Reduction and control of stimulated Brillouin scattering in polymer-coated chalcogenide optical microwires

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We investigate the onset of nonlinear effects in hybrid polymer-chalcogenide optical microwires and show that they provide an enhanced Kerr nonlinearity while simultaneously mitigating stimulated Brillouin scattering as compared to both chalcogenide and silica optical fibers. It is shown in particular that the polymer cladding surrounding the microwire significantly broadens the Brillouin linewidth and increases the threshold, thus enabling Kerr nonlinear applications. We also study the influence of the wire diameter on the Brillouin dynamics and demonstrate that the Brillouin frequency shift can be finely tuned over a wide radio-frequency range.

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One of the goals of modern nonlinear optics is the development of the ultimately fast, highly nonlinear optical fibered device in view of applications to all-optical processing for telecommunication networks, optical sensors, and lasers. Chalcogenide-glass optical fibers and waveguides are promising candidates in this regard as they exhibit both an ultrahigh nonlinear Kerr coefficient and enhanced stimulated Raman scattering [1,2]. In addition to providing broadband and large gain in telecommunication bands, chalcogenide glasses also allow for supercontinuum generation in the mid-infrared wavelength range up to 10 µm [3,4]. Among other nonlinear effects, stimulated Brillouin scattering (SBS) is a fundamental acousto-optical interaction also present in chalcogenide optical waveguides [5–7]. SBS has important implications in various fields ranging from microwave photonics to smart optical fiber sensors [8]. On the other hand, it can also be detrimental to other specific applications operating in the continuous-wave or narrow-linewidth regimes such as all-optical parametric processing and fiber lasers, by severely limiting the transmission of optical power [8,9].

In this Letter, we investigate the SBS in hybrid As2Se3-polymer optical microfibers [10–12] and report both the reduction and control of this nonlinear effect. Specifically, we show that hybrid microfibers exhibit a broadened Brillouin linewidth and a higher threshold compared to standard chalcogenide optical fibers, owing to the damping of acoustic waves in the polymer cladding. We also study the influence of the wire diameter on the Brillouin spectra and demonstrate that the Brillouin frequency can be widely tuned as the wire diameter increases, in good agreement with Brillouin theory and numerical simulations.

A typical As2Se3 optical microwire is shown schematically in Fig. 1(a). They are coated with a polymethyl methacrylate (PMMA) polymer and tapered using the procedure detailed in [11]. The polymer coating adds strength to the otherwise fragile microwire and prevents optical interaction with and damage from the outside environment. The microwires are butt-coupled to single-mode fibers (SMF-28) and permanently bonded with UV epoxy. They all have a 13 cm long uniform length and a total insertion loss between 4 and 6 dB, with 1 dB loss from the Fresnel reflections at the two As2Se3-silica interfaces, ~1 dB propagation loss, and the remaining ~1–2 dB loss from the modal mismatch at the input and output ends.

Figure 1(b) shows the experimental setup used for Brillouin backscattering measurements [13]. As a pump laser, we used a narrow-linewidth continuous-wave distributed-feedback (DFB) fiber laser operating at a vacuum wavelength of 1.55 µm. It was split into two beams using a 90:10 fiber coupler (FC). One beam was injected in the optical microwire through an optical circulator to generate Brillouin scattering, while the other beam served for detection. We then used a standard heterodyne technique in which the backscattered Brillouin signal from the microwire is mixed with the input laser coming from a second fiber coupler. The resulting beat signal was detected using a fast photodiode and the...
averaged Brillouin spectra were recorded using an electrical spectrum analyzer (ESA) and were studied as a function of the microwire diameter.

