Temperature dependence of the refractive index of optical fibers*

Wang Zhi-Yong(王智勇)†, Qiu Qi(邱 琪), and Shi Shuang-Jin(史双瑾)

School of Optoelectronic Information, University of Electronic Science and Technology of China, Chengdu 610054, *China*

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Many experimental investigations on the temperature dependence of the refractive index of optical fibers have been reported previously, however a satisfying theoretical explanation for it is still absent. In this paper, a theoretical model about the temperature dependence of the refractive index of optical fibers is presented and it is in agreement with the previous experimental results. This work is a significant reference for the research and development of temperature sensors based on optical fiber delay lines.

Keywords: refractive index, optical fiber, temperature sensor

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1. Introduction

Based on the fact that the larger the refractive index of a medium is, the slower the light travels in the medium, a fiber optic sensor can be devised by means of the temperature dependence of the refractive index of an optical fiber. Usually, an optical fiber has a core made of fused silica (a non-crystalline form of silicon dioxide SiO_2). In fact, the refractive indices of media and optical sensors are two important issues in many research studies,[1–12] while the investigations on the fiber optic sensors combine the two issues.

As far as the temperature dependence of the refractive index of fused silica is concerned, many experimental investigations on it have been presented (see for example, Refs. [13]–[17]), and they have shown that the refractive indices of fused silica are directly proportional to the temperature with the temperature coefficients lying in the range of $[5\times10^{-6}, 3\times10^{-5}]$ (K^{-1}) when the temperature is in the range from roughly 100 K to 800 K. On the other hand, the corresponding theoretical interpretation is still insufficient. For example, in Ref. [18], the author tried to develop the most elementary possible model for the temperature dependence of the refractive index of fused silica, and his model was based on a complex dielectric constant whose imaginary part was assumed to satisfy an analytical model. However, as we know, the fused silica is an insulating medium, its dielectric constant is usually taken as a real number, not to mention the validity of the analytical model yielded by the author's assumption. In fact, just as the author himself said, his theoretical result was about 3.4 times smaller than the measured value. In Ref. [19], the authors presented a theoretical interpretation for the temperature dependence of the refractive index of an optical fiber in terms of the thermal effects of the refractive index caused by the electron–phonon interaction. However, their investigation is independent of the structure details of solids, so their result should be valid for any solid, which does not agree with the fact that the refractive indices of some solids are not directly proportional to the temperature within the same temperature range. In contrast with those works, in this paper, starting from the first principle and taking into account as much as possible the structure details of fused silica, we present a theoretical investigation on the temperature dependence of the refractive index of fused silica. It should be pointed out that there have been a few theoretical investigations on the temperature dependences of the refractive indices of some monatomic crystals and diatomic ionic crystals.[1–7] However, those theoretical models are not valid for the fused silica. For example, contrary to the fused silica, those monatomic crystals or diatomic ionic crystals' refractive indices can be inversely (rather than directly) proportional to the temperature within the same temperature range.

2. Some general considerations

As for the amorphous phase of SiO_2, the fused silica consists of a non-repeating network of tetrahedra and differs from the crystalline phase only by the missing long-range order, but there remains significant ordering at length scales well beyond the Si–O bond length. One example of this ordering is found in the preference of the network to form rings of 6-tetrahedra.[20] As we know, the building block of the silica is the SiO_4 unit, each individual SiO_4 tetrahedron is connected with adjacent tetrahedra at the corners, forming a three-dimensional structure. The solid fused silica can equivalently be taken as consisting of nonlinear SiO_2 molecules (the length of the Si–O bond is about 0.16 nm), where the atoms are arranged in an aperiodic pattern void of long-range order.

Because of the absence of the simplifications associated with periodicity, up to now, people's understanding of amor-

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†Corresponding author. E-mail: zywang@uestc.edu.cn

phous materials still remains far from complete. However, the essential features of the electronic structure, and thereby also the macroscopic properties, are determined by short-range orders, so these properties are similar for solids in crystalline and amorphous states. Moreover, the medium made of fused silica can be divided into many small portions with the same size, all the portions are identical in the sense of average, and they can be called equivalent primitive cells.

At any given temperature, atoms in a solid vibrate with small amplitudes about their equilibrium positions, which implies that there is an attractive potential energy in the solid. On the other hand, the stability of the lattice requires a repulsive potential, and hence a repulsive potential energy. The atomic vibrations are also affected by the vibrations of adjacent atoms through bonding, and the result is elastic waves (phonons) that propagate through the solid. It is important to mention that, as a result of polarization, some of the energy may be converted into elastic deformations (phonons). In such a way, the phonon interactions in a medium make a contribution to the refractive index of the medium.

