Polarization study of photoinduced second-harmonic generation in glass optical fibers

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By studying the polarization properties of photoinduced second-harmonic generation in a germania-doped silica glass optical fiber we show that the induced $\chi^{(2)}$ is consistent with being due to the $\chi^{(3)}$ of the glass acted on by an internal dc electric field. Additional arguments are provided to support the view that the observed $\chi^{(2)}$ is not simply due to the microscopic $\chi^{(2)}$ of ordered dipoles.

Although it has been four years since the first report [1] of efficient second-harmonic generation (SHG) in glass optical fibers, there is in our opinion no convincing model for the origin of the effect. Those models which have been put forward can be roughly divided into two categories. In some models [2–5] it is proposed that an internal dc [6–8] electric field is set up which, acting on the $\chi^{(3)}$ of the glass, serves as an effective $\chi^{(2)}$. In other models [6–8] it is proposed that the macroscopic $\chi^{(2)}$ originates from an orienting of dipoles ("defects") in the glass (perhaps as a result of some internal dc electric field). In this paper we examine the polarization properties of the second-harmonic light generated by a germania-doped fiber. From the observed behavior we deduce that the effective $\chi^{(2)}$ of the fiber behaves as a $\chi^{(3)}$ of a nondispersive isotropic material (glass) acted on by a transverse dc electric field. Invoking also some supplementary arguments we therefore conclude that we are likely dealing with an electric-field-induced second-harmonic generation (EFISH) phenomenon. We do not assume any particular microscopic model for the origin of the $\chi^{(2)}$, or the phase matching, nor do we propose one. Rather, it is our intention to provide a clear focus in the search for a suitable model.

For a prepared fiber possessing a $\chi^{(2)}$ we may write

$$P_i^{2\omega} = \tfrac{1}{2}\chi_{ijk}^{(2)} E_j^{\omega} E_k^{\omega}, \qquad (1)$$

where $P^{2\omega}$ is the induced polarization at 2ω, E^{ω} is the electric field amplitude of the laser at the fundamental frequency ω, and the factor of $1/2$ is in accord with the convention of refs. [9,10], to which we adhere throughout this work. An SHG measurement is actually sensitive to an effective $\chi^{(2)}$, which represents a spatial average along the length, L, of the fiber,

$$\chi_{ijk}^{\mathrm{eff}} \equiv \frac{1}{L}\int_0^L \chi_{ijk}^{(2)}(z)\exp(-\mathrm{i}\Delta\kappa z)\,\mathrm{d}z\,. \qquad (2)$$

Here $\Delta\kappa = \kappa_{2\omega} - \kappa_{\omega}$ is the usual wavevector mismatch between the second harmonic and the fundamental. If the $\chi^{(2)}$ is phase-matched then $\chi^{(2)}(z) \propto \exp(\mathrm{i}\Delta\kappa z)$, and χ^{eff} is simply the amplitude of the $\chi^{(2)}$ grating. For our purposes we will ignore the distinction between $\chi^{(2)}$ and χ^{eff} on the assumption that all of the active tensor components of the $\chi^{(2)}$ have the same spatial dependence.

Since the longitudinal optical fields are negligible for our single mode fiber [11] there are at most six independent components of the $\chi^{(2)}$; they can be accessed in two experiments. Let us suppose that a fiber is prepared for SHG by external seeding in the usual fashion [6], with the linearly polarized fundamental and the seed second-harmonic beams copolarized (along the x-axis), and propagating in the z direction. After preparation, we measure the gen-

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erated SHG intensity (with no seed beam present) after an analyzing polarizer, while we vary the polarization angle, θ, of the fundamental, relative to the x-axis. We consider two cases; that of the analyzer parallel to the polarization of the original writing beam (along x), and that of the analyzer perpendicular (along y). We write

$$|E_x^{2\omega}|^2 \propto |\chi_{xxx}\cos^2\theta + 2\chi_{xxy}\cos\theta\sin\theta + \chi_{xyy}\sin^2\theta|^2 ,$$
$$|E_y^{2\omega}|^2 \propto |\chi_{yyy}\sin^2\theta + 2\chi_{yyx}\cos\theta\sin\theta + \chi_{yxx}\cos^2\theta|^2 , \quad (3)$$

where $E^{2\omega}$ is the electric field amplitude at 2ω. In principle these tensor elements can be complex quantities, but we will assume that we are sufficiently far from any resonances in the problem that they may all be taken to be real. Therefore the form of the $\chi^{(2)}$ tensor can be determined by studying the polarization properties of the SHG from the prepared fiber.

