# The Material Dispersion Zero in Infrared Optical Waveguide Materials

By K. NASSAU

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The wavelength where the material dispersion is zero, i.e., that wavelength for which a multimode lightguide would function at highest bandwidth, can be estimated using fundamental materials properties. Operation of lightguides at or near this wavelength is essential for the use of very long fiber lengths with high bandwidth. Calculations have been performed for large groups of metal oxides, fluorides, chlorides, and bromides. These data can be used to locate materials with potential for ultralow-loss fibers at longer wavelengths than are currently in use.

#### I. INTRODUCTION

Operation of optical waveguides at wavelengths up to 1.3  $\mu m$  and 1.6  $\mu m$  is being actively investigated and losses of a few tenths dB/km appear to be feasible. Operation at a longer wavelength has the attraction that much lower losses should be possible since the  $\lambda^{-4}$  relationship controls the scattering loss, subject of course to the achievement of adequately low intrinsic and impurity absorptions. This should permit large distances between repeaters.

The utilization of very long lengths of low-loss fibers is critically dependent on the material dispersion parameter  $(\lambda/c)(d^2n/d\lambda^2)$ , where n is the refractive index at wavelength  $\lambda$  and c is the velocity of light. At the wavelength  $\lambda_0$  where the material dispersion is zero, the delay distortion in multimode fibers is minimized and maximum bandwidth can be achieved. In single-mode fibers, zero total dispersion requires balancing the waveguide dispersion with the material dispersion; the maximum bandwidth then occurs at a wavelength longer than  $\lambda_0$ .

Several reports have discussed possible longer wavelength fiber materials, including the extended discussion of Goodman<sup>1</sup> for the 4-µm band, the polycrystalline materials Tl(Br,I) of Pinnow et al.<sup>2</sup> and Ag(Cl,Br) of Garfunkel et al.,<sup>3</sup> and ZnCl<sub>2</sub> glass of Van Uitert and

Wemple.<sup>4</sup> The use of fluoride glasses based on ZrF<sub>4</sub>, HfF<sub>4</sub>, and AlF<sub>3</sub>,<sup>5,6</sup> mainly derived from earlier work by Poulain and co-workers (e.g., Refs. 7 and 8) has also been studied. A limited consideration of a few<sup>1</sup> or only a single<sup>4</sup> value of the material dispersion was given in some of these reports, while others<sup>2,3,5,6</sup> ignored this parameter.

Wemple<sup>9</sup> has given a general technique for calculating an approximate value of the wavelength of the material dispersion zero, based on materials parameters which were discussed in detail in his previous report.<sup>10</sup> Using this approach it is possible to calculate  $\lambda_0$  values for large groups of potential optical waveguide materials and thus locate the most promising candidates for operation at desired wavelengths. In this study we focus our attention on metal oxides, fluorides, chlorides, and bromides which may have such a potential and from which glasses might be made. The reasons for concentrating on these four groupings and on glasses specifically, as well as further limiting factors, are discussed in the appendix.

These data can, of course, be used to direct the investigation of specific materials with potential for ultralow loss at longer wavelengths than are currently in use. They can also eliminate from consideration some already studied materials which may be transparent in this region, but which have too low a  $\lambda_0$  value.

#### II. THE CALCULATION OF $\lambda_0$

Based on considerations of optical oscillator strengths and excitation energies, Wemple<sup>9,10</sup> has developed a formalism from which  $\lambda_0$ , the wavelength for which  $d^2n/d\lambda^2=0$  is given as

$$\lambda_0 = hc \left( \frac{10^{-10} cn f \mu d^3}{4\pi e^2 Z E^3} \right)^{1/4}, \tag{1}$$

where E is the average electronic (Sellmeier) excitation gap (usually a few eV higher than the band gap), f is the normalized oscillator strength, Z is the formal valence of the anion A, n is the number of valence electrons on the anion in the compound, d is the anion-cation distance,  $\mu$  is the reduced mass of the anion-cation pair, h is Planck's constant, c is the velocity of light, and e is the charge on the electron.