In general, there are four types of dielectric behaviors.[21] 1) The polarization of the electronic cloud around the atoms: when an external electric field is applied, the electronic clouds are distorted, the resulting polarization is directly related to the dielectric constant. 2) The motion of the charged ions: this effect is primarily of interest in ionic crystals in which the positive and negative ions can move with respect to one another and thus polarize the crystal. 3) The rotation of molecules with permanent dipole moments: in an electric field, the dipoles tend to line up with the electric field, while the thermal effects tend to oppose this alignment, and so, the phenomenon is temperature dependent (this type of dielectric behavior is mostly relevant for liquids and gases). 4) The dielectric screening of a quasi-free electron gas (this phenomena is of interest for metals).

As far as the fused silica is concerned, it exhibits both ionic and covalent bondings. The electronegativity of silicon is 1.8 and that of oxygen is 3.5,[22] then the fraction of the covalent bonding is $\exp[-(3.5-1.8)^2/4]=0.486$. Therefore, the fused silica possesses the aforementioned dielectric behaviors 1) and 2). On the other hand, the fused silica under consideration is solid and dielectric, so its dielectric behaviors 3) and 4) can be ignored. In our case, a time-varying electric dipole moment can be caused by a fixed ion core with positive charge and a vibrating electron cloud with negative charge. Therefore, in the following we will study the temperature dependence of the refractive index of fused silica based on the dielectric behaviors 1) and 2) together.

3. Temperature dependence of the refractive index of fused silica

When an optical fiber made of fused silica is placed in an external electric field (e.g., the electric field component of an electromagnetic wave propagating along the optical fiber, it is uniform over an Si–O bond), the intensity of polarization is proportional to the total macroscopic field $\boldsymbol{E}$ in the medium $\boldsymbol{P}=\varepsilon_0\chi\boldsymbol{E}$, where χ is the electric susceptibility of the medium, ε_0 is the vacuum permittivity, and the relative permittivity of the material is $\varepsilon_{\rm r}=1+\chi$. Because the fused silica is a non-magnetic material, its refractive index is $n=\sqrt{\varepsilon_{\rm r}}=\sqrt{1+\chi}$. On the other hand, let N denote the average number of Si–O bonds along the direction of a local electric field $\boldsymbol{E}_{\rm c}$ per unit volume (for example, if the angle between the direction of $\boldsymbol{E}_{\rm c}$ and that of an Si–O bond is θ, then this Si–O bond contributes $\cos\theta$ to the number, it follows that it is not necessary for N to be an integer), we have $\boldsymbol{P}=N\alpha\boldsymbol{E}_{\rm c}$, where α is the polarizability. For convenience, let us assume that the space allotted to each Si–O bond is a sphere, approximately, we can obtain $\boldsymbol{E}=(1-N\alpha/3\varepsilon_0)\boldsymbol{E}_{\rm c}$. Using $\boldsymbol{P}=\varepsilon_0\chi\boldsymbol{E}=N\alpha\boldsymbol{E}_{\rm c}$, we can obtain the Lorentz–Lorenz formula (also named the Clausius–Mossotti relation)

$$\frac{3\varepsilon_0}{N}\left(\frac{n^2-1}{n^2+2}\right)=\alpha. \tag{1}$$

As mentioned before, the fused silica can equivalently be regarded as consisting of nonlinear SiO_2 molecules. For a nonlinear molecule consisting of l atoms, there are $3l-6$ displacements corresponding to the vibrations of the molecule, which implies that there are three displacements corresponding to the vibrations of a nonlinear SiO_2 molecule. Moreover, the potential energy of a nonlinear polyatomic molecule depends on all the displacements of the atoms from their equilibrium positions, in terms of a Taylor expansion relative to the equilibrium position of $\boldsymbol{x}=(x_1,x_2,\ldots)=(0,0,\ldots)\equiv 0$, we have

$$\begin{aligned} V &= V(0)+\sum_i(\partial V/\partial x_i)_0 x_i \\ &\quad +(1/2)\sum_{i,j}(\partial^2 V/\partial x_i\partial x_j)_0 x_i x_j+\cdots. \end{aligned} \tag{2}$$

