High Verdet constant Ga:S:La:O chalcogenide glasses for magneto-optical devices

Carmen B. Pedroso
Universidade Estadual de Campinas, UNICAMP
Instituto de Fisica Gleb Wataghin
13083-970 Campinas, São Paulo
Brazil

Egberto Munin
Universidade do Vale do Paraiba, UNIVAP
Av. Shishima Hifumi 2911
12244-000 São José dos Campos S.P.
Brazil
E-mail: munin@univap.br

Antonio Balbin Villaverde
Universidade Estadual de Campinas, UNICAMP
Instituto de Fisica Gleb Wataghin
13083-970 Campinas
São Paulo, Brazil

Januncio A. Medeiros Neto
Centro Federal de Educação Tecnológica do Paraná, CEFET-PR
Av. 7 de setembro 3165
80230-901 Curitiba, PR
Brazil

Norberto Aranha
Universidade Estadual Paulista, UNESP
Instituto de Quimica
CP 355
14800-900 Araraquara, S.P.
Brazil

Luis C. Barbosa
Universidade Estadual de Campinas, UNICAMP
Instituto de Fisica Gleb Wataghin
13083-970 Campinas
São Paulo, Brazil

Abstract. The magneto-optical rotation at room temperature was measured for three Ga:S:La:O chalcogenide glasses at several laser lines in the visible. The first sample was a binary system constituted by 70 mol % Ga_2S_3 and 30 mol % La_2O_3, whereas in the second and third ones the lanthanum oxide was partially substituted by lanthanum sulfide, keeping the amount of gallium sulfide fixed. A pulsed magnetic field between 50 and 80 kG was used for the Faraday rotation measurements. The Verdet constant for one of the ternary samples was found to be as high as 0.205 min G^{-1} cm^{-1} at 543 nm, indicating that these chalcogenide glasses are very promising for magneto-optical applications. The data for each sample were fitted using the expected analytical expression for the magneto-optical dispersion. Measurements of the refractive index of the glasses at 632.8 nm are also reported. Data on the magneto-optical properties of two high Verdet constant, heavy-metal oxide diamagnetic glasses are also included for comparison. [S0091-3286(99)00102-6]

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1 Introduction

Chalcogenide glasses containing sulfides of gallium and lanthanum (GLS glasses) are very promising candidates for many optical applications due to the several interesting optical properties that they present, such as low phonon energy, high transparency in the infrared up to approximately 10 μm, and high refractive index. Extensive research has been carried out in order to optimize these glasses, since they are also good host materials for rare-earth ions. These glasses, when they are adequately doped, can be used as a bulk laser material or as an efficient fiber optic amplifier operating in the region of the second telecommunication window at 1.3 μm.[1–3] Recently the second-harmonic generation in GLS glasses was investigated at 1.06 and 1.319 μm, using a Nd:YAG laser.[4] The high index of refraction of these chalcogenide glasses, higher than 2.3, indicates that they can also show a high value for their Verdet constant; therefore, they are good candidates for magneto-optical devices. Vitreous and crystalline materials with high magneto-optical rotation are very useful for building optical isolators, magnetic field sensors, transducers for measuring electric current in high-voltage transmission lines, and displacement sensors, among many other applications.[5–14] The magneto-optical effect is also widely used to study basic material properties; for instance, the magneto-optical dispersion in semiconductors and insulators gives us important information about the nature of the optical transitions and the magnitude of the energy gap between bands in those materials.[15–18] The investigation of the magneto-optical effect on optical glasses[19–23] has gained a new impetus in the last few years, due to the growing need for good materials to be used in magneto-optical devices.[16,24–26]

Although in paramagnetic glasses heavily doped with rare-earth ions the magnetically induced rotation of the light polarization is larger than in diamagnetic glasses, devices that use paramagnetic materials as magneto-optically active media have the drawback of requiring special care to compensate for thermal effects.[27,28] It is advantageous in certain applications to use diamagnetic materials as the magneto-optical medium because of the small intrinsic temperature dependence of their magneto-optical constants, allowing for much less temperature-sensitive devices. The largest diamagnetic Faraday rotations are found for high-refractive-index and high-optical-dispersion glasses.

