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Spectrum evolutions of spontaneous and pump-depleted stimulated Brillouin scatterings in liquid media*

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A theoretical model for calculating spontaneous and stimulated Brillouin scattering(SBS) spectra is described. An empirical formula for the Stokes output spectral linewidth, a function of spontaneous Brillouin linewidth and the exponential gain coefficient, is obtained by the calculated data fitting. The formula holds true for two cases involving pump undepletion and depletion. The lineshape change from spontaneous to highly pump-depleted SBS spectra is also investigated. The result shows that for the pump power below the SBS threshold, the Stokes output spectral lineshape evolves from Lorentzian to approximately Gaussian as the pump power increases. For the pump power near or beyond the threshold, the SBS spectrum is in the form of a steady Gaussian profile, and the spectral linewidth comes to a certain value about 7 times narrower than the spontaneous one. The theoretical results are experimentally demonstrated by using several common liquid media.

Keywords: Brillouin scattering, spectrum evolution, linewidth, lineshape
PACC: 4265C, 4265F

1. Introduction

Currently, stimulated Brillouin scattering (SBS) is attracting much attention because it opens up some new areas of applications such as Brillouin lidar system for monitoring ocean,[1,2] amplification of lidar return signal,[3,4] optical limiting for laser protection in high-power laser,[5] optical fibre sensor and slow light in the fibre.[6,7] Brillouin lidar and SBS in optical fibre can present useful information by measuring spontaneous or SBS spectrum. SBS is produced from the spontaneous Brillouin scattering in a medium. The lineshape and the linewidth of the SBS spectrum are related to those of the spontaneous process. The relations between them are of great importance for the application of a lidar system based on SBS and the investigation of new media.

The analytical solution for the spectral density of the Brillouin Stokes wave has been obtained for the case of pump undepletion.[8,9] According to Ref.[9], in the high-gain limit, the Stokes spectral linewidth is given by

$$\Delta v_{\mathrm{B}} = \Gamma_{\mathrm{B}} \left(\frac{\ln 2}{G} \right)^{1/2}, \tag{1}$$

where Γ_{B} is the spontaneous Brillouin linewidth, G is the exponential gain coefficient ($G = gI_{\mathrm{P}}L$, with g being the Brillouin gain coefficient, I_{P} the pump intensity and L the interaction length). The value of Δv_{B} tends to infinity as the value of G reduces. Thereby, expression (1) cannot be used to calculate the linewidth in the low-gain limit. Yeniay[10] derived an expression for the linewidth of the Stokes wave for the two limiting cases by using the spectral density equation in Ref.[8], that is

$$\Delta v_{\mathrm{B}} = \left[\frac{G\,(\Gamma_{\mathrm{B}})^2}{\ln\left(\dfrac{\mathrm{e}^G + 1}{2} \right)} - (\Gamma_{\mathrm{B}})^2 \right]^{1/2}. \tag{2}$$

Because the SBS linewidth is expected to reach zero for very high pump depletion, expressions (1) and (2) are useful only near or below the SBS threshold. In practical application, to achieve a high SBS energy reflectivity, the pump energy is usually tens to hundreds

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of times higher than the SBS threshold. However, in all previous work, to the best of our knowledge, the mathematical relation between the spontaneous and the SBS linewidths for high pump depletion has not been given theoretically. The process of spectrum evolution between them has not been adequately discussed, especially in liquid media.

In this paper, according to the transient SBS coupling wave equations,[9] considering the spectral distribution of Stokes wave of spontaneous Brillouin scattering, we develop a theoretical model for calculating spontaneous and SBS spectra, and obtain the empirical formula $\Delta v_{\rm B}(\Gamma_{\rm B}, G)$. The evolution characteristics from spontaneous to highly pump-depleted SBS spectra are investigated by using some common liquid media theoretically and experimentally.

2. Theory

2.1. Mathematical model

According to the localized nonfluctuating source model given by Zel'dovich *et al*,[11] the origin of SBS is ascribed to spontaneous scattering noise occurring in a thin region located near the rear of the SBS medium. The gain is maximum for Brillouin Stokes-shifted wave in the noise. Hence it will preferentially grow in the competition among other frequency scattering waves. Because the Stokes wave is nonmonochromatic, that is, it has a spectral bandwidth, we can consider it as a linear combination of monochromatic plane-wave components with different frequencies. They interact with the pump at the same time, and obtain different magnifications because of different gains and acoustic fields. As a result, the SBS spectrum is formed.

