A unified materials approach to mitigating optical nonlinearities in optical fiber. II. B. The optical fiber, material additivity and the nonlinear coefficients

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Abstract
The purpose of this paper, Part IIB in the trilogy [Int J Appl Glass Sci. 2018 (not available); Int J Appl Glass Sci. 2018 (not available); Int J Appl Glass Sci. 2018 (not available)], is to describe the continuum models employed to deduce the effects of differential thermal expansion in the clad optical fiber in addition to the nonlinear optical behaviors at high intensities of light. In this continuation of Part IIA, more of the state-of-the-art in W-S-based continuum material models are reviewed here with specific examples provided based on canonical material systems suggested from the findings of Part I and treated in detail in Part III. Part IIB pays particular attention to the optical fiber and its nonlinearities.

KEYWORDS
lasers, optical fibers, optical glasses, optical properties

INTRODUCTION
The first half of Paper II of the trilogy (Part A) focused on component additivity modeling and the determination of simple first-order material properties of multicomponent glass.\textsuperscript{1} In some instances, these properties can be used in tandem to calculate more complicated nonlinear gain coefficients. For example, the Brillouin gain coefficient is a function of several of the parameters described in Paper IIA. The purpose of the second half of Paper II is to review the state-of-the-art in modeling of these parameters in the optical fiber configuration (as opposed to bulk glass), and how these can then be applied to the reduction in several nonlinear gain coefficients (including the nonlinear refractive index $n_2$). Numerous examples are provided and methodologies to determine nonlinear optical coefficients are presented.

As in the previous work, the materials modeling described herein is not intended to provide specific insights into glass structure or chemistry. Furthermore, for several reasons, the optical fiber geometry itself also must be considered in modeling efforts. Firstly, the optical mode may overlap with regions of the fiber that possess differing compositions. Locally, this means that physical properties, such as the acoustic velocity or refractive index, may be varying across the spatial distribution of the mode. In that sense, the measured physical properties from a fiber are those corresponding to the optical mode rather than to those of a bulk material. Secondly, the cladded fiber geometry can play a significant role when the fiber system is subject to changes in temperature and a core-cladding thermal expansion coefficient (CTE) mismatch is present. The requisite corrections to the bulk
glass calculations (Paper IIA) in the fiber geometry are therefore illustrated in this work. Additionally, the direct measurements of selected properties, eg, fictive temperature, $T_f$, also are described and exemplified.

2 | PROPERTY DEDUCTION AND THE OPTICAL FIBER

2.1 | Fiber design modeling

While the previous work focuses specifically on material property additivity models, such models are guides for the subsequent design and optimization of high performance optical fibers. The performance of the fiber depends on how the electromagnetic modes interact with the materials comprising the core and cladding (clad). Obviously, for a fiber possessing a high numerical aperture, the modes are more tightly confined to the core and the fiber performance will be predicated more on the properties of the core glass. Accordingly, this section briefly discusses approaches for estimating the impact of the optical fiber design on its overall performance when coupled with the additivity models for material properties.

Given material transport facilitated by the high temperature processing of both the preform and the subsequent optical fiber, it is not unusual that the core possesses a graded composition in the radial direction. In such a case, all parameters deduced through measurements of the fiber necessarily are those associated with the waveguide modes, and not exclusively those of the core glass. In the limit of an acoustic or optical wave being tightly confined to the center of the core, the property values associated with the modes will approximate those of the bulk. However, in the weakly guiding limit, modeling of the fiber becomes more complicated because the material parameters vary across the core region, which must be taken into account when solving for mode values that best match the measured values.

Adding to the complexity, conventional fibers possess fairly small core sizes and, consequently, the composition is only known at a few locations across the core. In such cases, the core may be partitioned into sections, corresponding to the available measurements. Each layer possesses a specific composition and, therefore, differing properties. A schematic representation of this is shown in Figure 1 for a 4-layer step-wise approximation to the refractive index profile (RIP) of a fictitious fiber core. Beyond the outermost core layer resides the cladding, which most often is compositionally uniform (eg, pure silica). In this particular example, 4 regions are defined and the modes can be calculated by enforcing the appropriate electromagnetic and acoustic boundary conditions. In short, the boundary conditions are identified at each interface and from them a characteristic matrix for the system is determined. The eigenmodes of this characteristic matrix give the propagation constants for the system (acoustic and optical mode phase velocities, with the effective modal index derived from the latter). As an illustrative example, while the various layers shown in Figure 1 all have unique electromagnetic (EM) velocities, they come together to give rise to a single, observable EM velocity corresponding to that of the EM mode.

