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Brillouin scattering in planar waveguides

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Brillouin spectra have been measured in silica-titania planar waveguides of different thicknesses, obtained by radio-frequency sputtering. The laser beam was coupled to the waveguide with a prism and the scattered light was collected from the front surface. The spectra, which are different for excitation in the different modes of the waveguide, are accounted for by a model that considers the space distribution of the exciting field. This allows measuring the sound velocities in the film. The role of the finite thickness of the planar waveguide is discussed. [S0163-1829(98)50726-6]

In the last years, a considerable interest has been devoted to the study of planar waveguides, both passive and activated by rare-earth ions, due to their potential applications in the integrated optics.[1–3] The optical properties of the planar waveguide can be measured by means of the m-line technique,[4] which gives the main parameters as the effective refractive index of modes, their shapes, and the thickness of the films. Recently, waveguide Raman spectroscopy and waveguide luminescence spectroscopy have been used for the structural characterization of SiO_2-GeO_2 waveguide produced by dip-coating[5] and silica films containing Ag nanoparticles.[6,7]

Here, we will show that the spectroscopic characterization, which uses waveguided excitation, can be extended to include the study of the film by Brillouin scattering (BS). BS is widely used in transparent bulk materials to measure transversal (v_T) and longitudinal (v_L) sound velocities at frequencies of about 10^{10} Hz. Besides this, important information on the structure and dynamics of the system can be obtained, since from BS measurements the dynamical structure factor $S(\vec{q},\omega)$ can be derived.

In bulk materials the exchanged $\vec{q}$ value is determined by the experimental geometry: $q=2k\sin(\varphi/2)$ where $k=(2\pi/\lambda_0)n$ is the incident wave vector, λ_0 the wavelength in vacuum, n is the refractive index, and φ the scattering angle. Recently, BS measurements have been extended from the region of visible light ($q\sim 10^{-2}$ nm^{-1}, $\omega\sim 10^{10}$ Hz) to that of x rays ($q\sim 1$ nm^{-1}, $\omega\sim 10^{12}$ Hz).[8] In this way, the BS measurements allow us to determine the sound velocity and the scattering length of the involved vibrations in a wide $(\vec{q},\omega)$ range from the $S(\vec{q},\omega)$ peak and from its linewidth, respectively. This can be done as a function of some parameters as, for example, the temperature and the stress.[9,10]

For excitation in a planar waveguide, the peculiar distribution of the exciting field requires the development of an *ad hoc* model for BS; this is done in the present paper and applied to silica-titania planar waveguide of different thicknesses. This model allows to fit the Brillouin line shape, to

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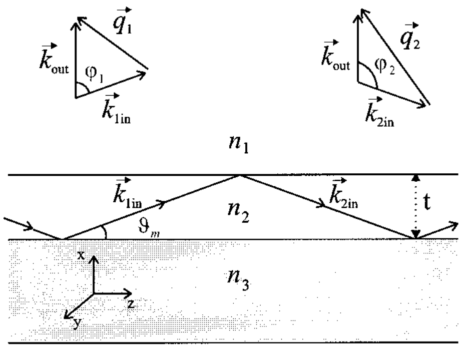

FIG. 1. Wave propagation in the planar waveguide. $\vec q_1$ and $\vec q_2$ are the exchanged wave vectors of the scattered light in the zig and zag paths.

deduce the sound velocity values and to compare them with the existing data for bulk materials.

An exciting light beam, confined in a planar waveguide, does not have a single well-defined wave vector. For the description of wave propagation, the ray-optic approach, reported in Fig. 1, can be used. The light propagates in the z direction and the plane waves have a zig-zag path in the x-z plane, undergoing total internal reflection at the boundary interfaces of the waveguide. In order that an electrical transverse mode TE (electric field parallel to the y axis) could propagate, the phases of the plane waves, reflecting at the interfaces, must satisfy a precise relation:[11]

$$2kn_2t\sin\vartheta_m-2\varphi_{23}-2\varphi_{21}=2m\pi, \tag{1}$$

where t is the thickness of the waveguiding region 2, ϑ_m is the angle of reflection with respect to the z direction, m is the mode number, and $2\varphi_{23}$ and $2\varphi_{21}$ are the phase changes suffered upon internal reflection at the interfaces. Thus, a discrete set of modes are obtained corresponding to the various ϑ_m values, limited by the thickness and the refractive index of the film. The m values ($m=0,1,2,\ldots m_{\max}$) give the number of nodes of the electric field along the x direction. For even m the electric fields of the zig and zag paths are in phase at the center of the guide, meanwhile for odd m they are in counterphase and the field has a node at the center of the guide. For each allowed mode, there is a corresponding propagation constant β_m given by

$$\beta_m=kn_2\cos\vartheta_m, \tag{2}$$

and the propagation velocity, parallel to the waveguide, is $v=ck/\beta$.