We assume that the Stokes wave is decomposed into M spectral component, and written as

$$E_{\rm S}(z,t)=\sum_{m=1}^{M}E_{{\rm S}m}{\rm e}^{{\rm i}(k_{{\rm S}m}z-2\pi v_{{\rm S}m}t)}, \tag{3}$$

where $E_{{\rm S}m}$ and $v_{{\rm S}m}$ denote the amplitude and the frequency of the mth component, respectively. For simplicity, we assume that the counterpropagating pump $E_{\rm P}(z,t)$ is a monochromatic beam with a frequency of $v_{\rm P}$. Considering the interaction among a pump field, N Stokes components and M acoustic fields ρ_m $(m=1,2,\ldots,M)$, we have $2M+1$ coupling wave equations as follows:

$$\frac{\partial E_{\rm P}}{\partial z}-\frac{n}{c}\frac{\partial E_{\rm P}}{\partial t}=-\sum_{m=1}^{M}{\rm i}g_{\rm o}\rho_m E_{{\rm S}m}+\frac{\alpha}{2}E_{\rm P}, \tag{4a}$$

$$\begin{aligned}
&\frac{\partial E_{\rm S1}}{\partial z}={\rm i}g_{\rm o}\rho_1^{*}E_{\rm P}-\frac{\alpha}{2}E_{\rm S1}, \qquad (4b)\\
&\cdots\cdots,\\
&\frac{\partial E_{{\rm S}M}}{\partial z}={\rm i}g_{{\rm o}M}\rho_M^{*}E_{\rm P}-\frac{\alpha}{2}E_{{\rm S}M};\\
&\frac{\partial \rho_1}{\partial t}+\frac{1}{2}\Gamma\rho_1={\rm i}g_a E_{\rm P}E_{\rm S1}^{*},\\
&\cdots\cdots,\\
&\frac{\partial \rho_M}{\partial t}+\frac{1}{2}\Gamma\rho_M={\rm i}g_a E_{\rm P}E_{{\rm S}M}^{*}, \qquad (4c)
\end{aligned}$$

where g_{om} and g_{am} are SBS coupling coefficients associated with the Brillouin medium, α is the absorption coefficient, and $\Gamma=1/\tau$ denotes the phonon decay rate, with τ being the phonon lifetime.

The mth solution of Eq.(4c) is

$$\rho_m(z,t)={\rm i}g_{am}\int_0^t E_{\rm P}E_{{\rm S}m}^{*}\exp\left[-\frac{\Gamma}{2}(t-\tau)\right]{\rm d}\tau. \tag{5}$$

Substituting expression (5) into Eqs.(4a) and (4b), taking into account the variation of the beam cross-section σ, and using the transforms $g_{om}g_{am}=\dfrac{g_m\Gamma}{4}\cdot\dfrac{nc}{8\pi}$, and $\dfrac{A_{{\rm P},{\rm S}m}}{\sqrt{\sigma}}=\sqrt{\dfrac{nc}{8\pi}}E_{{\rm P},{\rm S}m}$, we have

$$\left(\frac{\partial}{\partial z}-\frac{n}{c}\frac{\partial}{\partial t}\right)A_{\rm P}=\sum_{i=1}^{N}\frac{g_m\Gamma}{4\sigma}A_{{\rm S}m}\int_0^t A_{{\rm S}m}^{*}A_{\rm P}\exp\left[-\frac{\Gamma}{2}(t-\tau)\right]{\rm d}\tau+\frac{1}{2}\alpha A_{\rm P}, \tag{6a}$$

$$\begin{aligned}
\frac{\partial}{\partial z}A_{\rm S1}=&\frac{g_1\Gamma}{4\sigma}A_{\rm P}\int_0^t A_{\rm P}^{*}A_{\rm S1}\exp\left[-\frac{\Gamma}{2}(t-\tau)\right]{\rm d}\tau-\frac{1}{2}\alpha A_{\rm S1},\\
&\cdots\cdots,
\end{aligned}$$

