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Correlation effects on Raman scattering from low-energy vibrational modes in glasses. II. Experimental results

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New experimental results of low-frequency Raman scattering from different glasses are presented. They are compared to the vibration density of states obtained from inelastic neutron scattering to deduce the light-vibration coupling coefficient $C(\omega)$ as a function of the vibration frequency. It was found that $C(\omega) \propto \omega^2$ at very low frequency $\omega < 20$ cm^{-1} in inorganic glasses (SiO$_2$,B$_2$O$_3$) and that $C(\omega) \propto \omega$ at low frequency in polymer glasses and in inorganic glasses for $\omega > 20$ cm^{-1}. Our experimental results for glasses are interpreted by assuming no correlation at very low-frequency and only radial correlation at low-frequency. A full correlation would exist in silica-aerogels in which the effect of random fluctuations would be negligible.

I. INTRODUCTION

The disorder in amorphous and heterogeneous systems has a strong effect on vibrational dynamics in the acoustical range. For this reason, inelastic neutron scattering (INS) and low frequency Raman light scattering (LFR) are now often used to study the structure of disordered systems. From INS one deduces the vibrational density of states (VDOS). On the other hand the intensity of Raman scattering $I(\omega)$, at the frequency $\omega/2\pi$, is proportional to the VDOS and to the coefficient of light-vibration coupling $C(\omega)$ (Refs. 1 and 2),

$$I(\omega) = C(\omega)g(\omega)[n(\omega)+1]/\omega. \qquad (1)$$

In this expression $I(\omega)$ is the intensity for Stokes scattering, $n(\omega)$ is the Bose factor, and $g(\omega)$ the VDOS. It is assumed that the VDOS observed in Raman scattering is identical to the VDOS from INS. This is valid in a frequency range where all vibration modes are localized, since Raman scattering does not occur from propagating modes because of the small light wave vector.

Information on the disordered structure can be obtained directly from the VDOS. A quasiuniversal feature of vitreous systems is the excess of VDOS in comparison with the Debye VDOS.[3,4] This VDOS excess is correlated to the excess of specific heat and also to the Boson peak in LFR.[4,5] It is certainly related to a universal characteristic of the glass structure. Information on the disordered structure can be obtained also from the coefficient $C(\omega)$ that can be deduced from comparison between INS and LFR.

Here we are particularly interested by the coefficient $C(\omega)$. It is now possible to determine $C(\omega)$ from LFR in several inorganic or polymeric glasses and other disordered systems, for which the VDOS has been determined by INS.

In this paper we publish new LFR results for different typical glasses. We show that structural relaxation motions bring a non-negligible contribution to the low-frequency light scattering in comparison with the LFR coming from vibrations, even at temperatures much lower than the glass-transition T_g. Consequently the coefficient $C(\omega)$ is deduced from LFR at low-temperature, and INS. The ex-

perimental $C(\omega)$ is interpreted by the preceding theory[6] (paper I) and disorder characteristics in glasses are emphasized.

II. EXPERIMENTAL TECHNIQUES

A. Raman scattering

The LFR was observed along a direction perpendicular to the excitation laser beam. The 5145 Å line of argon laser was used. The 6471 or 6725 Å lines of a krypton laser were used with samples which were fluorescent by excitation at a shorter wavelength. The scattered light was analyzed with a quintuple monochromator of high resolution and rejection rate, mounted by Dilor. This spectrometer with a 4 m focal length, allows observation of very low-frequency Raman scattering. Raman observations were carried out with samples at room temperature and at lower temperature in a helium cryostat.

B. Samples

Different glass samples were studied by Raman scattering. Suprasil silica was obtained from Heraus. B$_2$O$_3$ glass was prepared at the laboratory of Chimie des Matériaux, University of Rennes. The studied polymethylmethacrylate (PMMA) was atactic with an average molecular weight of 63 000, and a glass-transition temperature $T_g = 393$ K. The amorphous polybutadiene had an irregular microstructure of *cis*-, *trans*-, and vinyl units (about 47:46:7), a molecular weight of 10^5 and a DSC $T_g = 186$ K. LFR was also observed from the epoxy monomer diglycidyl ether of bisphenol A (DGEBA, T_g close to 273 K).

