Absence of a boson peak in anharmonic phonon models with Akhiezer-type damping

Andrij Shvaika,1 Mykola Shpot,1 Walter Schirmacher,2 Taras Bryk,1,3 and Giancarlo Ruocco4,5

1Institute for Condensed Matter Physics of the National Academy of Sciences of Ukraine, UA-79011 Lviv, Ukraine
2Institut für Physik, Universität Mainz, D-55099 Mainz, Germany
3Institute of Applied Mathematics and Fundamental Sciences, Lviv National Polytechnic University, UA-79013 Lviv, Ukraine
4Center for Life Nano Science @Sapienza, Istituto Italiano di Tecnologia, 295 Viale Regina Elena, I-00161, Roma, Italy
5Dipartimento di Fisica, Università di Roma “La Sapienza”, I-00185, Roma, Italy

(Dated: April 28, 2021)

In a recent article M. Baggioli and A. Zaccone (Phys. Rev. Lett. 112, 145501 (2019)) claimed that an anharmonic damping, leading to a sound attenuation proportional to \( \omega^2 \) (Akhiezer-type damping) would imply a boson peak, i.e. a maximum in the vibrational density of states, divided by the frequency squared (reduced density of states). This would apply both to glasses and crystals. Here we show that this is not the case. In a mathematically correct treatment of the model the reduced density of states monotonously decreases, i.e. there is no boson peak. We further show that the formula for the would-be boson peak, presented by the authors, corresponds to a very short one-dimensional damped oscillator system. The peaks they show correspond to resonances, which vanish in the thermodynamic limit.

One of the characteristic features of disordered solids, in particular, of glasses, is an anomalous excess of the vibrational density of states (DOS) \( g(\omega) \) beyond the Debye law \( g_D(\omega) \propto \omega^2 \), which appears as a peak in the reduced density of states \( g(\omega)/\omega^2 \). This peak has been called “boson peak” (BP) in the early literature on the spectral properties of glasses \([1]\), because the temperature dependence of the measured Raman spectra followed the boson occupation function \( n(\omega) = 1/[1 - \exp(-\hbar \omega/k_B T)] \), pointing to a temperature-independent spectrum. As all anharmonic phenomena are temperature dependent, the conclusion was, that the BP in glasses must be a harmonic phenomenon.

A vast number of suggestions for the origin of the BP has been published in the last 60 years (see e.g. \([2, 3]\) for bibliographies). Most authors attribute the anomaly to the frozen-in structural disorder, be it spatially fluctuating elastic constants \([3, 4]\) or quasi-localized soft-potential defects \([4, 5]\). Some other authors interpret the BP as a crystal-like van-Hove singularity, washed out by the structural disorder \([6, 7]\). All three mechanisms nowadays are known to exist separately \([8, 9]\), and are, as said above, of harmonic origin.

In spite of the established evidence for the harmonic origin of the BP, in a recent paper Baggioli and Zaccone (BZ) \([10]\) attributed it to the anharmonicity-caused damping of acoustic modes. Guided by the observation of boson-peak-like features in the temperature dependence of the specific heat of some crystals \([12, 17, 19]\), they claimed that the BP anomaly in glasses and crystals would be universally of anharmonic origin.

However, their treatment contains severe mathematical errors. We show in the remainder of this article that a correct treatment of their anharmonic phonon model does not produce a maximum in the reduced DOS \( g(\omega)/\omega^2 \), i.e. no BP, thus rendering all their conclusions irrelevant and useless for discussing the BP vibrational anomalies. In fact, as we shall show, the “boson peaks” they presented correspond to resonances of a one-dimensional system. These resonances only exist for very short system lengths and go away in the thermodynamic limit.

