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Influence of Acoustical Nonlinearities on Stimulated Brillouin Scattering

A stationary theory for stimulated Brillouin scattering in isotropic media is given for cases in which the generation of acoustical second harmonic due to elastic nonlinearities must be taken into account. On the base of measuring the coefficient of quadratic acoustical nonlinearity at hypersound frequencies by investigation of Brillouin scattering is shown.

1. Introduction

In experimental investigations of Stimulated Brillouin Scattering (SBS) the used intensities are often so high that the amplitudes of the participating acoustical waves lie beyond the range of validity for the linear stress-strain-relations, which is true not only for solids but even for fluids and gases. Qualitatively it is well known that the acoustical nonlinearity has a certain influence on the character of the scattered wave and, because of the generation of the acoustical second harmonic, gives a kind of additional acoustical absorption.

The investigation of acoustical nonlinearities is also of interest from the viewpoint of fluid theories, because its results may give some new insight in the structural behaviour of materials. Observations on fluids were made under static conditions and at frequencies limited to ultra sound.

In this paper a stationary theory of SBS for isotropic materials is given for the case in which the acoustical second harmonic is generated due to an acoustical nonlinearity. After a brief consideration of the nonlinear p-ϱ-relation (section II) the system of coupled differential equations is developed (III). An approximative solution by means of an iterative method is found in IV. In section V the way is shown, in which a second process of Brillouin scattering can be used to observe the very weak acoustical second harmonic. Thus, a possibility is given for measuring the coefficient of acoustical nonlinearity by means of SBS investigations. A numerical example gives some insight in the order of magnitude of the effect (VI).

2. Nonlinearity of p-ϱ-relation

Let us consider the relation between pressure and density for isotropic materials, especially for fluids. It is well known, that this relation, taken exactly, is not linear. With density ϱ and specific entropy s assumed as independent variables, and p_0, ϱ_0, s_0 considered as equilibrium values and with

$$p = p_0+\bar{p},$$
$$\varrho = \varrho_0+\bar{\varrho},$$
$$s = s_0+\bar{s},$$

one can be use under static conditions a Taylor-series for the environment of the equilibrium point:

$$p(\varrho, s) = p_0(\varrho_0, s_0)+\left(\frac{\partial p}{\partial \varrho}\right)_{s_0}\bar{\varrho}+\left(\frac{\partial p}{\partial s}\right)_{\varrho_0}\bar{s}+ +\frac{1}{2!}\left(\frac{\partial^2 p}{\partial \varrho^2}\right)_{s_0}\bar{\varrho}^2+\ldots \quad (1)$$

In the past years several papers were concerned with the nonlinear terms of this equation, both from theoretical and experimental points of view [1], [2], [3], [4].

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For adiabatic sound propagation, equation (1) may be simplified to

$$\bar{p}(\bar{\varrho}) = \left(\frac{\partial p}{\partial \varrho}\right)_{s_0} \bar{\varrho} + \frac{1}{2!}\left(\frac{\partial^2 p}{\partial \varrho^2}\right)_s \bar{\varrho}^2 + \ldots$$

$$= a\frac{\bar{\varrho}}{\varrho_0} + b\,\frac{1}{2}\left(\frac{\bar{\varrho}}{\varrho_0}\right)^2 + \ldots, \qquad (2)$$

where

$$\frac{a}{\varrho_0} = \left(\frac{\partial p}{\partial \varrho}\right)_{s_0} = v_0^2.$$

Here v_0 means the velocity of sound in equilibrium state.

It is possible to derive the following equation by thermodynamical considerations [1]:

$$\frac{b}{a} = 2\varrho_0 v_0 \left(\frac{\partial v}{\partial p}\right)_T + 2v_0 T\beta \frac{1}{c_p}\left(\frac{\partial v}{\partial T}\right)_p. \qquad (3)$$

(with β volume expansion coefficient and c_p specific heat at constant pressure). Hence, it follows that the nonlinearity-coefficient b/a can be evaluated by measurements of the pressure- and the temperature-dependences of sound velocity.

But one has to be expect, that the relation between p and ϱ will be noninstantaneous (and probably non-local too). This means that formula (1) is correct only for the static case or for a plane sound wave with a definite frequency and definite wave vector; the nonlinearity coefficients are functions of frequency. Therefore, it will be of interest to find the values of the coefficients of acoustical anharmonicity for several different frequencies. It is quite clear that acoustical anharmonicity will be observable only at very strong sound amplitudes. Within the range of hypersound this can be realized in the process of SBS.

