Ultrasonic absorption in semiquantum liquids

L. L. Buishvili

Institute of Physics, Georgian Academy of Sciences

A. I. Tugushi

Tbilisi State University

(Submitted 1 July 1982)
Zh. Eksp. Teor. Fiz. **84**, 970–974 (March 1983)

The influence of nonlinear saturation effects on ultrasonic absorption processes in semiquantum liquids is investigated, and the relaxation absorption is studied. It is shown that in the case $\omega\tau \gg 1$ (ω is the ultrasonic frequency, τ is the time of quantum delocalization of the liquid particles) the absorption coefficient at both low and high ultrasonic intensities is proportional to ω^2. In the case $\omega\tau \ll 1$, the absorption coefficient is proportional to ω and depends weakly on the intensity of the incident sound wave. The relaxation absorption coefficient at $\omega T_1 \ll 1$ (T_1 is the relaxation time of the elementary excitations) is directly proportional and, at $\omega T_1 \gg 1$, inversely proportional, to ω.

PACS numbers: 67.90. + z, 62.80. + f, 43.35.Bf

As is well known, liquids in the temperature range $T_d < T < \Theta$ (Θ is the Debye temperature, T_d is the quantum energy uncertainty connected with the liquid-particles delocalization due to tunnel transitions) are called semiquantum liquids. The specific properties of the semiquantum liquids (SQL) lead to universal temperature dependences of their thermodynamic quantities and kinetic coefficients.[1–4] In Ref. 3, the ultrasonic absorption coefficient in SQL was obtained under the assumption of weak intensity of the incident wave, so that nonlinear saturation effects were excluded. The purpose of the present work is the study of the effect of nonlinear saturation in the ultrasonic absorption processes, and also the study of the so-called relaxation absorption.[5]

For the calculation of the ultrasonic absorption coefficient, we use a model in which the tunnel transitions of the liquid particles in a nonsynmetric potential well created by the surrounding particles play the role of elementary excitations. These elementary excitations in turn can be regarded as a set of two-level systems (TLS), the splitting energy of which extends from $\varepsilon_{\min} = 0$ to the characteristic energy of interaction of the liquid particles $\varepsilon_{\max} = U$. Here it is most remarkable that the function that characterizes the probability density distribution of the energy $\nu(\varepsilon) = \nu_0 = \mathrm{const}$ does not depend on the energy over the region of interest to us.

This model, which was suggested in Refs. 1–3, is analogous in many ways to the model that gives a good description of the low-temperature properties of glass.[6–8] The difference from the case of glass is that the tunneling probability in liquids is greater; therefore, the smallness of the concentration of the excitations is due to the smallness of the temperature in comparison with the interaction energy U. Thanks to the large tunneling probability, the uniform level width of the TLS in SQL is governed in fact by the tunneling frequency. Moreover, in consideration of saturation questions, the relaxation of the TLS is important. In view of the high mobility of the liquid particles, the relaxation mechanism of the TLS can be due to collisions of the particles with the walls of the container, in contrast with the phonon relaxation mechanism in amorphous solids.

We shall assume that an ultrasonic wave with frequency ω interacts with a single TLS of energy $E = \varepsilon n/2$ (n is the difference in populations between the upper and lower levels, ε is the energy of splitting of the TLS). Moreover, the TLS undergoes a relaxational interaction with a characteristic relaxation time T_1.

The population difference n obeys the following equation

$$\frac{dn}{dt} = -2Wn - \frac{n-n_0}{T_1}, \quad n_0 = -\frac{1}{2}\,\mathrm{th}\,\frac{\varepsilon}{2T}, \tag{1}$$

where n_0 is the equilibrium population difference, W is the probability of transitions between the states of the TLS and is due to interaction of the TLS with ultrasound.

The change in energy of the phonon system (of the ultrasound) under stationary conditions has the form

$$\frac{dE_{\text{ph}}}{dt} = \varepsilon W n_{\text{st}}, \quad n_{\text{st}} = \frac{n_0}{1+2WT_1}$$

or, taking into consideration the fact that $E_{\text{ph}} = \hbar\omega N_{\text{ph}}$ we obtain for the change in the number of phonons per unit time,

$$\frac{dN_{\text{ph}}}{dt} = \frac{\varepsilon}{\hbar\omega}\,\frac{Wn_0}{1+2WT_1}. \tag{2}$$

Equation (2) was obtained under the assumption of the presence of a single TLS with splitting energy ε. To account for the fact that the spectrum of splitting of the TLS in a SQL extends from $\varepsilon = 0$ to $\varepsilon = U$, it is necessary to sum Eq. (2):

$$\frac{\overline{dN_{\text{ph}}}}{dt} = \int_0^U \nu(\varepsilon)\,\frac{dN_{\text{ph}}}{dt}\,d\varepsilon. \tag{3}$$