Figure 2(a) shows the SBS spectrum for three microwires with an increasing diameter of 0.7 μm (blue line), 0.9 μm (green line), and 1 μm (red line), respectively. Figure 2(b) reports the corresponding Brillouin frequency shift versus effective refractive index \( n_{\text{eff}} \) of the fundamental optical mode propagating in the microwire. \( n_{\text{eff}} \) was calculated using a finite element method (FEM) from the As\textsubscript{2}Se\textsubscript{3} refractive index [5]. The weak frequency peak seen around 8.2 GHz in Fig. 2(a) results from SBS in the short untapered sections of microfiber. This peak nearly corresponds to the SBS frequency in the bulk As\textsubscript{2}Se\textsubscript{3} material. It is plotted as a black cross in Fig. 2(b). The solid line in Fig. 2(b) is a plot of the phase-matching condition for Brillouin frequency that reads as \( \nu_B = 2n_{\text{eff}}V_L/\lambda \), where \( \nu_B \) is the frequency of the acoustic phonon, \( V_L \) is the longitudinal acoustic velocity (\( V_L = 2250 \text{ ms}^{-1} \) in As\textsubscript{2}Se\textsubscript{3} glass [5]), and \( \lambda \) is the vacuum optical wavelength [8]. As can be seen, the agreement between the theoretical phase-matching condition and the experimental data is very good. The Brillouin frequency shift is a linear function of the effective refractive index, as expected from the above phase-matching relation. In addition, a direct comparison between the three spectra of Fig. 2 reveals that the Brillouin frequency shift significantly increases from 6.8 to 7.6 GHz as the wire diameter, or equivalently the effective index, increases. This means that the SBS frequency shift can be widely tuned and finely controlled over a few GHz frequency range by the diameter of the microwire, unlike standard optical fibers in which the Brillouin frequency shift is fixed. This large tunability could be useful for applications such as microwave photonics.

We then compared the observed Brillouin spectra to numerical simulations based a recent theoretical model of SBS (for details, see [14,15]). With respect to standard three-wave mixing theory, this new model is mainly based on directly solving the elasto-dynamic equation subject to an electrostriction stress distribution. It provides an excellent estimate of the theoretical Brillouin spectrum by computing the elastic energy of acoustic phonons generated by light through electrostriction. In our simulations, we considered the microwire as a rod-type cylinder and took into account all elastic and optical parameters of As\textsubscript{2}Se\textsubscript{3}. The elastic energy, measured in dB, is plotted in Fig. 3 versus the acoustic frequency and for a wire diameter varying from 0.6 to 1 μm. For further comparison, the experimental Brillouin spectra already shown in Fig. 2(a) are superimposed on Fig. 3. As can be seen, there is a good agreement between numerical simulations and the measured spectra. The slight frequency difference can be interpreted as the result of wire diameter uncertainty that we estimated to about 10%. The numerical simulation results of Fig. 3 also show that, for a given wire diameter, multiple acoustic modes with widely spaced frequencies can be simultaneously excited in the optical microwire, as previously demonstrated in small-core photonic crystal fibers [16,17]. This means that, in such small submicrometer waveguides, the Brillouin dynamics is completely different from standard optical fibers, since it is not only the signature of the bulk longitudinal phonons that leads to a single Brillouin peak. In tiny waveguides, instead, the waveguide boundaries induce a strong coupling of shear and longitudinal phonons, resulting in much richer dynamics of light interaction with hybrid acoustic modes.

**Fig. 2.** (a) Brillouin spectra measured in three chalcogenide-based optical microwires with an increasing diameter of 0.7 μm (blue line), 0.9 μm (green line), and 1 μm (red line), respectively. (b) Brillouin frequency shift versus the effective refractive index (crosses, experiment; solid line, theory).

**Fig. 3.** Numerical simulations. Color plot of elastic energy of acoustic modes generated by electrostriction versus the acoustic frequency and for a wire core varying from 0.6 to 1 μm from top to bottom. The experimental Brillouin spectra are superimposed for the sake of comparison. The black curve shows the phase-matching condition.
with avoided crossings, as those observed in Fig. 3 [15]. This topic undoubtedly deserves more extensive coverage and will be investigated in details in further work.

