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Pressure dependence of the boson peak for repulsive homogeneous potentials

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Isotropic potentials used for studying glasses computationally are often positive homogeneous functions of the coordinates of the constituent particles or they reduce effectively to such forms at higher pressures. For such cases we show that the position and the intensity of the Boson peak, when it exists, scales asymptotically with pressure (P) as P^{δ} and $P^{-3\delta}$, respectively, where $\delta = (2+m)/[2(S+m)]$, $(-m)$ is the degree of homogeneity, and S is the spatial dimension. In the same limit the shape of the *entire* vibrational spectrum saturates and the Boson peak frequency, the Debye frequency, and the average frequency become proportional to each other. Our numerical studies on single-component hyperquenched Lennard-Jones glass in three dimensions support the predicted scaling laws and are largely consistent with the predictions of one existing theory [W. Schirmacher *et al.*, Phys. Rev. Lett. **98**, 025501 (2007)] for the normalized boson peak intensity. However, the scaling exponent for the variation of the boson peak location and intensity with applied pressure is substantially different in this case from that found from laboratory experiments.

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I. INTRODUCTION

Due to the absence of long-range positional order, understanding the vibrational spectra of glassy materials presents extremely difficult challenges.[1,2] One example that vividly illustrates these difficulties is the apparently universal existence of a "boson peak" (BP). This refers to the presence of an excess of low-frequency vibrational modes, typically in the THz region, when compared with the predictions of the Debye model of the vibrational spectrum. If the vibrational density of states (VDOS) at frequency ω is denoted by $G(\omega)$ this excess manifests itself as a peak in the plot of $G(\omega)/\omega^2$, also called the reduced density of states (RDOS), as a function of ω. A large number of experimental, theoretical, and numerical studies have been conducted over the past several decades to understand the exact physical origin of this boson peak.[3–19] Although substantial progress has been made, the issue is far from closure. To discriminate between the various explanations that have been proposed it is necessary to expand the scope of the phenomenology so that additional constraints can be placed on the theories. For this reason several variations have been performed on the original experiments involving boson peaks. The most important of these are perhaps the influence of temperature and pressure (or densification) on the various quantifiers of the boson peak.[20–29] Here we concentrate only on the latter.

The aspects that are studied in these experiments are typically the location (ω_{BP}) and the intensity (I_{BP}) of the (boson) peak of the RDOS function as well as the shape of the RDOS function around the peak. The most important common observations are that (1) ω_{BP} keeps increasing and I_{BP} keeps decreasing with increasing applied pressure, and (2) the rescaled RDOS function obtained by normalizing the frequency by ω_{BP} and the original RDOS by I_{BP} remains invariant with respect to the applied pressure *in the proximity of* the boson peak. What the experiments do not agree upon is whether the invariance around the boson peak mentioned above would hold if the Debye frequency (ω_D), rather than ω_{BP}, is used as the unit of frequency.[20,27,29–31] This issue is critical from the point of view of understanding the underlying basic physics since invariance under rescaling by ω_D would imply that the RDOS around the boson peak is controlled by the property of elasticity, which operates at length scales significantly larger than the interatomic distances.

Understanding the difference of outcome in this regard (and perhaps in other respects) is made difficult by the complex structural nature of these glasses, the presence of bonding interactions, etc. For example, let us consider the most detailed theory presently available for the evolution of the boson peak frequency with pressure. This theory[32] is applicable to cases where the boson peak owes its origin purely due to disorder. In particular it is not applicable to situations where the boson peak merely reflects the broadening of some low-lying vibrational modes due to disorder. The complexities introduced by the factors present in experimental situations, as described above, make it difficult to decide the extent of applicability of a theory such as this with complete certainty.