The sum is over all $3l$ displacements of the l atoms, so some displacements (those corresponding to the translation and rotation of the molecule as a whole) will turn out to have zero force constant. Therefore, for small displacements from the equilibrium positions, we have

$$V=(1/2)\sum_{i,j}k_{ij}x_i x_j,\quad k_{ij}=(\partial^2 V/\partial x_i\partial x_j)_0, \tag{3}$$

where k_{ij}'s are the generalized force constants. In terms of the mass-weighted coordinates $q_i=\sqrt{m_i}x_i$, with m_i the mass of the atom being displaced by x_i, the potential energy can be

rewritten as

$$V = (1/2)\sum_{i,j} K_{ij} q_i q_j,$$
$$K_{ij} = k_{ij}/\sqrt{m_i m_j} = (\partial^2 V/\partial q_i \partial q_j)_0. \quad (4)$$

The total kinetic energy becomes

$$T = (1/2)\sum_i m_i \dot{x}_i^2 = (1/2)\sum_i \dot{q}_i^2, \quad (5)$$

where the dot represents the differentiation with respect to time. The classical expression for the total energy is therefore

$$H = (1/2)\sum_i \dot{q}_i^2 + (1/2)\sum_{i,j} K_{ij} q_i q_j. \quad (6)$$

The Hamiltonian can be rewritten in terms of the normal coordinates to remove the cross terms (we just pay attention to the projections of the normal coordinates onto the direction of the local electric field $\boldsymbol{E}_c$ so that the triatomic molecule SO_2 is equivalent to a linear molecule), namely,

$$\begin{cases} Q_1 = (\sqrt{m_O} q_1 + \sqrt{m_S} q_2 + \sqrt{m_O} q_3)/\sqrt{m}, & \kappa_1 = 0, \\ Q_2 = (q_1 - q_3)/\sqrt{2}, & \kappa_2 = k/m_O, \\ Q_3 = (\sqrt{m_S} q_1 - 2\sqrt{m_O} q_2 + \sqrt{m_S} q_3)/\sqrt{2m}, & \\ \qquad \kappa_3 = km/m_S m_O, & \end{cases} \quad (7)$$

where $m = m_S + 2m_O$ is the total mass of a SO_2 molecule. For the moment, the total energy can be expressed in the form

$$H = (1/2)\sum_i \dot{Q}_i^2 + (1/2)\sum_i \kappa_i Q_i^2. \quad (8)$$

As a result, we can study the vibrations in fused silica starting from the model of harmonic oscillators (or with an anharmonic term as a correction).

The fused silica exhibits both ionic and covalent bondings, the interaction energy of a Si–O bond can be expressed as

$$u(r) = -\frac{Mq^2}{4\pi\varepsilon_0}\left(\frac{1}{r} - \frac{r_0^{s-1}}{sr^s}\right), \quad (9)$$

where r_0 is the equilibrium distance between two adjacent Si and O atoms (it is equal to the length of the Si–O bond, i.e., $r_0 = 1.6 \times 10^{-10}$ m),[23] $M = 1.638$ is the Madelung constant, and s is the repulsive-energy parameter. There are two valence electrons in the Si–O bond, and the fraction of the ionic bonding is 0.514, then the total electric quantity can be taken as $q = 0.514 \times 2 \times 1.602 \times 10^{-19}$ C $\approx 1.647 \times 10^{-19}$ C. As we know, the repulsive-energy parameter satisfies the formula

$$s = 1 + 36\pi\varepsilon_0 \beta r_0^4 B_m / Mq^2, \quad (10)$$

where $\beta = 3.34$ and $B_m = 3.7 \times 10^{10}$ Pa (i.e., the bulk modulus) for silica. Substituting all the known quantities and $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m into Eq. (10), we have $s \approx 2.825$. The wavelength of the electromagnetic wave propagating along the optical fiber is $\lambda = 633$ nm.

When the distance between two adjacent Si and O atoms becomes $r = r_0 + \Delta r$ with Δr being very small, by means of the Taylor expansion, the interaction energy $u(r)$ can be written approximately as

$$u(r) \approx u(r_0) + \frac{Mq^2(s-1)}{8\pi\varepsilon_0 r_0^3}(\Delta r)^2 - \frac{Mq^2(s-1)(s+4)}{24\pi\varepsilon_0 r_0^4}(\Delta r)^3. \quad (11)$$