Most often n = 8 for closed-shell anions, and eq. (1), rewritten in practical units, then reduces to

$$\lambda_0 = 2.96 \left( \frac{d^2 f \mu}{E^3 Z} \right)^{1/4},\tag{2}$$

where  $\lambda_0$  is in  $\mu$ m, d is in Å, and E and f are in eV. Here Z=1 for halides, 2 for oxides and chalcogenides, and 3 for pnictides; the reduced mass for a compound written as  $AB_b$  (i.e., with one cation A and with

b not necessarily an integer) of atomic weights  $M_A$  and  $M_B$  is given by

$$\mu = \frac{M_A M_B}{M_A + b M_B}. (3)$$

For compound glasses, eq. (4) can be used to determine values for use in eqs. (2) and (3) for a mixture of i components, containing x mole fraction each of  $PR_r$  where P is a cation, R is an anion, and r is the number of anions per cation (not necessarily integral) with  $\sum_i x_i = 1$ :

$$f = \sum x f_{PR_r},$$

$$E = \sum x E_{PR_r},$$

$$Z_A = \sum x Z_{PR_r},$$

$$M_A = \sum x M_P,$$

$$b = \sum x r,$$

$$M_B = \frac{1}{b} \sum x r M_R,$$

$$d = \frac{1}{b} \sum x r d_{PR}.$$
(4)

In these equations intensive properties, such as energies, are averaged according to the composition, but extensive properties, such as the bond lengths and the effective cation masses, are additionally weighted by the number of anion bonds per cation; b is the composition-weighted average of this last parameter.

Values of the required parameter were taken from Wemple's collection of data<sup>10</sup> when available or from the literature. Otherwise estimates for E and f (to the nearest ½ eV and so listed in Tables I and II) were made following Wemple's principles, allowing for coordination number, bond length anomalies, ionicity, anion contact, and shallow electron cores.<sup>10</sup> This last factor is significant for the  $d^{10}$  core of the Ag halides where N is effectively 14 rather than 8, introducing a correction factor for  $\lambda_0$  in eq. (2) of  $(14/8)^{1/4} = 1.15$ ; smaller corrections apply elsewhere, for example to the  $s^2$  cores of the Tl and Pb halides.

Wemple has also pointed out that although amorphous nonvitreous solids differ significantly from crystals in the applicability of the approach used, glasses do not; they can be viewed as loosely-packed versions of the crystals to a good approximation.<sup>10</sup>

# III. VALUES OF $\lambda_0$ FOR OXIDES, FLUORIDES, CHLORIDES, AND BROMIDES

The E and f values used and the  $\lambda_0$  wavelengths obtained are listed for oxides in Table I and for fluorides, chlorides, and bromides in Table

Table I—Parameters for the calculation of  $\lambda_{\text{0}}$  for oxides

Oxide	E	f	$\lambda_0$	Oxide	E	f	λο
$MoO_3$	6	4	2.8	$Sc_2O_3$	11	3	1.8
$WO_3$	6	4	3.0	$Y_2O_3$	9	4	2.4
$P_2P_5$	13.5	4 5	1.3	$La_2O_3$	8	4	2.9
$As_2O_5$	11	5	1.8	ZnO	6.1	3.1	2.7
$Nb_2O_5$	$\begin{array}{c} 11 \\ 7 \end{array}$	4	2.6	PbO	4.7	3.8	3.9
$Ta_2O_5$	7	4	2.7	BeO	13.7	5.1	1.2
$SiO_2$	13.3	5	1.3	MgO	11.4	2.8	1.6
$GeO_2$	11.0	4.0	1.7	CaO	9.9	2.8	2.0
$SnO_2$	8.1	3.0	2.2	SrO	8.3	2.5	2.5
$TeO_2$	6.3	3.9	2.8	BaO	7.1	2.4	3.0
$TiO_2$	5.5	3.8	2.8	$\mathbf{Tl_2O}$	4	4 5	5.3
$ZrO_2$	11	4	2.0	Li <sub>2</sub> O	12	5	1.6
$HfO_2$	10	4	2.2	Na <sub>2</sub> O	11	4	2.1
$ThO_2$	10	4	2.3	$K_2O$	11	4 3	2.3
$B_2O_3$	12.4	6.8	1.3	$Rb_2O$	10	3	2.5
$Al_2O_3$	13.4	3.8	1.4	$Cs_2O$	10	3	2.7
$Ga_2O_3$	$9\frac{1}{2}$	4	2.0				
$In_2O_3$	7	4	2.8	GaAs	3.7	4.5	6.3
$As_2O_3$	11	4 5	1.9	ZnTe	4.4	5.2	6.6
$Sb_2O_3$	7	5	2.9	BN	10.6	4.5	1.2
Bi <sub>2</sub> O <sub>3</sub>	5	5 5	3.9				