In the BS calculations, the zig ($\vec k_{1in}$) and zag ($\vec k_{2in}$) paths cannot be considered separately, unless they produce nonoverlapping spectral intensity distributions. The electric field of the laser excitation has to be considered as the sum of two fields propagating in the waveguides with wave vectors $\vec k_{1in}$ and $\vec k_{2in}$ as shown in Fig. 1. Being $\vec k_{out}$ determined by the geometry of detection of the scattered light, two values of exchanged wave vector will be present:

$$\vec q_1=\vec k_{out}-\vec k_{in}\qquad \vec q_2=\vec k_{out}-\vec k_{2in}.$$

Therefore, the BS intensity will be given by the time space Fourier transform of the correlation function of the electrical polarizability density tensor $\overline{P}(\vec r,t)$, but with two space Fourier components:[12]

$$I_{yy}(\vec q_1,\vec q_2,\omega)\approx\int e^{i\omega t}dt\int\int d\vec r_1 d\vec r_2\langle(e^{-i\vec q_1[\vec r_1(t)-\vec r_2(o)]}+e^{i\gamma}e^{-i\vec q_2[\vec r_1(t)-\vec r_2(0)]})P_{yy}(\vec r_1,t)P_{yy}(\vec r_2,0)\rangle, \tag{3}$$

where γ is the relative phase of the zig and zag fields in the m mode of the guide. If the acoustic vibrations, which scatter the light, are phonons of wave vector $\vec k_p$, by developing Eq. (3) and reporting only the interesting dependencies, we will find

$$I(\vec q_1,\vec q_2,\omega)\propto\frac{T}{\omega^2}\left|\int q_1e^{i(\vec k_p-\vec q_1)\vec r}+e^{i\gamma}q_2e^{i(\vec k_p-\vec q_2)\vec r}d\vec r\right|^2, \tag{4}$$

where T is the temperature, $\omega=v_{L,T}\cdot k_p$, and $v_{L,T}$ is the longitudinal, transverse sound velocity.

For a thick guide only phonons with $\vec k_{p1}=\vec q_1$ and $\vec k_{p2}=\vec q_2$ will produce scattering at the frequencies $\omega_{L,T}=v_{L,T}\cdot q_i$. Therefore, the Brillouin spectrum of a glass waveguide will show four peaks in the Stokes and four in the anti-Stokes spectrum. Two peaks are due to longitudinal phonons and the other two to transverse phonons. If the waveguide stands perpendicularly to the detection system, the corresponding frequencies will be

$$\omega_{L,T}^{(+,-)}=\left(\frac{2n_2\omega_0}{c}\right)v_{L,T}\sin\left(\frac{\frac{\pi}{2}\pm\vartheta_m}{2}\right), \tag{5}$$

where ω_0 is the laser frequency. The components of the doublet have the same intensity since the q^2 dependence is compensated by the ω^{-2} one.

However, if the guide thickness is comparable to the wavelength of the exciting light, the Brillouin linewidths are expected to be larger than in the bulk case. In fact, the usual sources of broadening are the dynamical effects, the instrumental resolution and the angle width of light collection, that makes $\vec k_{out}$ and $\vec q$ not well defined. Besides these effects, in the case of planar waveguides, the finite thickness contributes to the line broadening. By performing the integral in Eq. (4) in the scattering volume of a uniform planar waveguide of thickness t, with large dimensions along the y and z directions, the contributions of a phonon with $\vec k_p(k_{px},k_{py},k_{pz})$ to the scattered intensity is given by

$$I(k_{px},\omega)\propto\frac{T}{\omega^2}\left(q_1\frac{\sin\left[(k_{px}-q_{1x})\frac{t}{2}\right]}{(k_{px}-q_{1x})}+(-1)^m q_2\frac{\sin\left[(k_{px}-q_{2x})\frac{t}{2}\right]}{(k_{px}-q_{2x})}\right)^2 \tag{6}$$

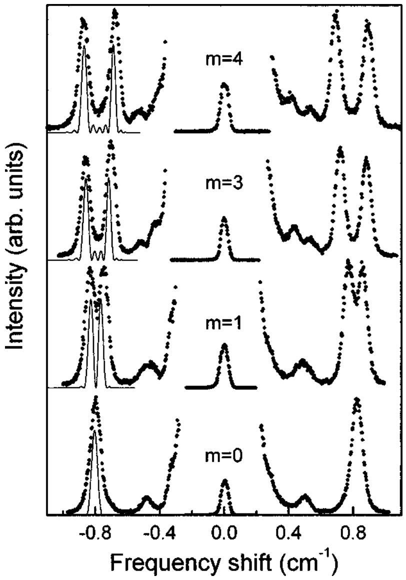

FIG. 2. Brillouin spectra of a SiO_2 (87 mol %)–TiO_2 (13 mol %) thick planar waveguide ($t=3.7$ μm) obtained by excitation in different TE_m modes and detection perpendicular to the plane of the guide.