$$\frac{\partial}{\partial z}A_{\mathrm{S}M} = \frac{g_M \Gamma}{4\sigma} A_{\mathrm{P}} \int_0^t A_{\mathrm{P}}^* A_{\mathrm{S}M} \exp\left[-\frac{\Gamma}{2}(t-\tau)\right] \mathrm{d}\tau - \frac{1}{2}\alpha A_{\mathrm{S}M}, \tag{6b}$$

where $g_m = g\dfrac{1}{1+\dfrac{(\nu_{\mathrm{P}}-\nu_{\mathrm{S}m}-\nu_{\mathrm{B}})^2}{(\Gamma_{\mathrm{B}}/2)^2}}$, with g being the maximum gain coefficient for $\nu_{\mathrm{P}}-\nu_{\mathrm{S}m}=\nu_{\mathrm{B}}$, ν_{B} the Brillouin shift of SBS medium.

By implicit finite difference in time and backward difference in space, Eqs.(6a) and (6b) are discretized. Boundary conditions are $A_{\mathrm{P}}(z=L,t) = A_{\mathrm{P}}(t)$ and $A_{\mathrm{S}m}(z=0,\nu_{\mathrm{S}m}) = A_{\mathrm{S}m}(\nu_{\mathrm{S}m})$. The pump beam $A_{\mathrm{P}}(t)$ is in the form of Gaussian pulse, that is, $A_{\mathrm{P}}(t) = A_{\mathrm{P}0}\exp\left\{-2\ln 2\left[(t-t_0)/t_{\mathrm{P}}\right]^2\right\}$. The spectral shape of the spontaneous Brillouin scattering is Lorentzian with a linewidth of Γ_{B}. The ith Stokes component $A_{\mathrm{S}m}(\nu_{\mathrm{S}m})$ can be expressed as

$$A_{\mathrm{S}m}(\nu_{\mathrm{S}m}) = A_{\mathrm{S}0}\left[\frac{1}{1+\dfrac{(\nu_{\mathrm{S}m}-\nu_{\mathrm{S}0})^2}{(\Gamma_{\mathrm{B}}/2)^2}}\right],$$

where $\nu_{\mathrm{S}0}$ is the central frequency of the Stokes wave. The beam power is $P_{\mathrm{P,S}i} = |A_{\mathrm{P,S}m}|^2$, thus forming a complete mathematical model.

3. The relation between spontaneous and pump-depleted SBS spectrum

Firstly, we numerically simulate the Stokes output spectra in the range from spontaneous to highly pump-depleted SBSs, taking the heavy fluorocarbon liquid FC-72 for example. The following parameters are used in numerical simulation: L=50 cm, α=0 cm^{-1}, the lens focus f=15 cm, the distance between the lens and the cell d=10 cm, the wavelength of the incident pump pulse is 532 nm with an 8ns FWHM and a 0.45mrad divergence angle. The parameters of media are listed in Table 1. The pump beam has a Gaussian intensity profile with different peak powers: 10^{-4}, 10^{-3}, 10^{-2}, 10^{-1}, 1 and 10 MW. Using the above parameters, the SBS threshold power is calculated according to the following formula given in Ref.[12].

$$P_{\mathrm{P}} = \frac{G\lambda}{2g}. \tag{7}$$

At room temperature the threshold for SBS in organic liquids corresponds to G in a range of 20–25.[9] Here, we choose G=25. The SBS threshold of FC-72 is approximately 0.1 MW.

Table 1. The SBS parameters of some media at 532 nm.

	n	Γ_{B}/GHz	g/(cm/GW)	τ/ns
CS_2	1.63	0.199	68	1.59
Acetone	1.36	0.477	15.8	0.67
water	1.32	0.681	3.8	0.47
FC-72	1.25	1.080	6	0.29
HT-70	1.28	1.412	5.7	0.23

Figure 1 shows the Stokes output spectra for various pump powers. As seen in Fig.1, for a pump power (P_{P}) of 10^{-4} MW, the backscattered Stokes output has a Lorentzian spectral profile with a linewidth of 1.08 GHz, which is characteristic of spontaneous Brillouin scattering in FC-72(Brillouin shift of about 2.39 GHz). As the pump power increases, the linewidth of Stokes output spectrum gradually narrows and the lineshape changes from Lorentzian to approximately Gaussian. For pump powers near or higher than the SBS threshold, the SBS spectrum has a steady Gaussian profile. The reason for this evolution is that N spectral components of spontaneous Brillouin Stokes wave are amplified due to different gains in the process of producing SBS. Because the