Table II—Parameters for the calculation of  $\lambda_0$  for halides

	X = F			X = Cl				X = Br		
	E	f	$\lambda_0$	E	$\cdot f$	$\lambda_0$	E	f	λο	
LiX	16.5	3.8	1.3	11.0	4.8	2.3	9.5	4.8	2.7	
NaX	15.1	2.8	1.7	10.5	3.5	2.9	9.1		3.7	
KX	14.7	3.1	2.1	10.5	3.1	3.3	9.2		4.2	
RbX	14	$3\frac{1}{2}$	2.4	10.5	3.3	3.7	9.3	3.3	4.8	
CsX	13.5	4.9	3.3	10.6	3.5	4.1	9.4	3.4	5.3	
AgX* TlX*	$10\frac{1}{2}$	3	3.0	7	3	5.1	$\frac{5\frac{1}{2}}{5.2}$	3.1	6.7	
TÏX*	9	3 3	3.5	5.5	2.9	6.6	5.2	3.3	8.5	
D <sub>2</sub> V	16	5	1.2	11	5	1.8	$9\frac{1}{2}$	5	2.1	
$MgX_2$	16.8	3.9	1.4	11	4	2.4	$9\frac{1}{2}$	4	2.9	
CaX	15.7	3.1	1.7	$10\frac{1}{2}$	3	2.7	9	3	3.4	
SrX <sub>2</sub>	15	3	1.9	$10\frac{7}{2}$	3 3 3 3	3.1	9	3	4.0	
BaX	13.8	3.1	2.3	$10\frac{7}{2}$	3	3.5	9	3	4.5	
ZnX <sub>2</sub> *	13	3	2.1	9	3	3.5	$7\frac{1}{2}$	3	4.6	
CdX	11	3	2.3	$7\frac{1}{2}$	3	4.0	6	3	5.4	
MgX <sub>2</sub> CaX <sub>2</sub> SrX <sub>2</sub> BaX <sub>2</sub> ZnX <sub>2</sub> * CdX <sub>2</sub> HgX <sub>2</sub> SnX <sub>2</sub> * PbX <sub>2</sub> *	9	3	2.8	_	_	_	_	_		
SnX <sub>2</sub> *	10	3	2.6	7	3	4.4	$5\frac{1}{2}$	3	6.1	
PhX <sub>2</sub> *	8.4	2.8	3.3	$6\frac{1}{2}$	- 3	5.1	5	3	7.4	
$ScX_3$	16	3	1.4	10	3	2.6	81	3	3.2	
$YX_3$	16	3	1.6	10	3	2.9	$8\frac{1}{2}$ $8\frac{1}{2}$	3	3.7	
$LaX_3$	16	3	1.8	10	3	3.3	81	3	4.3	
LuXa	16	3	1.7	_		_		_	_	
AlX	16	3	1.2	<u> </u>			· · · · · · · · · · · · · · · · · · ·	44 <u>21</u> 4	2 + 1 <u>7 +</u>	
GaX <sub>3</sub>	15	3	1.5					<u></u>	·	
AlX <sub>3</sub> GaX <sub>3</sub> InX <sub>3</sub>	14	3 3 3	1.8	7	3	4.4	$5\frac{1}{2}$	3	5.3	
$TlX_3$	13	3	2.0			_				
RiX <sub>2</sub> *	8	3	3.2	$6\frac{1}{2}$	3	4.8	5	3	6.9	
TiX.	14	3	1.5	_			_			
ZrX.	14	3	1.7	10	3	2.7	9	3	3.3	
TiX₄ ZrX₄ HfX₄	14	3	1.8	10	3	3.0	9	3	3.8	
ThX₄	14	3	1.9	10	3	3.2	9	3	4.0	
SnX <sub>4</sub>	13	3	1.8	_	_	_	_		_	
PbX <sub>4</sub>	13	3	1.9	_	_	_	_			

<sup>\*</sup> Corrections applied for shallow electron cores.

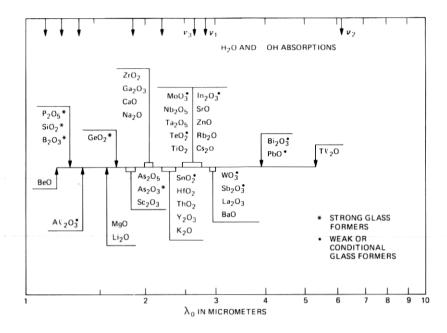


Fig. 1—Material dispersion crossover wavelengths for oxides. Also shown are the  $\nu_1$ ,  $\nu_2$ , and  $\nu_3$  fundamental water absorptions; "water" in fused silica shows  $\nu_3$  as well as the indicated absorptions at lower wavelengths.