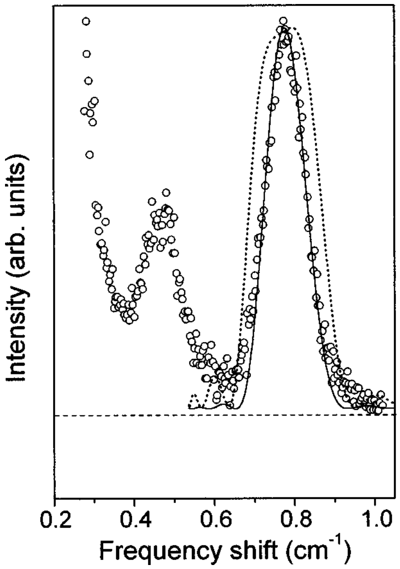

FIG. 3. Brillouin spectrum of a SiO_2 (91.3 mol %)–TiO_2 (8.7 mol %) thin monomode planar waveguide ($t=1.3$ μm). The horizontal dashed line gives an estimation of the flat luminescence background. The solid line is the spectrum of the longitudinal phonons, calculated by Eq. (6). The dotted line is the calculated spectrum in absence of interference between the field scattered by the zig and zag excitations.

with $k_{py}=0$, $k_{pz}=q_z=q_{1z}=q_{2z}$, $\omega_{L,T}=v_{L,T}(k_{px}^2+q_z^2)^{1/2}$ and where the $(-1)^m$ phase coefficient takes into account the fact that the two exciting fields are in phase at the center of the guide for even m and in counterphase for odd m.

This means that phonons with wave vectors in the two ranges, $k_{px}=q_{ix}\pm\Delta k_{px}$, $(i=1,2)$ with $\Delta k_{px}\cong 2/t$, contribute to the scattering and this produces a broadening of the Brillouin lines. Only if $|q_{1x}-q_{2x}|\gg 2/t$, the two terms in the sum do not interfere and the scattering is the same as that produced by two independent laser beams in the zig and zag paths. In general, the two scattered fields interfere, giving rise to a complicate lineshape, which depends on ϑ_m and on the scattering angle φ, longer than the t thickness of the waveguide.

Even in absence of interference effects between the two fields, the finite thickness of the guide produces a finite linewidth. To give an idea of the importance of this width, we can calculate it in the limit case for $\vartheta_m\ll 1$, $q_x\cong q_z$, $\varphi\cong\pi/2$, $\Delta k_x/k_x\ll 1$. It is easy to see that, in this case

$$\frac{\Delta\omega}{\omega}\approx 0.44\frac{t}{\lambda_0 n_2},$$

where λ_0 is the wavelength of the incident radiation in vacuum.

Finally, we want to recall the limits of the present model. The fields of the zig and zag paths are assumed to be plane waves traveling in a homogeneous medium with a constant profile of the refractive index and undergoing total reflection at the interfaces. Scattering from the substrate, actually produced by the evanescent wave or from losses, is neglected. The guide is assumed to be homogeneous (in particular $t=\mathrm{const}$), transparent and with flat interfaces; the phase of the two fields is taken constant along the path in the z direction.

The above prediction has been tested by taking the Brillouin spectra of two silica-titania planar waveguides. The first one suffers six longitudinal modes. In Fig. 2 we show the Brillouin spectra obtained by exciting with an argon laser ($\lambda=514.5$ nm) in different TE modes and by coupling with a gallium gadolinium garnet prism.[5–7] The diffused light, collected in a direction nearly perpendicular to the waveguide plane, was dispersed by a double monochromator with a focal length of 2 m operating at the 11th order of the gratings.