BZ consider anharmonically damped longitudinal \((\alpha = L)\) and transverse \((\alpha = T)\) phonons, obeying hydrodynamic equations

\[
\frac{\partial^2}{\partial t^2} u_\alpha(r, t) = \nabla^2 \left( c_\alpha^2 + D_\alpha \frac{\partial}{\partial t} \right) u_\alpha(r, t) \tag{1}
\]

with \( \alpha = (L, T) \), \( c_\alpha \) the sound velocities and \( D_\alpha \) the damping coefficients. Eq. (1) reads in wavenumber and frequency space

\[
\left[ \omega^2 - q^2 (c_\alpha^2 - i\omega D_\alpha) \right] u_\alpha(q, \omega) = 0 \tag{2}
\]

A damping term \( \propto \omega^2 \) corresponds to sound attenuation according to \( \Gamma(\omega) \propto \omega^2 \), which has been derived for crystals from an anharmonic Hamiltonian by Akhiezer \([20]\) and for amorphous solids by Tomaras et al. \([21]\). Both theories give a linear temperature increase of the coefficients \( D_\alpha \).

It has been pointed out by some of the present authors \([1]\) that in the frequency range near and below the boson peak a relationship between the excess over the Debye density of states and the sound attenuation exists, i.e. \( \Delta g(\omega) = g(\omega) - g_D(\omega) \propto \Gamma(\omega) \). Therefore, for a sound attenuation proportional to \( \omega^2 \) in addition to the Debye \( \omega^2 \) law, a second \( \omega^2 \) contribution can be expected, if Akhiezer-type anharmonic is efficient. Because this contribution, divided by \( \omega^2 \) is constant, i.e. does not
increase with frequency, it does not lead to a BP. This will be demonstrated below in detail.

However, BZ claim to be able to produce boson peaks, i.e., peaks in the reduced DOS \(g(\omega)/\omega^2\) from their anharmonic model. They proceed by relating the vibrational DOS in the usual way to the Green’s functions. In three dimensions \((d = 3)\) one has

\[
g(\omega) = \frac{1}{3} \left( g_L(\omega) + 2g_T(\omega) \right)
\]

\[
= \frac{1}{3} \frac{2\omega}{\pi} \text{Im} \left\{ G_L(\omega) + 2G_T(\omega) \right\}
\]

with the local Green’s functions

\[
G_\alpha(\omega) = \frac{1}{N} \sum_q \omega^2 - c_\alpha^2(\omega)q^2,
\]

where \(c_\alpha^2(\omega) = \nu_\alpha^2 - i\omega D_\alpha\).

The standard procedure for the \(q\) sum is to consider a cubic sample of size \(L\) with periodic boundary conditions. This gives a triple sum over discrete components \(q_i = \nu_i \Delta q, i = x, y, z\) with \(\nu_i \in \mathbb{Z}\) and \(\Delta q = 2\pi/L\). In the limit \(L \to \infty\), it transforms to the standard integral via \(N^{-1} \sum_q \to N^{-1} (L/2\pi)^3 \int dq_x \int dq_y \int dq_z = 3q_D^{-3} \int_0^{q_D} dq q^2\) with the Debye wavenumber \(q_D = (6\pi^2 N/L^3)^{1/3}, N\) being the number of atoms.

In their paper BZ say that the \(q\) integral for calculating \(G_\alpha(\omega)\) would be elementary, and is given by

\[
G^{3d}_\alpha(\omega) = -\frac{3}{\xi_\alpha^2(\omega)} \left( 1 + \frac{\eta_\alpha(\omega)}{2} \ln \frac{\eta_\alpha(\omega) - 1}{\eta_\alpha(\omega) + 1} \right)
\]

with \(\xi_\alpha(\omega) = q_D c_\alpha(\omega)\) and \(\eta_\alpha(\omega) = \omega/\xi_\alpha(\omega)\).