2.2 | Determination of optical properties

The refractive index of an optical fiber is the most straightforward property to characterize. Numerous commercial systems exist and this can even be performed as a service; eg, Ref. 4. Given a measurement of the composition (such as via electron probe microanalysis), the refractive index can be plotted as a function of composition. For a binary silicate system, for example, if refractive index and density values for SiO$_2$ are assumed, then the refractive index and density of the bulk “dopant” (eg BaO) can be used as fit parameters when applying Equation 3 from Ref. 1 to the data. Importantly, since each layer of the approximation in Figure 1 can be compositionally unique, the refractive index as a function of temperature may also differ for each layer. However, this enables the calculation of the modal thermo-optic coefficient (TOC), and in the reverse direction, the determination of the TOC of said dopant(s), again utilizing the simple additivity model described in Ref. 1.
2.3 | Determination of acoustic properties

As outlined in other publications, the acoustic properties are determined from measurements of the Brillouin gain spectrum; eg, Ref. 5. In short, a heterodyne process is utilized whereby a pump signal is launched into the fiber and a spontaneously Brillouin back-scattered signal is mixed with a portion of the pump signal to produce a Doppler-shifted gain spectrum. Since the acoustic wave involved in the Brillouin scattering process is Bragg-matched to the optical wave, the peak frequency obeys the relationship $\nu_B = 2 n_m V_m/\lambda$, where $n_m$ and $V_m$ are the optical mode index and acoustic mode velocity (P-wave), respectively, and $\lambda$ is the vacuum optical wavelength. Measurement of $\nu_B$ then allows for the determination of $V_m$, and in a way similar to that of the refractive index and TOC, the acoustic velocity and its dependence on temperature (or strain as described next) for “dopants” where these values are unknown, can be determined.

2.4 | Determination of strain-optic and stress-optic coefficients

The elasto-optic (stress and strain optic) coefficients of bulk glasses can be measured by applying stress to the glass and measuring changes in the refractive index.6-8 When one is utilizing optical fibers, it is convenient to measure the elasto-optic coefficients on the fiber itself using a fiber ring laser-based strain sensor for the determination of the strain optic coefficient9 and/or a twisted-fiber method for $p_{44}$.10,11 Specific details are provided in Refs. 12, 13 with brief generalities provided below.

With respect to the determination of the strain-optic coefficient, $\varepsilon$OC, the fiber is integrated as part of a ring laser cavity which has a free spectral range (FSR) given by $\text{FSR} = c/d n$, where $c$ is the speed of light, $n$ is the mode refractive index and $d$ is the cavity length. If axial strain is applied to the test fiber, both the cavity length and index will change, thus resulting in a change in the FSR. The elongated length $d$ with the application of strain is easily determined and measurements of the changes in FSR as a function of strain reveals the dependence of the modal index on strain, and hence the fiber $\varepsilon$OC.9 Measurement of the thermo-optic coefficient of a fiber is achieved using the same approach, however in this case, the free-spectral range ($\Delta$FSR) of a laser cavity is measured by heating (or cooling) the test fiber under controlled conditions.

With respect to the determination of stress-optic coefficient, $p_{44}$, a mechanical twist is applied to the fiber, which rotates the (linear) polarization state of the light propagating within. Assuming that no linear birefringence is already present or otherwise induced in the twisted fiber, the resultant optical activity, $\alpha$, is given by $\alpha = g \tau$, where $\tau$ is the twist rate (rad/m) and $g$ is,9

$$g = -\frac{n^2}{2} (p_{11} - p_{12}) = -n^2 p_{44}. \quad (1)$$

Here, $n$ is the mode index of the fiber at the wavelength being employed. Based on a measurement of polarization rotation versus the number of twists per meter, $p_{44}$ is easily determined. Knowing $\varepsilon$OC and $p_{44}$ then gives a system of 2 equations and 2 unknowns (with the Poisson’s ratio assumed to follow from simple additivity) whereby $p_{12}$ as seen by the mode can be determined. Once again, methodologies for determining the $\varepsilon$OC values of dopants involving the segmented core of Figure 1 may be utilized. Measurements of $p_{44}$ using twist requires that the fiber be free of birefringence. Should this condition not be satisfied, an alternative method for the determination of $p_{12}$ requires a direct measurement of the Brillouin gain coefficient, which is proportional to $p_{12}$.12

