics much more modestly: Mandel'shtam himself did not write anything more about this mechanism; and, if we had at our disposal only the few lines quoted from the 1936 paper, we might not have been able to guess what he had in mind. However, in the same year, he proposed to S. M. Rytov and his thesis students, V. V. Vladimirskii and M. D. Galanin, a study of a suspension of heavy particles in a liquid as a model of a liquid with a second relaxation mechanism. The calculation carried out by them gave an anomalous absorption at low frequencies, and, characteristic for relaxation processes, a slower growth rate in the absorption coefficient at higher frequencies. The students measured the absorption of ultrasound in an emulsion of mercury in water and obtained good agreement between calculations and experiment.4-6

Mandel'shtam's model introduces a new object into acoustics, a microscopically inhomogeneous medium (a medium with small inhomogeneities separated by distances that are small in comparison to the wavelength and sufficiently uniformly distributed so that the medium can be considered macroscopically uniform over regions that, although still rather small in comparison with the wavelength, are large enough to contain many such inhomogeneities). A microscopically inhomogeneous medium behaves acoustically as a uniform medium, but its acoustic and overall, macroscopic properties are sometimes affected by the properties of the components in an unexpected manner and, in general, cannot be described in terms of an average of the properties of these components. For example, the density appearing in the expression for the speed of sound cannot be determined by weighing a container filled with the medium. In this sense, inertial and gravitational masses are not equal in microscopically inhomogeneous media and many properties of such media turn out to be dependent.

Mandel'shtam's first acquaintance with the propagation of waves in microscopically inhomogeneous media dated further back than 1936. His first work in this field, On Optically Uniform and Turbid Media,<sup>7</sup> was his dissertation for the right to teach as privat-docent at the University of Strasbourg (1907); it is true that it concerned light waves rather than acoustic waves and examined scattering rather than absorption, but physicists' interests in those times were still focused on media. They had been concerned with this subject for a

long time and, from our modern view point, treated it rather naively. Thus, John Herschel, in his Essay on Sound cites an article published in the Transactions of the Royal Society of London for 1708 to 1709,<sup>8</sup> in which Derham found that fog, rain, and especially falling snow, strongly impeded the propagation of sound in the atmosphere. In England, this article was taken on faith and for more than 150 years was responsible for increased attention to light signaling and doubts about the reliability of sirens and other sound signals in fog. The inability of sound to penetrate fog "is verified by daily experience," some British authors wrote as recently as 100 years ago, referring, in particular, to the fact that at the time of the famous London fogs the knocking of carriages could not be heard as clearly as usual (I think that the coachmen fearing collisions, simply held back the horses,...). They said that the inability of fog to allow sound to pass through it is caused by reflection and loss of the oscillatory motion on countless boundaries between air and water drops (this opinion, in particular, was even shared by John Herschel), in other words, by the scattering of sound on drops. Even Osborne Reynolds proceeded from the general point of view on the unfavorable role of fog, although he correctly assumed that the friction between the air and drops must play a larger role than scattering.

The problem was solved only by Tyndall,<sup>9</sup> the scientific advisor to Trinity House. Together with "Senior Fellows" of this organization, he carried out actual experiments in the sea and became convinced that there is no impenetrability through fog and rain and that it is in fact through fog that audibility is best, since air is then uniform throughout its entire bulk, while with a clear sky and bright sunlight, the audibility at great distances is sharply decreased with sound being reflected backwards to the source from large masses of unevenly heated and moving air.

And yet, almost 200 years after Derham, the propagation of waves in microscopically inhomogeneous medium was still described incorrectly, in this case for light waves: Planck asserted that a light wave must decay as a result of scattering on the molecules of the medium, even in a macroscopically strictly uniform ("optically uniform") medium. In essence, this was a repetition of the error of those who attemped to explain Derham's assertion theoretically, never offered explanations never suspecting it to be mistaken. In his dissertation, Mandel'shtam showed that there is no such decay, but a debate with Planck then arose; which required more papers by Mandel'shtam to be resolved.

Apparently, this problem is really a difficult one, but for Mandel'shtam it was completely clear.

Let us return to the problems concerning the absorption and dispersion of the speed of sound in a microscopically inhomogeneous medium. Mandel'shtam's model is not the only known medium with a second relaxation mechanism: other examples are polycrystals, liquids with bubbles, emulsions, the components of which have the same densities but different thermodynamic characteristics, etc.