II; the more readily accessible values of d and  $\mu$  are omitted. The results are graphically presented in Figs. 1 and 2. A definite precision cannot be given for  $\lambda_0$ ; we feel that most values are accurate to  $\pm 10$  percent, although in a few instances the uncertainty may well be a little bigger. Note that since the fourth root is involved in eq. (2), the results are only moderately sensitive to d and E and even less so to f and  $\mu$ .

Most oxides with a value of E less than  $4\frac{1}{2}$  eV were excluded, since such substances are deeply colored and would also be expected to show significant intrinsic absorption in the wavelength region of interest. An exception was made with  $T1_2O$  (black,  $E\approx 4$  eV), which is often an ingredient in glasses, in small amounts, since it reduces the melt viscosity and assists in the removal of bubbles. Oxides were also generally excluded, which would be expected to decompose under conditions required for glass melting. The oxides which are strong glass formers are marked, as are those which are weak or conditional glass formers, i.e., those which do not form glass by themselves but will do so in combination with other similar compounds. 11

The halides of Table II and Fig. 2 were selected in an analogous way. Here BeF<sub>2</sub> and ZnCl<sub>2</sub> are the only strong glass formers; the

occurrence of glasses based on BeF<sub>2</sub> and other fluorides has been summarized by Sun.<sup>12</sup> Glasses based on the weakly glass forming ZrF<sub>4</sub>, HfF<sub>4</sub>, and AlF<sub>3</sub> have been studied by Poulain and others.<sup>5-8</sup>

Figure 1 also includes the fundamental vibrations of the free  $H_2O$  molecule,  $^{13}$  marked  $\nu_1$ ,  $\nu_2$ , and  $\nu_3$ , as well as the positions of the dominant "water," i.e., OH, absorptions in fused silica, consisting of  $\nu_3$  and its overtones, and combinations with  $SiO_4$  vibrations.  $^{14}$  These absorptions tend to become more intense and broader with increasing wavelength. In view of the difficulty of eliminating all traces of water, operation in windows between these absorptions may be desirable. For fused silica these windows occur at about 1.3, 1.6, and 2.5  $\mu$ m; since  $SiO_4$  vibrations are involved, shifts in both position and intensity must be expected in moving to different systems, although differences may not be large with related oxides, such as  $GeO_2$ . With halides, however, significant difference may exist.

Another factor sometimes considered is the location of the windows in the atmosphere occurring at about 1.3, 1.6, 2.3, 3.5, to 4, and 8 to 13  $\mu$ m. It was primarily this consideration that caused Goodman to limit his discussion to materials for the 4- $\mu$ m band. This does not seem to be a meaningful limitation, however, since the path of optical waveguide communication systems does not normally include any atmospheric links.

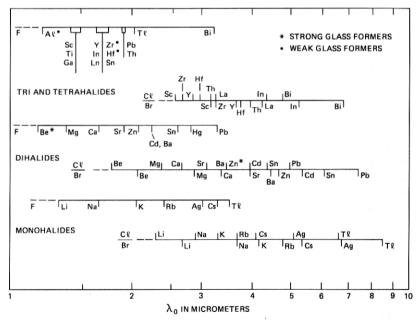


Fig. 2—Material dispersion crossover wavelengths for fluorides, chlorides, and bromides.

#### IV. DISCUSSION

In the case of the oxides of Table I and Fig. 1,  $\lambda_0$  extends only to about 5  $\mu$ m, with very limited choice above 3  $\mu$ m. The use of chalcogenides, such as ZnTe, and pnictides, such as BN and GaAs, also included for reference in Table I, would extend  $\lambda_0$  to much longer wavelengths for heavier anions, but the reduced bandgap of such compounds would also raise the intrinsic absorption (also see the appendix).

The only strong glass formers among halides are BeF<sub>2</sub> and ZnCl<sub>2</sub>,<sup>11</sup> the former being highly toxic and the latter hygroscopic. Figure 2 shows a wide range of  $\lambda_0$  among fluorides, chlorides, and bromides with values up to 8  $\mu$ m. The effect of a change in the halogen can be seen in the sequences of fluoride, chloride, bromide, and iodide for Li ( $\lambda_0$  = 1.3, 2.3, 2.7, and 3.4  $\mu$ m) and for divalent Pb ( $\lambda_0$  = 3.3, 5.1, 7.4, and 13.9  $\mu$ m).