In the present work, magneto-optical dispersion measurements on binary and ternary GLS glasses are presented and discussed. We studied GLS glasses with three different compositions: (i) 70 Ga_2S_3 : 30 La_2O_3, (ii) 70 Ga_2S_3 : 20 La_2S_3 : 10 La_2O_3, and (iii) 70 Ga_2S_3 : 25 La_2S_3 : 5 La_2O_3, with all the concentrations expressed in mole percent. The Verdet dispersion of these samples was measured at room temperature in the visible region of the spectrum using three He-Ne lasers operating at the wavelengths 543.5 nm (green line), 594 nm (yellow line), and 632.8 nm (red line), and using a semiconductor laser at 675.5 nm. The measurements were carried out using the technique of pulsed magnetic fields, which are generated by a capacitive discharge over an air-core solenoid. We are also including in this study, for the sake of comparison, magneto-optical dispersion measurements (at room temperature and at the same wavelengths) of two glass systems containing heavy-metal oxides: 60 Bi_2O_3 : 25 CdO : 15 GeO_2 (BCG ternary glass) and 10 Bi_2O_3 : 50 PbO : 32 GeO_2 : 8 B_2O_3 (BPGB quaternary glass). These two glasses are known to have a high Verdet constant.[25] Data on the refractive index at 632.8 nm for all the glasses are also presented.

2 Magneto-Optical Rotation: Theory

The magneto-optical effect known as the Faraday effect consists in the rotation of the plane of polarization of light when transmitted through a sample of material subjected to a magnetic field. It is a nonreciprocal phenomenon in which the rotation angle θ is given as a function of the strength of the applied magnetic field H and the length l of the sample by the following relation:

$$\theta = VlH, \tag{1}$$

when the light propagates parallel to the magnetic field direction. The coefficient V is known as the Verdet constant and is related to the light wavelength and the optical dispersion of the material by the so-called modified Becquerel equation,[21,29]

$$V = \frac{e}{2mc^2}\,\gamma\lambda\,\frac{dn}{d\lambda}, \tag{2}$$

where e and m are the electron charge and mass, and c the velocity of light. The Verdet constant V is a characteristic of each material. The factor γ, first introduced into the Becquerel equation by Darwin and Watson,[29] is referred to as the magneto-optic anomaly factor. Ramaseshan[30] has considered the relationship between this anomaly factor and the nature of the electronic bonding in certain cubic crystals. He inferred that γ would have a value close to one for ionic bonding and a deviation of its value from unity would indicate a certain amount of covalent character. Other groups pursued no further investigations for a long period of time after Ramaseshan's work trying to correlate γ with some other material properties. Recent studies, undertaken by us, showed that for substances having the general formulas AB_2 and A_2B there is a correlation between the magneto-optic anomaly factor γ and the difference of electronegativity between the ions A and B in the bond.[31]

The optical dispersion can be expressed by the Sellmeir equation

$$n^2 - 1 = A + \frac{B\lambda^2}{\lambda^2 - \lambda_0^2}, \tag{3}$$

where A and B are constants and λ_0 is the wavelength corresponding to the average position of the ultraviolet absorption bands in the material. The constant A accounts for the contribution to the refractive index from the infrared absorption bands. If we calculate the index derivative that appears in Eq. (2), using the expression for the refractive index n as a function of the wavelength given in the Eq. (3), we obtain the following relation:

$$nV = \frac{e}{4mc^2}\,\gamma\lambda\,\frac{d(n^2)}{d\lambda} = \frac{|e|}{2mc^2}\,\gamma\,\frac{B\lambda_0^2\lambda^2}{(\lambda^2-\lambda_0^2)^2}, \tag{4}$$