On examining these models, one becomes convinced that the first and second relaxation mechanisms are different in principle. In the" real" (Kneser) mechanism the relaxation processes evolve locally in each element of the medium, whether it is the exchange of energy between degrees of freedom in each individual molecule of the gas or the change in the parameter, characterizing, together with the density and the temperature, the state of the liquid at a given point, as in the theory of Mandel'shtam and Leontovich. In the second mechanism the processes are nonlocal and consist in the exchange of some property between different elements of the medium as a result of diffusion. Thus, with the passage of a wave in a suspension, the heavy particles, lagging behind the medium, create viscous waves in the latter, waves that emanate from the boundaries of the particles and tend to equalize the speeds of the different components. This is in fact a nonlocal process (exchange of momentum between different particles of the medium). It is precisely the delay in the exchange relative to the sound wave that leads to the additional absorption and dispersion of the speed of sound.

When Academician N. N. Andreev noted that sour cream must absorb sound strongly, it became clear that even though there is no effect due to the difference in the densities (the components of this emulsion-water and fat drops-have almost the same density), the interplay of forces is due to the difference in the adiabatic heating of the components on compression and rarefraction: while the fat droplets are strongly heated and cooled on compression and rarefaction, the temperature of the water hardly changes at all. The temperature jumps forming at the boundaries of the drops are smoothed out by temperature waves, propagating from the boundary into the water and into the drop. Such a redistribution in the temperature, by way of a heat exchange between different parts of the medium, is also a nonlocal process, which likewise obeys the diffusion equation and leads again to anomalous absorption and dispersion.<sup>10</sup>

In this way, even though the behavior of a microscopically inhomogeneous medium to a certain degree imitates the "real" relaxation process, in essence the two are completely different. This is expressed formally by the fact that a "real" relaxation process is described by ordinary differential equations, while the nonlocal character of the second type of relaxation requires partial differential equations. As a result, with all the qualitative similarities between the effect of local and nonlocal relaxation, the quantitative relationships turn



FIG. 1. The dashed lines refer to "real" relaxation, while the continuous lines refer to nonlocal relaxation.

out to be completely different (it may be recalled that Mandel'shtam in fact spoke only about a similarity between the absorption laws). This difference can be seen in Fig. 1, which shows the behavior of the dispersion in the speed of sound (a) and the absorption over a single wavelength (b) with the same dispersive jump  $v_{\infty}/v_0$ = 1.4 for both types of relaxation mechanisms. The dimensionless quantity  $\omega \tau$  ( $\omega$  is the frequency,  $\tau$  is the relaxation time) is chosen as the argument. The difference between "Kneser" and "non-Kneser" liquids is especially large for large values of  $\omega \tau$ : for "real" relaxations, the deviation of the speed of sound from its highfrequency asymptotic value and the absorption over a single wavelength decrease rapidly as  $1/(\omega \tau)^2$  and  $1/\omega \tau$ , respectively, while for a two-component medium with a nonlocal relaxation mechanism both these quantities decrease more slowly as  $1/\sqrt{\omega\tau}$ . For this reason, in particular, the dispersive range is much wider for nonlocal relaxation than for Kneser-type relaxation.

It is curious that the properties of the nonlocal relaxation mechanism in microscopically inhomogeneous media were understood by acousticians only with great difficulty. Thus, in 1938 Zener, studying absorption in polycrystals, overlooked heat exchange between neighboring crystallites and obtained an erroneous frequency dependence for the absorption of sound (this result was even cited in Mechanics of Continuous Media, 1944). British acousticians also failed to take local heat exchange into account; they studied in great detail the increased attenuation in emulsions with components of the same density, devising clever but incorrect explanations for the observed anomalous absorption. As soon as the heat exchange due to thermal waves between components of an emulsion was taken into account the disagreement between theory and experiment was resolved.11

With time, everyone became accustomed to the fact that the only noteworthy thing about microscopically inhomogeneous media, in addition to their intrinsic interest, was that they gave a graphic model for relaxation of a pure liquid, and that the components in such a model should be different substances. While the microscopically inhomogeneous medium was valued as a model of a "real" relaxing liquid, the "real" relaxation theory reigned supreme in the theory of pure liquids. However, changes appeared soon.

It became clear that a large class of liquids does not fit in any way within the framework of a theory of the Kneser type. Such liquids include high-viscosity liquids, glycerin, triacetin, butanedial, and many other organic liquids (as well as some inorganic liquids, such as molten glass). These are supercooled liquids, the viscosity of which depends strongly on temperature: it is sufficient to cool them by several tens of degrees below room temperature for their viscosity to increase by 5, 7, or 10 orders of magnitude. In these media, weakly damped and strongly dispersive transverse waves are possible. The dispersion in the speed of sound in these liquids attains 50 percent or more rather than 10% to 15% as in low-viscosity liquids. The dispersive frequency range in these liquids is large, of several orders of magnitude and is the same for sound, transverse, and electromagnetic waves. All this constitutes a completely different behavior from that of low-viscosity liquids, for which the theory of Mandel'shtam and Leontovich was mainly developed. However, the Mandel'shtam-Brillouin doublet is clearly observed even in them and, therefore, they, too, are relaxing liquids.