Using eqs. (4) to calculate  $\lambda_0$  for mixtures should be an improvement over assuming linearity between values of  $\lambda_0$  for the end members in binary mixtures, and can also be used for more complex compositions. Some sample calculations, e.g., for LiTlCl<sub>2</sub>, SiGeO<sub>4</sub>, Mg<sub>2</sub>OCl<sub>2</sub>, etc., show that the two approaches range from agreement to producing a 15% difference, in either direction.

Keeping in mind the limitations discussed in the appendix, Figs. 1 and 2 indicate a number of compositions worthy of investigation. Note that Goodman1 limited his thinking to wavelengths of 31 to 4 µm because this is an atmospheric transmission window and it might represent the limit of room temperature sources and detectors. The first constraint is not relevant to a normal fiber system containing no atmospheric linkages and the latter could also be unimportant if. sav. a sufficiently low loss permitted a trans-Atlantic link without repeaters: Operation of the two cooled terminals would seem to be a reasonable price for such a system. In the oxides of Fig. 1, the currently used compositions based on SiO2 and involving relatively small amounts of other oxides such as  $GeO_2$  provide a  $\lambda_0$  near 1.3  $\mu$ m, conveniently centered in a "water" window of SiO2. Use of the next window at about 1.6 µm could be possible in SiO<sub>2</sub> with a larger addition of GeO<sub>2</sub>, with smaller amounts of many other possible oxides, or in a single mode fiber at zero total dispersion by balancing the waveguide and materials dispersions. <sup>16</sup> The limiting scattering loss for pure  $GeO_2$  at its  $\lambda_0$  of about 1.7 µm has been estimated at 0.15 dB/km.17 Note that widely scattered  $\lambda_0$  values for the SiO<sub>2</sub>-GeO<sub>2</sub> system have been reported. apparently none above 16 percent GeO2, however.18

Shifting our focus to wavelengths above 2  $\mu m$  necessitates omitting the light Si atom. One possibility now centers about compositions containing the strong glass former GeO<sub>2</sub>; combinations of this with

many of the weak or conditional glass formers in Fig. 1, such as PbO, <sup>19</sup>  $Bi_2O_3$ , <sup>20</sup> and PbO plus  $In_2O_3$ , <sup>21</sup> are known to give glasses. The other possibility is to use mixtures containing only the weak or conditional glass formers by themselves. The scope for a  $\lambda_0$  above 3  $\mu$ m appears to be quite restricted, with the availability of only  $Bi_2O_3$ , PbO, and the very toxic  $Tl_2O$ , all with relatively small values of E.

The highest value of  $\lambda_0$  among the fluorides of Table II and Fig. 2 is the 3.5  $\mu$ m of TlF. The glass-forming fluorides have relatively low  $\lambda_0$  values, varying from the 1.2  $\mu$ m of BeF<sub>2</sub> and AlF<sub>3</sub> to the 1.7 to 1.8  $\mu$ m of ZrF<sub>4</sub> and HfF<sub>4</sub>. Fluoride glasses based on some of these compounds have recently been investigated for waveguide use in the infrared. Calculations, however show relatively low  $\lambda_0$  values of 1.91, 1.81, and 1.79  $\mu$ m for three typical compositions: 57.5 HfF<sub>4</sub>, 33.75 BaF<sub>2</sub>, 8.75 LaF<sub>2</sub><sup>5</sup>; 40 BaF<sub>2</sub>, 60 ZrF<sub>4</sub>, and 10 GdF<sub>3</sub>, 60 ZrF<sub>4</sub>, 30 BaF<sub>2</sub>, <sup>6</sup> respectively. Thus  $\lambda_0$  values of fluoride glasses will probably not be significantly larger than the 1.8  $\mu$ m of GeO<sub>2</sub> with its much lower reactivity and simpler chemistry. In the absence of the discovery of new glass-forming fluoride systems, prospects for fluoride glasses with  $\lambda_0$  in the region beyond 2  $\mu$ m are therefore not very promising.