Let us consider what one might expect to find in different cases. First, suppose, as other workers have [6–8], that the SHG is ultimately caused by the macroscopic orientation of dipoles, each possessing some microscopic $\chi^{(2)}$. If there is to be some preferred axis (as there must ultimately be, once a macroscopic $\chi^{(2)}$ is generated), then one might expect it to be the x-axis, as this axis is distinguished during the writing process in our experiment. In this case oriented dipoles would point preferentially along the $\pm x$-axis. Such dipoles may have a complicated microscopic symmetry, but if they are distributed symmetrically about this preferred direction (as would be the case for a "gas" of orientable dipoles), it is straightforward to show [12] that $\chi^{(2)}_{xxy} = \chi^{(2)}_{yxx} = \chi^{(2)}_{yyy} = 0$. This would also be the case for perfectly oriented dipoles if the dipoles had the symmetry of a pencil ($C_{\infty v}$) or, more generally, whenever the x–z plane of the fiber is a mirror plane. Specific relationships between the nonzero components that remain depend on the microscopic symmetry of the orientable dipoles, and on their degree of alignment.

As an alternative approach, we may assume that there is an internal dc electric field, E^{dc}, that acts on the $\chi^{(3)}$ of the glass, yielding an effective $\chi^{(2)}$ in the EFISH sense. In this case we write

$$\tfrac{1}{2}\chi^{(2)}_{ijk}(\omega,\omega) = \tfrac{3}{2}\chi^{(3)}_{ijkl}(\omega,\omega,0)E_l^{dc} . \quad (4)$$

It again seems reasonable to assume that such a dc electric field points along the $\pm x$-axis. Since the glass is macroscopically isotropic (with regards its $\chi^{(3)}$), we again have [12] $\chi^{(2)}_{xxy} = \chi^{(2)}_{yxx} = \chi^{(2)}_{yyy} = 0$. It can also be shown [10] that $\chi^{(2)}_{xxx} = 2\chi^{(2)}_{yyx} + \chi^{(2)}_{xyy}$. If we further assume that the $\chi^{(3)}$ possesses Kleinman symmetry [10] (valid for a nondispersive medium) we can finally make the specific predictions that $\chi^{(2)}_{yyx} = \chi^{(2)}_{xyy}$, and also $\chi^{(2)}_{xxx} = 3\chi^{(2)}_{xyy}$. These predictions will be verified below.

The fiber that we have studied is a step-index germania-doped silica fiber (Lightwave Technologies type F0500C), of 3 μm core diameter, with a numerical aperture of 0.11. The cladding is pure silica, the core is 3 mol% GeO_2 doped silica. The fiber is single mode at both the fundamental wavelength (1.064 μm) *and* the second-harmonic wavelength (0.532 μm). This is essential to being able to do good polarization studies. Propagation in the fundamental mode (LP_{01}) far from the cutoff wavelength (440 nm) also ensures that the optical fields are very close to transverse [11]. By using a short length (10.5 cm) of fiber, mounting it without bends or twists, and paying careful attention to removing the cladding modes (using glycerin on the outside of the fiber as a cladding mode stripper), the fiber maintains polarization at both ω and 2ω. This was determined by measuring the extinction coefficient of the system (fiber plus microscope objectives) between crossed polarizers. The extinction ratio was 10^{-3} at ω, and 6×10^{-3} at 2ω.

The fiber was prepared for second-harmonic generation in the usual way [6]. The laser (Quantronix 416) was operated cw mode-locked at 76 MHz, with 110 ps pulses at 1.064 μm. The fiber was seeded with the second harmonic of the YAG, generated in a KTP crystal. The seed average powers were 0.9 W at 1.064 μm (corresponding to a peak power of order 100 W), and 0.02 W at 0.532 μm, both measured after the fiber. The fiber was seeded for two hours, with both the infrared and the seed green copolarized, as ensured by the polarizer placed before the fiber, which also serves to define the x-axis. We have never observed this fiber to self-prepare (that is, to generate second-harmonic without first seeding with green). This is consistent with previous work [6], in which

it was pointed out that quadrupole SHG, which may serve as a seed in a self-preparing fiber, is not supported in a single mode fiber. By operating cw mode-locked, keeping the fiber short and the peak power low, it was possible to avoid cross-phase modulation effects [13] that would affect the accuracy of the polarization measurements. However, these preparation conditions result in only weak generated second harmonic, necessitating the use of a chopper and lock-in amplifier for accurate measurements during the reading phase. To study the prepared fiber a Schott Glass OG560 long wave pass filter was inserted before the fiber, in order to block the seed green light. As a result, the "reading" infrared power (measured after the fiber) was 0.65 W, resulting in a peak measured conversion efficiency to the second harmonic of 2×10^{-7}.

