PHYSICAL REVIEW B VOLUME 45, NUMBER 5 1 FEBRUARY 1992-I

Local vibrational states of glasses

Tao Pang*
Department of Physics, University of California, Berkeley, California 94720

(Received 6 May 1991; revised manuscript received 19 September 1991)

A glass system is argued to be a collection of clusters. The average density of vibrational states from these clusters is calculated and found to be universal. The theoretical result is then compared with the density of vibrational states obtained from recent inelastic-neutron-scattering experiments and a good agreement is found in the intermediate-energy region of 1.5–15 meV.

The interesting debate[1] about what is the correct picture to describe low-temperature properties of insulating glasses has shed some doubts on the widely accepted two-level tunneling description.[2,3] More importantly, the strong argument made there shows that anomalies in the specific heat and thermal conductivity[4] in glasses are more or less universal in the moderate-temperature region around 10 K. Most recently, the density of vibrational states (DVS) obtained from inelastic-neutron-scattering experiments[5] and the Raman spectra[5,6] for various glasses are found to be universal in the energy region about 1.5–15 meV.[5,6]

However, what causes the universal structure in the DVS and Raman spectra is still unclear. The distinct feature of glass systems is their structures in the intermediate distances around few tens of Å. If a dimension analysis is made, one will find that the peak positions in the DVS do correspond to the intermediate length scales. The length scale corresponding to a given energy $\hbar\omega$ is $L \sim c/\omega$ where c is the sound velocity in the glass. All data of the Raman spectra for various glass systems can be well described by log-normal functions.[7] This indicates that if the broad peaks in the Raman spectra result from the structures with intermediate length scales, the size distribution of these structures will be log-normal, too.[7] For convenience, let us call these structures with intermediate length scales *clusters*. The vibrational properties of a single cluster may be inferred from the vibrational properties of microcrystals in a glass system. Experiments[8] have shown that the microcrystals in glass systems have similar but sharper peaks in the Raman spectra with peak positions at $\omega \sim c/L$ with L as the diameters of the microcrystals. The characteristic length of clusters in a glass system can also be estimated from the sharp peaks of x-ray experiments.[9] An interesting discussion[10] attributes the universal structure in the DVS and Raman spectra to a special kind of one-dimensional clusters in glasses, the so-called Rivier lines formed by odd member rings in the glass structures.[11,12]

In the present work we assume that the clusters in glasses are three-dimensional structures with a log-normal size distribution. Later we will show that the dimensionality of the clusters is not important. With help of this log-normal distribution, we derive a simple expression for the DVS in glasses. The obtained DVS is universal with a characteristic energy scale. We then compare the theoretical DVS with those found from the inelastic-neutron-scattering experiments and find that the theoretical expression for the DVS represents experimental data very well in the energy region of 1.5–15 meV.

As discussed above, a glass system is considered a collection of clusters with various sizes. It is not easy to identify one specific cluster in the system since each cluster may percolate into several other clusters to keep the whole system homogeneous. However, even though the clear separation between two clusters is very difficult, one can still assign a characteristic length L for each cluster, which is approximately the diameter across the cluster. The DVS of the whole system is then given by the average of the DVS in all clusters:

$$g(\omega) = \int g_L(\omega) P_L \, dL \ , \tag{1}$$

where $g_L(\omega)$ is the DVS in a cluster with characteristic length L. If size effect is ignored, the density of vibrational states in a cluster will have the Debye form

$$g_L(\omega) \sim \omega^{D-1} \ , \tag{2}$$

at the low-energy region. Here D is the dimension of the cluster. Of course, Eq. (2) cannot be true if the size effect dominates the spectra. As evidenced from the experiments,[8] the dispersion in the clusters should be more like those obtained from the vibrations of an elastic sphere.[13] There is a low-energy cutoff in $g_L(\omega)$ due to the finite-size effect: α/L and α is a constant close to c, the average sound velocity. This cutoff means that, when the energy is less than α/L, the vibration cannot exist in the cluster. If the cluster is taken as a cube with volume L^3 and the dispersion relation is assumed to be linear, α is given by $\alpha = 2\pi c$; or if the cluster is assumed as an elastic sphere with diameter L, α is given by $\alpha = 0.35c$.[13] P_L in Eq. (1) is the probability of a cluster with characteristic length L forming in a specific glass system and is normalized to 1,

$$\int P_L \, dL = 1 \ . \tag{3}$$

A trivial case of Eq. (1) is for the lattice system, where one has only one cluster with $L \to \infty$ and Eq. (1) reduces to the Debye form of Eq. (2). Hereafter we will exclude such case and assume that the cluster distribution has a peak at a characteristic length L_0 and that the probability goes to zero as $L \rightarrow \infty$. Many random processes[14,15] have shown that the clusters formed have log-normal size distributions and the log-normal fit for the Raman spectra[7] provides another strong evidence. This motivates our assumption that the size distribution of clusters in glass systems is log-normal:

$$P_L=\frac{1}{\sqrt{2\pi}\sigma L}\exp\left[-\frac{(\ln L-\ln L_0)^2}{2\sigma^2}\right], \qquad (4)$$

with σ the half width of the log-normal distribution.