where n and γ are slightly dependent on the wavelength. We take the absolute value of e because the Verdet constant is defined as positive for a diamagnetic material. Since we are interested in calculating the value of V for only a short range of wavelengths, far from the absorption bands, we will neglect the dependence of n and γ on the wavelength in a first approximation, considering them as constants. We should also take into account the effect of the infrared absorption in the expression of V, adding to Eq. (4) a constant term, which will be denoted by K_1. So we finally obtain the following relationship between the Verdet constant of a diamagnetic material and the wavelength of the incident radiation:

$$V = K_1 + \frac{K_2\lambda_0^2\lambda^2}{(\lambda^2-\lambda_0^2)^2}, \tag{5}$$

where the new constant K_2 is given by:

$$K_2 = \frac{e}{2mc^2}\,\gamma\,\frac{B}{n}. \tag{6}$$

The Verdet constant V is expressed in min G^{-1} cm^{-1}, and λ in nanometers. The corresponding units for the constants are K_1, K_2 (min G^{-1} cm^{-1}) and λ_0 (nm).

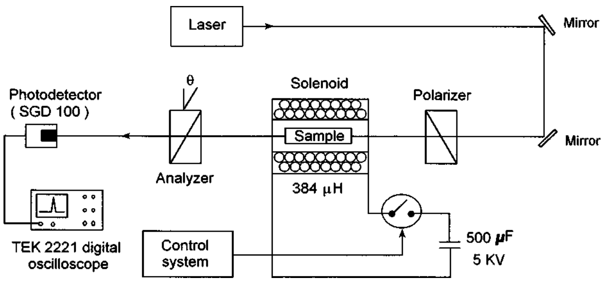

Fig. 1 Basic experimental setup for the measurement of the Faraday rotation with a pulsed magnetic field.

3 Experimental Results

The magneto-optical rotation of our glass samples was measured at room temperature by using a pulsed magnetic field. The basic experimental setup is shown in Fig. 1. The pulsed magnetic field is produced by discharging a 500-μF capacitor bank through an air-core solenoid with a measured inductance of 384 μH (the resulting pulse duration was about 1.5 ms). To prevent sample heating, the system was operated at a low repetition rate and the solenoid was cooled by forced air. Also, the sample was mounted mechanically isolated from the magnet, avoiding in this way sample vibrations during the high-energy discharges. The rotation angle was measured by the null technique, which has been described elsewhere.[15,31] The magnet was calibrated using a sample of HOYA FR-5 whose dimension was approximately the same as that of the glass samples. We take for the Verdet constant of the HOYA FR-5 glass[32,33] the value of -0.245 min G^{-1} cm^{-1} at 632.8 nm and at room temperature. All the measurements were carried out with magnetic field intensities in the range of 50 to 80 kG. The glass refractive indices were derived from the measurement of the Brewster angle at 632.8 nm. At the Brewster angle θ_B the reflection of the p waves on the material surface is zero, i.e., the reflected light from the surface is totally polarized. The angle θ_B is related to the material refractive index n by the following relation[34]: $\tan \theta_B = n$. So from the measurement of the angle θ_B it is possible to determine the value of n for the material.

The magneto-optical rotation of the glass samples was determined as a function of the wavelength. Measurements of the Faraday rotation were taken at 543.5, 594, and 632.8 nm, corresponding to emission lines of green, yellow, and red He-Ne lasers, respectively, and 675.5 nm from a red diode laser.