Many acousticians, even in the face of such large discrepancies, did not want to give up the familiar local theories. Their perseverance was costing them dearly for they had to postulate just two or three relaxation mechanisms, but an entire spectrum of such mechanisms for high-viscosity liquids, and furthermore, to find a separate spectrum for sound waves, a separate spectrum for transverse waves, and a separate spectrum for electromagnetic waves. This manner of fitting theory to experiment did not explain anything and the authors themselves hardly took their constructions seriously.

In the meantime, experimentalists established (mainly, I. G. Mikha'lov<sup>12</sup> for absorption, and I. L. Fabelinskiĭ and coworkers<sup>13</sup> for dispersion) that for the most diverse highly viscous liquids the same square root law  $(1/\sqrt{\omega\tau})$  is valid as for microscopically inhomogeneous media with a nonlocal type of relaxation process. The following proposition arose: Could it be that high-viscosity liquids are in reality microscopically inhomogeneous media with their own nonlocal relaxation mechanism? Did not this mean that it was time to stop looking at twocomponent media as just a model and take the above few lines from the Mandel'shtam-Leontovich article in the literal sense? That is, to say, just as they, in all probability, were written?

A purely phenomenological approach was attempted first<sup>14</sup>: it was assumed that a high-viscosity liquid is really something like an emulsion, that a wave disrupts the equilibrium between drops and the surrounding medium according to some parameter, on which the specific volume of the medium depends (similar to temperature in a real emulsion), that the disrupted equilibrium is equalized by way of diffusion of this parameter between components, and that the delay of this equalization in fact leads to anomalous absorption and dispersion of the speed of sound. The theory was constructed in such a way that it did not contain a single arbitrary parameter: all quantities entering into the theory were required to be obtained from independent experiments. The scheme for the theory closely followed the calculations of a real emulsion whose components had different thermodynamic properties. The following data were taken from experiments: (1) the thermal behavior of the dispersion jump, (2) the absorption coefficient for sound at low frequencies (in the range of applicability of the quadratic growth with frequency), and (3) the temperature behavior of a classical viscosity.

Figures 2 to 6 show a comparison of the theory with experiment. Figures 2 to 5 show experimental points along the theoretical curves for the speed of sound and the absorption coefficient for some high-viscosity liquids. The continuous lines are the theoretical curves and the cross marks and circles are the experimental



FIG. 2. Theoretical curves and experimental points for the temperature dependence of the speed of sound v (1) and the absorption coefficient  $\delta$  (2) for butanediol 1, 3 at a frequency of 22.5 MHz.<sup>15</sup>

data. The dashed lines are obtained from experimental data according to asymptotic values of the speed of sound for low  $(v_0)$  and high  $(v_{\infty})$  frequencies. For butanediol and glycerin, the emulsion nonlocal theory was made more precise by taking relaxing shear stresses into account. Figure 6 compares theory with experiment for the dielectric permeability (for glycerin). The experimental data shown were not obtained by changing the frequency, but rather by varying the relaxation time with a change in the temperature of the liquid. The relaxation time  $\tau$  is proportional to the classical viscosity of the liquid, while the viscosity, as already mentioned, depends strongly on the temperature. This allows passage through the entire dispersive range relative to the parameter  $\omega \tau$  at a fixed frequency by simply heating and cooling the liquid, which greatly simplifies the experiment. Establishing the proportionality of  $\tau$  and the coefficient of viscosity. of course, required experiments at different frequencies. The graphs are constructed in accordance with the actual conduct of the experiments: the quantities are given as a function of temperature, taken along the abscissa. Comparison of the theoretical curves with the experimental points showed that a satisfactory agreement is observed over practically the entire dispersive range. The same kind of agreement occurs for many other liquids, as well as for the frequency dependence of the change of the liquid parameters at a fixed temperature.