Taking into account the relation $\nu(\varepsilon) = \nu_0$ and Eq. (1), we obtain

$$\frac{\overline{dN_{\text{ph}}}}{dt} = -\frac{\nu_0}{\hbar\omega}\int_0^U \frac{\varepsilon\,\mathrm{th}\,(\varepsilon/2T)\,W}{1+2WT_1}\,d\varepsilon. \tag{4}$$

The probability W, of transitions between the states of the TLS, has the form[8]

$$W=(B^2N_{\mathrm{ph}}\hbar\omega/\rho c^2\hbar^2)\,\delta(\varepsilon/\hbar-\omega), \tag{5}$$

where ω is the frequency of the ultrasound, ρ is the density of the liquid, c is the speed of sound, N_{ph} is determined by the intensity of the ultrasonic wave incident on the system, and B is a constant of the interaction.

With account of the finite width of the levels of the TLS, which is due to the finiteness of the lifetime τ of the localized excitation ($\hbar/\tau\sim T_d$), and under the assumption that the distribution over the uniform width is Lorentzian, Eq. (5) takes the form

$$\begin{gathered} W=(B^2N_{\mathrm{ph}}\hbar\omega/\rho c^2\hbar^2)\,g(\varepsilon/\hbar-\omega), \\ g(\varepsilon/\hbar-\omega)=(\tau/\pi)\,[1+(\varepsilon/\hbar-\omega)^2\tau^2]^{-1}. \end{gathered} \tag{6}$$

The relaxation of the TLS, as was noted above, can be either via a phonon mechanism or via the mechanism of collision of liquid particles with the walls of the container.

The phonon relaxation time of the TLS is calculated by analogy with what was done in the theory of electron paramagnetic resonance,[5] and has the form

$$\frac{1}{T_{1\mathrm{ph}}}=\frac{\varepsilon^3B^2\,\mathrm{cth}\,(\varepsilon/2T)}{2\pi\rho c^5\hbar^5}+\alpha T^7, \tag{7}$$

where α is a constant expressed in terms of the interaction constant, the density of the material, and the speed of sound.[5] The first term of Eq. (7) is due to single-phonon processes, the second to two-phonon processes. It is obvious that the first term is much more significant for the TLS with high splitting energy, the second for a TLS with small splitting energy. Since the spectrum of the splitting of the levels of the TLS is broad, it is necessary to keep both terms in Eq. (7).

We now calculate the time of relaxation due to collisions with the walls of the container. Taking into account the diffuse character of the particle motion, we can write the following for the relaxation time:

$$T_1^*=L^2\tau/a^2; \tag{8}$$

here L gives the dimensions of the container, τ is the localization time, and a is the interatomic distance.

For the semiquantum liquids He^3 and He^4 we have $\hbar/\tau\sim T_d$, where T_d is determined by the degeneracy temperature: $T_d\sim 3$ K. For hydrogen, which is also an SQL, the time τ can be estimated with account of the connection between the coefficient of thermal conductivity $\varkappa$, the heat capacity and the coefficient of temperature conductivity $\chi{:}\varkappa = C\chi$. According to Refs. 1–3, $\tau\sim\hbar/\langle I\rangle$, where $\langle I\rangle$ is the value of the overlap integral. Estimates show that $\hbar/\tau\sim 10$ K. As is seen from Eq. (8), T_1^* does not depend on the temperature and is basically determined by the dimensions of the container.

The mean relaxation time of the TLS, which is governed by the interaction with phonons, is equal to $T_{1\mathrm{ph}}\sim 10^{-3}$–$10^{-2}$s in order of magnitude. If the dimensions of the container are $L\sim(10^3$–$10^4)a$, then the collision mechanism of relaxation will predominate.

We now analyze the possibility of achievement of saturation conditions in the case of ultrasonic absorption in an SQL. As the authors of Refs. 7 and 8 have noted, the powers usually encountered in experiments on ultrasonic absorption in amorphous bodies, exceed by five orders of magnitude the critical values, i.e., they are such that saturation is achieved. Assuming that the constants of interaction in amorphous bodies and semiquantum liquids are approximately the same, we note that the condition for achievement of saturation in SQL will be principally determined by the homogeneous width of the levels of the TLS, by the relaxation time T_1 and by the speed of sound.

