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**Nonlinear Index of Refraction of
Cu- and Pb-Implanted Fused Silica***

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ABSTRACT

We have measured the nonlinear index of refraction in high-purity fused silica implanted with Pb and Cu ions, using 100-ps pulses from a cw, mode-locked, frequency-doubled Nd:YAG laser ($\lambda = 532$ nm). The nonlinear index measured for the Pb-implanted silica is an order of magnitude larger than that for Cu:silica. Qualitative evidence from infrared reflectance spectra suggests that these differences may result from distinctive ways in which these implanted ions are incorporated into the glass network: the Cu appears to form of nm-size colloids, while the Pb may be incorporated into a Pb-O-Si glassy phase.

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Introduction

Optical signal processing will require nonlinear optical materials meeting exacting and not necessarily mutually intercompatible requirements: ultrafast switching times, high resistance to bulk and surface laser damage, low two-photon absorption, and, of course, large nonlinear indices of refraction, that is, large third-order optical nonlinearities.[1] In addition, nonlinear materials for optical switching should be manufacturable by processes compatible with microelectronics technology.

Recently we have initiated studies of the nonlinear optical behavior of ion-implanted fused silica. Our first studies [2] were carried out on Cu-implanted fused silica, because Cu is known to form colloids upon ion implantation in silica, and because noble-metal colloids - in particular, the gold colloids responsible for the beautiful colors of the ruby-gold glasses - are known to have large, ultrafast optical nonlinearities.[3] Apart from the known nonlinear behavior of metallic and dielectric nanoclusters, we are also motivated by the fact that ion implantation is already a standard technology for modifying the properties of semiconductors and optical waveguides, and thus fulfills one of the important criteria for a manufacturable waveguide or switching material.

In this paper, we compare the nonlinear index of refraction in Cu:silica and Pb:silica. We briefly consider whether the measured nonlinear index of refraction is thermal or electronic in origin, and adduce infrared reflectance spectra to show qualitatively that the implanted ions affect the Si-O stretch frequency in distinctive ways. We infer from these and transmission electron microscope measurements of Cu-implanted silica that the Cu and Pb ions are incorporated into the glass network in different phases, the former as metallic colloids of nm size, the latter in a lead silicate glassy phase.

Implantation and Optical Measurements

High-purity silica (Spectrosil A) substrates were implanted with Cu and Pb ions. The Cu-implanted samples were prepared at nominal doses of 0.5, 1, 3 and $6 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$ at an energy of 160 keV and at a current density of $7.5 \mu\text{A}\cdot\text{cm}^{-2}$. The Pb ions were implanted at 320 keV beam energy to doses of 1, 3, and $6 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$ at a current density of $5.1 \mu\text{A}\cdot\text{cm}^{-2}$; one sample was also implanted to a dose of $2.2 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$ at a current density of $1 \mu\text{A}\cdot\text{cm}^{-2}$. All implantations were carried out at room temperature. For the Cu ions and for the Pb ions implanted at 320 keV, ion backscattering with 2-MeV He $^{+}$ was used to determine the concentration and depth profiles of the implanted ions, as described elsewhere.[4] In the case of the Pb samples implanted at 100 keV, medium-energy ion backscattering at 270 keV, with time-of-flight detection, was used to measure the depth profile.[5]

The intensity-dependent component of the index of refraction for implanted samples was measured by observing variations in the far-field transmission of laser light as the sample was scanned through the focal spot of a long-focal-length lens, in one possible realization of the so-called "Z-scan" technique.[6] The laser source was a continuous-wave, mode-locked, frequency-doubled Nd:YAG laser producing a train of pulses at a wave-length of 532 nm, having a duration of 100 ps full-width-half-maximum and a pulse-repetition frequency of 76 MHz. The peak power in the second-harmonic beam ranged up to approximately 200 W, yielding maximum peak irradiances of order 10^{11} W $\cdot$ m $^{-2}$ in the 40- μ m diameter focal spot. The laser power was always below the threshold for thermal damage to the target, as verified by visible microscopy. However, the samples became warm to the touch after a few minutes of laser irradiation, consistent with measured absorptions in the range of 20-30%.[1]