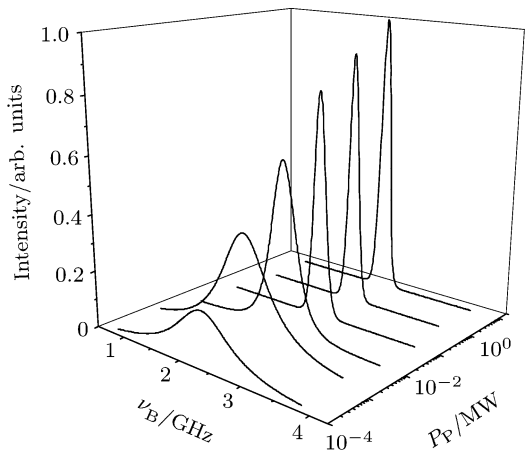


Fig.1. Stokes output spectra versus pump power.

gain curve has a Lorentzian shape,[3] the gain is maximum for spectral components in the proximity of the line centre, which are fully amplified. However, those components in the curve wings have lower gains, hence they cannot be efficiently amplified.

Secondly, to obtain the empirical formula $\Delta\nu_{\rm B}(\Gamma_{\rm B}, G)$, we calculate Stokes output spectral linewidths for different exponential gain coefficients in five types of media (CS_2, Acetone, water, FC-72 and HT-70) as shown in Table 2. Taking the entries for 75 sets of $\Gamma_{\rm B}$, ln$G$ and $\Delta\nu_{\rm B}$, we perform an eighth order polynomial fit[13] to determine a function $\Delta\nu_{\rm B}(\Gamma_{\rm B}, G)$ in the form

$$\Delta\nu_{\rm B} = \Gamma_{\rm B}\sum_{j=0}^{8} t_j\,(\ln G)^j. \tag{8}$$

This equation holds true for the two cases including pump undepletion and depletion. The coefficients t_j are given in Table 3.

Table 2. Data for $\Delta\nu_{\rm B}$(GHz) calculated from the model in this paper.

ln G	$\Gamma_{\rm B}$/GHz				
	0.199	0.477	0.681	1.080	1.412
–3.9121	0.1984	0.4791	0.677	1.0779	1.4099
–2.5253	0.1945	0.4723	0.666	1.0611	1.3983
–1.3862	0.1915	0.4587	0.6475	1.0146	1.3515
–0.6931	0.1834	0.4301	0.6101	0.9456	1.2664
0.0011	0.1622	0.3754	0.5221	0.8175	1.0857
0.6931	0.1254	0.2813	0.3945	0.6358	0.8025
1.6094	0.0856	0.1717	0.2645	0.4024	0.5136
2.4849	0.0547	0.1125	0.1698	0.2587	0.3265
3.2189	0.0391	0.0861	0.1256	0.1789	0.2416
3.9121	0.0328	0.0783	0.1122	0.1564	0.2187
4.8283	0.0306	0.0732	0.1048	0.1542	0.2104
5.5215	0.0295	0.0702	0.1012	0.1532	0.2087
6.2146	0.0287	0.0687	0.0987	0.1521	0.2047
6.9078	0.0283	0.0668	0.0954	0.1516	0.2001
7.8241	0.0281	0.0652	0.0932	0.1496	0.1991

Table 3. Coefficients calculated from expression (8).

t_0=0.756204	t_1=–0.221554	t_2=–3.907176×10^{-2}
t_3=1.605813×10^{-2}	t_4= 2.708147×10^{-3}	t_5=–9.179284×10^{-4}
t_6=–3.824570×10^{-5}	t_7=2.312603×10^{-5}	t_8=–1.435024×10^{-6}

Figure 2 shows Stokes output spectral linewidth of FC-72 versus the pump power(or ln$G$). The solid dots and solid line in Fig.2 denote the linewidths calculated from the above model and the result fitted by expression (8), respectively. The dotted line and dashed line show simulated results by using expressions (1) and (2), respectively. We see that the linewidths from expression (8) are in good agreement with those from expression (2) for pump powers less than the SBS threshold. After the SBS has taken place, the linewidths narrow slowly and reach a constant value in the highly pump-depleted regime, which is contrary to the theoretical prediction given by expressions (1) and (2). This is because after the pump depletion has occurred, most energy of the pump is transferred to the backscattered Stokes wave, thus the SBS reflectivity grows rapidly and tends to saturation, which further restricts SBS linewidth narrowing. As can be seen from Table 2, these constant linewidths for highly pump depletion are about 7 times narrower than spontaneous ones. The spectral lineshapes and linewidths for other media such as water and the perfluoropolyether liquid HT-70 as a function of pump power are similar to those of FC-72.