Let us assume at the moment that the DVS in a cluster is a constant, that is, $g_L(\omega)=g_0$. Later we will show that the resultant average density of vibrational states will not change qualitatively as long as $g_L(\omega)$ has a power-law behavior, $g_L(\omega)\sim\omega^\gamma$. It is straightforward to show using Eqs. (1) and (4) that when $g_L(\omega)=g_0$, one has

$$g(\omega)=\frac{g_0}{2}\,\mathrm{erfc}\left[\beta\ln\left[\frac{\omega_0}{\omega}\right]\right], \qquad (5)$$

where erfc(x) is the complementary error function

$$\mathrm{erfc}(x)=\frac{2}{\sqrt{\pi}}\int_x^\infty e^{-x^2}dx\ , \qquad (6)$$

and $\omega_0=a/L_0$ and $\beta=1/\sqrt{2}\sigma$.

Now if the DVS of a cluster has a power-law behavior, $g_L(\omega)\sim\omega^\gamma$ with $\gamma=D-1$ for the Debye case, we can express this power-law density of states in terms of the characteristic length of the cluster using $\omega\sim 1/L$ in the cluster[8,13] as $g_L(\omega)=g_\gamma(L_0/L)^\gamma$. It is straightforward to show that $g(\omega)$ is still given by Eq. (5) but with $g_0=g_\gamma e^{\sigma^2\gamma^2/2}$ and $\omega_0=ae^{2\gamma\sigma^2}/L_0$. Please note that β is assumed not to change with the dimension or the specific choice of $g_L(\omega)$. This is consistent with the later discovery in our numerical results that β is almost a constant for all glass systems (see the numbers for β in Table I below). So the expression for the DVS in Eq. (5) will be an identical curve for all glass systems if the amplitude is rescaled by g_0 and the energy is rescaled by ω_0. This is precisely what was observed in the inelastic-neutron-scattering experiments in the energy region of 1.5–15 meV.[5] Later we will show that the universal structure in the Raman spectra[5,6] in the corresponding energy region also originates from the cluster size distribution.

Now let us see how well the expression of Eq. (5) represents experimental results. In Fig. 1, we plot three sets of experimental data of the DVS for glass systems As_2S_3, SiO_2, and $Mg_{70}Zn_{30}$ taken from Ref. 5 and the DVS of Eq. (5). g_0, β, and ω_0 for each case are optimized by a least-squares fit of Eq. (5) to the experimental data at given energy points in Fig. 1. It is very clear that the DVS from Eq. (5) represents all three sets of experimental data very well in the energy region of 1.5–15 meV. The optimized parameters β, and ω_0 for each case are given in Table I. It is important to point out that the