The width of the TLS in amorphous bodies is equal to $\Delta\omega\sim 10^7$ Hz in order of magnitude, and $\Delta\omega\sim 10^{10}$–10^{11} Hz in SQL. The ratio of relaxation times of of the order of unity. The speed of sound in the SQL is three times smaller than in solids. These estimates allow us to conclude that the critical sound intensity in SQL is two–three orders of magnitude greater than the corresponding value for amorphous bodies. The numerical value, for the critical intensity in SQL in the case in which $T_1\sim 10^{-3}$ s, is equal to 10^{-5}–10^{-6} W/cm^2 in order of magnitude, which corresponds to the actually achieved intensities.[8] Thus these very rough estimates indicate the possibility of achieving saturation conditions.

We shall assume that the relaxation time is determined by the collision mechanism and by Eq. (8). Then Eq. (4) yields

$$\begin{gathered} \frac{\overline{dN_{\mathrm{ph}}}}{dt}=-\frac{B^2N_{\mathrm{ph}}\nu_0\,\mathrm{th}\,(\hbar\omega/2T)}{4\pi\rho c^2\tau}\left\{\ln\frac{1+(U/\hbar-\omega)^2\tau^{*2}}{1+\omega^2\tau^{*2}}\right. \\ \left.+2\omega\tau^*(\mathrm{arctg}\,\tau^*(U/\hbar-\omega)+\mathrm{arctg}\,\tau^*\omega)\right\}, \\ \tau^{*2}=\tau^2\left(1+\frac{2B^2N_{\mathrm{ph}}\omega T_1\tau}{\pi\rho c^2\hbar}\right)^{-1}=\tau^2\left(1+\frac{E_{\mathrm{ph}}}{E_{\mathrm{cr}}}\right)^{-1} \end{gathered} \tag{9}$$

($\hbar/\tau^*$ is the width of the levels of the TLS under conditions of saturation).

We now consider the various limiting cases.

1. Let $\omega\tau^*\gg 1$; then

$$\frac{\overline{dN_{\mathrm{ph}}}}{dt}\approx-\frac{B^2N_{\mathrm{ph}}\nu_0}{2\rho c^2}\,\omega\,\mathrm{th}\,\frac{\hbar\omega}{2T}\left(1+\frac{E_{\mathrm{ph}}}{E_{\mathrm{cr}}}\right)^{-1/2}. \tag{10}$$

If the system is far from saturation, i.e., $E_{\mathrm{ph}}\ll E_{\mathrm{cr}}$, then Eq. (10) with account of the inequality $\omega\ll T$ gives

$$\frac{\overline{dN_{\mathrm{ph}}}}{dt}\approx-\frac{\hbar B^2N_{\mathrm{ph}}\nu_0\omega^2}{4\rho c^2T}. \tag{11}$$

The temperature and frequency dependences of Eq. (11) are identical with the result of Ref. 3.

In the case of strong saturation, i.e., $E_{\mathrm{ph}}\gg E_{\mathrm{cr}}$, we have

$$\frac{\overline{dN_{\mathrm{ph}}}}{dt}\approx-\frac{\hbar B^2N_{\mathrm{ph}}\nu_0\omega^2}{4\rho c^2T}\left(\frac{E_{\mathrm{cr}}}{E_{\mathrm{ph}}}\right)^{1/2}. \tag{12}$$

The frequency dependence of the absorption coefficient does not change and has the form

$$\frac{\overline{dN_{\mathrm{ph}}}}{dt}\sim\omega^2. \tag{13}$$

2. Let $\omega\tau^*\ll 1$ and $U\tau^*/\hbar\gg 1$ (this case actually exists in SQL). Equation (9) takes the form

$$\frac{dN_{\mathrm{ph}}}{dt} \approx -\frac{\hbar B^2 N_{\mathrm{ph}} \nu_0 \omega}{4\pi \rho c^2 T} \frac{1}{\tau} \ln \frac{U\tau^*}{\hbar}. \tag{14}$$

If the system is far from saturation, then $\tau^* = \tau$ and the absorption is proportional to ω/T; at strong saturation, an additional logarithmic dependence on the intensity also develops.

We note that when the relaxation of the TLS is determined by phonon interactions, no analytical calculation of integral (4) is possible. However, if we introduce the mean relaxation time, as is done in Ref. 8, the results of the frequency dependence will naturally be the same as in the collision relaxation mechanism.

To be specific, we apply the results that have been obtained to liquid helium. Above the degeneracy temperature of the helium, as is well-known,[1-3] the properties of an SQL should appear. The sound absorption coefficient in helium has been measured in Refs. 9–13, basically at low frequencies $\omega \sim 10$–100 MHz and low ultrasound intensities. As we saw above, the frequency dependence of the absorption coefficient is different, depending on the relation between ω and τ^* and also depending on the ultrasound intensity.

