Local structure and dynamics of tungsten oxide-based glasses: insights from concurrent neutron diffraction and Compton scattering

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Abstract
In this work, following our previous work on molybdate glasses, we employ a combination of neutron diffraction and neutron Compton scattering, augmented by ab initio harmonic lattice dynamics and Reverse Monte Carlo modelling to characterise the force-constant disorder in the tungsten oxide-based glasses. Specifically, we discuss the correlations between the average interatomic force constant magnitudes inferred from neutron Compton scattering and the glass formation ability, measured in terms of the value of the glass transition temperature, as well as the average bond-lengths and interatomic distances obtained from diffraction data analysis. Moreover, we provide a comparative analysis of the widths of force-constant distributions of individual atomic species in glasses and their precursor metal oxides based on the distributions of the widths of nuclear momentum distributions. Furthermore, we assess the degree of softening of atom-projected vibrational densities of states induced by the force-constant disorder in the glasses.

1. Introduction
Recently WO3-based glasses have attracted scientific interest because of their specific thermal, physical and optical properties due to the presence of WO3, rendering them attractive for engineering towards tailored advanced multifunctional devices [1—3]. Despite the flurry of research activity, there is still not enough local structural and dynamical data present in the literature that would shed more light on the structure-property relationships in these functional materials.

The information content inferred from the studies of disordered systems can be improved by simultaneously applying diffraction and spectroscopic methods and supplementing them with ab initio modelling to understand the correlation between the dynamical and structural properties [4—12]. Motivated by this, we have recently performed neutron diffraction measurements combined with Reverse Monte Carlo (RMC) modelling to calculate partial atomic pair correlation functions, nearest neighbouring distances, and coordination number distributions in many amorphous systems [4, 13—23].

Apart from the local structural information inferred from diffraction studies augmented with RMC, functional disordered materials are in need of local investigation of dynamical properties, and amongst them, most crucially, the interatomic force-constants. Force-constant values and their distributions in disordered systems are believed to be responsible for a plethora of mechanical and thermodynamic properties. In general, it is believed that the distribution of force-constant values leads to the broadening and softening of phonon modes [24, 25]. The most iconic manifestation of this phenomenon is the broadening/lowering of the lowest van Hove singularity in the corresponding reference ordered glass-precursor system due to the distribution of force
constants, believed to be responsible for the existence of the Boson peak in glasses \([26, 27]\). Boson peak, manifested by an excess of vibrational modes at low frequencies and the excess heat capacity at low temperatures exceeding the Debye predictions \([28]\), is believed to control or affect all anomalies and behaviours in the specific heat, heat conduction and low-temperature properties of solids \([29]\).

The \textit{a priori} knowledge of the mean values and distributions of interatomic force-constants is crucial in the context of modelling of materials properties. Experimental techniques capable of accessing the values of force constants in materials are indispensable sources of control and benchmarking of force fields. Historically, the interatomic force-constant values, inferred from spectroscopic studies, were employed to construct the first classical force fields for molecular dynamics (MD) simulations. With the advent of density functional theory (DFT), simulating a wide range of condensed matter systems became possible with a reasonable computational cost and accuracy. \textit{Ab initio} molecular dynamics (AIMD), offering a way to simulate the atomic motion using forces computed at the DFT level, has extended the capabilities of \textit{ab initio} materials modelling to systems subject to finite temperature and pressure conditions. Unfortunately, computational requirements limit the timescale and size of AIMD simulations to a few hundred atoms for a few hundred picoseconds of time, rendering the investigations of complicated disordered systems practically intractable. Thus, large-scale MD simulations must resort to empirically derived analytical interatomic force fields with a fixed functional form that are in their nature less accurate than their DFT-derived counterparts \([30]\). In an attempt to compute forces with \textit{ab initio} accuracy at the cost of classical interatomic force fields, computational protocols have recently been established that avoid the problems of machine learning (ML) algorithms and generate not only the force-constant values but also their predictive distributions \([30]\). Thus, it is desired to have a source of force-constant distributions derived directly from experiments that could provide an additional element of control of the accuracy and reliability of the active learning of MD models.

Motivated by these recent developments, in our investigations of amorphous systems, we have extended the portfolio of techniques by applying, apart from the usually employed inelastic neutron scattering spectroscopy (INS), Raman scattering (RS), and solid-state nuclear magnetic resonance (SS NMR) \([20, 31–37]\), the neutron Compton scattering (NCS) technique \([4, 8, 10]\). Due to its inherent isotopic-mass selective nature and the fact that its main observables are widths of nuclear momentum distributions characterising the degree of local atomic binding \([12, 38, 39]\), this technique has proven very useful for the characterisation of the force-constant distributions in glasses \([4]\). The advantage of NCS comes to the fore, especially in disordered systems, where positional, isotopic mass, and force-constant disorder renders vibrational densities of states (VDOSes) impossible to resolve by means of techniques such as INS and RS \([4, 40]\). Moreover, due to the fact that NCS effectively probes nuclear quantum dynamics and nuclear binding potentials at the scale of a fraction of an Angstrom \([12, 38, 39]\), this technique provides a dynamic counterpart to the local structural information inferred from diffraction augmented by the reverse Monte Carlo (RMC) analysis.

In the present work, we describe the results of neutron diffraction, augmented by RMC modelling and NCS measurements on tungsten glasses, as well as \textit{ab initio} modelling, in the framework of the periodic-DFT, on their precursor metal oxides, ZnO, Al\(_2\)O\(_3\), La\(_2\)O\(_3\), WO\(_3\), and Nd\(_2\)O\(_3\). Using the computational protocol established in our previous work on molybdate glasses \([4]\), we provide an assessment of the mean values and variances of the average effective inter-atomic mean forces. We use these variance values as upper-conservative bounds of the isotopic disorder-induced force-constant disorder. Moreover, by benchmarking the values of the average force constants and their variances obtained for the glasses against the \textit{ab initio} predictions for their counterparts obtained for the metal oxide precursors, we assess the degree of softening of atom-projected vibrational densities of states induced by the force-constant disorder in the glasses. Furthermore, we investigate correlations between the degree of the force-constant disorder and the bond length distributions in the glasses. Such an approach allows us to relate the structural disorder to changes in nuclear dynamics of micro-heterogenous disordered systems where direct \textit{ab initio} modelling of nuclear quantum dynamics is beyond the computational power of state-of-the-art high-performance computing. Finally, motivated by recent work on molecular dynamics simulations on atomic nanoclusters, we investigate the correlations between the glass formation ability, measured in terms of the value of the glass transition temperature, and the flatness of the potential energy landscape measured through the magnitude of the effective force constants obtained from NCS experiments. The positive correlations between parameters characterising the microscopic structure and dynamics and the macroscopic parameters characterising the glass behaviour obtained in this work show that some results obtained from studies of nanoscale glasses may be potentially transferable to bulk glasses, and, in the case of the parameters characterising the potential energy landscape, the central role in this process is played by NCS observables.
2. Experimental details