Both methods were utilized to generate the plots provided in Figure 5 in Companion Paper IIA.1,13,15,16 These measurements can be used to deduce $p_{12}$ values for individual glass constituents, which in turn can be utilized in glass design efforts. As will be discussed in greater detail in Companion Paper III,17 the compositions at which $p_{12} = 0$ are of particular interest as such glasses should exhibit no Brillouin scattering and reduced density-related Rayleigh scattering; the former implying no possibility for parasitic stimulated Brillouin scattering (SBS). Obviously, however, other practical considerations also exist, such as the glass-forming capability for these compositions and their refractive indices, which influence fiber design and fiber laser performance.

2.5 | Thermo-optic coefficient additivity in the presence of thermal expansion mismatch

When the optical fiber is heated, either externally or through the laser action itself, the core will attempt to expand in accordance with its coefficient of thermal expansion (CTE). However, if the CTE of the cladding is less than that of the core, the core’s thermal expansion is restricted and, therefore, the core will experience a compressive force; ie, a negative stress. This effective compressive force is exerted on the core in all 3 directions. For silica, most commonly employed in optical fibers, a positive strain (fiber stretch) results in a decrease in the refractive index since the Pockels coefficients are positive. Hence pressure (compressional force) should act to increase the refractive index of SiO2. Accordingly, if a glass additive contributes a negative thermo-optic coefficient, but also results in an increase in CTE, its negative-valued dn/dT contribution can be partly mitigated.

For a mixed oxide glass, the volume additivity of the refractive index, and of the thermo-optic coefficient, TOC, $dn/dT$, was described in equation 7 in Ref. 1. However,
that expression must be modified to include the effect of a CTE mismatch between fiber core and cladding as well as resultant strains that are introduced. Given that strain occurs in all 3 directions (x, y, and z), with 2 perpendicular (relative to a linear optical polarization) contributions and 1 parallel contribution, the expression for the refractive index of the \( i \)th component is as follows:

\[
n_i(T) = n_{0,i} + \frac{dn}{dT_i}(T - T_0) + \frac{1}{2} n_{0,i}(\alpha_{\text{core}} - \alpha_{\text{clad}})(T - T_0) \left[ 2(p_{12,i} - v'p_{11,i} + p_{12,i}) + (p_{11,i} - 2v'p_{12,i}) \right]
\]

(2)

where \( \alpha \) is again the linear coefficient of thermal expansion. The value for the core can be found using the model present in Ref. 1. It is useful to point out that when \( \alpha_{\text{core}} > \alpha_{\text{clad}} \) the refractive index increases as the temperature is increased from \( T_0 \) for cases where the Pockels term is positive (such as it is for silica). This effect of differential CTE on TOC is shown in Figure 2 for some representative binary silicate systems. In order to convert from the normal strain relationships, the substitution \(-\epsilon_{x,y,z} \rightarrow (\alpha_{\text{core}} - \alpha_{\text{clad}})(T - T_0)\) has been made. Again, the strain value is considered a stretch, but, for \( \alpha_{\text{core}} > \alpha_{\text{clad}} \), the CTE mismatch results in effective compression, hence the change in sign.

Finally, the refractive index as a function of temperature can be computed (and an effective \( dn/dT \) value determined) over a given range of temperatures. It has clearly been assumed here that there is a linear dependence of the refractive index on temperature over the range of temperatures modeled. A fully analogous corrected equation can be determined for the acoustic velocity as well.

### 2.6 Fictive temperature

As noted in Companion Paper 1, the fictive temperature, \( T_f \), influences the density-related component of Rayleigh scattering. As such, heat treatments to facilitate relaxation of the as-formed glass have been used to reduce \( T_f \) and Rayleigh scattering losses in optical fibers.

