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Effect of fictive temperature on medium range order in v-SiO_2

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Abstract

The first-order Raman spectra of vitreous SiO_2 between 10 and 650 cm^{-1} have been measured as a function of sample fictive temperature, T_F, achieved by laser quenching. The changes observed in the high frequency range of the spectra, > 100 cm^{-1}, which is related to the short range order, are correlated with the shift of the 'Boson peak' (BP) which is present in the low frequency range, < 100 cm^{-1}, and characteristic of the medium-range order. Based on the non-continuous glassy structure model, these observations are attributed to a decrease in the size of domains, associated with a modification of the Si–O–Si angle distribution, with the increase of T_F. © 1997 Elsevier Science B.V.

1. Introduction

The properties of glasses at low excitation energies differ from those of crystalline solids and appear to be universal [1,2]. These properties result from structures on the scale of a few nanometers which are involved in the medium range order. Furthermore, it is known [3–6] that various parameters such as the composition, the chemical preparation, the OH impurity concentration and the fictive temperature affect the vitreous structure. The structure of this medium range order is not well determined.

Experimental investigations show the existence of a non-Debye density of vibrational state, $g(\omega)$, with a universal shape in glasses [7,8]. Most of the models relate this to the vibrational excitations localised on a nanometer scale but the source of these vibrational excitations is not completely explained.

The aim of this paper is to relate the changes observed in the first-order Raman spectra to structural modifications of the silicate network as a function of the fictive temperature, T_F (T_F is a temperature, greater than the glass transition temperature, at which the glass is allowed to reach an equilibrium state before a rapid quenching to room temperature).

The Raman spectra of vitreous silica may be divided into three parts: the very-low frequency range (< 10 cm^{-1}), the low frequency range (between 10 and 100 cm^{-1}) and the high frequency range (> 100 cm^{-1}). The region corresponding to 'quasi-elastic light scattering excess' (QLSE), is temperature dependent. A description in terms of relaxational modes was developed in order to explain neutron and inelastic light scattering [9]. The low frequency contains a band around 80 cm^{-1} usually called the 'Boson peak' (BP). The high frequency range is made up of a band and two defect lines, D_1 and D_2. The VV polarised band around 430 cm^{-1} is attributed to a Si–O–Si stretching mode whereas the 490 cm^{-1} (D_1) and 606 cm^{-1} (D_2) lines are at-

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