POSSIBILITIES OF ACTIVE NONLINEAR SPECTROSCOPY OF INHOMOGENEOUS CONDENSED MEDIA[1])

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Methods of the nonlinear acoustical spectroscopy of gaseous bubbles in liquids are investigated. At first there are studied the experimental possibilities of acoustical analogies of the nonlinear optical phenomena: the enforced sound scattering and the nonlinear active spectroscopy of bubbles. In addition the method for the observation of the enforced scattering on the individual bubble is proposed and the acousto-electrical and acousto-optical methods of the bubbles spectroscopy are described. The possibility of the using the searched methods for the spectroscopy of other inhomogeneities in a condensed matter is mentioned in conclusion.

ВОЗМОЖНОСТИ НЕЛИНЕЙНОЙ АКТИВНОЙ СПЕКТРОСКОПИИ НЕОДНОРОДНЫХ КОНДЕНСИРОВАННЫХ СРЕД

В работе рассмотрены некоторые методы нелинейной акустической спектроскопии газовых пузырьков в жидкости. Изучены в первую очередь экспериментальные возможности наблюдения акустических аналогов нелинейных явлений: вынужденного комбинационного рассеяния звука и нелинейной активной спектроскопии пузырьков. Предложен также метод наблюдения вынужденного рассеяния на отдельных пузырьках и описаны комбинированные акустоэлектрические и акустооптические методы спектроскопии пузырьков. В заключение отмечена возможность использования обсуждаемых методов для спектроскопии других неоднородностей в конденсированных средах.

I. INTRODUCTION

When speaking about nonlinear acoustic spectroscopy of air bubbles and some other inhomogeneities in condensed media we imply methods used to measure the parameters of air bubbles in the observation of nonlinear effects induced by a high-power ultrasonic field. Only two methods of nonlinear bubble spectroscopy have been virtually studied up to now, namely those involving generation of the second harmonic and the Raman frequencies [1—3]. Then, first observation of an echo-type phenomenon in bubbles have been reported [4, 5]. However, potentials afforded by nonlinear spectroscopy have been hardly revealed conclusively. Below we will consider some variants of nonlinear bubble spectroscopy using analogies to the phenomena of nonlinear optics.

We bear in mind primarily the optical phenomenon of stimulated Raman scattering [6], which arises in the interaction of the pump wave (of frequency ω) with the Stokes component of the field (of frequency ω_s) through low-frequency molecular vibrations (of frequency $\Omega = \omega - \omega_s \ll \omega$). An acoustic analog to this phenomenon is the stimulated Raman-type scattering of sound, in which the pump and the Stokes component interact through bubble vibrations.

Another optical phenomenon having an acoustic analog is active spectroscopy of the Raman scattering. In this case, bubble vibrations are preexcited at its eigenfrequency by two pumps, which facilitates the display of these vibrations due to the irradiation by a third frequency.

In acoustics, however, some kinds of stimulated scattering have no optical analogs. One of them, stimulated scattering by individual bubbles, will be discussed below.

And finally, mention will be made of some possibilities of spectroscopy related to the use of combined a acousto-electrical and acousto-optical methods.

II. STIMULATED RAMAN-TYPE SCATTERING OF SOUND BY AIR BUBBLES

The onset of stimulated Raman-type scattering of sound can be represented as follows. Let an intense pump wave of a frequency ω_p propagate in a medium containing gas bubbles. Being scattered by the thermal fluctuations of bubbles which are close to the resonant frequency Ω, the pump wave gives rise to the Stokes ($\omega_s = \omega_p - \Omega$) and anti-Stokes ($\omega_a = \omega_p + \Omega$) components of the field. The Stokes component, which predominates over the anti-Stokes component, interacts with the pump wave and contributes to the rise of bubble vibrations. Enhanced bubble vibrations, in turn, foster pump scattering and increase the Stokes wave amplitude. The result is generation of the Stokes component. It is this generation that is called stimulated scattering.