The intensity on the sample was varied by translating the sample on a motorized, high-precision translation stage through the focal spot of a 150-mm focal length lens - hence the rubric, "Z-scan." The translation stage had a resolution of 25 μm and a repeatability of 1.3 μm . Signals from a beamsplitter positioned before the focusing lens were monitored on a power meter to provide normalization, while the signal transmitted through the sample was monitored by a power meter at the end of the translation, situated behind a beam-limiting aperture. Signals from both power meters were fed into an analog-to-digital converter and stored in a Macintosh IICx microcomputer. Long-term thermal drifts were monitored by comparing Z-scans in which the sample was translated forward and backward; in the measurements reported here, the differences between the two scans were less than one per cent.

For a material with a positive nonlinear index of refraction, moving the sample initially toward, and then away, from the focal spot causes an initial decrease in the normalized far-field intensity, followed by a crossing through unity at the focal spot and then an increase in intensity. Beyond about 40 mm on either side of the focal spot, the nonlinear index is sufficiently small that the far-field intensity is essentially that produced by the linear index of refraction.

In order to characterize the electronic structure of the implanted layers, infrared reflectance spectra were measured with a Bomem MB100 Fourier-transform spectrometer, using an SiC light source, KBr beamsplitter, and Hg-Cd-Te detector permitting coverage of the spectrum from 450 to 4000 cm^{-1} . The samples were mounted on a laboratory-built reflection accessory having an angle of incidence of 15°. The raw spectra of all samples were divided by the reference signal to provide a consistent normalization. Reflectance spectra were converted to extinction coefficients by a Kramers-Kronig transformation.

Experimental Results

The Z-scan transmission data for Cu:silica at an average laser power of 450 mW (peak irradiance of $5 \cdot 10^{10} \text{ W} \cdot \text{m}^{-2}$) are shown in Figure 1. The shape of the spectrum indicates a material with positive self-focusing. The abscissa is relative distance from the focal spot; the crossing through 1 occurs at the focal point of the focusing lens placed in front of the sample. The nonlinear phase impressed on the beam as it traverses the sample is proportional to the difference between the peak and valley components, in this case, of order 0.45. Corresponding data for one of the Pb-implanted fused silica samples are shown in Figure 2, where the difference in peak and valley transmission is 1.35. The average laser power in this measurement was about 300 mW, giving a peak irradiance of $3 \cdot 10^{10} \text{ W} \cdot \text{m}^{-2}$. The detailed differences between the Z-scan transmission curves for Pb:*a*-SiO₂ and Cu:*a*-SiO₂ are not well understood at present, and probably have their origins in the differing morphologies of the implanted layers. Small bumps in the Z-scan spectra for Cu:*a*-SiO₂ appeared in earlier work [2], and are thought to be connected with double-sided implantation and with small alignment errors in the beam axis, for example.

Infrared reflectance spectra from 1400 to 800 cm⁻¹ are shown in Figure 3 for unimplanted fused silica as well as for the Cu- and Pb-implanted samples. The primary spectral feature in the reflectance of the unimplanted silica, labeled curve (a) in each Figure, is the peak at 1111 cm⁻¹ which we attribute to the Si-O stretching vibration. In the upper portion of Figure 3, the curves labeled (b), (c), (d) and (e) are reflectance spectra for Cu:*a*-SiO₂ at doses of 6, 3, 1 and $0.5 \cdot 10^{16} \text{ ions} \cdot \text{cm}^{-2}$, respectively. The reflectance spectra are not only shifted and strongly broadened on the low-wavenumber side, but there is a significant increase in reflectance with increasing ion dose. Reflectance spectra for Pb:*a*-

SiO₂ are shown in the lower part of Fig. 3, with curves (b), (c) and (d) matching implanted doses of 1, 3 and 6·10¹⁶ ions·cm⁻², respectively. In this case, the reflectance *decreases* with increasing dose, and the shape of the low-frequency shoulder is quite different from that in the Cu-implanted samples.