Most simplistically, \( T_f \) is the temperature where the structure of an amorphous material is the same as that of the equilibrium liquid (molten) state. In other words, to first order, \( T_f \) represents the temperature where the nonequilibrium structure is that of the equilibrium melt.

For completeness, it is worth mentioning that care should be taken in how broadly one applies the fictive temperature alone as an indicator of Rayleigh scattering and other physical or scattering-related properties. Firstly, \( T_f \) is known to influence density with a monotonic proportionality, which also therefore influences refractive index.

Secondly, as noted by Mauro et al, the use of a fictive temperature representation is rigorously valid only for a glass that experienced an infinitely fast quenching from the melt such that the structure of the liquid phase was instantaneously frozen in. Even in the case of the high quench rates (\( >2000 \text{ K/s} \)) associated with the molten core method, used to make the majority of the fibers treated in this trilogy, it is very unlikely that this qualifies as “infinitely fast.” Perhaps more importantly is that Mauro also points out that while the macroscopic view of \( T_f \) reasonably estimates state properties (eg, enthalpy and molar volume), it does not at all accurately estimate probability distributions at lower temperature such as fluctuations in enthalpy and volume (which do influence Rayleigh scattering) or corresponding changes in heat capacity and thermal expansion, which could influence TMI. Readers are directed to Refs. 24, 25. If interested in more detailed and thoughtful discussions of the inherent inequalities between glassy and liquid states.

The fictive temperature can be estimated using methods that probe the glass structure, such as Raman and infrared (IR) spectroscopies. More specifically, the micro-Raman approach correlates the fictive temperature to shifts in the Si-O-Si stretching vibration at wavenumbers of about 440 cm\(^{-1}\), whereas the IR approach utilizes shifts in the Si-O-Si asymmetric stretch at wavenumbers of about 1120 cm\(^{-1}\). An example of this latter approach is provided in Figure 3, which compares the peak position of the Si-O-Si asymmetric stretch in a conventional telecommunications optical fiber to a molten core-derived oxyfluoride core.

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**FIGURE 2** Thermo-optic coefficient (TOC, \( dn/dT \)) as a function of non-Si\(_2\)O\(_3\) content for SiO\(_2\)-clad optical fibers in the P\(_2\)O\(_5\)-SiO\(_2\), B\(_2\)O\(_3\)-SiO\(_2\), and BaO-SiO\(_2\) systems with and without taking into account differential thermal expansion between core and clad. The \( dn/dT = 0 \) line is provided as a guide and shows those compositions where stimulated thermal Rayleigh scattering should be neglected. [Color figure can be viewed at wileyonlinelibrary.com]
fiber. Based on Equation 2 from Ref. 27, the computed $T_f$ for the core and clad are between about 1450 and 1670°C, corresponding to the IR peak positions of 1121 and 1119.5 cm$^{-1}$, respectively. These values are within the range expected for silica optical fibers.

For completeness, it is noted that the slope of the change in Si-O-Si peak position with fictive temperature depends on the glass “dependency of volume with temperature,$^3$ but often with modified coefficients to improve data fitting.$^36$ This alternative formula is given as follows:

$$n_2(10^{-13}\text{ esu}) = 391 \frac{(n_d - 1)}{v_d^{4/3}}. \quad (3)$$

While several empirical relations can be found in Ref. 33 for the nonlinear refractive index, the version embodied by Equation 3 (Equation 39 in Ref. 33) seems to offer the best fit to a wide set of $n_2$ data across compositional ranges, and has gained broad acceptance.$^34$ An alternative expression, based on a derivation from physical properties (equation 38 in Ref. 33) also is frequently found in the literature, but often with modified coefficients to improve data fitting.$^36$ This alternative formula is given as follows:$^33$

$$n_2(10^{-13}\text{ esu}) = \frac{68(n_d - 1)(n_d^2 + 2)^2}{v_d \left[1.517 + \frac{(n_d^2 + 2)(n_d + 1)}{600} \right]^{1/2}}. \quad (4)$$

The terms in Equations 3 and 4 are the refractive indices ($n_f$, $n_d$, $n_c$) and Abbé number, $v_d = (n_d - 1)/$ $(n_f-n_c)$, at the historically based wavelengths of the hydrogen F line (486.13 nm), the helium d line (587.56 nm), and the hydrogen C line (656.27 nm).