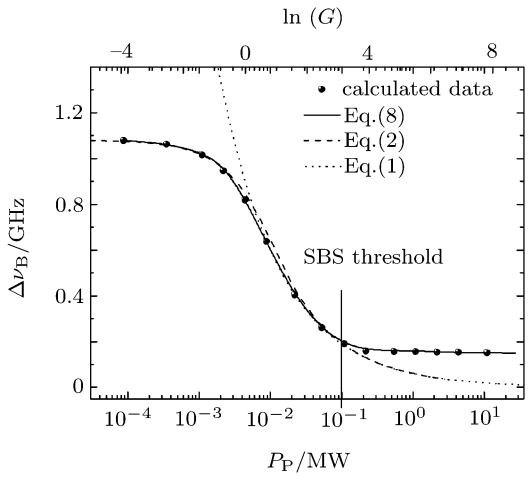


Fig.2. Stokes output spectral linewidths versus pump power.

4. Experiment

Figure 3 shows the experimental set-up. The laser used was an injection-seeded, Q-switched, pulsed Nd:YAG laser (Continuum Powerlite Precision II

9010) with a linewidth of 90MHz and repetition rate of 10Hz. After frequency doubling, the output wavelength was 532 nm, the pulse width was 7–8 ns. The pump energy was continuously varied by adjusting an attenuator, which consisted of a rotatable half-wave plate ($\lambda/2$) and a polarizer (P). The pump beam was focused into a 50cm-long generator cell containing FC-72 by a lens L with a focal length (f) of 15cm. The distance between the lens and the cell was 10cm. The Stokes light produced was attenuated by neutral density filters(ND) and directed into a confocal scanning Fabry-Perot interferometer (Burleigh SA$^{\mathrm{PLUS}}$) with a free spectral range of 2 GHz (the maximum resolution is 6.7 MHz). The interferometer was driven by a ramp generator (Burleigh RG-93) with a longest scanning period of 10 s. This scanning time is so short for the laser with a 10-Hz repetition rate that a high-accuracy spectrum cannot be obtained from the samples. Therefore, we specially designed a ramp generator with an adjustable period. The scanning time was chosen as 500 s. The signal detected by a CR115 photomultiplier tube (PMT) was integrated and averaged over 10 laser shots by a gated integrator and Boxcar averager (Stanford Research System SR-250).

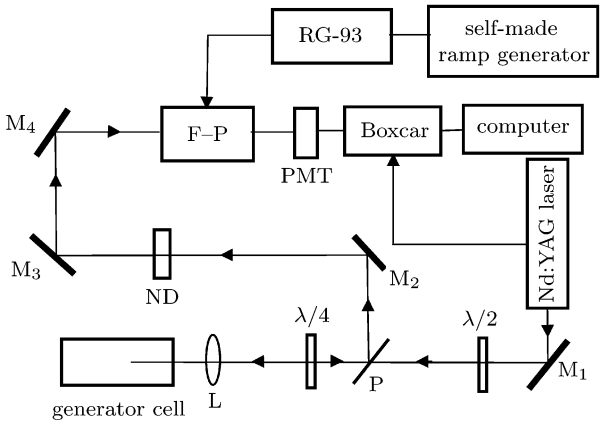


Fig.3. Experimental set-up.

We experimentally studied the SBS spectrum evolutions for various pump energies as shown in Fig.4. These spectra are the averaged results for five groups of measured data under the same condition. The solid lines are the fitted curves by Gaussian functions, indicating that these SBS spectra have Gaussian lineshapes. Fitted results were used for determining the SBS linewidth.[14] For the pump energies of 1 mJ, 5 mJ, 15 mJ, 45 mJ and 68 mJ, their SBS linewidths are determined to be approximately 206 (the energy reflectivity of 15%), 188(66%), 176(78%), 175(80%) and 173 MHz(81%), respectively. As can be seen, under the condition of pump depletion, the linewidth progressively narrows slowly and comes to a steady value in the highly pump-depleted regime. These are well in qualitative agreement with the results predicted by the theory.