Extinction coefficients for Cu- and Pb-implanted samples were computed numerically from the reflectance spectra by a Kramers-Kronig transformation, and are displayed in Figure 4. The labeling of the curves corresponds to the same ion doses as in Figure 3. Once again, the Pb:*a*-SiO₂ sample shows a uniform shift of the Si-O stretch-mode peak to lower wave-number (1103 cm⁻¹) following implantation, and a slight but perceptible decrease in extinction coefficient with increasing dose. In the Cu:*a*-SiO₂ sample, on the other hand, the extinction coefficient *increases* with increasing ion dose, and the peak of the Si-O stretch mode shifts progressively from 1103 cm⁻¹ at the lowest dose to 1092 cm⁻¹ at the highest. More will be said about the negative values of the extinction coefficient in the **Discussion** section.

Discussion

The intensity-dependent change in refractive index $\Delta n = \gamma I$ can be computed from the Z-scan measurement because the difference in peak and valley far-field transmission is proportional to the nonlinear phase shift at the focus,

$$\Delta\Phi_o = g(r_a) \cdot \Delta T_{P-V} = \left(\frac{2\pi}{\lambda} \right) \cdot I_o \cdot \left(\frac{1 - e^{-\alpha L}}{\alpha} \right) \cdot \gamma \quad (1)$$

as shown in Ref. 6. The symbols have the following meanings: $g(r_a)$ is a geometrical scale factor which depends on the size of the aperture at the far-field detector, r_a ; ΔT_{P-V} is the difference between the peak and valley transmission in the transmitted signal; λ is the wavelength of the laser; I_o is the

peak irradiance at the focal spot; and α is the small-signal (Beer's law) absorption in the implanted layer of thickness L .

Owing to the extremely large small-signal absorption of the implanted layer - $\alpha \approx 10^4 \text{ cm}^{-1}$ - the penultimate factor in Eq. (1) is approximately equal to L , and the nonlinear index is given very nearly by

$$\gamma \cong \frac{g(r_a) \cdot \Delta T_{P-V}}{I_0 \cdot L \cdot (2\pi/\lambda)} \quad (2)$$

We calibrated our method by measuring the thermal nonlinear index of CS_2 in a 1-mm thick quartz cuvette; we find a change in index of $\Delta n \approx 10^{-3}$, in excellent agreement with the value measured by others [6] and with the value computed from the thermo-optic coefficient for CS_2 .

Using Eq. (2), and mean thicknesses of approximately 150 and 40 nm for the Cu- and Pb-implanted samples, respectively, we find the nonlinear index of refraction for the two materials to be:

$$\gamma(\text{Pb}:a\text{-SiO}_2) \approx 3 \cdot 10^{-10} \text{ m}^2 \cdot \text{W}^{-1} \quad \gamma(\text{Cu}:a\text{-SiO}_2) \approx 2 \cdot 10^{-11} \text{ m}^2 \cdot \text{W}^{-1}$$

The computed nonlinear phases are dependent on the precise area of the focal spot and of the geometrical factor $g(r_a)$, which are subject to rather large errors. Hence, the calculated values for γ may be in errors by as much as 50%. The dominant effect in the calculation is the enormous absorption of the extremely thin implanted layer which justifies the approximations of Eq. (2). It is also noteworthy that the γ for Pb:silica is larger than that of Cu:silica even though the implanted-ion dose is a factor of three smaller for Pb:silica.

In general, there are four possible sources for a nonlinear index of refraction:[7] (1) the electronic response to the applied laser field; (2) the ionic response to the applied field; (3) electrostrictive effects; and (4) localized heating due to absorption of the laser light in a small volume. Since the density of implanted ions in the silica substrate may be as high as 10% by volume, it is

not clear whether the change in index should be attributed to the thermo-optic effect or whether it could be partly electronic in nature. In general, thermal processes should be associated with time scales on the order of the focal spot diameter divided by the speed of sound. Hence, we expect the characteristic decay for thermal excitation in these materials to be of order 20-40 ns, depending on our assumptions about the velocity of sound in the implanted layer. The interpulse spacing from the mode-locked laser is 13.2 ns, thus locating our experiments in an intermediate zone where thermal effects are present, and perhaps even dominant, but where electronic processes cannot be ruled out.