2.1. Sample preparation
The glass samples with the nominal compositions of 75WO3-5ZnO-10La2O3-10Al2O3, 80WO3-10La2O3-10Al2O3, 75WO3-5ZnO-10Nd2O3-10Al2O3, 65WO3-20ZnO-5La2O3-10Al2O3 (hereafter referred as W75Zn5La10Al10, W75Zn5Nd10Al10, W80Zn5Nd10Al10 and W80Zn5Nd10Al10 (mol%) were prepared from reagent grade powders (WO3, ZnO, La2O3, Nd2O3, Al2O3). The powders were melted in a platinum crucible under atmospheric conditions at 1200 °C–1300 °C for 30 min. The melted mixture was kept at the melting temperature for two hours. The melt was quenched by pouring it on a stainless steel plate.

The raw metal oxide systems (WO3, ZnO, La2O3, Nd2O3, Al2O3) were prepared and measured in the form of fine powder samples. The WO3, of purity of 99.9% (CAS: 1314–35–8, LOT: BCBV1199) obtained from Sigma-Aldrich, the ZnO, of purity of 99.9% (CAS: 1314–13–2, LOT: MFCD00011300), the Nd2O3, of purity of 85% (CAS: 1313–97–9, LOT: M20C022), the La2O3, of purity of 99.5% (1312–81–8, LOT: SLCC7064) and the Al2O3 of 99.5% purity (CAS 1344–28–1, LOT: Q30E25) obtained from Alfa Aesar. In all cases, the isotopic mass composition corresponded to the natural abundance of all isotopes.

2.2. Neutron diffraction experiments
Neutron diffraction (ND) measurements were performed on the PSD diffractometer (λn = 1.069 Å) at the Budapest Neutron Centre [41]. The powder specimens of about 3–4 g each were filled in thin-walled cylindrical vanadium sample holders of 8 mm diameter. The raw data were corrected for detector efficiency, background scattering and absorption effects. The total structure factor, \( S(Q) \), was calculated by local software packages.

2.3. Reverse monte carlo simulations
The Reverse Monte Carlo (RMC) simulation is a powerful technique to build 3D structural models in accordance with experimental data, in particular total structure factors (\( S(Q) \)) obtained from diffraction experiments [42–48]. The \( S(Q) \) data recorded for the tungsten oxide-based glasses were modelled using the RMC++ software [49]. Details of the modelling protocol were described in our earlier work [23], and thus here, only the main framework of the analysis will be outlined. The main observable in total scattering experiments, the total structure factor, \( S(Q) \), is expressed throughout the analysis as a linear combination of the partial structure factors, \( S_{ij}(Q) \), weighted by the factors, \( w_{ij} \), according to:

\[
S(Q) = \sum_{ij} w_{ij} S_{ij}(Q)
\]

where \( c_i, c_j \) are the molar fractions, \( b_{ij} \) the coherent neutron scattering lengths, \( k \) is the number of the elements in the sample, and \( Q \) is the momentum transfer. Importantly, the whole analysis is performed under the assumption that the neutron scattering lengths of all elements are constant in the entire \( Q \)-range [50]. In the RMC procedure, starting from the initial atomic configuration, the initial atomic configurations are iteratively adjusted for increasingly improved convergence with the \( S(Q) \) provided by the experiment. For the starting configuration, we have used the initial atomic configuration constructed according to the protocol already established in our earlier work on similar glassy systems [4, 51]. During the RMC runs, cut-off constraints were used, constraining the minimum inter-atomic distances between two atoms. Several RMC runs were performed for each sample under investigation. During each run, the cut-off distances were modified, assuring realistic pair-distribution functions of interatomic distances, \( g_{ij}(r) \). About 30 atomic configurations were obtained from the RMC calculations of each sample corresponding to more than 1 100 000 accepted atomic movements inside the simulation box.

2.4. Neutron compton scattering
NCS experiments were performed at VESUVIO spectrometer at the ISIS neutron and muon spallation source at the STFC Rutherford Appleton Laboratory, Harwell, Oxfordshire, in the UK [9, 12, 39, 52, 53]. The measurements of all four tungsten glass samples were carried out at room temperature. The powder samples were placed into thin-walled flat aluminium cells. The cells were assembled out of two flat (one front and one backside) walls, each of cross-section of 64 square centimetres, fully exposed to the incident VESUVIO neutron beam when placed perpendicular to its direction. The general setup of VESUVIO was described elsewhere [9, 12, 39, 52, 53]. The mass-resolved NCS spectra, recorded in the neutron time-of-flight (TOF) domain by detectors placed at scattering angles between 130 and 170 degrees (referred to as the backscattering regime),
containing the recoil peaks of W, Zn, Nd, La and O as well as Al from the sample containers were treated in a protocol detailed in our previous work [4].

The NCS spectra were assumed to consist of recoil peaks with underlying purely Gaussian nuclear momentum distributions (NMDs) with standard deviations \( \sigma_M \) (where \( M \) is the mass of a given nucleus), hereinafter referred to as the NMD widths. Crucially for the assessment of the force-constant disorder, the values of \( \sigma_M \) obtained from NCS experiments can be used to calculate the magnitudes of the effective force constants, \( k_M \), acting on nuclei in tungsten-oxide based glasses. In order to obtain the values of \( k_M \), one can employ the theory of the mean force function (MF), with the MF defined as the average force acting on an atomic particle by keeping all other particles in the system fixed [54]. In the case of an isotropic harmonic potential, the MF is linear with atomic displacement, with the force-constant magnitude for a nucleus of mass \( M \) at temperature, \( T \) the magnitude of the force constant can be calculated using the formula [54]:

\[
k_M = \left( \frac{k_B T}{\hbar^2} \right)^2 M - k_B T \sigma_M^2
\]

(3)

where \( k_B \) is the Boltzmann constant. Importantly, formula (3) is not valid in the limit of \( T \to 0 \) due to the limitations of the sequence of Feynman–Trotter approximations to the thermal Feynman path integral for a general non-relativistic system characterised by a smooth, single-minimum interaction potential that underlies a momentum distribution of a given nucleus [54]. However, as the formula (3) has been successfully applied to the description of the mean force in polycrystalline and amorphous ice at 100 K [54], it will be used here to describe the mean forces of glasses and their parent metal oxides at 300 K.