In the case of chlorides, bromides, and iodides there is only one known glass former, ZnCl<sub>2</sub>. Some ZnCl<sub>2</sub> fiber has been prepared,<sup>4</sup> but because of its hygroscopicity, only very limited studies have been performed so far. Weak glass formers leading to mixed halide glasses may exist here, as yet undiscovered. The existence of glass formation in mixed oxyhalides based on a combination of Figs. 1 and 2 is another possibility by analogy with lead oxyfluoride glass.<sup>22</sup>

# V. MATERIALS PROPERTIES

Given the above conditions, a potentially useful glass for a low-loss infrared optical waveguide would need to be readily melted and worked, stable to devitrification and phase separation, capable of being adequately purified at a not unreasonable cost, and have the necessary mechanical and chemical properties, particularly adequate strength and low hygroscopicity. The ability to modify the refractive index without large changes in the thermal expansion is also needed for index profile adjustment to obtain minimum mode dispersion in multimode fibers.

In view of these considerations, Tables I and II have been generally limited to those oxides, fluorides, chlorides, and bromides with  $E>4\frac{1}{2}$  which can be melted without excessive decomposition. Fluorides seem to have  $\lambda_0$  values that are too low and tend to be highly reactive and iodides are too unstable to be serious contenders, except possibly in small amounts for refractive index adjustment. A large hygroscopicity, as in  $ZnCl_2$ , can be a barrier, but such a substance may still be a useful

component in a mixed composition. The high toxicity of some elements, such as Tl, As, and some fluorides, are strong minuses. Chalcogenides and pnictides tend to be soft, brittle, somewhat reactive during preparation, and have low E and bandgap values.

#### VI. SUMMARY

We have calculated the wavelength for the material dispersion zero, i.e., that wavelength for which a multimode lightguide would function at highest bandwidth, for oxides and halides using fundamental properties. The results, as presented in the tables and figures, can be used to direct the search for long-wave lightguides, subject to a number of materials considerations.

#### VII. ACKNOWLEDGMENTS

It is a pleasure to thank S. Wemple for extended discussions and for his cooperation in estimating E and f values, and D. L. Wood for helpful comments on the manuscript.

#### **APPENDIX**

### Material Constraints on Ultralow-Loss Infrared Optical Waveguides

## A1. Types of fibers

Many types of optical waveguide fibers have been proposed, including liquid filled ones.<sup>23</sup> Glass has demonstrated its utility, but the use of single crystals and even of polycrystalline materials and plastically deformable Ag and Tl halides has also been proposed. In view of the basic stepped nature of single crystal surfaces and of the inherent scattering at grain and subgrain boundaries in the other nonvitreous materials, the applicability of material other than glass to ultralowloss fibers remains highly questionable.

#### A2. Bulk losses

Four fundamental types of losses are important<sup>24</sup>:

- (i) the electronic transition ultraviolet absorption tail loss (Urbach behavior, main optical absorption edge);
- (ii) the scattering loss, derived from both intrinsic and extrinsic inhomogeneities, dominant on the UV side caused by the  $\lambda^{-4}$  Rayleigh variation (Raman and Brillouin effects are not important<sup>24</sup>);
- (iii) the impurity losses, predominantly from "water" and 3d transition metal impurities, although rare earth metals may be more important in the infrared<sup>1</sup>; and
- (iv) the vibrational states IR absorption tail losses (reststrahlen, highest energy optical phonon).

Both the UV and the IR loss regions show an exponential drop-off

with energy, thus creating a "window," with minimum loss at the crossover point (in the absence of impurity losses). Values as low as 10<sup>-6</sup> dB/km are indicated by extrapolations (which are not necessarily valid). Ideally, this minimum-loss wavelength would also be close to the location of  $\lambda_0$  for an ultralow-loss long-distance fiber link.

A number of general principles for minimizing the various losses can he listed. For minimum uv tail and scattering losses, a large bandgap in a material with a low glass-transition temperature, a one or few component system, low isothermal compressibility, an open structure with a low density of anions and a low formal valence on the anion. and low coordination with a short nearest-neighbor bond length might be best, 4,25 For a minimum IR tail, a large bandgap, heavy atoms with weak (covalent) bonding, and low melting and glass-transition temperatures would lead to lower frequency vibrational states; most important is a very low anharmonicity so as to minimize overtones. Low impurity levels and a good homogeneity are obvious. Inevitably some of these preferences are mutually incompatible, particularly the high bandgap favoring both low uv and IR tails, as against a low E (a function of the bandgap) lowering  $\lambda_0$  in eqs. (1) and (2).

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