In the reading phase of the experiment two cases were considered. In the first case (corresponding to eq. (3a)), the analyzing polarizer, placed after the fiber, was oriented to be precisely parallel to the polarizer which preceded the fiber. Then a computer controlled $\lambda/2$ plate (at 1.064 μm) was used to rotate the polarization of the incident fundamental in 10° increments. For each incident polarization angle the time averaged second-harmonic signal, as measured with the lock-in amplifier, was recorded by the computer. In the second case (corresponding to eq. (3b)) the analyzer was set to be precisely perpendicular to the polarizer, and the incident polarization was again rotated by the computer in the same way as for the first case. In this way it was possible to determine the generated second-harmonic intensity as a function of the infrared polarization angle, θ.

The data for the case of the analyzer parallel to the polarizer, and then perpendicular to the polarizer, are shown in fig. 1 and 2 respectively. The solid line fits are discussed below. In each case the polarization angle (at ω) on the horizontal axis refers to the reading infrared beam, as measured relative to the writing polarization angle (at 0°). While the generated second-harmonic intensity, shown on the vertical axis, is given in arbitrary units, the vertical axes in figs. 1 and 2 may be directly compared. The absolute signal in the perpendicular case is an order of magnitude smaller than in the parallel case. We have performed a nonlinear least squares fit of eq. (3a) to the

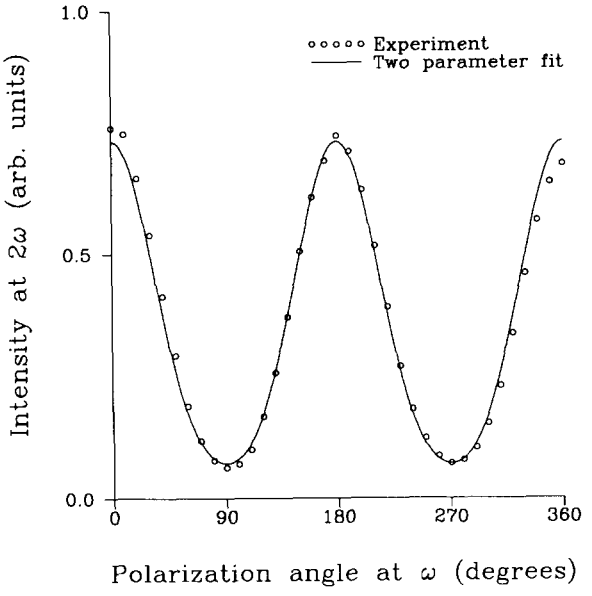

Fig. 1. Second-harmonic signal as a function of the infrared polarization angle for the analyzer set parallel to the polarizer. The solid line fit is based on eq. (3).

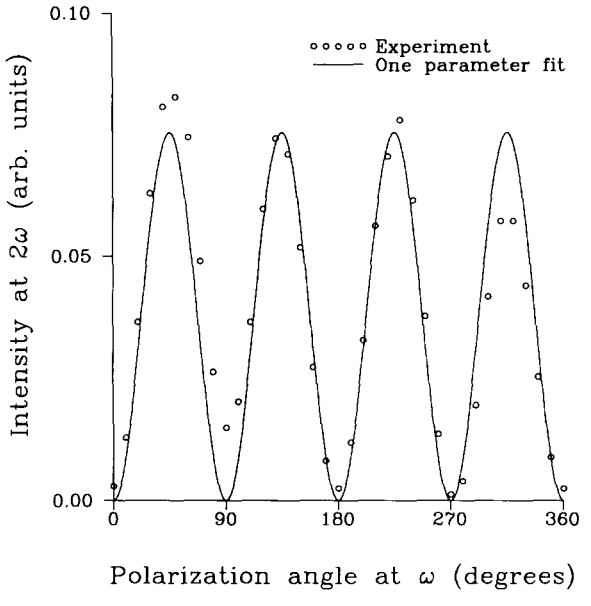

Fig. 2. As in fig. 1 but now the analyzer is perpendicular to the polarizer.

data of fig. 1, and of eq. (3b) to the data of fig. 2. In each case, assuming $\chi^{(2)}$ is real, we have a three-parameter fit.