III. RAMAN SCATTERING RESULTS

In this paper the polarized Raman spectra are presented. It was observed that the depolarization ratio is frequency independent in the considered spectral ranges for the different samples. In Fig. 1 the LFR spectra of SiO$_2$ glass at 55 and 300 K are compared. The spectrum at 300 K is divided by the factor $[n(\omega)+1]$ for $T=300$ K and multiplied by the same population factor for $T=55$ K. By
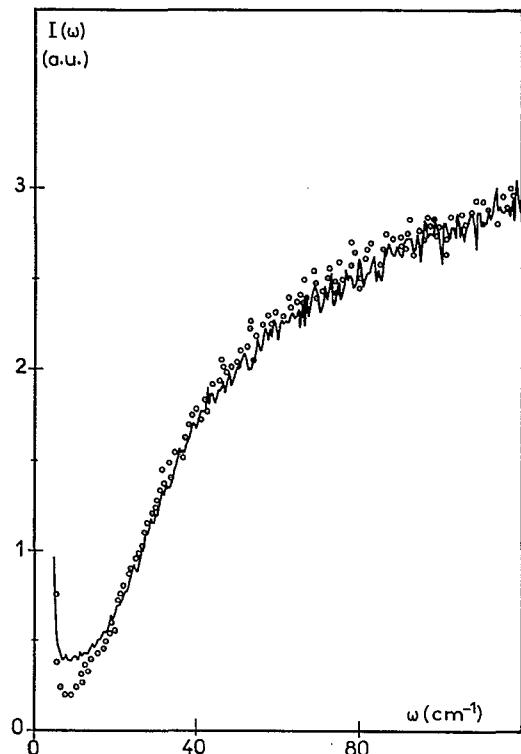
FIG. 1. Intensity of LFR from v-SiO$_2$. Circles, at 55 K; solid line, at 300 K, divided by $n(\omega)+1$ at 300 K and multiplied by the same factor at 55 K.
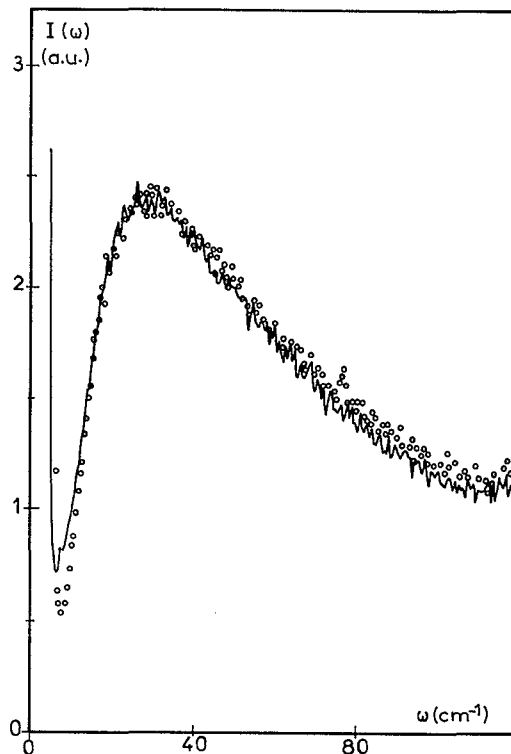

FIG. 2. Intensity of LFR from v-B$_2$O$_3$. Circles, at 57 K; solid line at 300 K, divided by $n(\omega)+1$ at 300 K and multiplied by the same factor at 57 K.

INS (Ref. 3) it was shown that the VDOS does not change from $T=55$ to 300 K. Consequently it is deduced that the scattering excess at $T=300$ K with respect the scattering at 55 K comes from structural relaxation. The same observation was made for vitreous B$_2$O$_3$ (Fig. 2). Therefore we will compare the VDOS obtained from INS to Raman scattering at low temperature. This scattering excess, which increases with the temperature more rapidly than $n(\omega)+1$ was observed in several glasses.[7–10]

In Fig. 3 the reduced Stokes LFR $I(\omega)/[n(\omega)+1]$ at $T=80$ K of polybutadiene is compared to the VDOS obtained from INS at the same temperature. At frequencies higher than 10 cm^{-1} the similarity of the two spectra are remarkable.

The reduced LFR of DGEBA and PMMA are shown in Figs. 4 and 5, respectively.