The inequality $\omega\tau^* \gg 1$ for helium is realized in the high-frequency region, where it amounts to $\omega \sim 10^4$–10^5 MHz. In this frequency range, the absorption will be determined by Eq. (11) or Eq. (12), depending on the intensity of the incident sound wave.

At low frequencies ($\omega\tau^* \ll 1$), i.e., for helium, $\omega < 10^4$ MHz, the absorption should be proportional to ω at small ultrasound intensities. At high intensities, i.e., upon achievement of saturation, as is seen from Eq. (14), the absorption coefficient changes insignificantly. In the experiments of Refs. 10–13, there are no data on the frequency dependence; therefore, a comparison with experiment cannot be made. Furthermore, the authors of these papers unfortunately do not give the values of the ultrasound intensities; however, they note that the intensities were specially chosen low enough to avoid nonlinear effects in the absorption.

We now proceed to the relaxation absorption of the ultrasound. In the Hamiltonian of the interaction of the ultrasound with the TLS, along with the nondiagonal terms that produce the transitions between the states of the TLS, there are also diagonal terms, which lead to harmonic modulation of the splitting of the TLS:

$$\varepsilon = \varepsilon_0 + {}^1/_2 A\left(e^{i\omega t} + e^{-i\omega t}\right),$$

where ε_0 is the splitting in the absence of the interaction, and A is a quantity determined by the coupling constant. This interaction leads to absorption of a completely different nature, unconnected with transitions between the levels of the TLS and therefore called relaxational.

For the absorption of energy per unit time, averaged over a single period, we easily obtain the following by making use of Ref. 5:

$$\frac{dE}{dt} = \frac{\omega T_1}{(1+\omega^2 T_1^2)} \frac{A^2}{8T\,\mathrm{ch}^2(\varepsilon/2T)}. \tag{15}$$

Here it is taken into account that the average change in the difference of populations is due only to relaxation processes. Averaging (15) over all possible splittings, we get for the change in the number of phonons per unit time:

$$\frac{\overline{dN_{\mathrm{ph}}}}{dt} = \frac{\nu_0 A^2 \omega T_1}{4\hbar (1+\omega^2 T_1^2)} \mathrm{th} \frac{U}{2T}. \tag{16}$$

If $\omega T_1 \ll 1$, this derivative is proportional to ω while in the opposite limiting case the absorption is inversely proportional to the frequency.

The authors sincerely thank A. F. Andreev for valuable advice and useful discussions, both of which stimulated this research.

[1] A. F. Andreev, Pis'ma Zh. Eksp. Teor. Fiz. **28**, 603 (1978) [JETP Lett. **28**, 556 (1978)].

[2] A. A. Andreev, Usp. Fiz. Nauk **127**, 724 (1979) [Sov. Phys. Uspekhi **22**, 287 (1979)].

[3] A. F. Andreev and Yu. A. Kosevich, Zh. Eksp. Teor. Fiz. **77**, 2518 (1979) [Sov. Phys. JETP **50**, 1218 (1979)].

[4] L. L. Buishvili and A. I. Tugushi, Zh. Eksp. Teor. Fiz. **79**, 1898 (1980) [Sov. Phys. JETP **52**, 958 (1980)].

[5] S. A. Al'tshuler and B. M. Kozyrev, Electron Paramagnetic Resonance, Academic (1964).

[6] W. A. Phillips, J. Low Temp. Phys. **7**, 351 (1972); P. W. Anderson, B. J. Halperin, and C. M. Varma, Phil. Mag. **25**, 1 (1979).

[7] J. Z. Jäckle, Z. Phys. **257**, 272 (1972).

[8] S. Hunklinger and W. Arnold, Physical Acoustics (Ed. by R. N. Thurston and W. P. Mason) **12**, 155 (1976).

[9] B. N. Esel'son, V. N. Grigor'ev, V. G. Ivantsov and E. Ya. Rudavskiĭ, Svoĭstva zhidkogo i tverdogo geliya (Properties of Liquid and Solid Helium) Moscow, Izd. standartov, 1978.

[10] B. N. Esel'son, V. G. Ivantsov, V. A. Koval', E. Ya. Rudavskiĭ and N. A. Serbin, Svoĭstva zhidkogo i tverdogo geliya. Rastvory He^3–He^4 (Properties of Liquid and Solid Helium. He^3–He^4 Solutions). Kiev, Naukova dumka, 1982.

[11] N. E. Dyumin, B. N. Esel'son and E. Ya. Rudavskiĭ, Ukr. Fiz. Zh. **19**, 818 (1974).

[12] J. A. Newell and J. Wilks, Phil. Mag. **4**, 745 (1959).

[13] K. Dransfeld, J. A. Newell and J. Wilks, Proc. Roy. Soc. (London) **243A**, 500 (1958).

Translated by R. T. Beyer