Because of these complications in using the existing experiments to validate theoretical ideas it would be desirable to study systems where the boson peak, without any ambiguity, is entirely due to disorder. Presently such structural glasses cannot be realized in laboratory experiments since the cooling rate required is well beyond existing capabilities. The only alternative is to generate such glasses in a computer. This is the route we take here. The model potentials used in the present work do not necessarily represent realistically any particular material (especially at high pressures). Hence we emphasise that our work has the limited objective of helping generate and validate *theoretical* understanding regarding the pressure evolution of (i) the shape of the reduced vibrational density of states function and (ii) the location and intensity of the boson peak—in a situation where we know unambiguously that the boson peak is due entirely to disorder. For these reasons any direct comparison of our results with any laboratory experiment that is presently realized (or is realizable in the near future) is not permissible.

We study model glassy systems where the interparticle interactions are isotropic and are described by positive homogeneous potentials—at least at higher pressures. This type includes in its scope most of the model studies of glasses with isotropic potentials that we are aware of (in particular, it includes the system (Ref. 8 of this paper) that was referred to in Ref. 32 as a model candidate for its application). The particles need not have identical mass but all pairs must interact via positive homogenous potentials with identical degree of homogeneity (to be defined later). We show that it is possible to derive, through rather simple arguments, the asymptotic pressure dependence of the location as well as the intensity of the boson peak (the procedure does not require a knowledge of the origin of the boson peak)—when it exists. Asymptotically both these quantities have power-law dependence on pressure and the exponent of the power law can be calculated exactly. We also show that, for these potentials, the shape of the *entire* vibrational spectrum (not just the region around the boson peak) can be made invariant with respect to pressure if the unit of frequency is taken to be either ω_{BP} or ω_D—since these two frequencies become proportional to each other at higher pressures. The actual asymptotic shape depends on the specific choice of the positive homogenous potential and can only be calculated numerically.

In this paper we choose the potential to be the familiar Lennard-Jones potential and present the results of a numerical study of the pressure evolution of the vibrational spectrum for the single-component hyperquenched glass in three dimensions interacting via this potential. The results demonstrate the validity of our analytical calculations (although our analytical results are applicable to much more complex systems also). They are also in reasonable agreement with an existing theory by Schirmacher *et al.*[7] as far as the normalized intensity of the boson peak is concerned. However, the phenomenology regarding the evolution of the location and intensity of the boson peak is quite different from that found in the laboratory experiments. All our calculations are done at zero temperature. But this should not be a serious compromise at the high pressures we are interested in as long as the temperature stays modest. In Sec. II we introduce the positive homogenous potentials. The existence of asymptotic shape invariance (with respect to pressure) for the reduced density of states is shown. Also the pressure scaling exponents for the location and the intensity of the boson peak are calculated. In Sec. III detailed results are presented for the numerical study of the pressure evolution of the reduced density of states for the hyperquenched Lennard-Jones glass. Section IV contains a discussion of the results.

II. SCALING AND SHAPE INVARIANCE: ANALYTICAL RESULTS

A positive homogeneous function V describing the interactions among the particles in an amorphous system, which may have one or more species, will have (by definition) the following property: $V(\alpha\vec{r}_1,\alpha\vec{r}_2,\ldots,\alpha\vec{r}_i,\ldots) = \alpha^k V(\vec{r}_1,\vec{r}_2,\ldots,\vec{r}_i,\ldots)$, where α is a positive real number (in practice, in a large enough range around unity) and k is the degree of homogeneity. The simplest example is when the potential energy is of the form of a sum over pairs and every pair interacts with a soft potential proportional to the inverse mth power of the pair distance. Evidently $k = -m$ for such a case. Examination of almost all isotropic potentials used in the literature in recent times[6,8,14,16,19,33–36] will show them to be positive homogenous potentials at higher pressures—with the degree of homogeneity being controlled by the repulsive part of the pair potential. In principle, even potential-energy functions containing many-body terms can be homogeneous functions but instances of this are not likely to be common. In the following the degree of homogeneity will be taken to be a negative integer $-m$.