IV. COMPARISON BETWEEN RAMAN SCATTERING AND NEUTRON SCATTERING: DETERMINATION OF $C(\omega)$

From Eq. (1) the comparison between the VDOS and the reduced LFR intensity, $I(\omega)/[n(\omega)+1]$, gives directly the light-vibration coupling coefficient $C(\omega)$. The case of vitreous silica that was very much studied by INS is very interesting. From the more recent published INS data, the VDOS below 20 cm^{-1} varies like ω^2.[11] At higher frequencies its power-law frequency dependence ω^x is less simple; x shows a steep increase from 2 to 3 at 20 cm^{-1} and decreases progressively in the 30–60 cm^{-1} spectral range.

In Fig. 6 the VDOS of vitreous silica[11] is compared, to the reduced intensity of LFR, $I(\omega)/[n(\omega)+1]$. In the 8–20 cm^{-1} range the VDOS is multiplied by ω and by $\omega^{0.05}$ at higher frequencies. From expression (1), it means that $C(\omega)\propto\omega^2$ at low-frequency and $C(\omega)\propto\omega$ at higher energy. A roughly linear behavior was observed by Mali-
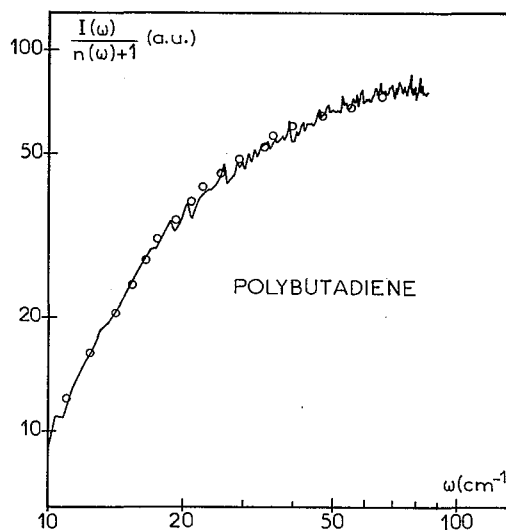

FIG. 3. Log–log plot of the LFR reduced intensity from polybutadiene at 80 K. The circles correspond to the VDOS obtained from INS (Ref. 13) at the same temperature.
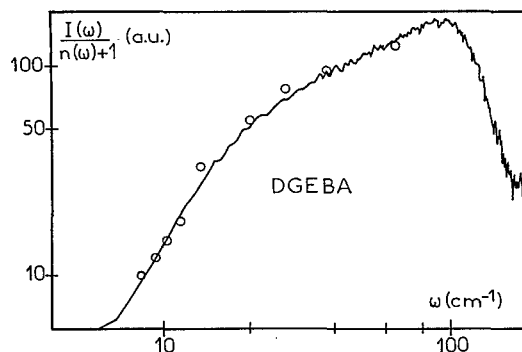
FIG. 4. Log–log plot of the LFR reduced intensity from DGEBA at 80 K. The circles correspond to the VDOS obtained from INS at 50 K (Ref. 15) multiplied by $\omega^{-0.05}$.

novsky *et al.*[4] The power-law $C(\omega) \propto \omega^2$ occurs when the VDOS $N(\omega) \propto \omega^2 = \omega^{d-1}$, d the Euclidean dimension. This behavior is exactly that is expected in a Debye regime without correlation.[6]