GLS glass samples were fabricated using a new technique that is effective in reducing all hydrogen impurities, yielding glasses with high optical quality. The conventional way of melting chalcogenide glasses is by batching the stoichiometric glass composition in a graphite crucible placed inside a fused silica tube, which is sealed after evacuating to a pressure of about 10^{-5} torr and heated to a temperature between 1100 and 1200°C. For the Ga:La:S:O glass system the highest temperature of the melt is limited to 1000°C because of the formation of sulfur dioxide and the consequent explosion of the sealed ampoule. In the new technique that problem is avoided by using a horizontal tube furnace with silica liner. Melting is done under flowing argon, and to avoid any loss of sulfur during the melting a graphite crucible containing typically 5 to 20 g of pure sulfur was inserted away from the melting zone and kept at temperature sufficiently high to create a vapor that is carried across the melting zone by the argon flow. A bubbler containing 2 to 4 in. of silicon oil provides a back pressure within the tube. In all, three GLS samples were prepared with the following compositions in mole percent: GLS-1, 70 $Ga_2S_3 : 30\ La_2O_3$; GLS-2, 70 $Ga_2S_3 : 20\ La_2S_3 : 10\ La_2O_3$; and GLS-3, 70 $Ga_2S_3 : 25\ La_2S_3 : 5\ La_2O_3$. The lengths of the three samples were approximately 5 mm for the GLS-1 and 10 mm for the GLS-2 and 3. The glasses were transparent, no striae or scattering centers being observed by visual inspection. The optical quality of these GLS glasses can surely be improved when they are made on a larger scale and under the controlled processes of a factory. High values of the Verdet constants together with good optical quality of our samples indicate that the GLS glasses are promising candidates for the construction of magnetooptical devices.

Glass samples were characterized by x-ray diffractometry (XRD), differential thermal analysis (DTA), and Raman spectroscopy. The XRD patterns were obtained with 3X-DA Shimadzu diffractometer, with a Ni filter and Cu $K\alpha$ 1.5418-Å radiation. The samples showed the typical halo ($2\theta \approx 28$ deg) in the XRD patterns, indicating that they were amorphous. From the DTA curves of the samples we found a glass transition temperature T_g of about 596°C and two crystallization temperatures T_c at 753 and 779°C. Raman spectra of samples in the vitreous and crystalline phases show significant differences between them. The Raman spectrum in the crystalline phase shows sharp peaks, whereas in the glass phase there appears a very broad band. Glass samples were crystallized at their surface by chemical attack with HCl. All three analysis methods indicated that our samples were glasses, without the formation of microcrystals.

BCG and BPGB glasses[25] were fabricated by melting the mixture of highly pure oxides in a high-purity alumina crucible placed in a Super Khantal electric furnace at 1160 to 1200°C (BCG glass) or 1000°C (BPGB glass), both for 30 min. After melting, glasses undergo annealing at approximately 300°C. The compositions (in mole percent) for these two last glasses were BCG, 60 $Bi_2O_3 : 25\ CdO : 15\ GeO_2$, and BPGB, 10 $Bi_2O_3 : 50\ PbO : 32\ GeO_2 : 8\ B_2O_3$.

Values of the Verdet constant and the refractive index for our glass samples, at 632.8 nm, are given in Table 1. All the measurements were carried out at room temperature. We also include in Table 1 the batch composition of each glass sample. The first sample is a binary glass system containing gallium sulfide and lanthanum oxide. In the second and third samples the lanthanum oxide is partially substituted by lanthanum sulfide, forming ternary systems. For the sake of comparison we have also added to Table 1 the corresponding information about two heavy-metal oxide glasses that are known to be good magneto-optical rotator materials.[25] The value of the refractive index of the BCG sample was calculated by interpolation of the data given in Ref. 25.

Table 1 The refractive index and Verdet constant, both measured at the wavelength 632.8 nm, and the batch composition of the GLS glass system. We have also included for comparison similar information on two heavy-metal oxide glasses. Estimated errors are ±1% (relative) for the Verdet constant and ±0.05 (absolute) for the index of refraction.

Sample	Composition (mol %)	*n*	*V* (min/G cm)
GLS-1	70 Ga_2S_3 30 L_2O_3	2.3	0.125_5
GLS-2	70 Ga_2S_3 20 La_2S_3 10 La_2O_3	2.4	0.115
GLS-3	70 Ga_2S_3 25 La_2S_3 5 La_2O_3	2.4	0.136_5
BCG	60 Bi_2O_3 25 CdO 15 GeO_2	2.0_5[a]	0.151
BPGB	10 Bi_2O_3 50 PbO 32 GeO_2 8 B_2O_3	2.0_5	0.113_5

[a]Value obtained from the data in Ref. 25 by interpolation.