To derive the pressure dependence of the boson peak and to demonstrate asymptotic shape invariance of the vibrational spectrum we follow the route that is taken in the numerical approach to this problem, i.e., approximate the amorphous state as a periodic crystal with a large number (N) of particles in the unit cell and then take the limit of $N \to \infty$. For a given finite value of N the amorphous states at pressure P are represented by the appropriate local minima of the enthalpy function $U + PV$ where U is the energy per unit cell of (generalized) volume V. The minima are defined with respect to the variation of the S sides of the unit cell ($\vec{a}_1$, $\vec{a}_2, \ldots$, $\vec{a}_S$ with $S > 1$) and the positions of the particles within the unit cell. Consider now one such local minimum (in the large N limit vibrational properties will be the same for all local minima). For this solid (in S dimension) the VDOS, in general, is computed by finding the SN eigenvalues of a generalized eigenvalue problem where one side of the equation contains a fixed mass matrix and the other side has the dynamical matrix $D(\vec{k})$. And this has to be done for all $\vec{k} \in$ the first Brillouin zone (FBZ).

We now show that, when the potential is a positive homogeneous function, the frequencies thus obtained will satisfy the following scaling law:

$$\omega_i(\vec{k}) = W(P) F_i(n^{-\frac{1}{S}}\vec{k}), \tag{1}$$

where the branch index $i = 1,2,3,\ldots,SN$ (with the convention that $i = 1,\ldots,S$ corresponds to the acoustic branches); n is the number density of particles and $W(P)$ is a characteristic scale of frequency that depends on pressure. The functions F_i are independent of pressure. Since the vibrational frequencies are the square roots of the eigenvalues, validity of the scaling law of Eq. (1) would be demonstrated if we can show that $D(\vec{k})$ itself has the structure of a function of $n^{-1/S}\vec{k}$ multiplied by a function of pressure only. For this purpose let us consider the geometry of any particular local minimum representing the amorphous solid at a pressure P_1 that is high enough (if necessary) to permit the homogeneous potential description. Now examine the configuration obtained from this one by shrinking it as a whole by a factor of $\beta > 1$. Since the first derivative of a homogeneous function of degree k with respect to any of its arguments is also a homogeneous function [but of degree $(k - 1)$] this contracted geometry is again an equilibrium arrangement corresponding to a higher pressure of $P_2 = \theta P_1$ where $\theta = \beta^{(m+S)}$. Thus the geometry of the solid is independent of the applied pressure, which controls only the *scale* of the interparticle distances and this scale varies as $P^{-[1/(m+S)]}$.

Now the dynamical matrix $D(\vec{k})$ is a Fourier transform of a derivative matrix $\mathbf{D}(\vec{R})$—the latter being the matrix of second derivatives of the potential energy with respect to the coordinates of two particles, which are situated in two unit cells separated by the Bravais lattice vector $\vec{R}$. In fact $D(\vec{k}) \equiv \sum_{\vec{R}} \mathbf{D}(\vec{R}) e^{-i\vec{k}\cdot\vec{R}} = \sum_{n_1,n_2,\ldots,n_S} \mathbf{T}(n_1,n_2,\ldots,n_S) e^{-i\vec{k}\cdot(n_1\vec{a}_1+n_2\vec{a}_2+\cdots+n_S\vec{a}_S)}$, where the values of $n_1,n_2,\ldots,n_S$ span all integers. The elements of the $\mathbf{T}$ matrix are homogeneous functions of degree $-(m+2)$ and are inversely proportional to the $(m+2)$th power of the characteristic length scale. Thus the $\mathbf{T}$ matrix has the form of a product of $P^{[(m+2)/(m+S)]}$ and a matrix that depends only on $(n_1,n_2,\ldots,n_S)$ and the geometry of arrangement of the particles within the unit cell (but not on the length scale). The vectors $\vec{a}_1,\vec{a}_2,\ldots,\vec{a}_S$ are all proportional to $n^{-1/S}$. Hence $\vec{k}\cdot\vec{R}$ is actually a dot product between $n^{-1/S}\vec{k}$ and a vector that depends only on $(n_1,n_2,\ldots,n_S)$ and the (pressure independent) *directions* of $\vec{a}_1,\vec{a}_2,\ldots,\vec{a}_S$. Thus $D(\vec{k})$ is indeed a product of a function of pressure (that defines the scale of the vibrational frequency) and a function of the product $n^{-1/S}\vec{k}$. Equation (1) follows immediately from this—with the additional specification that $W(P)$ is actually proportional to $P^{[(m+2)/2(m+S)]}$.