The same behavior is observed for vitreous B_2O_3. Hannon *et al.*[12] observed by INS that the VDOS of B_2O_3 is proportional to ω^2 up to 16 cm^{-1}. The reduced Raman intensity $I(\omega)/[n(\omega)+1]$ is proportional to ω^3 and therefore $C(\omega) \propto \omega^2$. The plot of VDOS at low frequency given by Hannon *et al.*[12(b)] is compared to the reduced intensity of LFR, $I(\omega)/[n(\omega)+1]$ in Fig. 7. In the 7–16 cm^{-1} spectral range the VDOS is multiplied by ω and by $\omega^{-0.2}$ from 20 to 100 cm^{-1}. This comparison shows that $C(\omega) \propto \omega^2$ for $\omega < 16$ cm^{-1} and that $C(\omega) \propto \omega^{0.8}$ for $\omega > 20$ cm^{-1}. On the other hand we notice that at frequencies higher than the spectral range where the ω^2 dependence of VDOS and $C(\omega)$ exists, the slopes of $I(\omega)/[n(\omega)+1]$ and of $N(\omega)$ decrease with the frequency, contrary to that observed for silica.
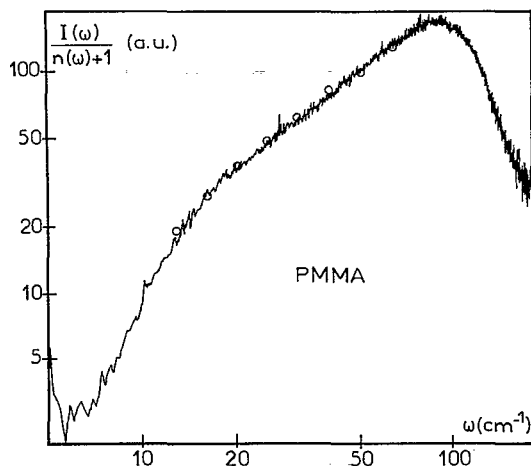

FIG. 6. Log–log plot of the LFR reduced intensity from v-SiO$_2$ at 55 K. The circles correspond to the VDOS obtained from INS at room temperature (Ref. 11), multiplied by ω up to the dashed horizontal line, and by $\omega^{0.05}$ at higher frequency.

In Fig. 3 it can be observed that there is an excellent correspondence between the reduced LFR intensity $I(\omega)/[n(\omega)+1]$ and the VDOS from INS (Ref. 13) for the polybutadiene. This means that $C(\omega) \propto \omega$, while the VDOS is not frequency power-law dependent.

The LFR reduced intensities of PMMA and DGEBA are also compared to the respective VDOS in the spectral range explored by INS (Refs. 14 and 15) (Figs. 4 and 5). A good agreement is obtained for PMMA with $C(\omega) \propto \omega$. In DGEBA the best agreement, is for $C(\omega) \propto \omega^{0.95}$.

V. DISCUSSION

A. Effect of structural relaxation on inelastic light scattering

In Figs. 1 and 2 it is shown that there exists a quasielastic light scattering that is not a Raman scattering from

FIG. 5. Log–log plot of the LFR reduced intensity from PMMA at 62 K. The circles correspond to the VDOS obtained from INS at room temperature (Ref. 14).
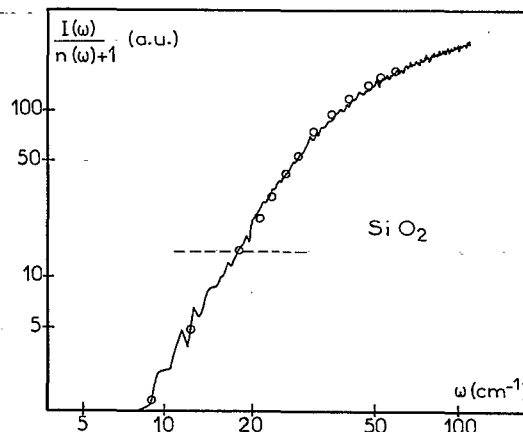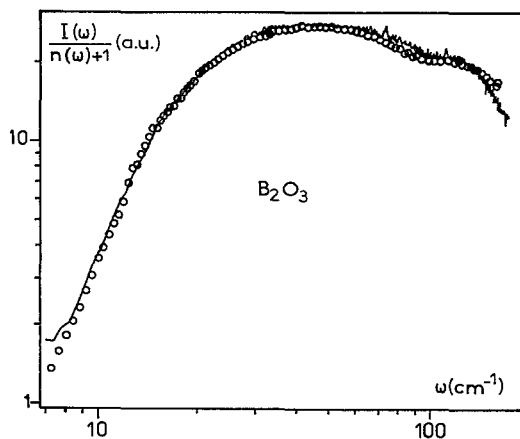

FIG. 7. Log–log plot of the LFR reduced intensity of v-B$_2$O$_3$ at 62 K. The circles correspond to the VDOS obtained from INS at room temperature [Ref. 12(b)] multiplied by ω for $\omega < 16$ cm^{-1}, and by $\omega^{-0.2}$ for $\omega > 20$ cm^{-1}.