According to Ref. 33, Equation 3 provides a better fit for materials that possess $n_2$ values greater than $\sim 2 \times 10^{-13}$ esu. It should be pointed out, however, that neither equation provides any direct dependence of $n_2$ (in esu units) on the specific optical wavelength; only on a measure of visible chromatic dispersion.

### 3.1 Nonlinear refractive index

Nonlinear optical effects, such as self-phase modulation (SPM) and four-wave mixing (FWM), can manifest at high optical powers in a fiber-based system.$^32$ In SPM, both spectral and temporal modifications to optical pulses can arise. In FWM, which is among the strongest parametric wave-mixing nonlinearities, new photon frequencies are created from those of the pump. In both cases, the effect is proportional to the nonlinear refractive index, $n_2$. The Reader is directed to Companion Paper 1$^14$ for more detail on these effects, their physical origin and material influences.

Similarly to the thermo-optic coefficient, to first order, the nonlinear refractive index can be introduced parametrically into the refractive index additivity model as $n = n_0 + n_2 I$, where $n_0$ is the linear refractive index, $n_2$ is the nonlinear refractive index and $I$ is the light intensity. The simplest and widely employed model for estimating $n_2$ is the “BGO” model, (so named for the authors: Boling, Glass, and Owyoung$^{33}$), reproduced here (in electrostatic units as was standard at the time of its derivation) as follows:

$$n_2(10^{-13}\text{ esu}) = 391 \frac{(n_d - 1)}{v_d^{4/3}}. \quad (3)$$

Not all properties of interest in this trilogy are easily modeled using additivity approaches; these include the fictive temperature described above and the Raman gain coefficient. This section describes the approach employed to determine selected quantities directly from measurements on the as-drawn optical fibers.
In order to estimate \( n_2 \) for an optical fiber, the refractive index profile (RIP) can be measured at multiple wavelengths. This enables the determination of a set of Sellmeier coefficients for the fiber from which \( n_0, n_r, \) and \( n_c \) can be computed. With Equation 3 these values enable the calculation of \( n_2 \). Furthermore, using the additive model enables the determination of the Sellmeier coefficients and effective \( n_2 \) of individual glass constituents, allowing for the calculation of \( n_2 \) for arbitrary compositions.

With respect to additivity of the nonlinear refractive index, as noted above, \( n = n_0 + n_2 I \). Accordingly, the refractive index of a binary glass mixture is given as follows:

\[
 n = m(n_i + n_2 I) + (1 - m)(n_j + n_2 J) = mn_i + (1 - m)n_j + (mn_2 I + (1 - m)n_2 J)I
\]

and, converting [esu] to [m²/W]:

\[
 n_2 \left( \frac{m^2}{W} \right) = \frac{40\pi}{c_0} \left( \frac{m n_2 (\text{esu})}{n_i} + (1 - m) \frac{n_2 (\text{esu})}{n_j} \right).
\]

Designations of binary components 1 and 2 have been termed \( i \) and \( j \) in Equation 3 simply to remove confusion with \( n_2 \). This equation can be rewritten to possess linear and nonlinear components that can be added separately. Therefore, Equation 5a suggests simple linear additive modeling of the nonlinear refractive index in a similar fashion to the linear refractive index. When the Sellmeier coefficients of a pure compound are known, it is possible to know the evolution of the linear refractive index as a function of wavelength, and therefore determine the \( n_2 \) value for each individual compound using Equations 5a and 5b. Thus, in a binary system, \( n_2 \) for each end compound can be determined, and, using Equation 5a, the \( n_2 \) aggregate value of the glass can be computed. Figure 4 shows the calculated values of \( n_2 \) as a function of GeO₂ content in a SiO₂-GeO₂ binary glass. These results agree with Nakajima et al.\(^{37} \) Accordingly, hereafter, when \( n_2 \) compound-specific data is not available (which is the case for most compounds), Equation 39 from BGO [Equation 3 above] will be employed using linear refractive index values.