The shape of the FBZ does not change with pressure but its linear size is proportional to $n^{1/S}$. It follows that the normalized VDOS function $G(\omega)$ will have the following form:

$$G(\omega) = [1/W(P)]H[\omega/W(P)], \tag{2}$$

where the normalized function H is independent of pressure. The boson peak, if it exists, will show up as a peak in the plot of $H(x)/x^2$ vs x. Clearly, in that case ω_{BP} is proportional to P^{δ} with $\delta = (m+2)/2(m+S)$. Similarly, I_{BP} scales as $W(P)^{-3}$.

The expression for δ shows that it has a universal value of 1/2 in two dimensions. In three dimensions δ is bounded above by 1/2 and its dependence on m is very weak. For example, varying m from 7 to 20 causes δ to change merely from 0.45 to 0.48. From Eq. (1) it can be seen that the speed of sound is proportional to $n^{-1/S}W(P)$ and, consequently, that the Debye frequency (ω_D) is also proportional to $W(P)$. Thus ω_{BP}, ω_D, and the average frequency are all proportional to each other. Given this mutual proportionality and the form of $G(\omega)$ in Eq. (2) it should be obvious that shape invariance of the rescaled RDOS can be realized in the *entire* spectrum (not just around the boson peak) by scaling the frequency either by ω_{BP} or by ω_D. As mentioned earlier this is not always the case in experiments.

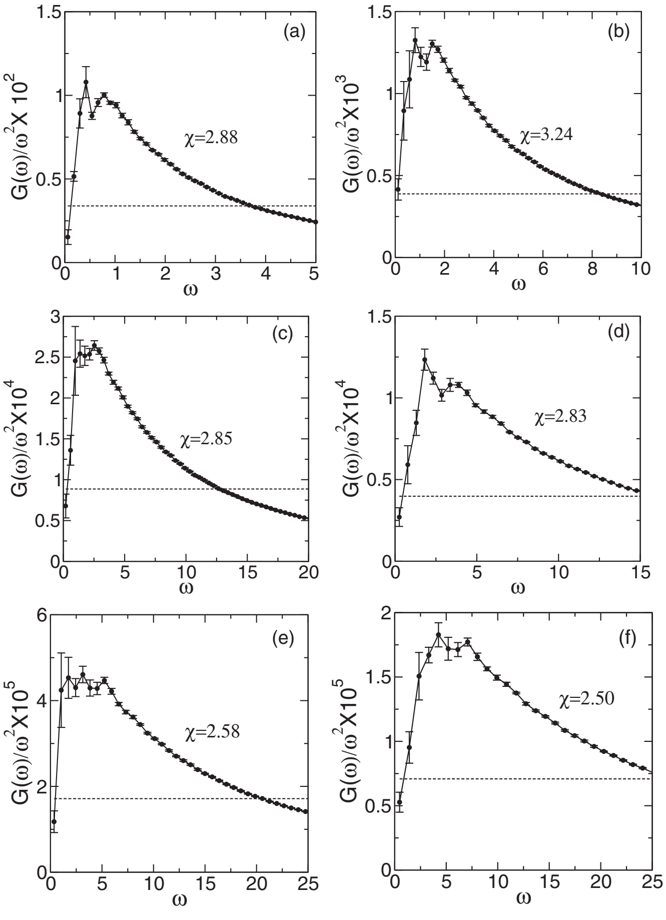

FIG. 1. Reduced density of states $[G(\omega)/\omega^2]$ is plotted as function of frequency (ω). The Debye value is shown as a horizontal dashed line. Also shown is the value (χ) of $G(\omega_{\mathrm{BP}})/G_{\mathrm{Debye}}(\omega_{\mathrm{BP}})$. (a) $P = 1$, (b) $P = 16$, (c) $P = 64$, (d) $P = 128$, (e) $P = 256$, and (f) $P = 512$.

