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BRILLOUIN SCATTERING OBSERVATION OF PHONON SCATTERING BY
INHOMOGENEITIES IN PHASE SEPARATED GLASSES

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Résumé.- Nous avons étudié par diffusion Brillouin l'atténuation et la
vitesse des ondes hypersoniques dans un verre formé en trempant une solu­
tion aqueuse de LiCl (contenu d'électrolyte : 10 moles pour cent).
Ce matériau a été utilisé parce que la séparation de phase peut y être
facilement réalisée par des traitements thermiques : nous l'avons détectée
en mesurant l'intensité de la diffusion Rayleigh. Nous avons observé une
variation des propriétés élastiques avec le traitement thermique.
Les différences observées entre les échantillons sont indépendantes de la
temperatur et paraissent liées à la diffusion des phonons acoustiques par
les inhomogénéités structurales.

Abstract.- We have studied by Brillouin scattering the attenuation and
velocity of hypersonic waves in a glass formed by quenching an aqueous
solution of LiCl (molar electrolyte content : 10 %). This material has
been used because the phase separation in this material can be easily
controlled by heat treatments and detected by measuring the Rayleigh scat­
tering intensity. The elastic properties appear to depend on heat treatment.
The differences observed for
various samples are independent of temperature and seems likely to be due
to scattering of acoustic phonons by structural inhomogeneity.

Fluctuations of the macroscopic physical properties as refractive index,
density, elastic constants are known for long time to scatter energy of light and
acoustic waves in inhomogeneous materials. Such fluctuations are present in glas­
ses, and can be easily detected even in perfectly transparent materials from elas­
tic Rayleigh scattering of light. Microscopic fluctuations are also present in
glasses and have been proposed as a possible scattering mechanism for phonons [1].
However, all the existing Brillouin scattering measurements in glasses have demons­
trated that the structural scattering of high-frequency phonons, if it must exists
in theory is a very small effect with is at maximum of the same order as the expe­
rimental uncertainty [2]. As the amplitude of the fluctuations must be a determi­ning
factor on this process, we have studied the glass forming mixture LiCl :
H2O , in which a phase separation has been detected in elastic neutron scattering
experiment for appropriate electrolyte concentration [3]. Recent studies [4] have
demonstrated that an homogeneous nucleation take place in this glass, leading to
the appearance of cubic ice crystals in the vitreous matrix. The glass transition
of this mixture is around 140 K , near the crystallization temperature, and the
process of crystallization can be easily controlled by varying the heat treatments.
The increase of the intensity of Rayleigh scattering can be used as a test of the
increase of the size of domains.

In order to obtain a glass, the solution containing 10 % LiCl in water
was first rapidly quenched into the same cryostat used for light scattering measu­
rements. Cracks related to internal stresses were then present in the material, but
some regions of the sample were transparent enough to allow performing Brillouin
scattering measurements. After this first study, the sample was put a short time
at a temperature of 138 K , 2 K below the glass transition temperature, in order
to relax the stresses. A perfectly transparent sample was then obtained, on which
Brillouin scattering measurements were performed. We found very small differences between the quenched and the relaxed samples. The sample was then held for a few hours at 140 K: the phase separation then appeared and was detected by observing the Rayleigh scattering of light. At the end of the thermal treatment, the intensity of the Rayleigh line was about 1000 times that in the relaxed sample. However, the sample was transparent enough to keep a good definition of the wave vector of the incident and scattered light and to be sure that a parasitic broadening due to a spreading of light was not present.

The measurements on the phase separated glass are compared to the original sample in Figs 1 and 2. The velocity \( v \) of acoustic waves is related to the measured frequency shift \( \nu_B \) by \( \nu_B = (2nv/\lambda_0) \sin(\theta/2) \) where \( n \) is the refractive index, \( \lambda_0 \) the wavelength of incident light in vacuum and \( \theta \) the scattering angle. The attenuation \( \alpha \) can be deduced from the observed half-width at half-maximum \( \Gamma \) of the Brillouin line from the relation: \( \Gamma = \alpha v/4\pi \). The most striking feature is a difference in the attenuation between the two sample: this difference remains constant between 10 K and the phase transition. The amplitude of the excess attenuation of 20 GHz longitudinal acoustic phonons is of about 700 cm\(^{-1}\), which is well above experimental uncertainty.
A glass containing large density or concentration fluctuations, which are related to the intensity of the Rayleigh scattering of light, can be thought to exhibit correlatively large fluctuations of elastic constants, and therefore to present, the best conditions for the observation of the phonon scattering by static inhomogeneities. A constant excess attenuation between 10 K and the glass transition seems very difficult to explain by dynamical effects. It seems therefore likely that the difference observed between the two samples is due to static structural effects.

On the other hand, this contribution is small, even in a very inhomogeneous glass. The order of magnitude of our result indicates that this effect must be negligible for more homogeneous glasses.

References.-