The magneto-optical dispersion is shown in Table 2 for the three GLS glass samples. Measurements were taken in the visible from 543 to 675 nm. Also included in Table 2 are the corresponding values for the magneto-optical dispersion of the two heavy-metal oxide glasses. The estimated relative error is ±1% for the Verdet constant, whereas we have an absolute error for the refractive index measurements of ±0.05.

4 Discussion

Two of the GLS glass samples (GLS-2 and 3) were doped with a very low Pr^{3+} ion concentration: 500 ppm, since both samples were originally prepared to study the feasibility of building fiber optics amplifiers from these glasses. Let us estimate how much such a low doping can affect our measurements of the glass Verdet constant. Paramagnetic ions give a contribution to the total Verdet constant of the glass that is opposite in sign to that from the glass host. Let us now make a quick estimate of how much the Pr^{3+} ions contribute to the Verdet constant of the GLS glasses. Petrovskii et al.[26] observed that the Verdet constant of highly concentrated rare-earth-doped glasses is almost independent of the host glass. They found that the ratio V/C (where V is the Verdet constant and C is the ion concentration in the glass) changes by no more than 10% for different hosts and ion concentrations. From their data on V/C for Pr^{3+} at 306, 342, and 400 nm we found by extrapolation the value of $V/C=0.04\times10^{-21}$ min cm^2 G^{-1} ion^{-1} at 543 nm. Since the Pr^{3+} concentration in our samples corresponds to approximately 9×10^{18} ions/cm^3, the total contribution of the Pr^{3+} ions to the Verdet constant should be less than 3.6×10^{-4} min G^{-1} cm^{-1} at 543 nm, i.e., 500 times smaller than the contribution of the glass host. So the fact that our samples were doped is irrelevant as far as the Verdet constant measurement is concerned.

Our magneto-optical dispersion data for the GLS, BCG, and BPGB glass samples were fitted with the theoretical expression given in Eq. (5). Values of the fitting parameters K_1, K_2, and λ_0 are also shown in Table 2. We found that Eq. (5) fits the data within the experimental error of ±1%. The magneto-optical dispersion data as well as the fitting curves are displayed in Fig. 2 for the samples GLS-1, BCG, and BPGB, and in Fig. 3 for the two ternary glass samples: GLS-2 and GLS-3, Equation (5) with the values of the parameters, K_1, K_2, and λ_0 given in Table 2 can be used to determine the value of the Verdet constant for other wavelengths within the studied range. It is observed that our GLS glasses are good magneto-optical rotator materials, presenting values of the Verdet constant that are comparable to those of the best diamagnetic materials. For instance, we obtained for the Verdet constant of the GLS-3 glass sample a value as high as 0.207 min G^{-1} cm^{-1} at the wavelength 543 nm.

The binary glass system GLS-1 shows a Verdet constant that is between the corresponding values for the GLS-2 and GLS-3 samples, which are ternary systems. The refractive index and the parameter λ_0 for the GLS-1 are smaller than

Table 2 Magneto-optical dispersion of the GLS glass system. The parameters K_1, K_2, and λ_0 were obtained by fitting Eq. (5) to the magneto-optical dispersion data. We have also included for comparison similar information on two heavy-metal oxide glasses. The estimated error for the Verdet constant is ±1%.

Sample	Verdet constant (min/G cm)				K_1 (10^{-3} min/G cm)	K_2 (10^{-2} min/G cm)	λ_0 (nm)
	543 nm	594 nm	633 nm	675 nm			
GLS-1	0.189	0.149	0.125_5	0.108	7.2	63.8	236
GLS-2	0.173	0.133_5	0.115	0.0980	25.3	25.3	294
GLS-3	0.207_5	0.161	0.136_5	0.117_5	22.3	39.0	277
BCG[a]	0.228	0.174_5	0.151	0.126	20.5	44.2	276
BPGB[a]	0.168	0.131	0.113_5	0.095_0	7.02	54.1	239

[a]Reference 25.