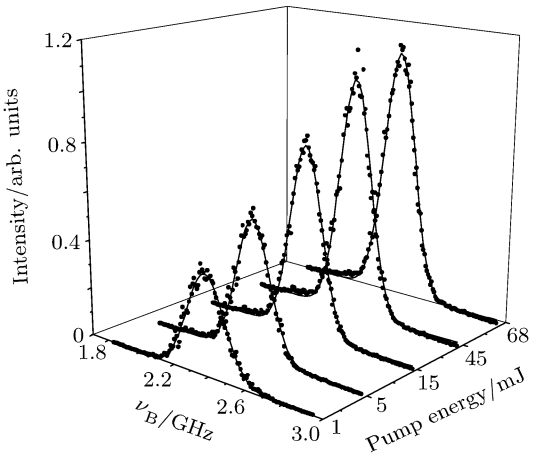


Fig.4. SBS spectra versus pump energy.

However, for pump energies higher than 70mJ, the optical breakdown occurred at the focal point of lens. The corresponding SBS spectrum presented several peaks and was very unstable, and its intensity also decreased compared with the spectra without breakdown as shown in Fig.5. Here the decrease and the instability of SBS energy reflectivity caused by optical breakdown[15,16] may be used to explain the reduced intensity and the spectral fluctuation. In addition, we find that the serious modulation appeared in SBS temporal waveshape after the breakdown occurred. According to Refs.[17, 18], the temporal modulation is due to the relaxation oscillation produced by a fast energy interchange mechanism between the Stokes and pump field via the acoustic field when the pump energy is high enough. This further damaged the SBS laser mode. Therefore, in SBS experiments, the optical breakdown should be avoided as much as possible by using appropriate methods such as medium purification.[16] Because the optical breakdown is not included in the above model, the spectrum shown in Fig.5 cannot be numerically simulated. This is also the drawback of the theoretical model.

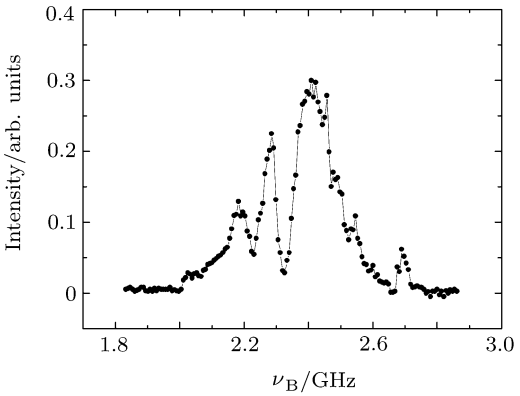


Fig.5. SBS spectrum with optical breakdown.

The spontaneous Brillouin scattering spectrum of FC-72 cannot be measured by using our current equipment. However, for pump powers less than the SBS threshold, the measurement results in fibre[10] can validate the linewidth change in Fig.2. In the experiment, we also measured the SBS spectra of water and HT-70 under pump depletion. Their SBS linewidths are 139 MHz and 226 MHz, respectively. Because of the bandwidths of the pump and the detector, the measured values are slightly larger than the calculated ones.

5. Conclusion

We have developed a model to calculate spontaneous and SBS spectra and numerically investigated the spectral linewidth and lineshape as a function of the pump power. We find that Brillouin Stokes output spectrum transforms from Lorentzian to approximately Gaussian, eventually to a steady Gaussian profile as the pump power increases, and the spectral linewidth narrows and eventually reaches a constant value. We have obtained a rigorous theoretical formula $\Delta\nu_{\rm B}(\Gamma_{\rm B}, G)$, which covers a range from the pump undepletion to high depletion. This is expected to be used for calculating spontaneous Brillouin linewidth of new media or mixture media by measuring SBS linewidth. The theoretical predictions are experimentally demonstrated by using FC-72, water and HT-70 as Brillouin media. Meantime, we also find that the SBS spectrum shows a serious aberration after optical breakdown occurs.

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