### 3.2 Raman scattering

Although a general description of the material influences on Raman scattering was provided in Companion Paper 1,\(^{14} \) and approaches to computing absolute Raman intensities have been reported in the literature,\(^{38,39} \) the modeling of Raman gain coefficient in multicomponent glasses is beyond the simple additivity models treated here. One important reason for this is that fibers fabricated using the molten core method have been shown to possess more disordered structures than conventionally prepared optical fibers.\(^{40} \) As a result, the glass structure, and not just the molecular species, plays a considerable role in the resultant Raman properties. That said, as described in more detail below, the Raman spectra of the optical fiber core can be directly measured and meaningful trends with processing conditions and glass constituents can be deduced.

The relative Raman gain (\( g_R \)) of the fiber core material can be determined from direct measurement of its spontaneous Raman scattering cross section using micro-Raman spectroscopy. The procedure to correct the spectra is similar to that in Ref. 1, which accounts for reflection loss, internal solid angle variation, and the Bose-Einstein correction factor. The corrected spectra are normalized relative to the highest peak intensity of silica (typically around 440 cm\(^{-1} \)) since the Raman gain of silica is well known.\(^{41,42} \) This is especially convenient since pure silica is the cladding material used for the molten core fabricated fibers often invoked in this Trilogy. Thus, a comparative Raman analysis of the intrinsically low nonlinearity core materials can be directly made relative to the cladding of the same fiber, which experienced the exact same thermal history during preform heat-up and fiber draw.

As an example, corrected and normalized Raman scattering spectra are shown in Figure 5 for silica and fibers in the BaO-SiO₂ and Al₂O₃-SiO₂ families. The reductions in the magnitude of \( g_R \) relative to pure silica are explained by the high cooling rates associated with the fiber drawing process, which enables the formation of a more highly disordered core glass (as evidenced by the broadened Si-O spectral features) and a reduced overlap of the individual
Raman peaks between the differing constituents. A more detailed discussion of the most promising material systems for realizing intrinsically low Raman response is provided in the Companion Paper III.

3.3 | Brillouin scattering

Brillouin scattering was described in some detail in Companion Paper I. The Brillouin gain coefficient, BGC, is given by $2\pi n^2 P_{12}/c\rho V_t\Delta V_B$. Calculating this value is somewhat straightforward. In short, the models presented in Ref. 1 can be used to calculate the relevant terms and a global BGC can be computed. Measurements of this value can include deducing the individual terms comprising the BGC equation and substituting into the equation. Alternatively, BGC can be measured directly. Most typically, this is done by comparing the strength of the Brillouin backscattered signal from a fiber to that of a control fiber of known BGC.

4 | SOME CANONICAL EXAMPLES

This section provides specific modeling results, overlaid with measured property values, for selected optical nonlinearities of interest to this work. Companion Paper III in this trilogy will delve with greater detail into which glass families warrant further attention for optical fibers that exhibit simultaneous reductions to all of the parasitic nonlinearities that limit scaling to higher powers in laser and communication systems.

4.1 | Brillouin scattering

As noted in Companion Paper I, Brillouin scattering is an inelastic interaction between acoustic phonons of the material and the optical signal. In its spontaneous form, Brillouin scattering materially depends on the refractive index, transverse ($P_{12}$) photoelastic coefficient, and adiabatic compressibility, which, in turn, are related to density and acoustic wave velocities. In its stimulated form (SBS), the gain is materially dependent upon the Brillouin linewidth in addition to the factors noted above.

As discussed in the previous section, each of the material properties relevant to Brillouin scattering can be calculated utilizing the models discussed in Ref. 1. Figure 6 provides a comparison of the Brillouin gain coefficient, relative to a conventional telecommunications-grade optical fiber, as a function of non-silica concentration for a variety of binary and ternary silicate core glass optical fibers (all employing pure silica cladding glasses).