The large difference in γ measured for the Pb- and Cu-implanted silica could result from differences in the electronic or thermal properties of the implanted species, or from differences in the electronic environment of the implanted ions. Some clues to this electronic environment can be obtained from infrared reflectance measurements contrasting samples with and without implanted ions, as exhibited in Figure 3.

The most prominent feature in the reflectance spectra of Figure 3 is the peak near 1100 cm^{-1} attributed to the Si-O stretching vibration. In unimplanted fused silica, this peak occurs at 1111 cm^{-1} ; ion implantation changes the amplitude of this peak and also produces a small, but clearly discernible, red shift.

The change in amplitude of the Si-O stretch peak corresponds to a decrease in the number of Si-O-Si linkages, due either to a chemical interaction with an implanted ion - *i.e.*, changing the oscillator strength of the bond - or to radiation-damage-induced bond reformation. As noted above, following an initial decrease in Si-O stretch amplitude in both Cu- and Pb-implanted samples, the

amplitude *decreases* slightly with increasing dose in the case of Pb:*a*-SiO₂, but *increases* significantly with increasing dose in the case of Cu:*a*-SiO₂.

We attribute the red shift of the Si-O stretching vibration in the Cu- and Pb-implanted samples both to changes in the Si-O-Si bond angle, and to a decrease in the Si-O stretching force constant. The red shift due to implantation of noble gas ions has been shown by a number of researchers to be consistent with with an ion-implantation-induced compaction of the glass by 1-1.5%.[8] However, others have shown that compaction alone cannot account for changes observed in peak shifts and intensities for reactive implanted ions. Webb and Townsend [9] and, more recently, Magruder *et al.* [10] have suggested that chemical interactions between the substrate and implanted ions may have to be invoked to explain the observed reflectance and absorption spectra of implanted materials.

The red shifts for the Cu- and Pb-implanted silica samples can be computed by using a Kramers-Kronig transformation to obtain the extinction coefficient k defined by $\mathbf{n} = n_0 - i k$, where $\mathbf{n}$ is the complex refractive index and n_0 the real part of the index of refraction.[11] The extinction coefficient is related to the reflectance r by

$$k = \frac{-2r \sin\vartheta}{1 + r^2 - 2r \cos\vartheta} \quad (3)$$

The phase difference ϑ between incident and reflected waves is given by

$$\vartheta(\omega) = -\frac{\omega}{\pi} \mathbf{P} \int_0^{\infty} \frac{\ln R(\omega')}{\omega'^2 - \omega^2} d\omega' \quad (4)$$

where $R = r^2$ is the experimentally measured reflectivity. Note that the range of integration is over all frequencies ω' .

The extinction coefficients derived from the numerical integration of Eq. (4) are shown in Figure 4. The peak of the Si-O stretch for the Pb-implanted

samples is at 1103 cm^{-1} , while the peaks for the Cu-implanted silica range from 1103 cm^{-1} at the lowest dose to 1092 cm^{-1} at the highest dose. These red shifts are consistent with an implantation-induced compaction in the range of 2-3% as reported for ion-implanted glasses [10]. Of greater significance, however, are the differences between the red-shift variations for the two materials seen as a function of implanted ion dose: The red shift for Cu:*a*-SiO₂ depends on dose, while that for Pb:*a*-SiO₂ does not.

We can understand the changes in both amplitude and redshift of the Si-O stretch with the help of recent transmission electron microscope measurements showing that Cu forms colloids upon implantation, and that the relative fraction of large Cu colloids increases significantly at higher doses.[12] Hence, for the Cu implants, Si-O bonds may be broken at low doses and the Si-O bond angle changed to accommodate Cu nanoclusters in the glass network. At higher doses, the mean size of the Cu nanoclusters increases, and Si-O bonds are reformed, as the Si and O do not interact chemically with the Cu. This accounts both for the progressive red shift and the eventual recovery in Si-O intensity with increasing dose. In Pb:*a*-SiO₂, on the other hand, the redshift of the Si-O stretch is independent of dose, suggesting that the dangling bonds due to implantation are terminated by forming Pb-O-Si bonds, simultaneously reducing the intensity of the Si-O stretching peak.