Stimulated Raman-type scattering of sound by bubbles is analysed in study [8] using a linear wave equation

$$\frac{\partial^2 p}{\partial x^2} = \frac{1}{c_0^2}\frac{\partial^2 P}{\partial t^2} - \varrho_0 n \frac{\partial^2 V}{\partial t^2} \qquad (1)$$

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and the equation for the gas bubble motion

$$\ddot{v} + \Omega_0^2 v + f\dot{v} - \alpha v^2 - \beta(2\ddot{v}v + \dot{v}^2) = -\varepsilon P \,. \tag{2}$$

Here P is the acoustical pressure, v is the variable portion of the bubble volume, Ω_0 and f are the eigenfrequency and the damping coefficient of bubble vibrations, respectively, n is the bubble content, α, β, and ε are the coefficients whose values are equal to $\alpha = \Omega^2(\gamma+1)/2v_0$, $\beta = 1/6v_0$, $\varepsilon = 4\pi R_0/\varrho_0$ and listed in [7].

Seeking solutions in the form of running waves with frequencies ω_p, ω_s, and $\Omega = \Omega_0$ we can show that the amplitude of the back-scattering wave, P_s, is

$$P_s = P_0 \exp(\mu x) \tag{3}$$

where

$$\mu = \frac{\varrho_0 n\omega_s^2 [\alpha - \beta(\omega_p^2 + \omega_s^2 - \omega_p\omega_s)]^2 \varepsilon^3 |P_p|^2}{2(\Omega^2 - \omega_p^2)^2 (\Omega^2 - \omega_s^2)^2 f\Omega K_2} - \mu_0$$

$$K_2 = \frac{\omega_s^2}{c_0^2} + \frac{\varrho_0 n\omega_s^2 \varepsilon}{\Omega^2 - \omega_s^2} \tag{4}$$

P_p is the pump wave amplitude, μ_0 is the absorption coefficient of the Stokes wave in the absebnce of pumping, R_0 is the equilibrium bubble Radius, ϱ_0 is the density of a liquid, c_0 is the speed of sound in the liquid.

Let us estimate the value of μ for a volume content of bubbles $nV_0 = 10^{-5}$ in water (V_0 is the bubble volume) at an eigenfrequency of the bubble $\Omega/2\pi =$ 20 kHz. We set also that $\omega_p = 1.5\ \Omega_0$ and $\omega_s = 0.5\ \Omega_0$. For the specified values of the parameters we obtain

$$\mu = (0.3\ I_p - 1.5 \times 10^{-3})\ \text{cm}^{-1} \tag{5}$$

where I_p is the pumping intensity (W/cm^2).

According to (5), the Stokes component is amplified when the pump wave intensity exceeds its thershold value $I_p > 5.2 \times 10^{-3}$ W/cm^2. To observe this amplification, a pump wave should be coupled to the cell with a liquid and gas bubbles and a weak wave of the Stokes frequency should be coupled in the counter direction. To observe a fairly stable effect, the bubbles must be of about the same size.

To observe the Stokes component generation from the spontaneous noise level, the gain of μL should run to 25—30. It is possible with a pumping intensity $I_p \simeq 1$ W/cm^2 and a cell length $L \simeq 1$ m. In that case, the amplification will occur at an acute angle to the direction of the pump wave.

III. ACTIVE SPECTROSCOPY OF BUBBLES

Let identical bubbles placed in a liquid be irradiated by two pump waves of frequencies ω_1 and ω_2, so that the frequency difference coincides with the bubble

eigenfrequency Ω_0: $\omega_1 - \omega_2 \simeq \Omega_0$. The stimulated bubble vibrations can be registered with a sounding signal of a frequency ω_3. The sounding signal scattered by vibrating bubbles will generate waves of frequencies $\omega_4 = \omega_3 \pm \Omega_0$. By varying one of the pump frequencies and measuring the amplitude of the scattered wave having the frequency ω_4, we can determine the size distribution of bubbles.

Assume that $dn = N(\Omega_0)\, d\Omega_0$ is the number of bubbles per unit volume, whose sigenfrequencies are within the interval $\Omega_0 < \Omega < \Omega_0 + d\Omega_0$. The analysis carried out in study [8] has revealed that in the collinear propagation of all the four waves the amplitude of the scattering wave p_4 of the frequency ω_4 is proportional to the bubble content, $N(\Omega_0)$, to the product of the pump amplitudes, $P_1 P_2^* P_3$, and to the interaction region length, L:

$$P_4 = -\frac{\varrho_0 c_0 \omega_4 L N(\Omega_0) \pi\varepsilon^2 x_1 x_2 P_1 P_2^* P_3}{4 \prod_{i=1}^{4} (\omega_i^2 - \Omega_0^2)\Omega_0} \tag{6}$$

$x_1 = \alpha - \beta(\omega_1^2 + \omega_2^2 - \omega_1\omega_2)$, $x_2 = \alpha - \beta(\Omega^2 + \omega_3^2 + \Omega\omega_3)$. It is assumed that neither of the frequencies $\omega_{1.2.3.4}$ is in the resonant band of the bubbles.

