Available online at www.sciencedirect.com
ScienceDirect
Journal of Non-Crystalline Solids 353 (2007) 1824–1827
JOURNAL OF NON-CRYSTALLINE SOLIDS
www.elsevier.com/locate/jnoncrysol

Origin of boson peak in glasses: Role of Ioffe–Regel crossover and atomic soft modes

Michael I. Klinger *

Department of Physics, Bar-Ilan University, Ramat-Gan 52900, Tel Aviv, Israel

Available online 16 April 2007

Abstract

A theoretical '*complete soft-mode-dynamics*' model of the origin and properties of the boson peak accompanied by a high-frequency sound, observed in glasses of a certain type, is described. The origin is determined by interaction of non-acoustic vibrations with acoustic phonons and a Ioffe–Regel crossover for their inelastic scattering. The non-acoustic excitations are associated with vibrations of atomic soft-mode 'defects'. Two types of boson peak can be predicted in agreement with experiments.

PACS: 61.44.−n; 66.70.+f

Keywords: Glasses; Nitride glasses; Infrared glasses; Chalcogenides; Oxide glasses; Alkali silicates; Borates; Thermal properties; Thermal conductivity

1. Introduction

The purpose of this paper is to describe results of a recent theoretical '*complete soft-mode-dynamics*' model of the boson peak (BP) accompanied by a high-frequency sound (HFS), as well as of related vibrational dynamic and thermal properties, observed in glasses defined here as those of type I. The model is determined by a Ioffe–Regel crossover (IRC) originally defined [1] as a crossover from a weak *elastic* scattering of acoustic phonons to strong scattering. The present model extends the original definition of a IRC to *inelastic* scattering, which induces a strong hybridization of different types of excitations and gives rise to both BP and HFS. In general, experimentally observed universal dynamic and thermal properties of glasses at low frequencies and temperatures are rather due to the existence of both acoustic phonons and 'anomalous' non-acoustic vibrational excitations of moderately low frequencies $\nu \equiv \omega/2\pi$, $0.01\ \mathrm{THz} < \nu \lesssim \nu_M \approx 3\ \mathrm{THz}$. For glasses of type I, probably including v-SiO_2 and CKN [2], the properties at $\nu_{BP} \lesssim \nu \lesssim 3\nu_{BP}$ appear to contain both a broad asymmetric boson peak (BP) at $\nu = \nu_{BP} \sim 1$ THz, followed by a HFS with an acoustic-like dispersion law, $\nu^2(q) \approx a^2 + b^2q^2$, where $a = \mathrm{const} \neq b = \mathrm{const}$, $q = |q|$ and q is the wave vector. In general, the temperature dependent BP observed in inelastic photon (Raman, X-ray) or neutron scattering spectra of glasses is located around a frequency ν_{BP} independent of q. In the present model, for brevity, we focus on Raman scattering characteristics (total scattering intensity $I_R(\nu, T)$, reduced scattering intensity $I^r_R(\nu)$ and dynamic susceptibility $\chi''_R(\nu)$) [3], which appear to be most helpful for choosing the relevant model of BP in glasses. The characteristics are related to each other as follows: $I_R(\nu, T) = I^r_R(\nu)\nu\{1 + n(\nu, T)\} = \chi''_R(\nu)\{1 + n(\nu, T)\}$, where $n(\nu,T) = [1 + \exp(h\nu/kT)]^{-1}$ is the boson factor.

2. Soft-mode-dynamics model

The simplest soft-mode-dynamics (SMD) model of vibrational dynamics in glasses of type I is based on an isotropic elastic continuum, containing randomly distributed localized atomic soft-mode 'defects' of concentration c_{sm}, with independent quasi-local vibrational excitations [4].