vibrations. As shown by Buchenau et al.,[16] this quasielastic scattering comes from thermally activated structural relaxation. It is also observed by inelastic neutron scattering.[16] But we emphasize that the ratio of light scattering from relaxation motions over vibrational Raman scattering is in general much larger than the corresponding ratio in neutron scattering. The intensity of inelastic neutron scattering is the time Fourier transform of the structure factor.[17] On the other hand the inelastic light scattering is the space-time Fourier transform of the correlation function of the dielectric susceptibility fluctuations.[2] The amplitude of fluctuations is related to the amplitude of atomic displacement. Since the amplitude of a relaxation displacement is much larger than the amplitude of a vibrational displacement, the relative contribution of relaxation can be large in light scattering.

Figures 1 and 2 show that the thermally activated structural relaxation has an effect in inelastic light scattering up to 20 cm^{-1} in vitreous SiO_2, and 11 cm^{-1} in vitreous B_2O_3 at room temperature. Consequently, in order to determine the light-vibration coupling coefficient $C(\omega)$ from the comparison of Raman scattering with VDOS obtained from neutron scattering, it is necessary to use Raman scattering down to a frequency smaller than ≈ 10 cm^{-1} obtained at temperatures much lower than T_g (< 100 K in SiO_2 and B_2O_3).

B. Light-vibration coupling

1. Correlation in glasses

Two very important points follow from the comparison of Raman scattering with inelastic neutron scattering.

(1) At very low-frequency, where an ω^2-dependence of VDOS is observed, as in vitreous SiO_2 ($\omega < 20$ cm^{-1}) and B_2O_3 ($\omega < 16$ cm^{-1}), it is found $C(\omega) \propto \omega^2$. This is exactly what is expected in a nonfractal material, where $\tilde{d} = D = d$ for nonpropagating acoustical modes, in the case of no-correlation (Table I in paper I). The correlation length is smaller than the localization-length, or the phase of vibrational wave function is randomly fluctuating. Such a behavior of $C(\omega)$ at very low-frequency was shown in earlier models.[2,18] Also it was deduced from Raman experiments, for several glasses,[7,8] in particular for B_2O_3,[8] by assuming a Debye VDOS (ω^2) without experimental confirmation. However a recent comparison[19] between LFR and heat capacity shows that, even below $\omega = 20$ cm^{-1}, $C(\omega) \propto \omega$ for SiO_2 and B_2O_3.

(2) The light vibration coupling coefficient is found proportional or approximately to the frequency; $C(\omega) \propto \omega$, at frequencies higher than the ω^2 regime, whatever is the type of glasses (inorganic, organic or polymeric), and whatever is the ω-dependence of VDOS. This common character would be, a priori, explained by a common origin.

In the theory presented in paper I,[6] the radial correlation was distinguished from an angular correlation. The expression of the light-vibration coupling coefficient in spherical coordinates (r,θ,ϕ) is

$$C(\omega) \propto \int_0^\infty \nabla\bar{\psi}(r) r^{D-1} \int_0^\pi \exp\left(-\frac{r\theta}{a}\right) d\theta \int_0^{2\pi}$$

$$\times \exp\left(-\frac{r\phi \sin\theta}{a}\right) \sin\theta \, d\phi \cdot \int_0^\infty \nabla\bar{\psi}(r+\tau)$$

$$\times (r+\tau)^{D-1} \exp\left(-\frac{\tau}{b}\right) d\tau, \tag{2}$$

where $\bar{\psi}$ expresses the radial exponential decay of the vibrational wave function with a localization length l; a and b are the angular and radial correlation lengths, respectively, D is the fractal dimension. The presence of the correlation function in Eq. (2) was justified by the random fluctuations of the vibrational wave function.

With the dispersion law[20,21]

$$\omega \propto l^{-D/\tilde{d}}, \tag{3}$$

where $\tilde{d}$ is the spectral dimension, $C(\omega)$ becomes

$$C(\omega) \propto \omega \tag{4}$$

in the case of radial correlation ($a < l \leqslant b$) and $\tilde{d} = D = d$, for any the Euclidean dimension d.

The linear frequency dependence of the light-vibration coupling coefficient $C(\omega)$, experimentally observed in glasses, is interpreted by a radial correlation. Furthermore the glass structure is nonfractal on distances corresponding to the considered spectral range. This result appears very sensible. Indeed it is not surprising that correlation exists along a line in the glass and not between lines.