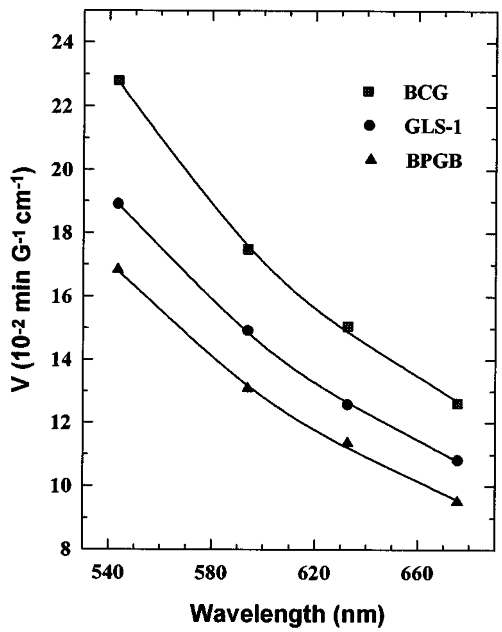

Fig. 2 Verdet constant as a function of the wavelength for the BCG, GLS-1, and BPGB glass samples. Solid lines represent the fitted curves obtained from Eq. (5) with the values of the parameters K_1, K_2, and λ_0 shown in Table 2. Vertical and horizontal error bars are smaller than the symbols representing the data.

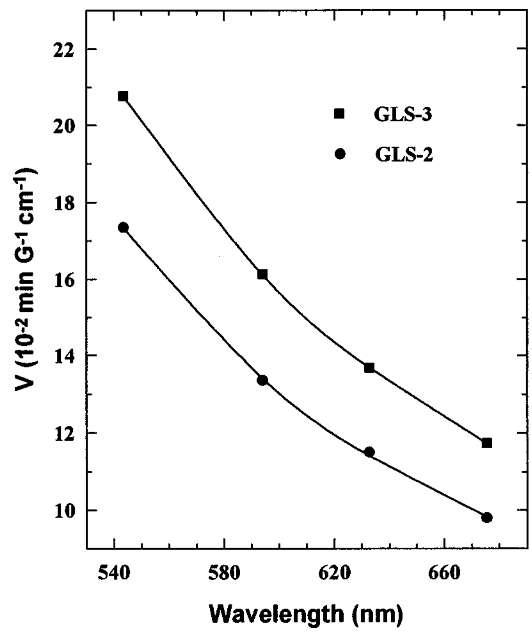

Fig. 3 Verdet constant as a function of the wavelength for the GLS-2 and GLS-3 glass samples. Solid lines represent the fitted curves obtained from Eq. (5) with the values of the parameters K_1, K_2, and λ_0 shown in Table 2. Vertical and horizontal error bars are smaller than the symbols representing the data.

for the other two glass systems, indicating that the absorption bands are shifted towards the ultraviolet for the GLS-1 glass. The concentration of Ga_2S_3 has been fixed at 70 mol % for all the three GLS samples, whereas the relative concentrations of La_2S_3 and La_2O_3 were varied from one sample to another. On increasing the amount of La_2S_3 in the ternary system, and therefore reducing the La_2O_3 concentration, the Verdet constant is increased by about 20%. It is not possible to make a straight comparison of the Verdet constant of the sample GLS-1 with those of samples GLS-2 and 3, since the glass structure is modified when we go from a binary to a ternary glass system. Such a structure change is corroborated by the differences observed in many of the glass optical properties. A thorough discussion on the glass formation and the optical properties of the ternary oxide sulfide system $La_2S_3:La_2O_3:Ga_2S_3$ is found in Ref. 3.

It is possible to estimate the value of γ for the GLS glasses. The parameter A in the Sellmeir equation (3) can be neglected in a first approximation, since it is small and it makes little contribution to the index of refraction in the visible. Taking $A=0$ in Eq. (3) and choosing the values $n=2.3$, $\lambda_0=236$ nm, $K_2=63.8\times10^{-2}$ min G^{-1} cm^{-1}, and $\lambda=632.8$ nm, we found from Eqs. (3) and (6) that $\gamma=0.39$ for the GLS-1 sample. That value, much less than 1, is characteristic of materials with covalent bonds and is very close to $\gamma=0.41$ reported for the BPGB glass in Ref. 25.