III. LENNARD-JONES GLASS: NUMERICAL RESULTS

We now present results of a numerical study of the evolution of the boson peak at zero temperature in the case of the single-component hyperquenched Lennard-Jones glass ($m = 12$) in three dimensions (the boson peak aspect of this problem was studied in Ref. 19 but only at $P = 0$; see also Ref. 36). In appropriate reduced units the expression for the pair potential in this case for a pair distance of r is $(1/r^{12} - 1/r^6)$ (please note the absence of the prefactor of 4). The mass of every particle is unity and the number (N) of particles in the unit cell is 3375. Calculations were done at $P = 1$, 16, 64, 128, 256, and 512 (for the Lennard-Jones parameters of argon typically used in the literature $P = 512$ corresponds to about 90 GPa). Amorphous geometries at $P = 1$ were obtained through hyperquenching of low-temperature uncorrelated liquid configurations. Configurations at higher pressures were derived from these through sequential minimization of the enthalpy function—as described earlier. For each pressure

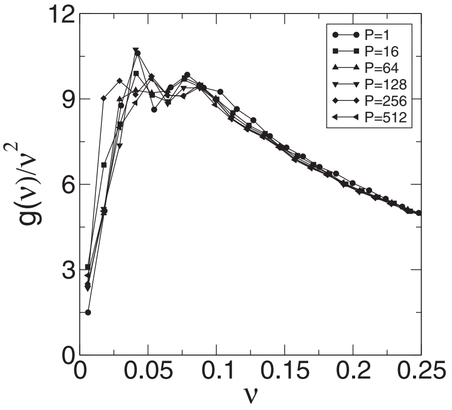

FIG. 2. Superposition of the plots of the reduced density of states [with normalized frequency (ν) as the argument] at various pressures is shown to demonstrate their asymptotic convergence.

TABLE I. Pressure dependence of various physical parameters for the Lennard-Jones glass.

Pressure (P)	Volume per particle (v)	Frequency of boson peak ($\omega_{\rm BP}$)	Intensity of boson peak ($I_{\rm BP}$)	Excess at boson peak (χ)	Strain parameter (ϵ)	Bulk modulus (B)
1	0.9238	0.60	0.0098	2.88	0.0171	24
16	0.7213	1.15	0.0013	3.24	0.0950	129
64	0.5861	1.94	0.00025	2.85	0.1555	418
128	0.5218	2.85	0.00011	2.83	0.1876	764
256	0.4619	3.48	0.000044	2.58	0.2199	1494
512	0.4072	5.66	0.000018	2.50	0.2520	2772

$G(\omega)$ was calculated by combining the spectra from 12 amorphous local minima. In Figs. 1(a)–1(f) we show the plots of the RDOS. Also shown is the (constant) value of the RDOS predicted by the Debye model. Next to each plot the value of $\chi = G(\omega_{\rm BP})/G_{\rm Debye}(\omega_{\rm BP})$ is also given. For homogeneous potentials χ should saturate at high pressures. From our numerics this limiting value would appear to be somewhat below 2.5—which is a little higher than the value of 2 predicted by a theory of Schirmacher *et al.*[7] that ascribes the origin of the boson peak to random spatial fluctuations of the elastic constants. It may be noted that Ref. 7 quoted a value of 2.4 for this parameter for the Lennard-Jones potential—although the value of pressure that the data related to was not mentioned.