The results (in arbitrary units) are, $\chi^{(2)}_{xxx} = 0.8549 \pm 0.0032$, $\chi^{(2)}_{xxy} = 0.0221 \pm 0.0036$, and $\chi^{(2)}_{xyy} = 0.2672 \pm 0.0068$, for the parallel case, and $\chi^{(2)}_{yyy} = 0.0229 \pm 0.0067$, $\chi^{(2)}_{yyx} = 0.2737 \pm 0.0035$, and $\chi^{(2)}_{yxx} = 0.0066 \pm 0.0067$ for the perpendicular case. The stated uncertainties come out of the fitting procedure. As we have noted above, one would expect that $\chi^{(2)}_{xxy} = \chi^{(2)}_{yyy} = \chi^{(2)}_{yxx} = 0$. The $\chi^{(2)}_{yxx}$ component is clearly zero to within the estimated experimental uncertainty. The $\chi^{(2)}_{xxy}$ and $\chi^{(2)}_{yyy}$ components are both less than 3% of the largest component. While this is still larger than the uncertainties estimated based on the quality of the fits, this uncertainty does not take into account possible systematic errors, discussed below. It appears that the assumption that these components may be neglected is reasonable. If we do so we are left with a simple one-parameter fit in the perpendicular case, and a two-parameter fit for the parallel case. The results of these fits are shown as solid lines in figs. 1 and 2, where now we find that $\chi^{(2)}_{xxx} = 0.8550 \pm 0.0046$, $\chi^{(2)}_{xyy} = 0.2679 \pm 0.0097$, and $\chi^{(2)}_{yyx} = 0.275 \pm 0.004$. Note that these are consistent with the values arrived from the full three-parameter fits. These values may now be compared with what we would expect for the specific case of the EFISH picture. The germania-doped silica glass may, in the first instance, be assumed to have a nondispersive $\chi^{(3)}$. This would lead to the so-called Kleinman symmetry, in which $\chi^{(2)}_{xyy} = \chi^{(2)}_{yyx}$. We see that this is consistent with our measurements. Further, by combining Kleinman symmetry with the isotropy of the $\chi^{(3)}$ for glass, we would expect [9] that $\chi^{(2)}_{xxx}/\chi^{(2)}_{xyy} = 3$. We have measured $\chi^{(2)}_{xxx}/\chi^{(2)}_{xyy} = 3.19 \pm 0.12$. Note that Kleinman symmetry is in fact not an exact symmetry, which may account for the small deviation from 3 in this ratio. For pure fused silica we have independently measured this ratio in a bulk ESHG experiment at 1.064 μm, using apparatus similar to that described in ref. [14], and obtained a value of 3.0 ± 0.1. We believe that our results are therefore consistent with the $\chi^{(2)}$ being in fact due to the $\chi^{(3)}$ of the glass times an internal dc electric field.

Our fits may be subject to residual systematic error that has not been taken into account in our estimates of the experimental uncertainty. During the reading phase the generated second harmonic can be elliptically polarized in the fiber, but the extinction coefficient is not very sensitive to small perturbations in elliptical polarization. Further, for the case of the analyzer perpendicular to the polarizer, the signal levels were particularly weak, making the measurement more sensitive to the presence of scattered light.

Tom et al. [9] carefully measured $\chi^{(2)}_{xxx} = 5.3 \times 10^{-12}$ esu for an externally seeded fiber in a low peak power experiment using a cw mode-locked YAG laser. Using also $\chi^{(3)}_{xxxx} = 1.3 \times 10^{-14}$ esu for fused silica [14] (derived from nonlinear refractive index measurements), we estimate (from eq. (4)) that the dc electric field should be of order 4×10^4 V/cm. For the case of the laser Q-switched and model-locked [9] this value would be of order 10^5 V/cm. Thus far there does not appear to be a consistent model which explains the origin of such large dc electric field amplitudes.

