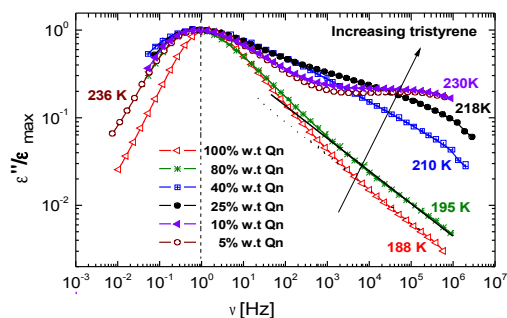# Interdependence of Primary and Johari-Goldstein Secondary Relaxations in Glass-Forming Systems

An important aspect concerning the relaxation dynamics of glass forming materials is the investigation of the molecular origin of secondary relaxation and its possible connection to the structural relaxation and thus to the glass transition phenomenon [1]. Binary mixtures of rigid polar molecules dissolved in apolar solvents are excellent systems to deepen the understanding of the molecular slowing down and of the origin of secondary relaxation in glassy systems [2-4]. In fact, these systems are ideal to study the secondary Johari-Goldstein, JG, β-relaxation process, which is a local process with intermolecular origin. The JG relaxation is most easily detectable in liquid composed by rigid molecules where secondary relaxation related to intramolecular degrees of freedom are absent [1,2]. In binary mixtures, the different size of the two components often prevent the system to crystallize, allowing its study in a broad temperature interval from below to well above the glass transition temperature T_g.

The molecular dynamics of polar rigid molecules (heterocyclic compounds: *tert*-butylpyridine, Quinaldine, QN) dissolved in an apolar solvent (tristyrene, 3Styr) with higher T_g investigated by means of dielectric spectroscopy at ambient and elevated pressures. By changing in a systematic way the relative concentration of the components it was observed a transition from a relaxation scenario with a structural α-process and an excess wing (in the case of the neat polar systems), to that with a structural process and a well resolved β-secondary process (in Fig. 1 representative spectra of the Qn 3Styr mixture are reported). Moreover, we observed, by means of dielectric spectroscopy under high pressure, that the secondary relaxation time is constant [5,6]. As a consequence the structural and secondary JG relaxation process have the same frequency separation in spectra measured at different value of temperature and pressure but the same value of τ_α (Fig. 2). These results support the idea that the Johari-Goldstein relaxation acts as a precursor of the structural relaxation and therefore of the glass transition phenomenon [7].
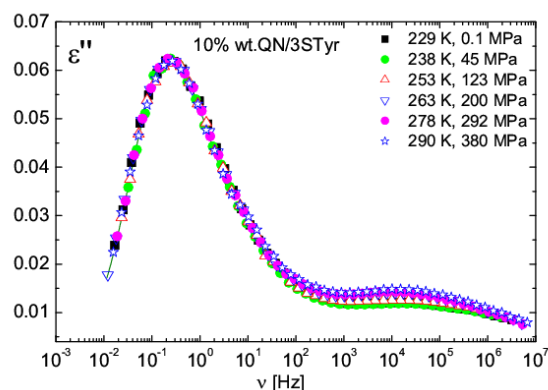
2: Master plot of different loss spectra for 10%QN in 3Styr with the same value of τ_α=0.67s corresponding to different combinations of temperature and pressure values. The line is a guide for eyes.

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1: Bi-logarithmic plot of the normalized dielectric loss vs. frequency of mixtures of Quinaldine in 3Styr at different concentrations and temperature (as reported in the figure).

JG secondary relaxation in binary mixtures is very sensitive to volume reduction [5,6] and that its pressure dependence can be rationalised according to CM [1]. In particular we found that, at a fixed value of the structural relaxation time, τ_α, the dispersion of α-relaxation is constant, independent of thermodynamic conditions (T and P): that is, the shape of the structural relaxation function depends only on the α-relaxation time. At the same time we found also the JG