1. Background and Context

Glasses are usually produced by cooling a liquid fast enough to avoid crystallization. If so, the liquid enters into the supercooled region where the viscosity increases by many orders of magnitude as the temperature is decreased by a relatively small fraction of the melting value T_m [1]. When the system becomes arrested on the experimental time-scale, the glass transition is reached. Below the glass transition temperature T_g, the system is frozen into an amorphous, liquid-like structure, and the atoms can only vibrate around their disordered equilibrium positions.

The study of the vibrational dynamics of glasses has pointed out the presence of many intriguing peculiarities with respect to their ordered crystalline counterparts. One of the most striking features is the occurrence of a broad peak in the low-energy part of the reduced vibrational density of states $g(\omega)/\omega^2$. This peak is named Boson Peak (BP) and originates from an excess of modes in the vibrational density of states (VDOS) with respect to the Debye ω^2-prediction. The BP has a direct relation to the low-temperature thermal properties and represents a universal feature of glasses and, more generally, of disordered solids [2]. Despite several decades of investigation, some questions remain open. Notably: i) the nature of its modes, collective *vs* localised excitations, and ii) the mechanism of the low-frequency piling-up. Moreover, a recent study pointed out the existence of a relation between the behaviour of the viscosity as the supercooled liquid approaches the glass transition and the vibrational properties of its parental glass [1]. This result relies on a link between the BP and the fragility parameter of the supercooled liquids, whose origin might be embedded in the nature of the non-ergodicity factor [3,4]. The latter quantity is the infinite-time limit of the density-density auto-correlation function and describes the dynamics across the glass transition, thus providing an insight into its complex phenomenology [5].

To understand the BP nature, an effective approach is the systematic study of the BP as a function of parameters such as temperature, press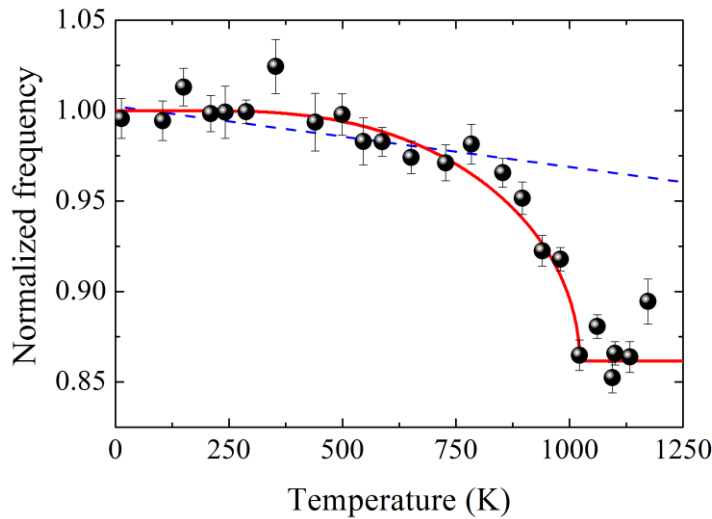ure, density, etc. [6-9]. By tuning these parameters, the BP shifts in energy and changes its intensity. Actually, in many systems the BP shape was found invariant, meaning that the intensity modification is only apparent, due to the peak shift. Furthermore, in some cases, the shift can be explained by simply considering the variation of the elastic medium plus a disorder-induced broadening of crystal-like features [9]. Interestingly, vitreous germania (v-GeO$_2$) displays a high-temperature critical-like crossover from a Debye to a non-Debye regime as shown in Fig. 1 [8]. This finding suggests a more complex scenario where the BP evolution is governed by at least two competing mechanisms, whose origin and weight have yet to be ascertained.

Figure 1. Debye to non-Debye crossover in v-GeO$_2$. Black dots represent ω_{BP} as obtained by Raman scattering; the blue dashed line indicates the calculated Debye frequency, ω_{DB}; both data are normalised to their lowest temperature value. The red line highlights the critical behaviour and it is obtained with a mean-field approach. Data are from Ref. [8].

2. Proposed experiment

Vitreous silica v-SiO$_2$ is the archetype of strong glasses and has the same corner-sharing tetrahedral structure as GeO$_2$. However, $T_g \sim 1500$ K and $T_m = 1998$ K have strongly limited the investigations of the vibrational dynamics in the supercooled and liquid phases of this system. We thus propose to map the BP T-evolution from room temperature up to the liquid state, accessing the almost unexplored supercooled liquid region. In particular, we want to investigate if the Debye to non-Debye

crossover and the critical shape observed in v-GeO$_2$ are system-dependent properties or a more general BP feature, and thus a possible signature of its underlying nature.

3. Summary of previous beamtime or characterisation
The v-SiO$_2$ sample is promptly available in our laboratory and it has been extensively characterised via several techniques (Raman and Brillouin spectroscopy, X-ray diffraction, and inelastic X-ray scattering). In particular, we measured the VDOS at room temperature in absolute units using nuclear resonance analysis of inelastic x-ray scattering carried out at the beamline ID18 at ESRF (experiment n. SC/4955, Ref. 10). The *T*-evolution of the collective excitations and the VDOS were already measured up to fairly high temperature (1620 K) via inelastic X-ray scattering and inelastic neutron scattering, respectively [11].

4. Justification of beamtime request
We plan to use LET to study the VDOS at room temperature and then at 8 temperatures between T_g and T_m. To the best of our knowledge, the RISO furnace, available at LET, is the only furnace in Europe that allows to reach the sample environment conditions for the proposed neutron scattering study. We will use an incident energy of about 25 meV with a resolution (full width at half maximum, FWHM) of 500 µeV. By exploiting the repetition rate multiplier (RRM) at LET, we will simultaneously investigate the $S(q,\hbar\omega)$ with relatively high resolution (< 50 µeV). Such a setup will allow us to determine the non-ergodicity factor from the ratio $S(q,0)/S(q)$, both during the VDOS measurements and during the heating ramps, thus providing an overall characterisation of the dynamics between T_g and T_m, and possibly above T_m.

The sample will be a thick hollow cylinder of SiO$_2$ (Suprasil, purchased from Silo, Florence) obtained by piling up small glassy rings. The sample holder will be a niobium hollow cylinder with a screw cap. This arrangement fully exploits the beam and reduces the multiple scattering. A properly designed vanadium sample will be also measured for normalisation. Data treatment will include normalisation, background subtraction, corrections for absorption and multiple scattering contributions through Monte-Carlo simulations. Starting from the properly normalised $S(q,\hbar\omega)$ we will obtain the generalised VDOS and thus the low-energy vibrational dynamics.

Considering the measurements of the VDOS at nine temperatures (4-6 hours/T, for both sample and empty cell), the time needed to heat up and cool down the furnace, and ancillary measurements (empty cell and vanadium), we ask for **5 days** of beamtime.

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