As mentioned in our previous work glasses [4], in an NCS experiment, each recoil peak needs to be fitted with an underlying momentum distribution defined for a single atomic species with a standard atomic weight and the entire effect of the isotopic mass disorder is contained in the uncertainty of the fitted NMD width value, which is broadened due to the combined effect of the isotopic mass distribution and the nuclear momentum distribution of each isotope of the same atomic species present in a sample. Thus, in what follows, we will account for the NMD width disorder in the glass samples under investigation by simply quoting the mean values of the NMD widths together with the experimental uncertainties, as obtained directly from NCS experiments. The uncertainties of NMD widths will be assumed to stem from normal distributions of NMD width values and thus can be interpreted as standard deviations, an assumption that is in agreement with standard NCS data analysis schemes [55–59]. As far as the distributions of force constants values are concerned, we will use the Monte Carlo data error propagation according to the following scheme. Initial normal distributions of NMD widths will be generated according to the mean and standard deviation values of NMD widths as obtained from NCS experiments. The distributions will be propagated using formula (3), and the output distributions will be analysed in order to obtain mean values and confidence intervals of the force constants, not assuming that the output distributions must necessarily be normal. For the entire analysis, a bespoke Monte Carlo Error Propagation Toolbox [60], implemented in Matlab computing environment, will be used.

2.5. Modelling

Following the methodology already established in our work on molybdate glasses [4], we characterise the degree of disorder in tungsten-oxide based glasses under investigation using the fact that disorder leads to broadening and softening of phonon dispersion relations and densities of states, leading to concomitant lowering of the values of nuclear kinetic energies and thus NMD widths [61–63]. Consequently, one can place the magnitude of an NMD width of each nucleus present in the glass on a scale between the NMD width value computed from the Maxwell–Boltzmann nuclear momentum distribution with no underlying potential and an \textit{ab initio} prediction for the NMD width of this nucleus in a parent metal oxide system [4]. In the case of a nucleus that is present in multiple parent metal-oxide systems (such as oxygen), for the upper end of the scale, one can use a compositional average of \textit{ab initio} predictions for NMD widths [4]. The details of the computational scheme adopted for the \textit{ab initio} harmonic lattice dynamics of the precursor metal oxide systems, \( \text{Al}_2\text{O}_3 \), \( \text{La}_2\text{O}_3 \), \( \text{ZnO} \), \( \text{WO}_3 \) and \( \text{Nd}_2\text{O}_3 \) as well as the validation of the obtained \textit{ab initio} predictions, are described in section 1 of the Supplementary Information ((available online at stacks.iop.org/JPCO/5/075013/mmedia) SI).

3. Results and discussion

3.1. Local structure of tungsten glasses from neutron diffraction

Figure 1 shows the experimental structure factors, \( S(Q) \), obtained from the ND experiments on four different tungsten-oxide based glass systems, along with the results of RMC simulations. The convergence of the RMC calculation is very good for all the systems under investigation, and the final simulated \( S(Q) \) curves match very
well the experimental data. The experimental data are similar for all the systems, indicating similar average local order, although they show differences in fine details.

Information about the individual O-coordination of the metal ions in the glass systems under investigation can be obtained from the RMC analysis of the partial atomic pair distribution functions. The correlation functions for W–O, Al–O, Zn–O, O–O, La–O and Nd–O are shown in figure S2 in the SI.

The partial interatomic distances, the lowest-distance peak positions and the average coordination numbers derived from the X–O and the O–O partial pair correlation functions are summarised in table S1 in the SI. The analysis of the data listed in table S1 reveals some important structural features related to the local structure of the tungsten-oxide based glasses under investigation. The first structural motif shared by all of them is the fact that two W–O bond lengths are present at 1.75–1.80/2.55 Å. This result is similar to the one reported for B2O3–Bi2O3–La2O3–WO3 glasses, where two distances at 1.7–1.8 and 2.6 Å were found and attributed to the tetrahedral and octahedral W-environments [64]. Similarly, the local structure of sodium tungstophosphate glasses, as investigated by EXAFS and NMR, revealed two W–O distances at 1.75 Å and 2.20 Å, attributed to tungsten atoms coordinated by four and six oxygen atoms, respectively [65]. Contrary to the above findings, in the case of the simplest tungsten-oxide based glass, WO3–TeO2, only a unimodal distribution was found with W–O distances in the range 1.69–1.75 Å [18]. Thus, this binary distribution of the W–O bond lengths seems to be the blueprint of multicomponent tungsten-oxide based glasses exhibiting a relatively more complicated local structure.

The average coordination number distributions obtained from the RMC analysis reveal that W atoms have the nearest-neighbour environment of 4 and 6 oxygen atoms. The average W–O coordination numbers obtained from RMC modelling are 4.94, 5.17, 4.87 and 4.22 (±0.05) for the W75La10, W80La10, W75Nd10 and W65Nd5 glasses, respectively (cf. figure S3(a) in the SI). These values confirm that the W–O network consists of WO4 and WO6 units. Importantly, however, the WO3 content influences the WO4/WO6 ratios. In the case of glasses with a higher WO3 content (W80Nd10), the WO6 unit concentration is higher than in glasses with lower WO3 content (W65Nd5), and this trend is reversed for the concentration of WO4 units, in agreement with results reported for tungsten-based glasses in the literature [1, 66].

The analysis of coordination numbers (cf. table S2 and figure S3 in the SI) reveals useful information on the possible linkages in the glass network. RMC calculations have revealed that for all glasses under investigation, the Al atoms are 4-fold with oxygen atoms (figure S3(b) in the SI). The average Al–O coordination numbers are 4.02, 4.00, 3.98 and 3.92 (±0.05) in the W75La10, W80La10, W75Nd10 and W65Nd5 glasses, respectively. The Al–O first-neighbour distributions exhibit characteristic distributions peaking at 1.80 ± 0.02 Å. On the whole, the first-neighbour distances and the coordination numbers obtained from the RMC analysis in this work point at AlO4 tetrahedron being the dominating structural motif. Moreover, our results are in agreement with results reported for La2O3–Al2O3–SiO2 glasses, where peaks in Al–O first-neighbour distributions were reported at 1.80/1.82 Å [67]. However, the Al–O first-neighbour distribution obtained in this work seems to be peaked at distances higher than in CaAl2O4, where the most likely Al-O distance is at 1.76 Å and Al on average is surrounded by four oxygen atoms [68, 69].