* Fax: +972 3 535329.
E-mail address: klingem@mail.biu.ac.il

0022-3093/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnoncrysol.2007.01.050

The general idea is that the soft-mode excitations coexist and interact with acoustic phonons in the THz range of frequencies v'. Most numerous are harmonic soft-mode excitations with their density-of states (DOS) $J_0(\varepsilon' \equiv v'^2) = g_0(v')/2v'$, or equivalent DOS $g_0(v') \propto c_{sm}$, at typical $c_{sm} \sim 10^{-2}$. The main contribution to dynamic and thermal properties comes from frequencies $v' \approx v_0$ around a typical, most probable, frequency $v_0 \sim 0.1 v_D \sim 1$ THz. The Hamiltonian $H = H_{ac} + H_{sm} + V_{ac-sm}$ includes terms describing an atomic soft mode x (H_{sm}) at its location $\mathbf{R}$ and for acoustic phonons (H_{ac}) with their displacement $u(\mathbf{R})$ in the continuum, as well as the interaction Hamiltonian approximated as $V_{ac-sm}(x) \propto \beta x e(R)$, with β the average coupling parameter of typical magnitude $|\beta| \approx 1$ eV ($\gg h v_D$). The interaction gives rise to essential changes in the excitation energy spectrum and a related IRC, as well as to interactions between soft modes. The first basic result of the model is the derived equation for the dispersion law $\varepsilon = \varepsilon(\mathbf{q};\ \varepsilon')$ of well-defined (wave-like) excitations of low eigen-frequencies $v = \varepsilon^{1/2}$:

$$\varepsilon(\varepsilon - \varepsilon') - s_0^2 q^2 (\varepsilon - \varepsilon' + \Delta(\varepsilon')) = 0. \tag{1}$$

In Eq. (1): $\Delta(\varepsilon') = \varepsilon' c_{sm} Q^2$, $\varepsilon' \approx \varepsilon_0 \equiv v_0^2$, $Q^2 \approx 10/\varepsilon_0$ for typical values of the model parameters and s_0 is a typical sound velocity. Eq. (1) describes the interaction rearranged excitation spectrum $\varepsilon = \varepsilon_{1,2}(q;\varepsilon')$ with two branches separated by a gap of width Δ of typical value $\Delta(\varepsilon_0) \approx 0.1\varepsilon_0$. The gap is a manifestation of a general quantum-mechanical phenomenon of repelling off each other two 'energy levels', sound-wave and soft-mode frequencies, which interact and intersect at $v_1^0(q') = s_0 q' = v'$. The spectrum results from strong hybridization of acoustic vibrations with soft-mode ones around and above the gap, due to a IRC from weak inelastic scattering of acoustic phonons by soft-mode vibrations to strong scattering. Indeed, the associated acoustic mean-free path $l^{(in)}_{ac-sm}(v')$ around the gap ($v' \approx v_0$) can be estimated in the general soft-mode model [5] as a short one close to the acoustic wave-length $\lambda_{ac}(v') = s_0/v'$, by the definition of the IRC and its frequency $v^{(in)}_{IR}$:

$$l^{(in)}_{ac-sm}(v) \approx \lambda_{ac}(v) = s_0/v, \quad \text{at} \quad v = v^{(in)}_{IR} \approx v_0 \sim 1 \text{ THz}. \tag{2}$$

This is the second basic result of the model under discussion. Then, the frequency $v^{(in)}_{IR}(\approx v_0)$ does not practically depend on q, in accord with the physical sense of a IRC, and the related half-widths $\gamma_{1,2}$ of 'eigen-frequencies' $v_{1,2} = \varepsilon_{1,2}^{1/2}$ are large, $\gamma_{1,2} \approx v^{(in)}_{IR} \approx v_0$. The spectral branch $\varepsilon_1(q;\varepsilon')$ below the lower gap edge is of 'pure' acoustic origin, $\varepsilon_1(q;\varepsilon') \propto s_0^2 q^2/(2\pi)^2$. The branch $\varepsilon_2(q;\ \varepsilon')$ above the upper gap edge, with an acoustic-like dispersion law $\varepsilon_2(q;\varepsilon') \propto \varepsilon' + s_0^2 q^2/(2\pi)^2$, describes wave-like excitations formed due to a strong hybridization of acoustic phonons with soft-mode vibrational excitations in the IRC for inelastic scattering. These wave-like excitations are similar to the observed HFS excitations [2] of relatively high v, at $v_0 < v \lesssim p_{max} v_0$ ($\ll v_D$) and an empirical $p_{max} \approx 3$.