At first glance, Figure 6 shows that conventional dopants into passive and active optical fibers, ie, GeO$_2$, P$_2$O$_5$, and B$_2$O$_3$, all lower the BGC relative to SiO$_2$. This is due to their acoustic damping terms, which are much larger than that of silica, since their $P_{12}$ values are not especially low. At low silica concentrations, these materials also show an upturn in BGC for the binary systems. This can be attributed to the fact that each of these network formers has an acoustic velocity that is less than that of silica. As the silica content is decreased, the acoustic velocity decreases, resulting in a concomitant decrease in the
Brillouin frequency shift. As discussed in Ref. 1 this causes a decrease in $\Delta \nu_B$, mitigating the reduction in gain due to the larger acoustic damping. This is also true of the alkaline earth silicate systems, but BGC is significantly reduced due to reduction in $p_{12}$. In the case of material systems possessing $Al_2O_3$, $MgO$, and $Li_2O$, they raise the acoustic velocity when added to silica, increasing the Brillouin frequency. This causes further broadening to $\Delta \nu_B$, and thus the BGC graph for these materials appears to drop monotonically (ignoring the zero-gain-coefficient regions) across the compositional range.

Two additional observations are worthy of note with specific material trends to be discussed in greater detail in Companion Paper III.\textsuperscript{17} First is the significant reductions in relative Brillouin gain coefficient at selected compositions. Though the minima as shown in Figure 6 attain values of greater than $-60$ dB (note the log scale), these zero Brillouin activity ("ZeBrA") compositions, in theory, should approach $-\infty$ (dB). Such points are associated with compositions where the effective $p_{12}$ photoelasticity goes to zero, hence, Brillouin scattering is no longer operative. This is achieved through the additive properties of glasses comprised of both $p_{12} > 0$ (eg, $SiO_2$) with $p_{12} < 0$ (eg, $BaO$) components. The exceptions in Figure 6 are the $Y_2O_3-Al_2O_3-SiO_2$ fibers whose core properties add as if comprised of $SiO_2$ and YAG ($Y_3Al_5O_{12}$), both of which exhibit $p_{12} > 0$ (though it is small for the latter), hence no ZeBrA composition exists. The second point of note is that, for all practical purposes, relative Brillouin reductions of greater than 3 dB (preferably 5 dB) are significant to the high power laser community. In this case, even in the YAG-derived fibers, reductions in relative Brillouin gain of about 6 dB, as has been experimentally validated,\textsuperscript{15} are useful. The results of Figure 6 show that high silica-content optical fibers with only 1 or 2 additional components can readily exhibit Brillouin reductions between 5 and 15 dB, all through judicious selection of the enabling materials, rather than the more complex and costly approach of geometric tailoring (eg, photonic crystal and/or microstructured fibers) of the fiber cross section.

4.2 | Transverse mode instability

Transverse mode instability (TMI), also known in the literature as "higher order mode instability" (HOMI), is presently the dominant limitation in scaling fiber-based lasers to higher output powers.\textsuperscript{47,48} As noted in Companion Paper I,\textsuperscript{14} TMI occurs in large mode area (LMA) multimode waveguides whereby, above a certain power threshold, the propagating optical modes becomes random and dynamic.\textsuperscript{49,50} It is generally accepted that TMI originates from stimulated thermal Rayleigh scattering (STRS), which is materially dependent on the thermal conductivity, heat capacity, and thermo-optic coefficient (dn/dT) of the glass comprising the core of the optical fiber.\textsuperscript{51}

While these material dependencies are discussed in greater detail in Companion Paper III,\textsuperscript{17} suffice it here to note that dn/dT is the one property of these three material influences that is most compositionally tailorable within a given glass family. In an analogous fashion to Brillouin scattering, components exist that possess positive- and negative-valued thermo-optic coefficients such that their additivity yields the potential for dn/dT = 0 compositions where STRS would be mitigated. Canonical examples of this are borosilicate and phosphosilicate binary glasses where both $B_2O_3$ and $P_2O_5$ possess dn/dT < 0 values, whereas that for $SiO_2$ is positive-valued. Examples of this additivity were already provided in Figure 2. Figure 7, on the other hand, shows example data obtained from two Yb-doped fibers using the ring laser measurement apparatus described in Section 2. The negative slope of the data means that the FSR is decreasing with temperature, implying that the CTE dominates the thermal dependence. Decreasing dn/dT lessens the slope, while a line with zero slope implies that the CTE and dn/dT have fully compensated each other. Interestingly, this suggests a novel way to thermally stabilize single frequency fiber lasers: by making the intracavity fiber system immune to changes in temperature.\textsuperscript{52}

Also, in a manner analogous to Brillouin scattering, while zero values exist in the operative material parameters,
actually obtaining those compositions is not usually critical in practice. Associated trade-offs between given material properties (e.g., $dn/dT$) and other material properties and fiber design and performance details are discussed further in Companion Paper III.17 Indeed, as relates to TMI, the threshold power should scale inversely with $dn/dT$ such that a 50% reduction in $dn/dT$ should manifest a 3 dB increase in TMI threshold. This would permit multiple kilowatt output powers from a single fiber.