5 Conclusions

We have measured the magneto-optical rotation for three chalcogenide glasses, containing gallium sulfide, lanthanum sulfide, and lanthanum oxide. One of the samples is a binary system without lanthanum sulfide. Measurements were taken in the visible from 543 to 675 nm. We found that the Verdet constant can be as high as 0.207 min G^{-1} cm^{-1} at the wavelength 543 nm. The glasses were transparent, with no striae or scattering centers detectable by visual inspection. The combination of high Verdet constant and good optical quality of the glasses made in our own facilities suggests that the GLS glasses are good candidates for magneto-optical devices. The linear refractive index was also measured at 632.8 nm. High values for the refractive index were observed: 2.3 for the binary sample and 2.4 for the two ternary glasses. No correlation could be established yet between Verdet constant and glass composition, except that the substitution of La_2O_3 by La_2S_3 (keeping constant the amount of Ga_2S_3) increases the Verdet constant of the glass. It will be necessary to analyze more samples, with different amounts of the three components, in order to find such correlation. A more detailed study is underway.

Agreement of $\pm1\%$ was obtained between the theoretical curves and the experimental data. That agreement is within our experimental error. Values of the fitting parameters were calculated for the three samples. The parameter λ_0 presents a value for the binary glass system that is lower than for the other two samples, indicating a shift of the absorption bands towards the ultraviolet.

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Carmen B. Pedroso received her BS degree in 1984, the MS in 1987 and the PhD degree in physics in 1993 from Universidade Estadual de Campinas, Brazil. During her PhD program she studied optical properties of composite materials applying Maxwell-Garnett and Lorentz models. Presently, her research interest includes the study of magnetooptical effect in solid state materials and the development of sensors based on optical technologies.

Egberto Munin received his BS, MS and PhD degrees in physics from the Universidade Estadual de Campinas, Brazil, in 1986, 1990, and 1994, respectively. His PhD program included a period of two years (1992–1993) as a visiting researcher at the Center for Research and Education in Optics and Lasers (CREOL) of the University of Central Florida. During that time he worked in collaboration with Dr. Michael Bass and crystal grower Dr. Bruce H. T. Chai, studying the optical properties of new solid state laser hosts. In 1996 he joined the Universidade do Vale do Paraiba, Brazil, where he is mainly involved with applications of the laser technology in medicine and biomedical optics.

Antonio Balbin Villaverde received his MS and PhD degrees in physics from the University of Buenos Aires-Argentina in 1967 and 1973, respectively. He spent two years from 1970 to 1972 at the University of Southern California working on his PhD program under the advisement of Prof. Larry DeShazer, on the subject damage in optical materials caused by high power laser irradiation. In 1974, he joined the Universidade Estadual de Campinas, Brazil, as an assistant professor. He was a visiting researcher at the Center for Laser Studies, University of Southern California (1984) and at the Center for Research and Education in Optics and Lasers (CREOL), University of Central Florida (1992–1993). His research interests include: solid-state lasers — spectroscopy and development, nonlinear optics, and magneto- and electro-optical effects.

Januncio A. Medeiros Neto: Biography not available.

Norberto Aranha received his BS and MS degrees in physics and the PhD degree in science from the Universidade Estadual de Campinas, Brazil, in 1984, 1987, and 1994, respectively. Since 1984 he has worked with several glass systems for optical communications, optical fibers, and waveguides. At the present time, he is working on biomaterials, bioceramics and bio-compatible coatings on titanium, for orthopedic and dental implants.

Luis C. Barbosa received his MS degree from the University of São Paulo, Brazil, in 1976, and the BS and PhD degrees from the Universidade Estadual de Campinas, Brazil, in 1972 and 1981, respectively. In 1981 he joined the Universidade Estadual de Campinas, where he is involved in the development of new glass materials for nonlinear optics and optical communication devices as well as rare earth doped glasses for optical amplification. He has published over 50 papers in scientific journals and holds 9 patents.