Brewer [5] reported on this fact and its experimental observation as early as about ten years ago. He has observed the generation of acoustical second harmonic in SBS, thus proving the existence of acoustical nonlinearity. Other authors [6], [7], [8] remarked that this generation of SH due to acoustical nonlinearity appears as an additional damping of the sound (ground-) wave in the scattering process. In the following sections a quantitative treatment of the generation of acoustical SH in SBS will be given for the stationary case. This will allow us to determine the nonlinearity coefficient b/a by means of SBS measurements.

3. Stationary theory — the ansatz

At present SBS has been observed mostly in backward-scattering, where the incident laser wave and the generated sound wave propagate forward and the scattered Stokes wave runs in the opposite direction. We identify the laser wave direction with the z-axis. Thus the problem will be spatially one-dimensional. Further we assume that the electromagnetic waves are linearly polarized in x-direction: $\boldsymbol{E} = E\cdot\boldsymbol{x}$. Then the linearized hydrodynamical basic equation (Newton's law with mass conservation) and the Maxwell equation take the form

$$\frac{\partial^2}{\partial t^2}\varrho(z,t) - \frac{\partial^2}{\partial z^2}p(z,t) - \frac{\eta}{\varrho_0}\frac{\partial^2}{\partial z^2}\frac{\partial}{\partial t}\varrho(z,t) = -\frac{\gamma^e}{8\pi}\frac{\partial^2}{\partial z^2}(E^2), \qquad (4)$$

$$\frac{\partial^2}{\partial t^2}E(z,t) - \left(\frac{c}{u}\right)^2 \frac{\partial^2}{\partial t^2}E(z,t) = \frac{\gamma^e}{n^2\varrho_0}\frac{\partial^2}{\partial t^2}(E\varrho) \qquad (5)$$

(γ^e-coupling coefficient). Here, the optical absorption is neglected and only the viscous part of acoustical damping is regarded, represented by an effective viscosity η, which is given from bulk and shear viscosity by

$$\eta = \eta_\beta + \tfrac{4}{3}\eta_{sh}.$$

We look for solutions of the coupled system (2), (4), (5) in the form of plane waves with amplitudes changing slowly in space. For this reason we make an ansatz in the usual form, but taking account of an acoustical second harmonic wave too:

$$\begin{aligned} E(\omega_L) &\equiv E_L = \tfrac{1}{2}\{\bar{E}_L(z)e^{i(\omega_L t - k_L z)} + \text{c.c.}\},\\ E(\omega_s) &\equiv E_s = \tfrac{1}{2}\{\bar{E}_s(z)e^{i(\omega_s t + k_s z)} + \text{c.c.}\},\\ \varrho(\omega_G) &\equiv \varrho_G = \tfrac{1}{2}\{\bar{\varrho}_G(z)e^{i(\omega t - k_G z)} + \text{c.c.}\},\\ \varrho(\omega_H) &\equiv \varrho_H = \tfrac{1}{2}\{\bar{\varrho}_H(z)e^{i(2\omega t - k_H z)} + \text{c.c.}\} \end{aligned} \qquad (6)$$

with

$$\begin{aligned} \omega &= \omega_L - \omega_S,\\ k_G &= k_L + k_s. \end{aligned} \qquad (7)$$

The condition of slowly varying amplitudes means

$$\left|\frac{1}{\bar{E}_L}\frac{d\bar{E}_L}{dz}\right| \ll k_L, \quad \left|\frac{1}{\bar{\varrho}_G}\frac{d\bar{\varrho}_G}{dz}\right| \ll k_G,$$
$$\left|\frac{1}{\bar{E}_s}\frac{d\bar{E}_s}{dz}\right| \ll k_s, \quad \left|\frac{1}{\bar{\varrho}_H}\frac{d\bar{\varrho}_H}{dz}\right| \ll k_H. \tag{8}$$

Substituting (6) into (4) and (5), and neglecting some terms because of (8) we get the system

$$\bar{E}'_L = A\bar{E}_s\bar{\varrho}_G, \tag{9a}$$

$$\bar{E}'_s = B\bar{E}_L\bar{\varrho}^*_G, \tag{9b}$$

$$\bar{\varrho}'_G + C\bar{\varrho}_G = D\bar{E}_L\bar{E}^*_s + E\bar{\varrho}_H\bar{\varrho}^*_G e^{-i\Delta k\cdot z}, \tag{9c}$$

$$\bar{\varrho}'_H + E\bar{\varrho}_H = G\bar{\varrho}^2_G e^{i\Delta k\cdot z} \tag{9d}$$