3. Results

In what follows, results of calculations of the total density of vibrational states (DOS), $J(\varepsilon = v^2)$, are briefly described. The general formula is as follows:

$$J(\varepsilon) = \int d\varepsilon' J_0(\varepsilon') \cdot I(\varepsilon;\varepsilon') = g(v)/2v, \tag{3}$$

where the lower limit of the integral is $\varepsilon'_{min} \approx 0.01\varepsilon_0$ while the upper limit is $\varepsilon'_{max} \approx \varepsilon_0$. For $\varepsilon_1(q) \ll \varepsilon_0$, as well as above the IRC region at $\varepsilon_2(q) > \varepsilon_0$, where the half-widths $\gamma_{1,2}$ of the eigen-frequencies $v_{1,2}$ are small, $\gamma_{1,2}/v_{1,2} \ll 1$ (well-defined excitations), a standard approximation $I_0(\varepsilon;\ \varepsilon')$ of the function $I(\varepsilon;\ \varepsilon')$ at a given ε' is as follows [4]:

$$I(\varepsilon;\varepsilon') \simeq I_0(\varepsilon;\varepsilon') = (a_1/2\pi)^3 \sum_j \int d^3\mathbf{q}\,\delta(\varepsilon - \varepsilon_j(q;\varepsilon')), \tag{4}$$

where $j = 1,2$ for the two spectral branches of Eq. (1). However, the approximation (4) fails in the IRC region characterized by large half-widths $\gamma_{1,2}$, or equivalently $\lambda_{ac}/l_{ac-sm} \approx 1$. In this region, the perturbation theoretical approach of scattering theory becomes irrelevant, and a consistent analytical theory of the effective eigen-frequencies $\Omega_{1,2} = v_{1,2} + i\gamma_{1,2}$ does not seem to be available so far. In this region, we must apply for numerical calculations of $I(\varepsilon;\varepsilon')$ a phenomenological approach, substituting a regular function, e.g., $\pi^{-1}\gamma_j^2\,[x^2 + (\gamma_j^2)^2]^{-1}$, for $\delta(x)$.

Numerical calculations of $J(\varepsilon)$ have been performed by applying, in addition to [4] with $n = 0.5$, both a simplest soft-mode vibrational DOS [5] $J_0(\varepsilon';n) = g_0(v';n)/2v' = \varepsilon_0^{-1}(\varepsilon'/\varepsilon_0)^n$, at $0.5 \leqslant n \leqslant 1.5$, and a different soft-mode vibrational DOS $J_0{*}(\varepsilon') = g_0{*}(v')/2v'$, with $g_0{*}(v')/(v')^2$ recently calculated [6] by accounting for the contribution

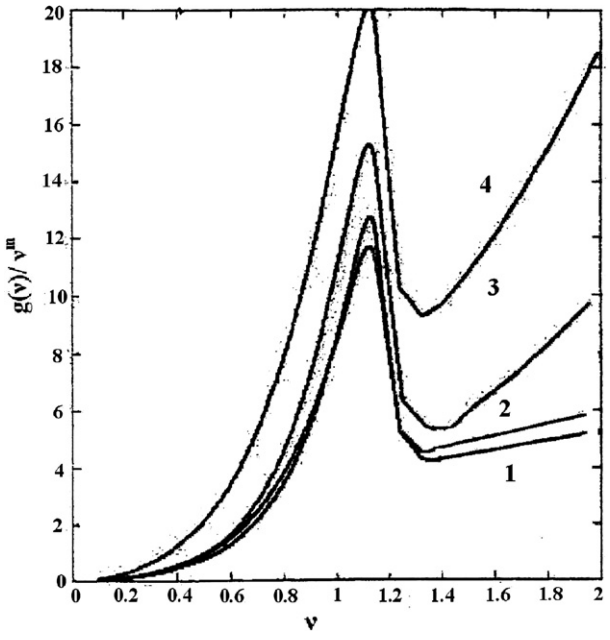

Fig. 1. Frequency dependence of functions $g(v)/v^m$ (in arbitrary units), at $J_0(\varepsilon') = J_0(\varepsilon';\ n = 0.5)$, curves 1 ($m = 2$) and 3 ($m = 1$), and at $J_0(\varepsilon') = J_0{*}(\varepsilon')$, curves 2 ($m = 2$) and 4 ($m = 1$).