The fact that $\tilde{d} = D = d$ means that the glass structure is nonfractal in the 3D space, but also that the elasticity is normal. The elastic force constant is proportional to l^{d-2}, with l the vibration localization length, and therefore the dispersion law is simply $\omega \propto l^{-1}$, whatever is d.

That appears to be surprising is that $C(\omega)$ has a frequency power-law dependence while VDOS does not. If we consider the case of SiO_2, we observe that for ω just > 20 cm^{-1}, the VDOS begins to increase more rapidly than ω^2 and then increases slowly for higher frequencies. It is possible to show that the VDOS increases with the localization of phonons and has a maximum at the frequency corresponding to the correlation length.[22,23] However we think that it is not possible to explain the behavior of VDOS by a simple continuous network (CRN) in particular in the case of SiO_2. We observe that the transition between the two vibration regimes, at 20 cm^{-1} for SiO_2, is abrupt, for the VDOS (Ref. 11) and $C(\omega)$. In a CRN model a progressive variation of VDOS would be expected when the vibration localization approaches the correlation length. Furthermore, if the glass structures was continuous, models of light-vibration coupling of the Martin–Brenig-type would be applicable,[18,24] and we would observe therefore a maximum of $C(\omega)$. This has not been observed in different studied glasses in which $C(\omega)$ is simply proportional to ω.

The experimental results can be explained if we assume that the glass structure is noncontinuous, and that there is

a weakening of atomic bonding in different points of the network which isolate small parts or clusters. At the crossover from the low-frequency vibration regime $[C(\omega) \propto \omega^2]$ to the higher-frequency regime, there is localization of vibration modes inside the clusters. This localization is necessarily accompanied by a change of the VDOS behavior. Due to a softening of the vibration modes, which have a large amplitude at the interface between clusters, and to a possible lower dimensionality of the clusters than the dimensionality $d=3$ of the glass at longer distances, we expect an increase of the frequency-dependence of VDOS at the crossover. The increase of VDOS at the crossover is at the expense of VDOS at higher frequencies. In consequence a decrease of the slope of VDOS against ω would occur at higher frequencies.

This description of vibration behavior is corroborated experimentally and by simulation. It was observed that when the disorder increases the VDOS at low-frequency increases at the expense of VDOS at higher frequency.[25,26] By computer simulation, Nagel et al.[27] showed that the low-frequency VDOS increases when the number of bonds taken out increases.

The hypothetical clusters can have any more a less complex shape. Furthermore, as noticed before, they can be one-dimensional, i.e., linear or filamentary, planar (2D) or volumic (3D). The effect of their size-distribution on the VDOS was studied in a recent paper.[5] It was shown that the mean cluster size $2a$ is given approximately by the following expression:

$$2a = Sv/\omega_0,$$

where v is the sound velocity in the glass, ω_0 the frequency at the inflection point of the VDOS or of the reduced Raman intensity $I(\omega)/[n(\omega)+1]$ against ω, and S is a shape factor. Without knowing the shape of clusters we choose $S = 0.65$, that is an intermediate value between 0.8 for a spherical shape and 0.5 for a linear one. In the case of silica $\omega_0 \approx 30$ cm^{-1} taking $v = 4000$ m/s, we find $2a = 29$ Å. This value is very close to the one given by the radial distribution function in x-ray diffraction.[28,29]

In polymer-glasses (PMMA, polybutadiene) and DGEBA, we do not explore the regime $C(\omega) \propto \omega^2$ in the LFR-INS comparison. Certainly because this regime is at very low frequency and is not observed with a sufficient precision by LFR, but also because INS data are not available at very low-frequency, like for PMMA.

Other explanations of $C(\omega) \propto \omega$ behavior could be suggested. It can be assumed that at the crossover there is a change of the spectral dimensionality $\tilde{d}$. Such an interpretation could be justified in the case of PMMA and DGEBA, as a frequency power law-dependance of $I(\omega)/[n(\omega)+1]$ is clearly observed in the 15–80 cm^{-1} spectral range. This would give $\tilde{d} \approx 1.5$ and 1.8 for DGEBA and PMMA, respectively. Taking $D = d = 3$, since experimental results showing a fractal structure of the glasses are not known, the LFR intensity would be interpreted assuming no-correlation. Although this interpretation was suggested in earlier studies,[4,30] the first one is preferred because it is general for common experimental results.