The RMC analysis reveals that the Zn–O distance appears at a significantly higher value of 1.95 ± 0.01 Å for all investigated glass samples. This value is in good agreement with our previous study on MoO3–ZnO–B2O3.
and with the results obtained for other glasses with ZnO, where the Zn–O distances were found at 1.93–1.95 Å [70], 1.94–1.97 Å [71], and 1.96 Å [72], respectively. For the Zn–O coordination number distributions, mainly 4-fold coordinated oxygen atoms are obtained, which points at the formation of ZnO$_4$ tetrahedral structural units in the tungsten oxide-based glasses under consideration.

The $g_{\text{O}-\text{O}}(r)$ distributions peak at 2.35 ± 0.05 and 2.75 ± 0.05 Å, showing the same O–O distances for all compositions with slight shifts of second nearest neighbour peaks. This trend confirms previous findings in the literature. Namely, the first peak position fairly well agrees with the $g_{\text{O}-\text{O}}(r) = 2.35$ Å distance characteristic for the boromolybdate glasses [23, 36, 73, 74] and borosilicate glasses [75], while the second peak agrees with that in LiTeVO (2.70–2.80 Å) [76], and is close to the second nearest neighbour O–O distances reported for MoZnBO (2.85/2.95 Å) [23]. Unfortunately, the peaks in the $g_{\text{O}-\text{O}}(r)$ distributions appear in the 2.30–2.85 Å interval, overlapping with the $g_{\text{La}-\text{O}}(r)$ and $g_{\text{Nd}-\text{O}}(r)$ distributions. Moreover, due to the limited Q-range of the present neutron diffraction experiment (dictated by the maximum value of the neutron momentum transfer, $Q_{\text{max}}$), the limited r-space resolution, $\Delta r = \frac{2\pi}{Q_{\text{max}}} \approx 0.6$, does not allow to resolve well the La–O, Nd–O and O–O first and second-neighbour atomic pair distributions. The average O–O coordination numbers are 7.35, 8.13, 7.38 and 7.17 ± 0.1 Å in the W75La10, W80La10, W75Nd10 and W65Nd5 glasses, respectively.

In the case of the La–O and Nd–O distributions, a marked coordination number dependence on the La- and Nd-concentration is evident from the RMC analysis. The La-O distribution is bimodal, with the first peak at shorter distances (2.35 ± 0.05 Å), followed by the second peak centred at 2.80–2.85 ± 0.05 Å. The first peak in the distribution appears at a shorter distance compared to the LaP$_2$O$_5$ glass, where it was reported at 2.45 Å [77]. However, it is in agreement with the results of the analysis of diffraction data presented for LaP$_3$O$_9$ glass, where the Nd–O distance distribution appears at a shorter distance compared to the LaP$_3$O$_9$ glass, where it was reported at 2.45 Å [77].

The RMC analysis reveals Nd–O first-neighbour distances at 2.30/2.35 ± 0.05 and 2.80 ± 0.05 Å, values very close to the ones obtained in our previous study on borosilicate glasses, where the Nd–O distance distributions peaked at 2.30 ± 0.02 Å/2.70 ± 0.05 Å [15]. They are, however, appearing at shorter distances than in the case of the SiO$_2$–B$_2$O$_3$–Na$_2$O–Al$_2$O$_3$–CaO–ZrO$_2$–Nd$_2$O$_3$ glass, where the Nd–O distance distribution shows a characteristic peak at 2.44 Å [78]. The actual Nd–O coordination numbers obtained from RMC modelling are 5.41 and 5.12 (±0.1) for W75La10 and W80La10, respectively (cf figure S3(e) in the SI), which are higher than reported earlier in [77].

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Like many other amorphous materials, tungsten-oxide based glasses lack long-range order, and their structures are made up of networks consisting of octahedral/tetrahedral/trigonal units. This network features long-range disorder and short and medium-range order. The short-range order in tungsten-oxide based glasses is manifested by the fact that the O–W–O bonds within [WO$_4$/WO$_5$] units remain largely unchanged throughout the entire network, and this is the case for both [AlO$_4$] and [ZnO$_4$] units. Medium-range-order concerns the arrangement of a group of structural units in an ordered manner, e.g. some multi-member rings have similar structures throughout the network. The RMC modelling is in principle capable of providing information about medium-range order by characterising second coordination spheres. In the case of the tungsten-oxide based glasses under consideration, the RMC analysis has been capable of revealing pronounced W–Al, W–Zn, W–La, W–Nd, Al–La and Al–Nd atomic correlations, as illustrated in figure S4 in the SI. Importantly, this has been possible despite the fact that the magnitude of the neutron diffraction signal carrying information about metal–metal atomic correlations is less than 2% of the total neutron diffraction signal magnitude. This is an important result, as the analysis of the metal–metal pair correlation functions gives useful information on the linkage of the basic structural units forming the glass network structure.

The W–Al distance distributions are broad, except in the case of the W75La10 sample, and peak at 2.80 ± 0.1 Å, at 2.75/3.0 ± 0.1 Å, at 2.80 ± 0.1 Å and 2.75/2.95 ± 0.1 Å in the W75La10, W80La10, W75Nd10 and W65Nd5 glasses, respectively. The W–Zn distance distributions peak at 2.80 ± 0.1 Å, at 2.80/3.00 ± 0.1 Å and at 2.85 ± 0.1 Å, in the W75La10, W75Nd10 and W65Nd5 glasses, respectively. The shapes of W–Al and W–Zn distributions suggest a possible connection between W- and Al/Zn-centred groups. The analysis of the distributions suggests the existence of linkages between the WO$_4$/WO$_5$ and AlO$_4$/ZnO$_4$ units contributing to the medium-range order in the tungsten-oxide glasses under investigation. On the whole, the network structure of all investigated glass systems appears stable and built up from WO$_4$/WO$_5$, AlO$_4$ and ZnO$_4$ units.

Significant correlations may be observed at higher r-values between La/Nd and the network- former W and Al atoms. In the case of W–La and W–Nd distributions, peaks were identified at relatively short distances, at 2.95 ± 0.1 Å and 3.0 ± 0.1 Å in the W75La10 and W80La10 glasses, respectively, and at 2.85/2.97 ± 0.1 Å and 3.0 ± 0.1 Å, in the W75Nd10 and W65Nd5 glasses, respectively. At similar values peaked the distributions of La–Al and Nd–Al distances, at 2.90 ± 0.1 Å and 2.95 ± 0.1 Å in the W75La10 and W80La10 glasses, respectively, and at 2.75/2.95 ± 0.1 Å and 2.80/2.95 ± 0.1 Å in the W75Nd10 and W65Nd5 glasses, respectively. Although these correlation functions are rather noisy because of the relatively low number of the contributing atoms in the
RMC simulation box, clear correlations may be observed between $2.75 - 3.0 \pm 0.1 \text{ Å}$. This indicates that La and Nd atoms are connected through an oxygen atom with the network former atoms, which build up the network structure. A correlation appears to exist in the location of La and Nd ions at relatively short distances, suggesting a well-defined medium-range order in the glass network.