4.3 | Self-phase modulation (SPM)

Self-phase modulation (SPM) is a nonlinear effect that leads to both spectral and temporal modifications to laser pulses at high energies, such as in mode-locked ultrafast laser systems. SPM originates from the optical Kerr effect whereby the refractive index is a function of intensity through the nonlinear refractive index, $n_2$. The time varying intensity associated with the pulse shape gives rise to a time-varying refractive index that modulates the phase of the optical signal. As an illustration, Figure 8 provides the idealized spectrum of a 100 ns, 1-kW-peak-power Gaussian pulse propagating through a conventional single mode fiber of different lengths. Here, the $n_2$ value for a conventional single mode fiber was taken to be $\sim 2 - 3 \times 10^{-20}$ m$^2$/W and the nonlinear phase change, $\gamma$, was computed using $\gamma = (2\pi n_2)/(\lambda_0 A_{\text{eff}})$, where $\lambda_0$ is the free space wavelength (1 $\mu$m for the purposes of Figure 8), and $A_{\text{eff}}$ is the effective area of the waveguide mode.53,54 Clearly apparent is the spectral distortion, which translates into a practical threshold value of $\gamma < 2\pi$ to avoid spectral broadening.

While SPM can be tailored geometrically, through enlarging $A_{\text{eff}}$ by fabricating a larger fiber, it can also be tailored materially through $n$ (which does also influence $A_{\text{eff}}$, $n_2$, and the group velocity dispersion of the fiber (through $n$). As observed conceptually in Figure 8, a reduction in $n_2$ to $1/2$ of its original value yields a spectral broadening due to SPM that is also reduced by $1/2$ (3 dB), which is quite consequential in practical systems.

5 | Compilation of Properties for Assorted Materials Studied to Date

Table 1 provides a list of properties, either measured directly or deduced using the models treated above, for a range of materials that have been studied to date. In the case of the deduced values, data on silica were used as known quantities and the properties of the additives to silica were employed as fitting parameters, and adjusted until modeling results matched measured data on actual fibers.

For completeness it is noted that the data obtained for SiO$_2$, GeO$_2$, P$_2$O$_5$, B$_2$O$_3$, and Yb$_2$O$_3$ were deduced from fibers that were fabricated utilizing conventional methods for telecommunications grade silica fibers; i.e., modified chemical vapor deposition (MCVD).9,55,56 The other fibers were fabricated utilizing the molten core approach.5,12,13,15,16,44,46 Care should be given in extrapolating property values significantly beyond the compositional ranges and fabrication conditions of the fibers/
glasses for which they were deduced since thermal histories, impurities, etc. may influence the resultant glass. As noted above, the continuum additivity approaches employed here are intended principally to provide a rapid and simple compositional guide for the subsequent fabrication of specific optical fiber core properties.

6 | CONCLUSIONS

Provided herein is a discussion of the use of simple additivity models for the deduction of the properties of multi-component silicate glass fibers (in contrast to bulk glass), central to optical nonlinearities, from the properties of amorphous or crystalline end-member compounds. The clad fiber geometry plays an important role since a core-cladding CTE mismatch can have a significant impact on the observed thermal coefficients. Furthermore, the mode spatial distribution, relative to the compositional profile, is also important to consider when modeling the fiber system and methods used to measure the physical properties of fiber were also outlined. Finally, a brief discussion of the modeling of selected nonlinear coefficients was provided. In general, over reasonable glass forming ranges in which the glass is presumed to remain compositionally uniform, homogeneous, and structurally similar, the models are found to be sufficiently accurate in order to guide the materials development of optical fibers that exhibit reduced optical nonlinearities.

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