The function \(G(\eta_\alpha)\) is analytical in the entire complex plane, except on the real axis. Moreover, this exact result does not give any maximum in the reduced density of states. This can be seen from Fig. 1 where we have plotted the transverse reduced DOS \(g_T(\omega)/g_0(\omega)\) with

\[
g_0(\omega) = \frac{3}{q^2 T} \omega^2.
\]

We see that the introduction of the anharmonic damping increases the initial \(\omega^2\) law — as anticipated above — and smoothes the sharp Debye cutoff. There is no trace of a boson-peak-like maximum. So the question arises, where BZ came up with (Eq. (7) in BZ) is rather intransparent and involves the digamma function \(\psi(z) = \text{d \ln} \Gamma(z)/\text{d} z\). In a less disguised form, identifying their variables \(x, y\) with \(x = -q_D \eta_\alpha(\omega), y = q_D \eta_T(\omega)\), and using \(1 + i = \sqrt{2i}\), an analogue of the Eq. (7) of BZ can be obtained with Green’s functions

\[
G^{BZ}_\alpha(\omega) = -\frac{q_D}{N 2\eta_\alpha(\omega) \xi_\alpha^2(\omega)} F(q_D, q_D \eta_\alpha(\omega)),
\]

where

\[
F(n, z) = (\psi(z) - \psi(-z) - \psi(1+n+z) + \psi(1+n-z)).
\]

Apparently, BZ used the well-known [22, entry 4.1.5.9] single sum (following from the recursion relation \(\psi(z+1) = \psi(z) + z^{-1}\))

\[
\sum_{n=0}^N \frac{1}{\nu^2 - z^2} = \frac{1}{2z} F(n, z),
\]

with \(n = q_D\), thus solving a sort of one-dimensional problem instead of the original three-dimensional one. There are more inconsistencies in the expression (7): First, the Green’s function must have the dimension of an inverse squared frequency, which does not hold for (7). Secondly, in (7), \(n\) is an integer and \(z\) a (dimensionless) complex number. If one wanted to apply the summation formula (11) to a one-dimensional problem, the corresponding Green’s function would be given, for large \(N\), by

\[
G^{1d}_\alpha(\omega) = \frac{2}{N} \sum_{\nu=0}^{N/2} \frac{\omega^2 - c^2(\omega) (\Delta q)^2 \nu^2}{\nu^2}
\]

\[
= -\frac{1}{2\eta_\alpha(\omega) \xi_\alpha^2(\omega)} F\left( \frac{N}{2}, \frac{N}{2} \eta_\alpha(\omega) \right).
\]

For large \(L\), i.e., small \(\Delta q\), by Stirling’s theorem \(\lim_{z \to \infty} \ln \Gamma(z) \to z \ln z \Rightarrow \lim_{z \to \infty} \psi(z) \to \ln z\), which gives the correct one-dimensional Green’s function

\[
G^{1d}_\alpha(\omega) \to \frac{1}{2\eta_\alpha(\omega) \xi_\alpha^2(\omega)} \ln \frac{\eta_\alpha(\omega) - 1}{\eta_\alpha(\omega) + 1}.
\]

In Fig. 2 we have plotted the transverse DOS, which results from the Green’s function in Eq. (11) for several
values of \( q_D/\Delta q = N/2 \), along with the result corresponding to \( N \to \infty \), Eq. (11). It is clearly seen that the peaks, which occur, are the resonances corresponding to the harmonics of a very short spring. Such resonances have nothing to do with the boson peak in a three-dimensional solid, discussed in the paper by BZ. Because in Figs. 1 and 4 of this paper only one maximum is visible, we suspect that the authors chose \( q_D/\Delta q = N/2 = 1 \), which corresponds to a diatomic molecule. It is clearly seen that for increasing \( N \) the resonances are shifted towards small frequencies, and gradually the \( N \to \infty \) result (11) is recovered. In this limit, as in three dimensions, no “boson peaks” appear.

It is a coincidence, that Eq. (11) for small \( N \) implies a DOS proportional to \( \omega^2 \). This frequency dependence comes from the Akhiezer law used as input and has nothing to do with three-dimensional Debye-type wave propagation, as discussed in the paper by BZ.

In summary, we have shown, that the “exact” results by BZ are not mathematically correct and correspond to the resonances of very small strings. The model considered by BZ, i.e. Debye waves with Akhiezer-type damping does not produce a boson peak.

Acknowledgment

TB was supported by NRFU grant 2020.02/0115.

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