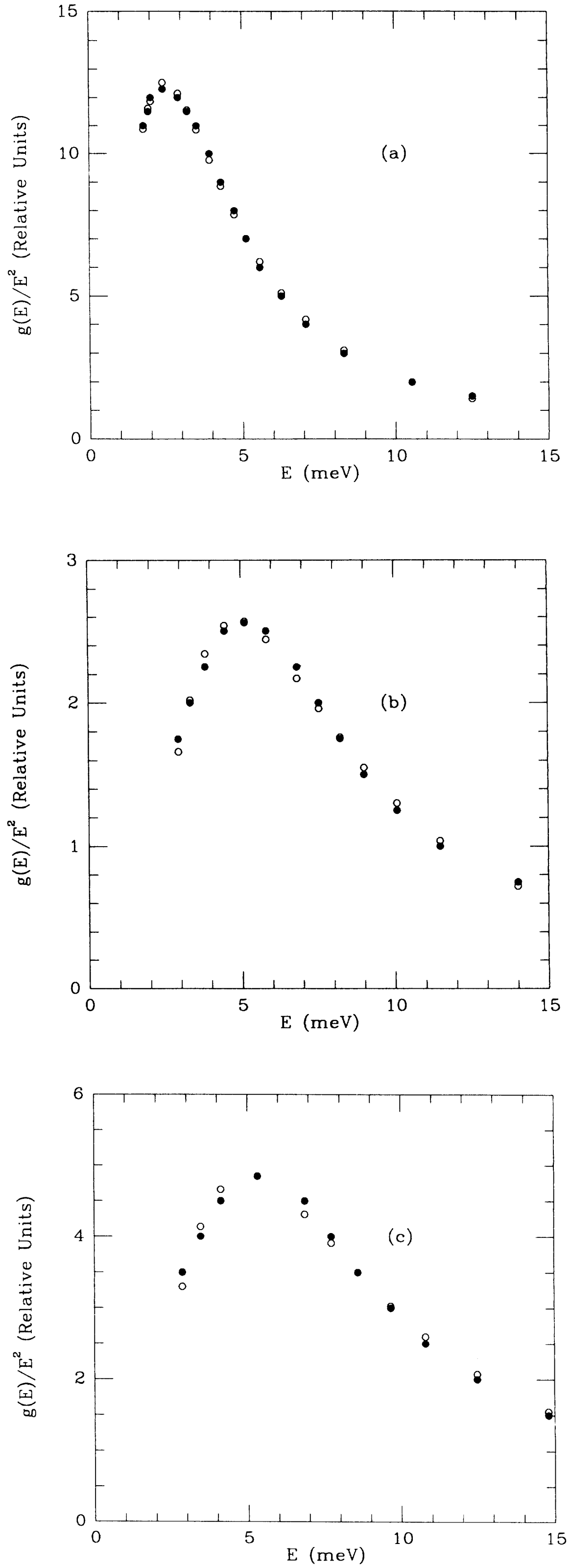

FIG. 1. Density of vibrational states observed in the inelastic-neutron-scattering experiments of Ref. 5 (●) compared with the theoretical expression in Eq. (5) (○) by a least-squares fit to determine g_0, β, and ω_0 in Eq. (5): (a) AsS_3, (b) SiO_2, and (c) $Mg_{70}Zn_{30}$.

TABLE I. The optimized β with ω_0 in Eq. (5) for various glass systems by a least-squares fit to the experimental data of Ref. 5.

	AsS_3	SiO_2	$Mg_{70}Zn_{30}$
β	1.32	1.52	1.29
$\hbar\omega_0$ (meV)	3.05	5.34	6.29

quantity β (or σ) for all three materials are almost the same.

In conclusion, the universal structure of the density of vibrational states in the glass systems is successfully attributed to the local vibrations of the clusters in the glass systems with structures at intermediate length scales. The simple expression obtained for the DVS of glasses is universal as long as the cluster size distribution is log-normal. In order to relate the universal DVS of glasses to the universal nature of the thermal properties, more work is still needed to find out the temperature dependence of the scattering length of phonons at this region.

I acknowledge the support of Miller Institute for Basic Research in Science at U. C. Berkeley.

---

[*] Present address: Department of Physics, University of Nevada, Las Vegas, Nevada 89154.

[1] C. C. Yu and A. J. Leggett, Comments Condens. Matter Phys. **14**, 231 (1988).

[2] P. W. Anderson, B. I. Halperin, and C. M. Varma, Philos. Mag. **25**, 1 (1972).

[3] W. A. Phillips, J. Low Temp. Phys. **7**, 351 (1972).

[4] J. J. Freeman and A. C. Anderson, Phys. Rev. B **34**, 5684 (1986).

[5] M. G. Zemlyanov, V. K. Malinovskiĭ, V. N. Novikov, P. P. Parshin, and A. P. Sokolov, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 521 (1989) [JETP Lett. **49**, 602 (1989)]; V. K. Malinovsky, V. N. Novikov, P. P. Parshin, A. P. Sokolov, and N. G. Zemlyanov, Europhys. Lett. **11**, 43 (1990).

[6] V. K. Malinovsky and A. P. Sokolov, Solid State Commun. **57**, 757 (1986).

[7] I. Pócsik and M. Koos, Solid State Commun. **74**, 1253 (1990).

[8] E. Duval, A. Boukenter, and B. Champagnon, Phys. Rev. Lett. **56**, 2052 (1986).

[9] J. C. Phillips, in *Solid State Physics,* edited by H. Ehrenreich and D. Turnbull (Academic, New York, 1983), Vol. 37.

[10] V. N. Novikov, Pis'ma Zh. Eksp. Teor. Fiz. **51**, 65 (1990) [JETP Lett. **51**, 77 (1990)].

[11] N. Rivier, Philos. Mag. A **40**, 859 (1979).

[12] See also N. Rivier, in *Topological Disorder in Condensed Matter,* edited by F. Yonezawa and T. Ninomiya (Springer-Verlag, Berlin, 1983).

[13] H. Lamb, Proc. London Math. Soc. **13**, 182 (1882); see also A. E. H. Love, *A Treatise on the Mathematic Theory of Elasticity* (Dover, New York, 1944), Chap. VII.

[14] C. G. Granqvist and R. A. Buhrman, J. Appl. Phys. **47**, 2200 (1976).

[15] A. N. Kolmogorov, J. Fluid Mech. **54**, 1 (1989).