Given the error bars in Figs. 1(a)–1(f) there can be no reasonable doubt about the existence of the boson peak (see also Ref. 19)—although the uncertainty in the location of $\omega_{\rm BP}$ is higher than desirable. This is confirmed further in Fig. 2 where superposed plots at various pressures are shown for $g(\nu)/\nu^2$ (focussing attention near the boson peak) where $g(\nu)$ is the normalized density of states but with $\nu \equiv \omega/\langle\omega\rangle$ where $\langle\omega\rangle$ is the spectral average of ω. While converting ω to ν the value of $\langle\omega\rangle$ specific to the local minimum is used. Although not seen fully in Fig. 2, there is indeed asymptotic convergence of the rescaled RDOS function over the *entire* range of ν. The effect of somewhat poor statistics is visible at the lowest frequencies. And this is further amplified due to division by the square of the rather low value of frequency in the proximity of the boson peak [in a superposed plot of $g(\nu)$ for all six pressures it is hard to distinguish between the six figures]. But there is little doubt that $\nu_{\rm BP}$ indeed becomes independent of pressure at higher pressures. From Fig. 1 it can be seen that $\omega_{\rm BP}$ and $I_{\rm BP}$ vary by a factor of about 9 and 600, respectively, across the range of pressures studied. These and other relevant data at various pressures are recorded in Table I. Figures 3(a) and 3(b) show the log-log plot of $\omega_{\rm BP}$ and $I_{\rm BP}$ against pressure. The best-fit values of the slope (using data for the three highest pressures only, since the predictions are asymptotic in nature) in Figs. 3(a) and 3(b) are 0.49 and -1.36, respectively. These values are to be compared with the analytical predictions of 0.467 and -1.4, respectively.

IV. DISCUSSION

This work is based on the observation that most of the isotropic potentials used in the literature for computational studies of glasses reduce to homogenous repulsive potentials at high pressures and hence it is enough to study the latter kind of potential if one is interested in high-pressure vibrational properties of these model potentials. We have demonstrated that the pressure scaling of the location and the intensity of the boson peak, when it exists (the proof of existence is numerical, not analytical), is of the power-law type and the exponent can be calculated without invoking the mechanism of BP formation. Since one motivation of this work is to generate data that unambiguously comply with the basic requirements of a theory such as Ref. 32, it is natural to ask how far our results are consistent with its predictions regarding the dependence of $\omega_{\rm BP}$ on pressure.

This theory, based on the soft potential model (SPM) of the origin of the boson peak, predicts a $P^{1/3}$ dependence of the boson peak frequency for an intermediate range of pressures where the pressure is high compared to an intrinsic and system dependent scale of P_0 but is low enough to satisfy the following two conditions assumed implicitly in the derivation of the predictions: (i) In the Hamiltonian the coupling between the strain induced by pressure and the displacement coordinates of the quasilocal vibrational (QLV) modes stays bilinear. (ii) The compressibility can be taken to be constant. In any comparison between experimental/computational data and a theory such as this, caution must be exercised in making sure that (A) the system is indeed of the type to which the theory is applicable, and (B) conditions under which the predictions are made are actually satisfied by the range of pressures used in the experiment/computation. As stated in the introduction of this paper our systems satisfy criterion (A) by design. Criterion (B) requires that the pressure should be in the range that satisfies the conditions (i) and (ii) stated above.

Validity of condition (i) stated above is hard to check *a priori* but the applicability of condition (ii) can be tested through a measurement/computation of the compressibility as a function of pressure. We have computed compressibility at the pressures used for the present calculation via interpolation through the pressure-volume data of a much denser set of pressures computed with $N = 343$. From the results available in Table I it is obvious that the variation of compressibility is very large over the range of pressures used by us. In fact if one were to demand only a small variation of the compressibility over the studied pressure range the latter would be so narrow that the resulting variation of $\omega_{\rm BP}$ over that range would be too small to permit any meaningful test of the $P^{1/3}$ prediction. This problem is not limited to our study. They are also known to be present in the experimental studies of amorphous silica[25] and PIB polymeric glass[27] (which are among those considered as confirmations of the $P^{1/3}$ law)—although to a lesser degree. For amorphous silica, over the studied range of ambient through 50 GPa, the compressibility varies by a factor of about 10. For the PIB study, which extends up to 1.4 GPa, this variation is by a factor of more than 2.5. We do not have access to similar compressibility data for glassy As_2S_3.[26] When compressibilty varies so much, what actually is the predicted pressure dependence of ω_{BP}?