For the moment, the function $U(x) = U(\Delta r) = u(r) - u(r_0)$ is the potential energy of an oscillator with the equilibrium position of $r = r_0$ and the displacement of $x = \Delta r$, that is,

$$U(x) = \frac{Mq^2(s-1)}{8\pi\varepsilon_0 r_0^3}x^2 - \frac{Mq^2(s-1)(s+4)}{24\pi\varepsilon_0 r_0^4}x^3 \equiv \frac{1}{2}ax^2 - bx^3, \quad (12)$$

where

$$a = \frac{Mq^2(s-1)}{4\pi\varepsilon_0 r_0^3} = 1.779 \times 10^2 \text{ J}\cdot\text{m}^{-2},$$
$$b = \frac{Mq^2(s-1)(s+4)}{24\pi\varepsilon_0 r_0^4} = 1.265 \times 10^{12} \text{ J}\cdot\text{m}^{-3}. \quad (13)$$

Because of $a > 0$ and $b > 0$, Eq. (12) shows that the anharmonic term $-b(\Delta r)^3$ contributes a repulsive energy, which is in agreement with the fact that, in a solid, it not only requires an attractive potential energy for atoms to vibrate about their equilibrium positions, but also requires a repulsive potential to keep the stability of the lattice. Under the local electric field $\boldsymbol{E}_c$, the total energy of an oscillator can be expressed as

$$H = p^2/2\mu + U(x) - qE_c x, \quad (14)$$

where $p = |\boldsymbol{p}|$, $\boldsymbol{p}$ is the momentum of the oscillator, and μ is the mass. Because N denotes the average number of Si–O bonds along the direction of $\boldsymbol{E}_c$ per unit volume, then there are N oscillators along the direction of $\boldsymbol{E}_c$ per unit volume (as mentioned before, it is not necessary for N to be an integer). According to statistical mechanics, under the conditions of thermal equilibrium, the probability that an oscillator has energy H is proportional to $\exp(-H/k_B T)$, where k_B is Boltzmann's constant and T is the absolute temperature. Then, the average intensity of polarization is

$$P = \frac{\int Nqx\exp(-H/k_B T)\mathrm{d}x}{\int \exp(-H/k_B T)\mathrm{d}x} = \frac{Nq\int x\exp[(-ax^2/2 + bx^3 + qxE_c)/k_B T]\mathrm{d}x}{\int \exp[(-ax^2/2 + bx^3 + qxE_c)/k_B T]\mathrm{d}x}. \quad (15)$$

For simplicity, let

$$\exp[(bx^3 + qxE_c)/k_B T] \approx 1 + (bx^3 + qxE_c)/k_B T$$

for small x. The integral $\int \exp(-ax^2/2k_B T)\mathrm{d}x$ is convergent for $x \in (-\infty, +\infty)$, and for large x, the integrand

$\exp(-ax^2/2k_BT)$ approaches to zero. According to quantum statistics, the displacement x in $\exp(-ax^2/2k_BT)$ (related to the resonant term), as the fluctuation of the equilibrium positions, is usually taken as $x \in (-\infty, +\infty)$. Then we have

$$P \approx \frac{Nq\int_{-\infty}^{+\infty} x[1+(bx^3+qxE_c)/k_BT]\exp(-ax^2/2k_BT)\mathrm{d}x}{\int_{-\infty}^{+\infty}[1+(bx^3+qxE_c)/k_BT]\exp(-ax^2/2k_BT)\mathrm{d}x}. \quad (16)$$

Using

$$\int_{-\infty}^{+\infty}\exp(-cx^2)\mathrm{d}x = \sqrt{\pi/c},$$

$$\int_{-\infty}^{+\infty} x^l\exp(-cx^2)\mathrm{d}x = \begin{cases} \dfrac{(l-1)!!}{\sqrt{(2c)^l}}\sqrt{\dfrac{\pi}{c}}, & l \text{ is even}, \\ 0, & l \text{ is odd}, \end{cases}$$

$$c>0,\ \ l=1,2,\ldots, \quad (17)$$

we can obtain

$$P = N\left(\frac{q^2}{a}+\frac{3qbk_B}{E_c}T\right)E_c \equiv N\alpha E_c, \quad (18)$$

where the polarizability is

$$\alpha = q^2/a + 3bqk_BT/a^2E_c \equiv A+BT, \quad (19)$$

with

$$A = q^2/a, \quad B = 3qbk_B/a^2E_c. \quad (20)$$

Using Eqs. (1) and (19), we have

$$\frac{3\varepsilon_0}{N}\left(\frac{n^2-1}{n^2+2}\right) = A+BT. \quad (21)$$

For $BT \ll A$ (as shown later), using Eq. (21) we have

$$\begin{aligned} n^2 &= 1+\frac{3N(A+BT)}{3\varepsilon_0-N(A+BT)} \\ &\approx 1+\frac{3NA}{3\varepsilon_0-NA}+\frac{3NBT}{3\varepsilon_0-NA}. \end{aligned} \quad (22)$$