There is no point group for which symmetry alone forces *uniformly* oriented dipoles, each possessing a microscopic $\chi^{(2)}$, to directly yield $\chi^{(2)}_{xxx}/\chi^{(2)}_{xyy} = 3$, although this ratio might arise fortuitously. On the other hand, one can always construct a distribution of dipoles for which this is so. Such a distribution would generally be highly artificial. However, there is an important case which might arise naturally in which the orienting of dipoles in the fiber could result in the same polarization properties as an EFISH process. Kielich [12] has studied the problem of a gas of noninteracting freely orientable dipoles (for example HCl gas at atmospheric pressure) in the presence of a static electric field. If the microscopic $\chi^{(2)}$ of the orientable dipoles obeys Kleinman symmetry, then in the weak dc electric field limit a polarization study would yield the same results as would an EFISH process. Although this calculation is not likely valid for a glass, where dipoles are not generally free, it is useful to consider this special case. Following Kielich we have, in our notation,

$$\chi^{(2)} = \left(\frac{\mu E_{\mathrm{dc}}}{10 k_{\mathrm{B}} T}\right)(N\beta)\,, \tag{5}$$

where k_{B} is Boltzmann's constant, T is the temperature, μ is the dipole moment, N is the number density of orientable dipoles, and β is the dominant component of the microscopic $\chi^{(2)}$. The first factor in eq. (5) is due to thermal disorientation in a gas. At room temperature, with the dc electric field es-

timated above (4×10^4 V/cm), and taking a conservatively large dipole moment corresponding to an electron and a proton separated by 1 Å, this factor is still only 1.6×10^{-3} (which justifies the assumption of a weak field limit). Using the results of Tom et al. [9] for the macroscopic $\chi^{(2)}$, and assuming that *every* SiO_2 and GeO_2 molecular unit in the glass is free to orient ($N=1\times10^{22}$ cm^{-3}), then the β would still need to be 3×10^{-31} esu, which is 5 times larger than the average β for crystal quartz (obtained by dividing the macroscopic $\chi^{(2)}$ of quartz by the number of SiO_2 molecular units)! This is clearly unrealistic. It is because of considerations such as these that we do not believe that orientation of "defects" (which would be present in even smaller numbers [15] than 10^{22} cm^{-3}) in the glass is the direct cause of the macroscopic $\chi^{(2)}$ that is induced, although it may be that some orientation of defects contributes to a macroscopic dc electric field.

In an early work [16] some qualitative polarization studies were carried out on a self-preparing fiber which was multimode at both ω and 2ω. It is difficult to draw specific conclusions from these results. Other workers [17], studying SHG in a high birefringence elliptical core bow-tie fiber, drew the "tentative conclusion" that no "unusual off-axis terms" in the $\chi^{(2)}$ tensor exist. This is consistent with our finding the $\chi^{(2)}_{xxy}$, $\chi^{(2)}_{yyy}$ and $\chi^{(2)}_{yxx}$ tensor elements are insignificant. More recently, the importance of self- and cross-phase modulation effects in determining the form of the phase match grating was clearly demonstrated by Ouellette [13]. As a by-product of that work it was estimated [#1] that the ratio $\chi^{(2)}_{xxx}/\chi^{(2)}_{xyy}=2.5$, in contrast to our value of 3.19 ± 0.12. However, the estimate of ref. [13] was obtained indirectly and required a model-dependent assumption about the form of the $\chi^{(2)}$ grating. Further, the fiber was multimode at 2ω, resulting in some depolarization of the green light, and rendering accurate polarization studies infeasible, as we have found in our own attempts with a multimode fiber. These points were already suggested in ref. [13]. We do not therefore believe that the discrepancy between our result and the cited estimate is significant. However, it is conceivable that at the much higher peak power densities used by Ouellette (about two orders of magnitude higher), the mechanism by which the $\chi^{(2)}$ is formed could be more complicated than at the low peak powers at which we have operated.

In a previous work [19] an external dc electric field was applied to a fiber during the preparation stage, in order to test a particular model for the origin of the $\chi^{(2)}$ grating (which would have predicted a small internal dc field). Although no statistically significant effect was observed, the applied field was only about 2500 V/cm in the fiber, which would not have adequately tested the presence of an internal dc field of order 4×10^4 V/cm. Further, as was noted in ref. [19], if there were free charge carriers present they could have shielded the externally applied field. There is therefore no contradiction with our present findings.

In conclusion we have demonstrated that the polarization properties of the photo-induced $\chi^{(2)}$ in a single mode germanosilicate fiber are consistent with being due to the $\chi^{(3)}$ of the glass, acted on by an internal dc electric field which is transverse to the fiber (along the axis defined by the optical electric field of the polarized seed light). We have not relied upon any particular microscopic theory. Our results are purely phenomenological in nature, and therefore should be quite general. It is hoped that these findings can now help to concentrate the search for a suitable model.

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[#1] In ref. [13] the stated ratio 6.3 actually applies to the ratio of intensities; the correct ratio should be 2.5 [18].

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