Finally, the parameter pertaining to the medium-range order in the tungsten-oxide based glasses is the metal-metal coordination number. The elucidation of this parameter from the RMC analysis is, however, not realistic because the second neighbours are connected through an oxygen atom.

On the whole, the tungsten-oxide based glasses under consideration contain a relatively high amount of WO$_3$, and their glass networks are built up by WO$_4$/WO$_6$ units closely linked to AlO$_4$ and ZnO$_4$ units. In all the glass systems under investigation, a trend is observed, whereby with increased WO$_3$ content, one observes an increased relative fraction of WO$_6$ and a decreased fraction of WO$_4$. The incorporation of La$_2$O$_3$ in both cases is stable, and the atomic parameters describing the distance distributions and coordination involving La atoms are close to the values reported in the literature. The La-O and Nd-O coordination numbers are below values usually expected for this type of glasses, which leads to a compact glassy network. The addition of La/Nd-oxide to the glass does not seem to change the basic topology of the glassy network.

3.2. Mass-selective nuclear quantum dynamics and force-constant disorder from neutron Compton scattering

Before we start our discussion of the results of the neutron Compton scattering experiments on tungsten oxide-based glasses, one important methodological remark is in order. As mentioned in sections 2.4 and 2.5, and following our methodology already established in our work on molybdate glasses, in the case of the nuclei present in the glasses, the NMD width and the force-constant values are obtained directly from the experimental NCS spectra, and no ab initio modelling of these observables is attempted. Conversely, in the case of the metal-oxide precursors, the values of these observables are obtained from the ab initio harmonic lattice dynamics calculations based on the optimised geometries. The reason for adopting such a strategy, explained in our previous work, is that, when calculating vibrational and elastic properties by first principles using harmonic lattice dynamics, the underlying structure must have been optimised and correspond to an equilibrium configuration. Such a configuration is defined by the optimised values of the unit-cell parameters and fractional coordinates, all fulfilling periodic boundary conditions imposed by the crystal symmetry and long-range order. Using the structure adopted from the solution of the RMC modelling for the glasses as an input for the harmonic lattice calculation scheme would certainly lead to imaginary mode frequencies as the initial structure is not properly equilibrated, and residual forces are still present between the atoms in the box representing the RMC solution. Moreover, the nature of the RMC solution is such that it does not represent any positional disorder that would have been tractable within the supercell approach to harmonic lattice dynamics. Going beyond the harmonic lattice dynamics and simulating the glasses starting from the RMC solution by means of the ab initio molecular dynamics and, at the same time, properly accounting for the nuclear quantum effects reflected in the values of the NMD widths, also seems like an unrealistic task. Thus, instead of following the fully computational path for the modelling of dynamic properties of nuclei in glasses, it seems more plausible to use the experimental observation that long-range disorder in glasses induces softening of vibrational properties as compared to their metal-oxide precursors and to characterise the degree of disorder-induced softening of vibrational properties by contrasting the experimental NMD width and force-constant values for glasses with their counterparts obtained from the ab initio harmonic lattice dynamic calculations for their metal-oxide precursors.

Figure 2 shows the mass-resolved NCS spectra of four tungsten-oxide based glasses under consideration. Owing to the technique of stoichiometric fixing, good-quality fits are obtained for individual atomic species. In table 1, the values and uncertainties of NMD widths fitted to Gaussian momentum distributions of atoms present in the glasses are contrasted with mean values and standard deviations of NMD widths obtained by averaging over the ab initio harmonic lattice predictions for different isotopic species of the same types of atoms in the metal-oxide precursors. As anticipated for disordered systems, a lowering of the values of the NMD widths and concomitant softening of the values of nuclear kinetic energies is present. The trends visible in the values of NMD widths are immediately reflected in the mean values of force constants and their distributions listed in table 2.

We start our discussion with the observation that the lowering of NMD widths and the concomitant softening of effective force constant values are not universally observed in all tungsten-oxide based glasses. Tungsten NMD widths in the glassy state exhibit the same values as their counterparts in tungsten trioxide. This lack of softening of the nuclear kinetic energy of tungsten may, however, be partially caused by the limited mass resolution of the NCS technique at these very high values of the atomic mass. Starting from neodymium, the degree of the softening increases with decreasing mean atomic weight, with aluminium exhibiting, on average,
25 per cent lower widths in the glassy state compared to the corundum structure of aluminium trioxide. Interestingly, in the case of oxygen, only about 10 per cent softening of the NMD width values is observed in glasses as compared to WO₃, Al₂O₃, and ZnO. Contrary to this trend, the NMD widths in glasses are almost equal to their counterparts in Nd₂O₃ and only slightly lower than in La₂O₃.

The softening of effective force constant values, observed in tungsten oxide-based glasses, has one more important connotation. Namely, in recent studies [79], it has been found that the flatness of the potential energy landscape (PEL) is very important for the formation of glass. By adjusting the flatness of the PEL, it was found that the atomic nanoclusters can undergo either a first-order solid-liquid phase transition or a glass transition.

Table 1. Mean values and standard deviations of NMD widths \( \sigma_M \), obtained by averaging the results for different isotopic species of the same types of atoms. The values for individual isotopic species were calculated from the atom-projected vibrational densities of states (apVDOSes) obtained from \textit{ab initio} harmonic lattice dynamics simulations performed for precursor metal-oxide systems. In tungsten-oxide based glasses, the values were obtained directly from NCS experiments. See section 1 of the SI for details.

| Systems   | Nd    | W     | Al   | Zn   | La   | O     |
|-----------|-------|-------|------|------|------|-------|
| Nd₂O₃     | 30.40 ± 0.29 | 35.50 ± 0.19 | 14.89 ± 0.00 | 21.11 ± 0.33 | 30.00 ± 0.00 | 10.88 ± 0.01 |
| WO₃       |       | 12.09 ± 0.01 |      |      |      |       |
| Al₂O₃     |       | 12.13 ± 0.00 |      |      |      |       |
| ZnO       |       | 11.59 ± 0.01 |      |      |      |       |
| La₂O₃     |       | 11.12 ± 0.00 |      |      |      |       |
| W75La10   | 35.5 ± 0.9 | 10.4 ± 0.5 | 20.5 ± 0.6 | 29.8 ± 0.7 | 10.9 ± 0.4 |       |
| W80La10   | 35.5 ± 0.9 | 11.9 ± 0.5 | 20.5 ± 0.6 | 29.8 ± 0.7 | 10.8 ± 0.4 |       |
| W75Nd10   | 30.3 ± 0.7 | 35.5 ± 0.9 | 10.5 ± 0.5 | 20.5 ± 0.6 | 11.1 ± 0.4 |       |
| W65Nd5    | 30.3 ± 0.7 | 35.5 ± 0.9 | 11.2 ± 0.5 | 20.4 ± 0.6 | 10.8 ± 0.4 |       |