Likewise, because of $BT \ll A$, the third term on the right-hand side of Eq. (22) is far smaller than the sum of the first and the second terms, then we can obtain

$$n \approx \sqrt{\frac{3\varepsilon_0+2NA}{3\varepsilon_0-NA}}+\frac{3NB}{2\sqrt{(3\varepsilon_0+2NA)(3\varepsilon_0-NA)}}T. \quad (23)$$

Substituting Eq. (20) into Eq. (23) yields

$$\begin{aligned} n \approx & \sqrt{\frac{3\varepsilon_0 a+2Nq^2}{3\varepsilon_0 a-Nq^2}} \\ & +\frac{9Nqbk_B}{2aE_c\sqrt{(3\varepsilon_0 a+2Nq^2)(3\varepsilon_0 a-Nq^2)}}T. \end{aligned} \quad (24)$$

Equation (24) implies that the refractive index of fused silica is directly proportional to the temperature T. Obviously, the temperature-dependent term vanishes for $b=0$, and it is related to the anharmonic term in Eq. (12), which is related to the phonon–phonon interactions.

Now, let us present a quantitative consideration. As mentioned before, N denotes the average number density of the Si–O bonds along a given direction (e.g., the direction of a local electric field $\boldsymbol{E}_c$), where the length of each Si–O bond is about $r_0 = 1.6\times10^{-10}$ m, then we can express N as $N = \eta/r_0^3$ with $0<\eta<1/3$. An advisable value is $\eta = 0.2$ for the moment $N = 4.883\times10^{28}$ m^{-3}. The amplitude of the local electric field is about $E_c = 1\times10^{11}$ V·m^{-1}. Substituting $\varepsilon_0 = 8.854\times10^{-12}$ F·m^{-1}, $a = 1.779\times10^2$ J·m^{-2}, $b = 1.265\times10^{12}$ J·m^{-3}, $q = 1.647\times10^{-19}$ C, $N = 4.883\times10^{28}$ m^{-3}, $k_B = 1.381\times10^{-23}$ J·K^{-1}, and $E_c = 1\times10^{11}$ V·m^{-1} into Eq. (24), we can obtain

$$n \approx 1.47269 + 7.09780\times10^{-6}T. \quad (25)$$

Equation (25) is in agreement with the previous experimental results. For example, the experimental result presented in Ref. [17] is

$$n \approx 1.47219\pm2.2\times10^{-4}+(7.83\pm0.99)\times10^{-6}T. \quad (26)$$

The theoretical and experimental results are compared in Fig. 1.

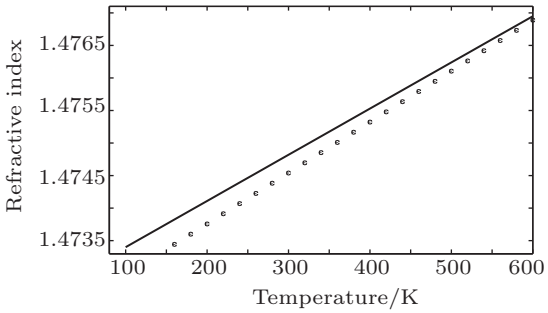

Fig. 1. Comparison between the theoretical (solid line) and experimental (dots) results.

Note that the temperature T must be lower than the maximal service temperature of fused silica (about 1200 K), which is the precondition of the investigation. In fact, in the previous experimental investigations, the temperature T is usually lower than 800 K. Using Eq. (20), we can show that the condition of $BT \ll A$ is valid for $T < 1200$ K, then we can safely apply $BT \ll A$ to obtain Eqs. (22) and (23).

4. Discussion and conclusion

Because of the absence of the simplifications associated with periodicity, up to now people's understanding of amorphous materials still remains far from complete. In particular, though there are many experimental investigations on the temperature dependence of the refractive index of fused silica, an

appropriate theoretical interpretation for the experimental results is still absent. In this paper, we present a theoretical interpretation for the experimental result. In high temperatures, the refractive index of fused silica is directly proportional to the absolute temperature. There are some differences among the results from different experimental reports, however most of them show $5 \times 10^{-6}\ \mathrm{K}^{-1} \leq \mathrm{d}n/\mathrm{d}T \leq 3 \times 10^{-5}\ \mathrm{K}^{-1}$. Therefore, our theoretical result is in good agreement with the previous experimental results.

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