Comment on "Universal Origin of Boson Peak Vibrational Anomalies in Ordered Crystals and in Amorphous Materials"

Recent unexpected results by Baggioli and Zaccone (BZ) [1] on the origin of the boson peak in glasses (and boson-peak-like features in crystals) attributed it to the anharmonicity-caused damping of acoustic modes. BZ claimed to obtain an exact expression for the vibrational density of states (VDOS) $g(\omega)$. However, the Letter contains mathematical errors that invalidate all obtained results and render their interpretation misleading.

BZ consider amplitudes $u_\alpha(\mathbf{q},\omega)$ of anharmonically damped longitudinal ($\alpha = L$) and transverse ($\alpha = T$) phonons, obeying hydrodynamic eigenvalue equations $[\omega^2 - c_\alpha^2(\omega)q^2]u_\alpha(\mathbf{q},\omega) = 0$ with $c_\alpha^2(\omega) = c_\alpha^2 - i\omega D_\alpha$, where c_α are the sound velocities and D_α are the damping coefficients. In *three* dimensions, the corresponding VDOS can be calculated in the usual way from minus the imaginary part of the sum of the longitudinal and twice the transverse Green's functions $G_\alpha(\omega) = N^{-1}\sum_{\mathbf{q}}[\omega^2 - c_\alpha^2(\omega)q^2]^{-1}$ yielding Eq. (6) of BZ. The standard procedure for the $\mathbf{q}$ 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 $N^{-1}\sum_{\mathbf{q}} \to N^{-1}(L/2\pi)^3 \int dq_x \int dq_y \times \int dq_z = 3q_D^{-3}\int_0^{q_D} dq q^2$ with the Debye wave number $q_D = (6\pi^2 N/L^3)^{1/3}$ and N being the number of atoms.

First of all, we do not agree with BZ that the integral resulting from the sum over momentum "is not analytical." The integral from $q = 0$ to $q = q_D$ is elementary, given by the analytical function

$$G_\alpha^{3d}(\omega) = -\frac{3}{\xi_\alpha^2(\omega)}\left(1 + \frac{\eta_\alpha(\omega)}{2}\ln\frac{\eta_\alpha(\omega) - 1}{\eta_\alpha(\omega) + 1}\right), \quad (1)$$

with $\xi_\alpha^2(\omega) = q_D^2 c_\alpha^2(\omega)$ and $\eta_\alpha(\omega) = \omega/\xi_\alpha(\omega)$. More important, using this exact expression, no peaks in the reduced VDOS $g(\omega)/\omega^2$ (boson peaks) are obtained [2].

Secondly, instead of using Eq. (1) for Green's functions, the authors claim that the $\mathbf{q}$ sum would be represented by an expression [Eq. (7) of BZ] involving the digamma function $\psi(z) = d\ln\Gamma(z)/dz$. In a less disguised form, identifying their variables x, y as $x = -q_D\eta_L(\omega)$, $y = q_D\eta_T(\omega)$, and using $1 + i = \sqrt{2i}$, Eq. (7) of BZ can be obtained with Green's functions

$$G_\alpha^{\mathrm{BZ}}(\omega) = -\frac{1}{N}\frac{q_D}{2\eta_\alpha(\omega)\xi_\alpha^2(\omega)}F(q_D, q_D\eta_\alpha(\omega)), \quad (2)$$

where $F(n,z) = \psi(z) - \psi(-z) - \psi(1+n+z) + \psi(1+n-z)$. Apparently, BZ used the well-known *single* sum [following from the recursion relation $\psi(z+1) = \psi(z) + z^{-1}$]

$$\sum_{\nu=0}^{n}\frac{1}{\nu^2 - z^2} = \frac{1}{2z}F(n,z), \quad (3)$$

with $n = q_D$, thus solving a sort of one-dimensional problem instead of the original three-dimensional one. Notice that n is an integer here and z a dimensionless complex number; i.e., correctly, one would have to replace n by $q_D/\Delta q$ and η_α by $\eta_\alpha/\Delta q$. Therefore, only in the one-dimensional case (where $q_D = N\pi/L$) one would get an expression like (2), which would read as

$$G_\alpha^{1d}(\omega) = -\frac{1}{2\eta_\alpha(\omega)\xi_\alpha^2(\omega)}F\left(\frac{N}{2}, \frac{N}{2}\eta_\alpha(\omega)\right). \quad (4)$$

For large L, i.e., small Δq, 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.

However, in three dimensions the summation cannot be done in this way and the results reported by BZ could only apply to one-dimensional systems. But, also in this case, there are severe drawbacks. Indeed, we found [2] that the "boson peaks" displayed in Figs. 1 and 4 of BZ can only be obtained with $\Delta q/q_D$ of order 1, i.e., L of order of an interatomic spacing. For very small $\Delta q/q_D$, i.e., for realistically large $L \gg q_D^{-1}$, there is no maximum in the reduced VDOS. The maxima in Figs. 1 and 4 of BZ are the main resonances of microscopically small strings and have nothing to do with vibrational properties of crystals or glasses [2].

We conclude that by a correct treatment of this anharmonic model [1] no boson peak is obtained; hence the complete discussion in the commented Letter becomes useless and misleading.

T. B. was supported by National Research Foundation of Ukraine Grant No. 2020.02/0115.

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0031-9007/21/127(17)/179601(2)

Received 27 April 2021; accepted 24 September 2021; published 21 October 2021

DOI: 10.1103/PhysRevLett.127.179601

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