Figure 2. Fits of the TOF spectra recorded at VESUVIO for tungsten-oxide based glasses: (a) 75WO₃-5ZnO-10La₂O₃-10Al₂O₃, (b) 80WO₃-10La₂O₃-10Al₂O₃, (c) 75WO₃-5ZnO-10Nd₂O₃-10Al₂O₃, (d) 65WO₃-20ZnO-5Nd₂O₃-10Al₂O₃. Recoil peaks have been colour-coded with peaks of oxygen, aluminium, zinc, lanthanum, tungsten, and neodymium plotted in cyan, grey, magenta, green, violet, and orange. Additionally, the recoil peaks due to the aluminium container are marked in blue. See text for details.

The recoil peaks have been colour-coded with isotopes of oxygen, aluminium, zinc, lanthanum, tungsten, and neodymium plotted in cyan, grey, magenta, green, violet, and orange. Additionally, the recoil peaks due to the aluminium container are marked in blue. See text for details.
which is independent of the structural symmetry [79]. Moreover, it was demonstrated, through molecular
dynamics simulations, that a relatively flat potential energy landscape is an intrinsic nature for glass transitions
[79]. In most molecular dynamics simulations, the total potential energy of a system is usually expressed as a sum
over specific interatomic potentials [79]. It is thus plausible to assume that the degree of flatness of the PEL
correlates with the values of the effective force constants felt by individual atomic species present in the glasses.
Moreover, as demonstrated in figure 7 in [79], for nanocluster glass systems, both the onset temperature of the
glass transition, \( T_s \), and the glass transition temperature, \( T_g \), decrease with the increasing degree of flatness of the PEL. In this context, a question arises whether this trend correlates with decreasing values of the effective force
constants of individual atomic species as obtained from the NCS experiments. To start with, it is worth
mentioning that, for the majority of the tungsten oxide-based glasses, when the WO3 is present in the glass in a
low concentration, it acts as a network modifier. However, in the case of tungsten oxide-based glasses with
higher WO3 content, WO3 can be regarded as glass-forming oxide. The increasing WO3 content strengthens the
glass network. In consequence, with higher tungsten concentration, the \( T_g \) is higher [80–82]. The observed
pronounced increase in the glass transition temperature with increasing WO3 content can be associated with the
formation of strong bridging W–O–W bonds, because at high WO3 concentration, the tungstate structural
units, start to polymerize. Thus, it is expected that W80La10 would have the highest \( T_g \) value, followed by
W75Nd10 and W75La10, with W65Nd5 having the lowest glass transition temperature. Inspection of table 2
shows that in the case of Nd, La and W, no trends are visible. However, in the case of Al, Zn, and O, the force-
constant values in W65Nd5 are lower than their counterparts in W75Nd10 and W75La10. Moreover, if one
sums the force-constant values of individual atomic species, the total force constant values are lowest in
W65Nd5, followed by W75Nd10, with W75La10 having the highest value of the total force constant.
Notwithstanding the above, both the total force-constant and the force-constant values for Al and O in
W80La10 are not the highest amongst all tungsten oxide-based glasses, signifying that in the case of W80La10,
the highest glass transition temperature anticipated does not necessarily correlate with the steepest PEL. On the
whole, in the tungsten oxide-based glasses under consideration, with the exception of W80La10, the glass
formation ability, measured in terms of the value of the glass transition temperature, does correlate positively
with the flatness of the PEL measured through the magnitude of the effective force constants obtained from NCS
experiments.

As discussed in our previous work on molybdate glasses [4], the values of NMD widths, being functions of
mean effective interatomic force constants, are very sensitive to the mean interatomic distances. Thus, in what
follows, we will briefly discuss the visible correlations between the average interatomic distances and the NMD
widths in glasses and their parent metal-oxide systems using the information obtained from our RMC analysis
and the available crystal structures of their precursor metal-oxides.

In our discussion of the results of the RMC analysis, we noted that universally in all glasses under
consideration, a structural motif is present, whereby two W–O bond lengths are present of 1.75–1.80/2.55 Å. In
the crystal structure of the monoclinic phase of WO3, the W–O bond lengths measured along the \( z \)-axis direction
are 1.785/2.245 Å [83]. For this crystal structure of tungsten trioxide, two types of W atoms and six types of O
atoms are present to form two types of the WO6 octahedron with W–O bond lengths of 1.871, 1.990, 1.908,
1.937, 1.785 and 2.245 Å, and W–O bond lengths of 1.856, 2.011, 1.909, 1.936, 1.785 and 2.244 Å. The octahedral
distortion is similar in both types of WO6, which introduces local dipole moments of 2.629D for the two
eighthedrons, respectively [83]. The average coordination number distributions, as revealed from our RMC
analysis, confirm that the W–O network in all glasses consists of WO4 and WO6 units. Thus, on the whole, the
local structure and coordination of tungsten atoms are very similar in tungsten-oxide based glasses and the

Table 2. Mean values and mean width of distributions of effective mean force constants, \( k_M \), obtained using NMD width values using formula (3). See sections 2.4 and 2.5 for details.

| Systems   | Nd    | W      | Al      | Zn      | La   | O      |
|-----------|-------|--------|---------|---------|------|--------|
| W65Nd5    | 0.38 ± 0.06 | 3.17 ± 0.05 | 1.41 ± 0.00 | 1.06 ± 0.05 | 1.05 ± 0.00 | 0.36 ± 0.01 |
| W65Nd5    | 3.17 ± 0.05 | 1.41 ± 0.00 | 1.06 ± 0.05 | 1.05 ± 0.00 | 0.36 ± 0.01 | 0.36 ± 0.01 |
| W75La10   | 0.30 ± 0.27 | 3.17 ± 0.05 | 1.47 ± 0.00 | 0.91 ± 0.00 | 0.91 ± 0.00 | 0.36 ± 0.01 |
| W75La10   | 3.17 ± 0.05 | 1.41 ± 0.00 | 1.06 ± 0.05 | 1.05 ± 0.00 | 0.36 ± 0.01 | 0.36 ± 0.01 |
| W75Nd10   | 0.30 ± 0.27 | 3.17 ± 0.05 | 1.47 ± 0.00 | 0.91 ± 0.00 | 0.91 ± 0.00 | 0.36 ± 0.01 |
| W75Nd10   | 3.17 ± 0.05 | 1.41 ± 0.00 | 1.06 ± 0.05 | 1.05 ± 0.00 | 0.36 ± 0.01 | 0.36 ± 0.01 |

\( k_M \) [eVÅ\(^{-2}\)]
crystal structure gamma phase of tungsten trioxide, as witnessed by similar values of tungsten NMD widths and force constants acting on tungsten in both types of systems (see table 2).