Estimates made using formula (6) for bubbles in water show that if the frequencies $\omega_{1.2.3.4}$ exceed the eigenfrequency Ω_0 by a factor not more than 5, we can expect a detectable scattered signal $P_4 \simeq 10$ Pa even at rather moderate pumping intensities $I_1 \sim I_2 \sim 0.01$ W/cm^2 and a weak sounding signal $P_3 \simeq 20$ Pa, $I_3 \simeq 10^{-8}$ W/cm^2 (the volume content is taken to be $nV_0 = 10^{-5}$). The above relationship of the intensities $I_{1.2.3}$ can be achieved, say, by means of amplitude-modulated pumping with a modulation frequency $\Omega_0/2$ at the frequency ω_3 which is attenuated at 50—60 db. The lateral components $\omega_1 = \omega_3 + \Omega_0/2$ and $\omega_2 = \omega_3 - \Omega_0/2$ will function as pump waves while the attenuation carrier — as a sounding signal.

The advantage of active bubble spectroscopy is the involvement of solely high frequencies, compared to Ω_0. This makes it possible to eliminate the increased background interference and reverberations typical of low frequencies.

Another advantage is related to the fact that the interaction of waves is determined only by the nonlinear properties of bubbles and it occurs without requiring the satisfaction of the spatial synchronism condition. An extra gain in the signal-to-noise ratio can be insured by synchronous detection. To this end, the frequency differences $\omega_1 - \omega_2 = \Omega$ and $\omega_4 - \omega_3 = \Omega$ should be provided by the same generator with a tunable frequency Ω. Active spectroscopy is also superior to stimulated scattering in the lack of the excitation threshold and in the possibility of studying the size distribution of all the bubbles.

IV. STIMULATED SCATTERING BY INDIVIDUAL BUBBLES

In acoustics, stimulated scattering arises not only from bubble nonlinearity but also from the nonlinear properties of the signal shaping unit. Let a bubble undergo spontaneous vibrations of frequency Ω_0. The incident ultrasonic wave of frequency ω_1, on scattering, is modulated to a frequency $\Omega \simeq \Omega_0$. If the modulated scattered signal is detected and the envelope is subsequently used to modulate the radiated field of the frequency ω_1, the system "radiator-bubble-detector-modulator-radiator" will operate under pulse conditions with a repetition period $T = 2\pi/\Omega$ coincident with the period of bubble eigenvibrations.

The above spectroscopic method has no nonlinear optical analogs.

V. COMBINED ACOUSTO-ELECTRICAL AND ACOUSTO-OPTICAL SPECTROSCOPIS METHODS

An important feature of many nonlinear acoustical phenomena is that they do not permit replacement of an acoustic field by an ac electric field of the same frequency. The electric field ensures a pondermotive force on bubbles and it can interact with the acoustic field through bubble vibrations.

Taking this fact into account, we can replace one-two acoustic vibrations involved in nonlinear spectroscopy by electric vibrations if we put the studied liquid under an electric field of a suitable frequency. For a magnetic liquid we can use, similarly, a variable magnetic field.

To display the resonant vibrations of bubbles in active spectroscopy, use can be also made of an optical method of registration. The main advantage of laser registration of bubble vibrations is that it provides separation of the intense and of the weak registering fields.

VI. CONCLUSIONS

It seems to the authors that the above methods suggest new opportunities of nonlinear acoustic spectroscopy of air bubbles in a liquid. Evidently, these methods can be extended to other inhomogeneities in condensed media either, say, to air bubbles in porous bodies, air cavities in polymers, defects in media, etc.

Further development of these methods may be an important and challenging trend among other approaches to a nondestructive control of condensed media.

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