A careful reading of Ref. 32 shows that the prediction can be written as a dependence of ω_{BP} on the strain parameter (ϵ)—the latter being a function of pressure. Again there is a system dependent scale ϵ_0 for the strain such that a model independent prediction exists only when $\epsilon \gg \epsilon_0$ and in this pressure range $\omega_{BP} \propto \epsilon^{1/3}$. For the laboratory experiments the strain parameter is not measured directly but it can be estimated if the compressibility is known as a function of pressure. In any case, on the basis of the fact that the compressibility rises monotonically with pressure, it can be inferred that ω_{BP} should rise slower (possibly substantially slower) than $P^{1/3}$. Hence if the pressure dependence of compressibility is not properly taken into account it can seriously compromise the process of confirming the theoretical predictions. Experiments, however, seem to show fairly close to $P^{1/3}$ dependence. These issues need to be understood more carefully.

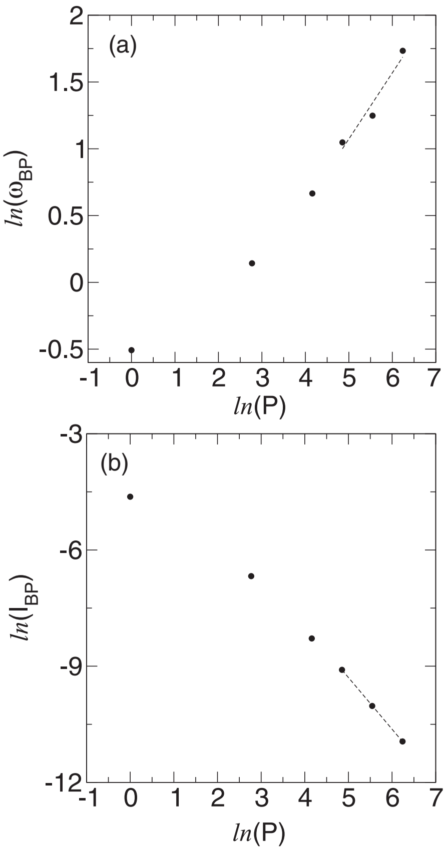

FIG. 3. Log-log plot against pressure (P) is shown for (a) frequency of the boson peak (ω_{BP}) and (b) intensity of the boson peak (I_{BP}).

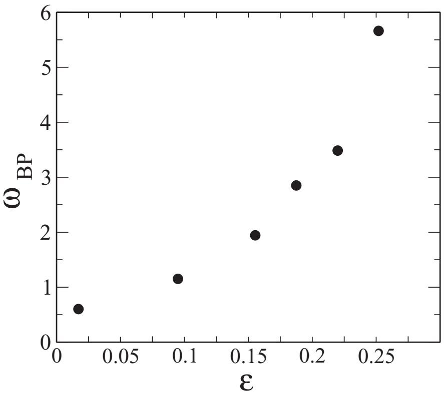

FIG. 4. Plot of the boson peak frequency (ω_{BP}) against the strain parameter (ϵ).

For the present study of homogenous repulsive potentials the predicted and observed pressure dependence of ω_{BP} is P^δ where δ is close to but a little lower than $1/2$. For the particular example of the Lennard-Jones potential we have available the pressure-volume data from which we can calculate the strain parameter as a function of pressure via the definition $\epsilon \equiv \{1 - [v(P)/v(0)]^{1/3}\}$ where $v(P)$ is the volume per particle at pressure P. The data for this is available in Table I. A plot of ω_{BP} vs strain is shown in Fig. 4. It should be obvious that the dependence of ω_{BP} on strain is strongly superlinear and is certainly far from being proportional to $\epsilon^{1/3}$. We conclude that our data cannot be fitted to the predictions of Ref. 32. A simple explanation for this observation may be that the values of the strain parameter for the higher pressures in our study are beyond the limit of validity of the bilinear coupling between the strain parameter and the displacement coordinate of the QLV.

The observations of the asymptotic shape invariance of the vibrational spectrum and the proportionality of Debye and boson peak frequencies are equally important in our view. This has been observed in some laboratory experiments. However, to the best of our knowledge, no explanation has been forwarded until now. Here we have pointed out that this is a generic feature of all homogeneous repulsive potentials. However, without further developments, this cannot be considered an explanation of any laboratory experiment. But at least we have demonstrated that the experimentally observed phenomenology is shared by most of the computationally studied models of glasses.

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