The Zn–O bond length in the wurtzite structure of zinc oxide is 1.98 Å [84], a value lower than the one obtained from our RMC analysis for all investigated glass samples (1.95 ± 0.01 Å). Due to the non-linear character of formula (3) and its temperature dependence, despite a relatively low degree of the softening of the NMD widths of zinc in glasses (at the level of 3 per cent), the reduction in the values of the mean force constant is quite substantial (ca. 50 per cent). The ideal wurtzite structure is composed of two interpenetrating hexagonal-close-packed (hcp) sublattices, each of which consists of one type of atom displaced with respect to each other along the threefold c-axis by the amount of \( u = 3/8 = 0.375 \) [85]. Each sublattice includes four atoms per unit cell, and every atom of one kind is surrounded by four atoms of the other kind, or vice versa, which are coordinated at the edges of a tetrahedron [85]. In a real ZnO crystal, the wurtzite structure deviates from the ideal arrangement by changing the c/a ratio, or u value, in such a way, that those four tetrahedral distances remain nearly constant through a distortion of tetrahedral angles due to long-range polar interactions [85]. The four-fold coordination of atoms in the wurtzite crystal structure agrees quite well with the results of our RMC analysis on the tungsten–oxide based glasses, whereby mainly 4-fold coordinated oxygen atoms are obtained, which points at ZnO4 tetrahedral structural units as the main structural motif. Thus, similarly as in the case of the tungsten trioxide, the local structure of zinc in the glasses is quite similar to its crystal counterpart with only the Zn–O distances differencing, which explains the force constant softening.

In the crystal structure of corundum, two nearest-neighbour Al–O distances are reported, with average values of 1.839 and 1.997 Å with four-fold coordinated oxygen and 6-fold coordinated aluminium atoms [86]. As mentioned in the pioneering work by Pauling et al [87], each metal atom is surrounded by six oxygen atoms, which are not at the corners of a regular octahedron. Three of these oxygen atoms are a few per cent nearer the metal atom than the other three. Moreover, each oxygen atom is surrounded by four metal atoms, two of which are nearer than the other two, with these atoms not being at the corners of a regular tetrahedron [87]. The latest synchrotron radiation x-ray powder diffraction study on corundum–type structures of metal oxides have shown that Al2O3 belongs to the group of structures that show a ‘negative’ monoclinic distortion which leads to relatively longer metal-metal distances and the absence of metal-metal bonds [88]. The structural information reported in the literature for the corundum–type Al2O3 needs to be contrasted with the local aluminium structure in the tungsten–oxide based glass, whereby mainly 4-fold coordinated oxygen atoms are obtained, which points at AlO4 tetrahedron being the dominating structural motif. The Al–O first-neighbour distributions exhibit characteristic distributions peaking at 1.80 ± 0.02 Å. Thus, contrary to the case of tungsten and zinc, the local structure of aluminium differs quite considerably between the corundum crystal structure and the glasses under investigation. Thus, the interplay between the different local coordination and Al–O distances in both structures does not allow for an easy interpretation of the trends visible in glasses as far as values of NMD widths and force-constants are concerned. The mean force constant values obtained for some glass systems are lower, whereas for the other glass systems higher, compared with their counterparts computed for the corundum structure.

Nd2O3 crystallises in the trigonal centrosymmetric space group P 3 1 m1 (164) [89]. The environment surrounding Nd3+ has C3v symmetry, consisting of 7 oxygen ions located at the vertex of a distorted cube along one diagonal. The C3 axis coincides with the c-axis, while one of the mirror planes lies perpendicular to the ab plane [89]. Three Nd–O nearest-neighbour distances are reported, 2.30, 2.41, and 2.66 Å [89]. The RMC analysis performed in this work on tungsten–oxide based glasses reveals a set of Nd–O first-neighbour distances at values similar to those obtained from the crystal structure analysis of Nd2O3. 2.30/2.35 ± 0.05 Å, and at larger values of 2.80 ± 0.05 Å. The Nd–O coordination numbers obtained from RMC modelling are 5.41 and 5.12 (±0.1) for the W75Nd10 and W65Nd5 glasses, respectively. Thus, although the coordination numbers are different in the glass and crystal structures, the differences between the largest first-neighbour distances in the glasses and in the Nd2O3 structure may explain the softening of NMD widths and force constant values in the glasses.

Two types of oxygen atoms are reported in the unit cell of the trigonal crystal structure of La2O3 (space group 164) [90]. The O(I) atom is octahedrally surrounded by six lanthanide atoms. The corresponding metal-oxygen bond lengths are fairly long (2.73 Å). The O(II) atoms are surrounded by lanthanide atoms at the corners of a slightly distorted tetrahedron. Two different Ln–O(II) bond lengths result from the symmetry deviation \( T_g \rightarrow C_{3v} \): one ‘short’ length corresponding to three bonds (2.36 Å) and one ‘intermediate’ length corresponding to the Ln–O(II) bond directed along the ternary axis (2.46 Å). The La–O distance distribution revealed from our RMC analysis in the glasses is bimodal, with the first peak at shorter distances (2.35 ± 0.05 Å), followed by the second peak centred at 2.80–2.85 ± 0.05 Å. Thus, the shorter La–O distances present in the glasses are at par with ‘short’ and even shorter than the ‘intermediate’ Ln–O(II) bond length in the crystal structure of La2O3. However, the long La–O distance in the glasses is longer than the Ln–O(I) bond length in the crystal. The La–O average coordination numbers inferred from our RMC analysis are 4.38 and 4.73 (±0.1) for W75La10 and
W80La10, respectively. These numbers are bracketed by the six-fold coordination of the O(I) atom and the four-fold coordination of O(II) atoms in the crystal structure. It might then be that an RMC analysis of total diffraction data in the trigonal crystal structure of La2O3 would have revealed similar average La–O coordination numbers as those obtained for the W75La10 and W80La10. On that basis, one could assume that a small degree of softening of the lanthanum NMD widths and average force constants, observed in both types of glasses, is a reflection of slightly longer average La–O distances, with the average La–O coordination numbers being similar in both types of structures.