with

$$\Delta k = k_H - 2k_G.$$

The coefficients in (9) have the following meaning

$$A = -i\frac{\gamma^e k_L}{4n^2\varrho_0} = -i|A|,$$

$$B = i\frac{\gamma^e k_s}{4n^2\varrho_0} = i|B|,$$

$$C = \frac{k_G\omega\eta_G}{2\varrho_0 v_G^2} = |C|,$$

$$D = -i\frac{\gamma^e k_G}{32\pi v_G^2} = -i|D|, \tag{10}$$

$$E = i\frac{b_H}{a_G}\frac{(k_H-k_G)^2}{8\varrho_0 k_G} = i|E|,$$

$$F = \frac{k_H 2\omega n_H}{2\varrho_0 v_H^2} = |F|,$$

$$G = i\frac{b_H}{a_H}\frac{k_G^2}{8\varrho_0 k_H} = i|G|.$$

For the sake of brevity we use the following abbreviations

$$I = \frac{nc}{8\pi}|E|^2,$$

$$g = \frac{8\pi}{nc}\left|\frac{BD}{C}\right| = |g|, \tag{11}$$

$$h = \frac{64\pi^2}{n^2c^2}\left|\frac{EGD^2}{FC^3}\right| = |h|.$$

Both the values of C and F for special substance and the type of solutions, which will be found in next section, justify a further neglecting of the terms $\bar{\varrho}'_G$ and $\bar{\varrho}'_H$ in (9c) and (9d). Thus the problem is reduced to solving a nonlinear system of only two differential equations of first order and two algebraic equations.

4. Iterative solution

In solving the remaining system of equations we begin with a z independent "zeroth" approximation of $\bar{E}_L$ which we call $\bar{E}^0_L = \bar{E}^{0*}_L$. Of course it is identical with the value measurable at the input window of the scattering cell.

Since there is no reasonable assumption of a "zeroth" approximation for $\bar{E}_S$, we make the first step of iteration in the form (the bars are omitted):

$$E^{I\prime}_L = AE^I_s\varrho^I_G,$$

$$E^{I\prime}_s = BE^0_L\varrho^{I*}_G,$$

$$\varrho^I_G = \frac{D}{C}E^0_L E^{I*}_s,$$

$$\varrho^I_H = \frac{G}{F}\varrho^{I2}_G e^{i\Delta k\cdot z},$$

$$E^I_L = \tfrac{1}{2}R^2E^0_L e^{-2gI^0_L z} + HE^0_L, \tag{12}$$

$$E^I_s = Re^{i\varphi}E^0_L e^{-gI^0_L z},$$

$$\varrho^I_G = Re^{-i\varphi}|E^0_L|^2\frac{D}{C}e^{-gI^0_L z},$$

$$\varrho^I_H = R^2e^{-i2\varphi}|E^0_L|^4\frac{DG}{CF}e^{-2gI^0_L z}e^{i\Delta k\cdot z}.$$

From the boundary condition it follows that $H = 1-\frac{1}{2}R^2$. The second constant of integration ($Re^{i\varphi}$) cannot be determined. Its meaning is obviously the amplitude conversion ratio at $z = 0$ in this step of approximation.

The second step of iteration gives

$$E^{II\prime}_s = BE^I_L\varrho^{II*}_G,$$

$$\varrho^{II}_G = \frac{D}{C}E^I_L E^{I*}_s + \frac{E}{C}\varrho^I_H\varrho^{I*}_G e^{-i\Delta k\cdot z},$$

$$\varrho^{II}_H = \frac{G}{F}\varrho^{II2}_G e^{i\Delta k\cdot z},$$

$$E^{II}_s = Re^{i\varphi}E^0_L\left\{\left(1-\frac{1}{2}R^2\right)^2 e^{-gI^0_L z} + \frac{R^2}{3}\left(1-\frac{1}{2}R^2\right)^2(1-hI^{02}_L)e^{-3gI^0_L z} + \frac{R^4}{5}\left(\frac{1}{2}-hI^{02}_L\right)e^{-5gI^0_L z}\right\}^8 + \text{const}.$$

$$\varrho_G^{\mathrm{II}} = R\mathrm{e}^{-i\varphi}|E_L^0|^2 \frac{D}{C}\left\{\left(1-\frac{1}{2}R^2\right)\mathrm{e}^{-gI_L^0 z}+\right.$$

$$\left.+R^2\left(\frac{1}{2}-hI_L^{02}\right)\mathrm{e}^{-3gI_L^0 z}\right\},$$

$$\varrho_H^{\mathrm{II}} = R^2\mathrm{e}^{i(\Delta k\cdot z-2\varphi)}|E_L^0|^4\frac{GD^2}{FC^2}\left\{\left(1-\frac{1}{2}R^2\right)^2\times\right.$$

$$\times\mathrm{e}^{-2gI_L^0 z}+R^2\left(1-\frac{1}{2}R^2\right)(1-2hI_L^{02})\mathrm{e}^{-4gI_L^0 z}+$$

$$\left.+R^4\left(\frac{1}{2}-hI_L^{02}\right)^2\mathrm{e}^{-6gI_L^0 z}\right\}. \quad (13)$$