After the satisfactory agreement of the phenomenological theory with experiment was established, the following molecular description was proposed for the phen-







FIG. 4. The same for triacetin at a frequency of 3 MHz.<sup>13</sup>

omenological picture observed.<sup>19, 20</sup> It was assumed that a high-viscosity liquid consists of disordered phases and ordered regions, which play the role of emulsion drops. The number and size of the ordered regions at equilibrium depend only on the temperature and the hydrostatic pressure. It was also assumed that the ordered regions have distinct boundaries (which is important for a nonlocal theory), as well as that the regions are small in comparison with the wavelength of light (otherwise the liquid would be turbid). Further, it was assumed that the parameter in the phenomenological theory is the concentration in the components of so-called holes (according to Frenkel) and that in the unperturbed liquid these concentrations have definite equilibrium values which vary with a change in the external conditions: a changed pressure (sound wave), shearing stress (transverse wave), and electric field (electromagnetic wave). External influences destroy the equilibrium and set off a diffusion of holes between components, analogous to heat exchange in an emulsion, striving to establish a new equilibrium state. The delay of this nonlocal process relative to the external influence is what leads to the anomalous absorption and dispersion.

Such a molecular picture automatically describes the experimentally observed coincidence in the dispersive range of waves of different types: sound, transverse, and electromagnetic. Subsequently, it was shown that this picture likewise explains the characteristics of nuclear magnetic resonance in high-viscosity liquids,<sup>21</sup> gives a graphic representation of the glass transition in high-viscosity liquids, and explains the jumps in the heat capacity and other parameters of these liquids observed at transition through the glass transition point.<sup>22</sup>

At the present time a great number of high-viscosity liquids have been investigated and for these liquids the nonlocal theory agrees well with experiment over a wide range of values of the parameter  $\omega\tau$ . Other proposed







FIG. 6. Theoretical curves and experimental points for the temperature dependence of the real  $\varepsilon'$  (1) and imaginary  $\varepsilon''$  (2) parts of the dielectric permeability of glycerin at a frequency of 30 MHz.<sup>17,18</sup>

theories either give an unsuitable frequency dependence for the acoustic characteristics or lead to quantities which are two orders of magnitude smaller than the observed values.

This kind of agreement between theory and experiment for a large number of high-viscosity liquids and different types of waves obviously corroborates the assumption as to the microscopically inhomogeneous structure of high-viscosity liquids. Nevertheless, one would naturally wish to verify by some independent method that a given pure transparent liquid is indeed similar to sour cream. For example, we could try to destroy the ordered regions or, vice versa, to create ordered regions larger than those present at equilibrium; the macroscopic properties of the liquid would have to change as a result, and these changes should be consistent with one another. Experiment would establish whether or not it is actually possible to obtain such changes.

It has been found that it is in fact possible to affect the ordered regions in a high-viscosity liquid. Such experiments were recently carried out at the suggestion of I. A. Chaban in the Physico-Technical Institute of the Academy of Sciences of the Turkoman SSR, where a great deal of attention has been directed in recent years toward the acoustical behavior of high-viscosity liquids.<sup>23</sup> The workers of this institute, B. Khemraev and V. A. Lysenko, used vacuum distillation of the liquid with subsequent condensation on a substrate, kept at some fixed temperature, as means for affecting the ordered regions. If distillation in fact destroys the equilibrium number and dimensions of the ordered regions, then the properties of the liquid must change and return to equilibrium values, and therefore, to the initial properties of the liquid, would take some time and occur gradually. The only unknown was whether or not there would be time for the experimentalist to discover this nonequilibrium composition of the two-component liquid. It should be noted that first nonsystematic indications of changed properties of the liquid occasioned by distillation had been mentioned earlier by Krivokhizha and Fabelinskii,<sup>24</sup> as well as Rank, Kless, and Fink.<sup>25</sup>

B. Khemraev and V. A. Lysenko have measured the acoustical and some other parameters of a distilled liquid immediately after distillation, and then repeated the measurements using the same procedure over the

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course of the next several days. They have not yet published their results but have given me permission to report here on the preliminary findings of their experiments. They have determined the temperature dependence of the speed of sound, the absorption coefficient, and the density for several high-viscosity liquids. They found that these quantities indeed change from one day to another and that after several days the measured quantities attain stable values. It was also observed that with subsequent repeated distillation of the given sample of liquid, the entire cycle repeats itself and the measurements again show the same values. In all cases, the error in the measurements was approximately an order of magnitude smaller than the differences observed between the results recorded immediately after distillation and after attainment of stable values. Thus, the expected effect was obtained experimentally.

The character of the observed changes in the properties of a liquid is consistent with a nonlocal relaxation theory. It is interesting to note that for glycerin under the same distillation conditions, no changes were observed in the properties of the liquid. This may be related with the fact that the viscosity of glycerin is an order of magnitude greater than for the other liquids studied, and therefore, to obtain this effect, condensation will have to be performed on a warmer substrate.

Such are the additional arguments in support of the assumption of a microscopically inhomogeneous structure of high-viscosity liquids.

It seems to me that after all, the fate of the second relaxation mechanism pointed out by L. I. Mandel'shtam turned out to be no less interesting than that of the "real" relaxation mechanism.

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