The negative values of the extinction coefficient near 1000 cm^{-1} are well-known artifacts of Kramers-Kronig analysis over finite data intervals. The primary culprit in the present case is the Si-O-Si bending mode peak at 468 cm^{-1} which is known to have a long tail extending toward the 1000 cm^{-1} region and which was not measured due to the cutoff of the spectrometer at 450 cm^{-1} . However, a consistency check of the entire reflectance spectrum for an

unimplanted SiO₂ sample showed that the position of the Si-O stretch-mode peak is not materially affected by the inclusion of the bending-mode data. Moreover, the denominator of Eq. (4) gives much greater weight to values of $R(\omega')$ nearest the peaks, as can be shown by integrating Eq. (4) by parts.[13] Hence, we assume that our fundamental conclusions are not altered materially by the artifact. Indeed, the artifact cannot logically affect the qualitative conclusion that the two materials have significant differences in electronic structure: If the 468 cm⁻¹ anomaly affects the Kramers-Kronig transform of both materials in the same way, the conclusion about their distinctive behavior in the network stands unaltered. On the other hand, if the bending-mode peak affects the two spectra differently, this may change the details of the redshifts, but will still support a conclusion that the red shifts, and hence the electronic structures of the implanted samples, differ.

Conclusions

We have implanted two ions with greatly differing electronic properties - Pb and Cu - into fused silica in order to create thin layers with unusual nonlinear optical properties. The size of the nonlinear index of refraction n_2 is large enough to generate an intensity-dependent index change of order unity at sufficiently high intensity for 100-ps pulses of 532-nm light. Infrared reflectance measurements indicate that the electronic structures of the two implanted materials are clearly distinct from one another: the Cu implanted ions aggregate as nanoclusters of Cu metal, while the Pb ions are apparently incorporated into the substrate as a mixed lead-silicate compound, perhaps a glassy phase. These materials exhibit nonlinear index values within two to three orders of magnitude of those found for resonant processes in multiple quantum wells, but it is not yet

clear whether the observed nonlinearity is purely thermal, and hence relatively slow, or whether it has a significant, ultrafast electronic component. However, while waveguide applications of these materials require picosecond switching times, a finding that the optical bistability is predominantly thermal would by no means rule out the application of ion-implanted glasses in useful technological areas, such as optical beam limiters for sensitive optical elements.

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12. For instance, at a dose of $6 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$, the range of colloid sizes is roughly 3-10 nm, while at a dose of $12 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$, the range is 5-25 nm. See R. H. Magruder and R. A. Zuhr, submitted to J. Non-Cryst. Sol.
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Figure Captions

1. Far-field transmission function of a Cu-implanted fused silica sample, dose $6 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$, as a function of distance from the Z-scan focus for 100-ps pulses of 532-nm light delivered at a pulse repetition frequency of 76 MHz.
2. Relative far-field transmission of a Pb-implanted fused silica, dose $2.2 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$, as a function of distance from the Z-scan focus for 100-ps pulses of 532-nm light delivered at a pulse repetition frequency of 76 MHz.
3. (Upper spectrum) Infrared reflectance measurements for Cu:*a*-SiO₂ samples of varying doses. The curve labelled (a) is for unimplanted fused silica. The reflectance curves labelled (b), (c), (d) and (e) are for samples implanted to doses of 6, 3, 1 and $0.5 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$, respectively. (Lower spectrum) Infrared reflectance measurements for Pb:*a*-SiO₂ samples of varying doses. The curve labelled (a) is for unimplanted fused silica. The reflectance curves labelled (b), (c), and (d) are for samples implanted to doses of 1, 3 and $6 \cdot 10^{16}$ ions $\cdot$ cm $^{-2}$, respectively.
4. Extinction coefficients as a function of wavelength and ion dose, extracted from the reflectance measurements shown in Figure 3 by means of a Kramers-Kronig transformation. (Upper spectrum) Extinction spectra for the Cu:*a*-SiO₂ sample. (Lower spectrum) Extinction spectra for the Pb:*a*-SiO₂ sample by a standard Kramers-Kronig analysis. The labeling of the curves corresponds to the same ion doses as in Figure 3.

FIGURE 1

FIGURE 2

Figure 3

Figure 4

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