5. Proposals for experimental observation

The intensity of the acoustic second harmonic will be very weak. As shown by BREWER [5], it may be, however, observed. For this purpose the scattering volume has to be irradiated with the second harmonic of the laser wave. This process may be also considered theoretically. At first we assume no dispersion for both optical and acoustical waves. Then the acoustic second harmonic has exactly the right frequency for phase-matched 180°-Brillouin-scattering of the optical second harmonic of the laser wave. The differential equation for the amplitude of the scattered wave is

$$E'_{2s} = B_2 E_{2L}\varrho_H^* \quad (14)$$

with

$$E(2\omega_s) \equiv E_{2s},$$

$$E(2\omega_L) \equiv E_{2L} = E_{2L}^0 = \mathrm{const},$$

$$B_2 \approx 2B.$$

We take $\varrho_H(z)$ from the second step of the interation process and get for $z=0$

$$\frac{E_{2s}(0)}{E_{2L}} \approx \mathrm{e}^{i2\varphi}\frac{8\pi}{nt}\left|\frac{GD}{FC}\right|R^2\left(1-\frac{1}{2}R^2\right)I_L^0. \quad (15)$$

This relation gives a possibility of determining the coefficient of acoustical nonlinearity by measurement of the scattering intensity in the neighbourhood of the optical second harmonic. Generally, the relative dispersion of optical waves will be different from that for acoustical ones. Then one may correct the phase-mismatch by oblique incidence of the optical second harmonic. But this gives a diminished and undetermined volume of interaction.

An other possibility could be the using of a Brillouin-scattered Stokes wave of the optical second harmonic. In this case, however, the scattering material should be chosen so that phase match be achieved.

6. Numerical example

We consider the situation for toluene. For this fluid we have $\gamma^e = 1.60$ and $b/a = 8.93$ (at low frequencies). The values for the other needed material quantities being known we get for ruby laser frequency

$$|A| \approx |B| = 2.8\ \mathrm{m^2/g},$$

$$C = 1.88\cdot 10^6\ \mathrm{m^{-1}},$$

$$|D| = 0.22\ \mathrm{s^2/m^3},$$

$$|E| = 35\ \mathrm{m^2/g},$$

$$F = 7.52\cdot 10^6\ \mathrm{m^{-1}},$$

$$|G| = 17.5\ \mathrm{m^2/g}.$$

For a 10 cm cell length, a 10^{10} W/cm² laser intensity and conversion ratio of intensity $R^2 = 1/9$, the quotient of scattered and incident intensity of the optical harmonic will be

$$\left|\frac{E_{2s}}{E_{2L}}\right|^2 \approx 2.2\cdot 10^{-8}.$$

This demonstrates that the effect is small but still measurable.

For many other fluids the b/a ratio is similar (≈ 10 for organic fluids, ≈ 5 for water). Therefore, we can expect to get a quantitative knowledge of acoustical anharmonicities at hypersound frequencies in the proposed way.

L'influence des non-linéarités acoustiques sur la dispersion stimulée de Brillouin

On a présenté la théorie stationnaire de la dispersion stimulée de Brillouin dans les milieux isotropes, en sonsidérant les cas où, vu les non-linéarités acoustiques, il faut tenir compte de la deuxième harmonique acoustique. On a montré qu' en partant de l'analyse de la dispersion de Brillouin, il est possible de mesurer, pour les fréquences ultra-sonores, le coefficient de non-linéarité acoustique quadratique.

Влияние акустических нелинейностей на стимулированное рассеяние Бриллюэна

Представлена стационарная теория стимулируемого рассеяния Бриллюэна в изотропной среде для тех случаев, в которых из-за акустической нелинейности нужно принимать во внимание генерацию второй акустической гармонической. Показана возможность измерения коэффициента квадратной акустической нелинейности для сверхзвуковых частот на основе анализа рассеяния Бриллюэна.

References

[1] Beyer R. T. J. Acoust. Soc. Am. **32**, 719 (1960).
[2] Coppens A. B. *et al.*, J. Acoust. Soc. Am. **38**, 797 (1965).
[3] Hawley S. *et al.*, J. Acoust. Soc. Am. **47**, 137 (1970).
[4] Kor S. K., Tandon U. S., Acustica **28**, 129 (1973).
[5] Brewer R. G., Appl. Phys. Letters **6**, 165 (1965).
[6] Polyakova A. L., JETP Pisma **4**, 132 (1966).
[7] Polyakova A. L., JETP Pisma **7**, 76 (1968).
[8] Fabelinsky I. L., Starunov V. S., Report at Conf. Interact. Light Sound, San Diego 1969, Moscow 1970 (Preprint).

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