Now back to Fig. 2 that also shows that the spectral widths of the Brillouin lines are surprisingly broad, reaching up to 110 MHz (FWHM) for the smallest 0.7 μm diameter wire. This is a significantly increased value compared to the 16 MHz linewidth previously measured in As2Se3 optical fibers [6]. Such strong spectral broadening of the Brillouin line cannot be accounted for solely by the diameter longitudinal variations of the microwire nor by the short tapered sections. Our heat-brush drawing technique indeed allows for a precise control of both the wire diameter and uniformity [18]. We did a preliminary measurement on a chalcogenide-air microwire that gave a uniformity error below 10% over the 2 cm long wire and the 10% error was given by the uncertainty of the measurement method (not actual variations in diameter). As well, from the many experiments we made on the microwires, a nonuniformity larger than 10% would have been noticed, e.g., in broadband flat parametric amplification experiments [12]. Instead, this linewidth broadening can be interpreted as the result of larger elastic losses and lower density of polymer relative to chalcogenide glass. The PMMA cladding surrounding the microwire has indeed a natural Brillouin linewidth of 120 MHz [19]. Consequently, the PMMA coating plays the role of an acoustic damper for the longitudinal acoustic waves involved in the SBS process. To verify this assertion, we first measured the SBS linewidth in a 15 μm large microwire in which the polymer has no influence on the acoustic properties. Figure 4(a) shows that the measured Brillouin spectrum in black is only 35 MHz wide in this case for a 8.51 GHz Brillouin frequency, which confirms the capability of the PMMA cladding to broaden and suppress Brillouin scattering in the chalcogenide microwire. Then, we have removed the PMMA cladding from the 0.9 μm diameter microwire by immersing it in an acetone bath. The results are shown in Fig. 4(b). These measurements were performed using a fast high-resolution optical spectrum analyzer (APEX technologies AP2040) with 20 MHz resolution in order to get a fast spectral measurement before the microwire is broken by mechanical stresses. That is why we can not record the same spectrum as in Fig. 2(a). Nevertheless, we can clearly see in Fig. 4(b) the spectral narrowing in red after removing the polymer coating. The central line narrows from about 200 MHz to less than 50 MHz, confirming the key role of the PMMA coating in broadening the SBS linewidth. It does not go back to the original bulk value, but it is very close to that of the large-core microwire shown in Fig. 4(b).

Table 1 compares the linear and nonlinear parameters of standard silica SMF-28, As2Se3 optical fibers, and the three hybrid As2Se3–PMMA microwires [12]. To estimate the SBS limitation, we used the Kerr-to-Brillouin nonlinear figure of merit (FOM) that has recently been introduced by Lee et al. [20]. This nonlinear FOM is written as \( B\text{-FOM} = \gamma \frac{L_{\text{eff}}}{P_{\text{th}}} \), where \( \gamma \) is the Kerr coefficient, \( L_{\text{eff}} \) is the effective length, and \( P_{\text{th}} \) is the SBS power threshold [8]. The power threshold \( P_{\text{th}} \) and the Brillouin gain \( g_B \) have been directly estimated from our measurements of the Brillouin frequency shift and of the Brillouin linewidth using standard SBS theory and the elastic parameters of As2Se3 [5,8]. It is worth mentioning that B-FOM is proportional to \( (n_2/g_B) \) and does not actually depend on the effective length \( L_{\text{eff}} \). As it can deduced from Table 1, chalcogenide-PMMA microwires exhibit higher Kerr-to-Brillouin FOM compared to both As2Se3 and silica fibers. For instance, the smallest 0.7 μm wire diameter microwire has a B-FOM 27 times higher than As2Se3 fibers and 115 times higher than silica fibers. Note also that the Kerr figure of merit (K-FOM), defined as \( \gamma L_{\text{eff}} \), is also very large for the microwires as compared to standard fibers.