2. Correlation in silica-aerogels

The silica-aerogels prepared by neutral hydrolysis of tetramethoxysilane have a fractal structure with a fractal dimensionality $D = 2.4$, as shown by small angle neutron scattering.[31]

A frequency power-law dependence of the reduced Raman scattering intensity $I(\omega)/[n(\omega)+1]$ was observed[31] over varying spectral range, dependent on the aerogel density. For an aerogel with a density of 0.2, a power law was found between 1 and 8 cm^{-1},[32] $I(\omega)/[n(\omega)+1] \propto \omega^a$, with $a = -0.36$ From inelastic neutron scattering Vacher et al.[33] deduced a VDOS, $N(\omega) \propto \omega^{\tilde{d}-1}$ with $\tilde{d} = 2.2$ in the same spectral range. Writing $C(\omega) \propto \omega^x$, $a = x + \tilde{d} - 2$, and therefore $x = -0.56$. From Table I of paper I, this value of x corresponds to a full correlation, $x = (2-D)\tilde{d}/D$. With $D = 2.4$ and $\tilde{d} = 2.2$, it is obtained $x = -0.37$. For the other cases of correlations the calculated values of x are positive and very different from the experimental one. The Raman scattering from neutral silica-aerogels is then in agreement with a full correlation.

The considered spectral range corresponds to a fractal arrangement on a length of ~ 100 Å. It means that the correlation lengths for the fluctuations, coming from disorder, are longer than 100 Å.

VI. CONCLUSION

Low-frequency Raman scattering was obtained for different types of glasses, with a spectrometer of high resolution and rejection rate. Raman scattering was measured close to the laser line. It was observed that a non-negligible contribution to light scattering, due to structural relaxation, adds to the Raman scattering up to more than 10 cm^{-1}, at room temperature. Consequently, it is necessary to measure Raman scattering at low-temperatures, at which the structural relaxation is negligible.

Comparison between LFR and VDOS, obtained from inelastic neutron scattering, allowed the determination of the light-vibration coupling coefficient $C(\omega)$. At very low-frequency it was found that $C(\omega) \propto \omega^2$ in the same spectral range where the VDOS determined by INS, is proportional to ω^2. This frequency power-law dependence was consistent with localized randomly fluctuating acoustical vibrations in a nonfractal disordered network. In this vibration regime, the vibration localization length is longer than correlation lengths characteristic of the glass. This vibration behavior was clearly observed in vitreous SiO_2 and B_2O_3.

At the crossover between the two vibration regimes the ω-dependencies of VDOS and $C(\omega)$ change. Above ω^2-dependence regime, the VDOS does no longer follow a frequency power-law-dependence and $C(\omega)$ becomes proportional to ω, $C(\omega) \propto \omega$. This behavior is explained by localization of vibrations in inhomogeneities or clusters that are isolated by a weakening of bonding, due to the disorder, in different point of the glass network. The consequence is a softening of vibration modes and an increase of VDOS at low-frequencies at the expense of the VDOS at higher frequencies. The interpretation of the VDOS excess

in silica by rotations of SiO_4-tetrahedra[3] is too specific for such a universal characteristic.

The law $C(\omega) \propto \omega$ is explained in our model by assuming a radial correlation without angular correlation in a nonfractal system with a normal elasticity, $\tilde{d} = D = d$. This is compatible with inhomogeneities which are one-, two-, or three-dimensional. It is the first time that a so clear and simple behavior of $C(\omega)$ is shown. The behavior, $C(\omega) \propto \omega$ is universal in glasses of different types, inorganic, organic, or polymeric. A similar behavior was seen in several glasses that we have not studied. A good example is that of borate glasses[34] for which the behavior of VDOS and $C(\omega)$ is very similar to that of silica.

A full correlation is found for silica-aerogels. This means that the effect of random fluctuations on Raman scattering is negligible and that gels are more ordered than glasses. This difference can be explained by the different ways to obtain an aerogel and a glass, condensation and aggregation or polymerization at relatively low-temperature for aerogels, and from melt for fused glasses.

ACKNOWLEDGMENTS

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