In the case of oxygen in the tungsten-oxide glasses, the average NMD width and force-constant values are a reflection of a complicated interplay of metal-oxygen bond softening and average coordination number modifications within a complicated network structure of the glasses, and it is extremely difficult to disentangle those trends and project them then back onto individual metal-oxygen atom pairs. The values of the force constants of the oxygen, calculated by averaging the force-constant values of individual metal-oxide systems obtained from ab initio simulations over the compositions of the glasses, yield 1.138, 1.281, 1.110, and 1.108, for W75La10, W80La10, W75Nd10 and W65Nd5, respectively. All these values are, on average, two times higher than the values of average force constants of oxygen in the glasses obtained directly from the NCS experiments.

As can be clearly seen from the inspection of table 2, the widths of force constant distributions in all glasses under consideration are a few times larger compared to their metal-oxide counterparts. Few remarks are here in order, however. Firstly, no positional disorder was taken into account during the calculation of apVDOSes in individual crystal systems, as periodic boundary conditions were imposed within the DFT calculation scheme. Secondly, the isotopic mass disorder was introduced a posteriori by replacing masses of individual isotopic species in the dynamical matrix. Finally, the NMD widths obtained directly from NCS experiments on glasses have to be taken as upper-conservative bounds of the isotopic-mass induced disorder as the recoil peaks fitted to NCS data do not resolve distributions of individual isotopic species. These factors most certainly overestimate the degree of force-constant disorder in glasses, measured in terms of the absolute values. However, comparing the relative values of the force constants between different atomic species and establishing trends from such comparisons should be, in principle, correct. Comparing the relative amounts, the degree of force constants disorder seems to be largest in the case of neodymium, followed by zinc, lanthanum, tungsten, oxygen and aluminium. Interestingly, this trend does not reflect the progression of the relative isotopic mass distributions of individual atomic species present in the glass directly but rather is a reflection of an interplay between these isotopic mass distributions and the degree of positional disorder influencing the bond-length disorder within the glass network.

We close our discussion by assessing the degree of the force-constant softening induced by disorder in the glasses using the scale of disorder introduced in our previous work on molybdate glasses [4]. Namely, one can place the magnitude of an NMD width of each nucleus present in the glass on a scale between the NMD width value computed from the Maxwell-Boltzmann nuclear momentum distribution with no underlying potential and thus no confinement, and an ab initio prediction for the NMD width of this nucleus in a parent metal oxide system [4]. In the case of a nucleus that is present in multiple parent metal-oxide systems (such as oxygen), for the upper end of the scale, one can use a compositional average of ab initio predictions for NMD widths [4]. Figure 3 shows the disorder scale for the tungsten-oxide based glasses based on the amount of the softening of the NMD widths of their constituent nuclei in (a) 65WO3–20ZnO–5Nd2O3–10Al2O3, b) 75WO3–5ZnO–10Nd2O3–10Al2O3, c) 75WO3–5ZnO–10La2O3–10Al2O3, and d) 80WO3–10La2O3–10Al2O3. The first observation is that, despite a relatively large amount of force-constant disorder of tungsten, neodymium, lanthanum and zinc, the degree of softening of their apVDOSes is relatively small based on the average NMD widths and, in consequence, the average force-constant values. In the case of tungsten nuclei in all four glasses under consideration, the NMD widths measured in NCS experiments are almost equal to the ab initio predictions for the parent metal oxide, WO3. On the contrary, in the case of aluminium and oxygen, the degree of softening is the largest. For oxygen, the degree of softening is similar in all four glass systems; however, in the case of aluminium, the largest degree of softening is observed in 75WO3–5ZnO–10La2O3–10Al2O3 and 75WO3–5ZnO–10Nd2O3–10Al2O3, followed by 65WO3–20ZnO–5Nd2O3–10Al2O3 and 80WO3–10La2O3–10Al2O3.

4. Summary and outlook

In this work, following our previous work on molybdate glasses, we have employed a combination of neutron diffraction and neutron Compton scattering, augmented by ab initio harmonic lattice dynamics and Reverse Monte Carlo modelling in order to characterise the force-constant disorder in the tungsten-oxide based glasses. We have tapped on the potential of both neutron techniques to investigate the structure and dynamics at the local scale and explored the correlations between the structural and dynamic observables that characterise the degree of disorder in the glass network. Specifically, we have discussed the correlations between that average...
interatomic force constant magnitudes inferred from neutron Compton scattering and average interatomic
distances obtained from diffraction data analysis. We have established that, in the case of similar atomic
coordination, clear correlations are visible between the interatomic distances and average force constant
distributions in the glasses and their precursor metal-oxide systems.

Moreover, we have shown that in the tungsten oxide-based glasses under consideration, with the exception
of W80La10, the glass formation ability, measured in terms of the value of the glass transition temperature, does
correlate positively with the flatness of the potential energy landscape measured through the magnitude of the
effective force constants obtained from NCS experiments. This is an important result in the light of recent
molecular dynamics simulations on atomic nanoclusters, as it shows that some results obtained from studies of
nanoscale glasses may be potentially transferable to bulk glasses, and, in the case of the parameters characterising
potential energy landscape, the central role in this process is played by NCS observables.

As far as the force-constant disorder is concerned, we have characterised the tungsten-oxide based glasses
using two different scales. On the first scale, we have compared the widths of force-constant distributions in
glasses and their precursor metal oxides based on the distributions of the widths of nuclear momentum
distributions of individual atomic species. The second scale was related to the softening of the NMD widths of their constituent
nuclei. The charts for each nucleus are composed of three bars. Each NMD width measured in an NCS
experiment (light magenta bar) is placed between the upper limit of the NMD scale set by the 
ab initio prediction (light cyan bar) and
the lower limit of the scale based on the Maxwell-Boltzmann distribution (violet bar). See text for details.

Figure 3. The disorder scale for the molybdate glasses based on the amount of the softening of the NMD widths of their constituent
nuclei: (a) 65WO3-20ZnO-5Nd2O3-10Al2O3, (b) 75WO3-5ZnO-10Nd2O3-10Al2O3, (c) 75WO3-5ZnO-10La2O3-10Al2O3, and (d)
80WO3-10La2O3-10Al2O3. The charts for each nucleus are composed of three bars. Each NMD width measured in an NCS
experiment (light magenta bar) is placed between the upper limit of the NMD scale set by the 
ab initio prediction (light cyan bar) and
the lower limit of the scale based on the Maxwell-Boltzmann distribution (violet bar). See text for details.
schemes would then benefit from robust convergence criteria that would include structural and dynamical constraints that are both local in nature and thus well suited for an RMC scheme used in diffraction data analysis.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Declaration of competing interest

The authors declare no competing financial interests.

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