In conclusion, we have investigated the relative magnitude of nonlinear effects in subwavelength-diameter As2Se3 microwires and demonstrated a strong increase in the Brillouin threshold as compared to As2Se3 fiber owing to acoustic damping in the PMMA coating. This work thus contributes to the design of a new class of Brillouin-less compact nonlinear fibered devices for nonlinear optical processing applications and widely tunable fiber lasers. We have also shown that the Brillouin frequency shift can be widely tuned simply by changing the microwire diameter, and that such tiny optical waveguides can exhibit hybrid acoustic modes with both shear and longitudinal phonons. Finally, this is also the first observation of backward Brillouin scattering in optical microwires with a few centimeter length of nonlinear propagation.
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References


Table 1. Comparison of Linear and Nonlinear Parameters of SMFs, As2Se3 Fibers, and Hybrid PMMA-As2Se3 Microwires

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>SMF-28</th>
<th>As2Se3 Fiber</th>
<th>Wire1</th>
<th>Wire2</th>
<th>Wire3</th>
</tr>
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<tbody>
<tr>
<td>Core diameter</td>
<td>μm</td>
<td>9.8</td>
<td>6</td>
<td>1</td>
<td>0.9</td>
<td>0.7</td>
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<td>Effective area at 1550 nm</td>
<td>μm²</td>
<td>85</td>
<td>39</td>
<td>0.483</td>
<td>0.404</td>
<td>0.273</td>
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<tr>
<td>Linear loss at 1550 nm</td>
<td>dB m⁻¹</td>
<td>0.0002</td>
<td>0.84</td>
<td>4</td>
<td>5</td>
<td>6</td>
</tr>
<tr>
<td>Length</td>
<td>m</td>
<td>1000</td>
<td>4.92</td>
<td>0.13</td>
<td>0.13</td>
<td>0.13</td>
</tr>
<tr>
<td>Effective length, L_eff</td>
<td>m</td>
<td>977.32</td>
<td>3.17</td>
<td>0.11</td>
<td>0.11</td>
<td>0.11</td>
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<tr>
<td>Effective index, n_eff</td>
<td></td>
<td>1.44</td>
<td>2.81</td>
<td>2.59</td>
<td>2.54</td>
<td>2.37</td>
</tr>
<tr>
<td>Nonlinear index, n2</td>
<td>m² W⁻¹×10⁻²⁰</td>
<td>2.5</td>
<td>2400</td>
<td>2400</td>
<td>2400</td>
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<tr>
<td>Nonlinear coefficient, γ</td>
<td>W⁻¹ m⁻¹</td>
<td>0.001</td>
<td>2</td>
<td>201</td>
<td>241</td>
<td>356</td>
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<td>Brillouin frequency shift, νB</td>
<td>GHz</td>
<td>11.1</td>
<td>7.95</td>
<td>7.53</td>
<td>7.37</td>
<td>6.87</td>
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<tr>
<td>Brillouin linewidth</td>
<td>MHz</td>
<td>27</td>
<td>13.2</td>
<td>64</td>
<td>65</td>
<td>110</td>
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<tr>
<td>Brillouin gain</td>
<td>m W⁻¹×10⁻¹¹</td>
<td>2.71</td>
<td>600</td>
<td>72.6</td>
<td>62</td>
<td>22.4</td>
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<tr>
<td>Brillouin threshold, P_th</td>
<td>dB m</td>
<td>20</td>
<td>18.1</td>
<td>22.3</td>
<td>22.3</td>
<td>25.1</td>
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<tr>
<td>K-FOM (γ·L_eff)</td>
<td>W⁻¹</td>
<td>1.17</td>
<td>7.92</td>
<td>25.97</td>
<td>30.55</td>
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<tr>
<td>B-FOM (γ·L_eff·P_th)</td>
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<td>0.08</td>
<td>0.34</td>
<td>2.81</td>
<td>3.29</td>
<td>9.13</td>
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</table>

*aThe measured values are in bold and those for As2Se3 fibers are taken from [6].