A He-Ne laser was used to determine the small α angle

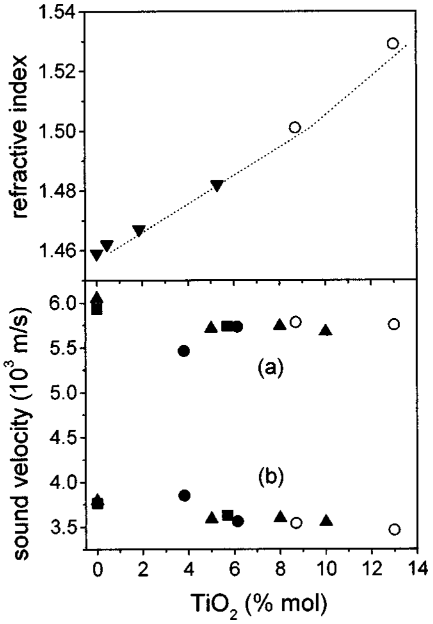

FIG. 4. Longitudinal, transverse sound velocities, and refractive index measured in our films (open circle) compared to the available data for bulk samples: square (Ref. 13), solid circle (Ref. 14), up triangle (Ref. 15), down triangle (Ref. 17). The dashed curve is taken from Ref. 16.

between the normal to the waveguide and the optical axis of the light detection system, in order to deduce the exchanged $\vec{q}$ values.

Two longitudinal and two transverse peaks are observed, with splitting which increases with the mode index, apart for the $m=0$ excitation, where no splitting is observed. The spectra calculated by using Eq. (6) are reported on the anti-Stokes spectrum of the longitudinal phonons. Note that the fit uses as free parameter only the sound velocity v_L. The values for $n_2=1.529\pm0.001$, $t=3.7\pm0.1$ μm, and $\vartheta_0=2.57°$, $\vartheta_1=5.14°$, $\vartheta_3=10.3°$, $\vartheta_4=12.87°$, have been obtained in an independent way with the m-line technique at $\lambda=543$ nm.

The overall best agreement is reached for $v_L=5.75\times10^3$ m/s and $v_T=3.46\times10^3$ m/s. There is some disagreement: the calculated splitting in the $m=1$ and $m=3$ modes is slightly smaller than the experimental one and the observed intensities of the lower energy component are systematically higher. Besides this, the model reproduces very well the experimental data.

The calculated line shapes cannot be easily tested since the experimental resolution is low, as can be seen from the linewidths of the elastic (filtered) peak. A better test of the calculated line shapes can be obtained by studying guides with smaller thicknesses and larger bandwidths.

Figure 3 shows the Brillouin spectrum, in the Stokes part, of a thinner monomode waveguide. Here the Brillouin lines are quite broad unsplitted bands. The flat background (dashed line) is due to the low energy broad band luminescence which was not filtered and which appears at lower diffraction orders together with the Brillouin light ($\lambda=514.5$ nm) of the 11th order of the gratings.

The Brillouin band, due to the longitudinal phonons, has been fitted by using Eq. (6), with $v_L=5.78\times10^3$ m/s, with the fixed parameters $m=0$, $\vartheta_0=6.45°$, $n=1.501$, $t=1.3$ μm. The line shape is very well reproduced by Eq. (6). Note that in this case the experimental resolution gives no problems as its linewidth is about $\frac{1}{4}$ of that of the Brillouin peak. The goodness of the agreement between the observed and the calculated spectrum better outcomes, if we compare (dotted line) the calculated line shape for the case of the two beams, in the zig-zag paths, considered independently, thus taking the sum of the squares instead of the square of the sum in Eq. (6). The Brillouin spectrum is not the sum of the two spectra taken at different exchanged $\vec{q}$ values, but the actual electric field distribution within the guide has to be taken into account to reproduce the experimental data. The fit of the Brillouin band, due to the transverse phonons, gives $v_T=(3.54\pm0.03)\times10^3$ m/s.

The sound velocities of the two waveguides are compared in Fig. 4 with the available data of silica-titania bulk glasses, obtained by different techniques.[13–15] The composition, 13 mol % TiO_2–87 mol % SiO_2 for the thick guide and 8.7 mol % TiO_2–91.3 mol % SiO_2 for the thin one, were measured by energy dispersive spectrometry (EDS). The measured refractive index of the two films are also reported and compared with the bulk data.[16,17] Sound velocities and refractive indexes of the films are very close to those of the bulk at the same compositions. Furthermore, Raman spectra, taken by waveguide excitation, show a good intermingling of the SiO_2 and TiO_2 components, with large intensities of the Si-O-Ti bands.[18] Therefore, these characterizations show that the film has a compact glass structure with vibrational dynamics very similar to that of the bulk glass reported by Bihuniak and Condrate.[17]

In conclusion, we have shown that the BS lines are structured. The line shape reflects the actual field distribution in the guide. In any case, the sound velocities can be measured with good precision. BS and Raman scattering could become a powerful tool for the structural characterization of the waveguides, for the control of basic components miscibility and porosity, especially in waveguides, produced by sol-gel route, where the sound velocity gives information about the densification degree.

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