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Lamb–Mössbauer factors as a local probe of floppy modes in network glasses

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Abstract

The temperature dependence of the Lamb–Mössbauer factor, $f(T)$, in a solid provides the first inverse, $\langle 1/\omega \rangle$, and second inverse, $\langle 1/\omega^2 \rangle$, moments of the vibrational density of states. In network glasses, these moments serve as local probes of low-frequency vibrational excitations, such as floppy modes, and provide a means to establish the rigidity percolation threshold. Lamb–Mössbauer results on prototypical chalcogenide glasses (Ge_xSe_{1-x}) correlate well with those of Raman scattering, inelastic neutron scattering and Mössbauer hyperfine structure experiments in indicating that the rigidity percolation threshold occurs near $\langle r \rangle_c = 2.46(4)$. These observations provide experimental support for predictions of the Phillips–Thorpe constraint theory, when provision is made for a small but finite concentration of broken bond-bending constraints around chalcogen sites.

1. Introduction

Vibrational excitations in glasses and corresponding crystals display similarities as well as differences. The similarities, in general, can be traced to elements of short-range order (SRO), i.e., to the nature of the building blocks of the network. Elastic scattering experiments [1] using a beam of X-rays or neutrons have shown over the years that elements of SRO between glasses and crystals are similar although not without exceptions [2]. On the other hand, differences in vibrational excitations between glasses and crystals possessing the same SRO arise in general from atomic correlations extending to larger distances. Glasses differ from crystals in that their networks do not possess long-range order. The vibrational features intrinsic to glasses, particularly in the low-frequency regime, continue to attract interest [3] because these are tied to elements of medium-range order (MRO), i.e., to the way and extent building blocks are correlated both spatially and vibrationally in a glass network.

Traditionally, the experimental methods used to establish the vibrational density of states (VDOS) in a glass have included inelastic neutron scattering [4], Raman scattering [5] and IR reflectance [6]. In this paper we introduce a new method – vibrational spectroscopy based on measurements of the Lamb–Mössbauer factors [7]. The Lamb–Mössbauer factor, also known as the f-factor, physically measures the probability for recoilless (zero phonon) emission or absorption of a γ-ray by a resonant atomic nucleus

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