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Soft localized vibrations in glasses and undercooled liquids

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ABSTRACT

Many unsolved riddles in the glassy and liquid state of matter seem to be connected with the low-frequency excitations in the millielectron volt range. In glasses, an interpretation of neutron scattering and specific heat data in terms of the soft-potential model (an extension of the tunnelling model) indicates a relatively high number of atoms partaking in a soft vibrational or tunnelling mode. The estimated number of about 100 participating atoms has been confirmed recently by numerical work in a glass of soft spheres. In the undercooled liquid, the number of soft modes seems to increase with increasing temperature. An explanation for their localization is attempted in terms of the balance between their vibrational entropy and the energy needed for their destabilization.

§1. INTRODUCTION

Our understanding of disordered solids is still poor compared with the understanding of crystalline solids. The anomalous behaviour of glasses at low temperatures (Phillips 1981) has never been convincingly explained. Experimentally, one finds both two-level states (Phillips 1981) and soft harmonic vibrations (Buchenau *et al.* 1986) coexisting with the sound waves in glasses. These findings support the soft-potential model (Karpov, Klinger and Ignatiev 1983, Galperin, Karpov and Kozub 1989), an extension of the well known tunnelling model (Anderson, Halperin and Varma 1972, Phillips 1972) for the two-level states in glasses. The soft-potential model describes both the tunneling and the soft vibrational motion in terms of soft anharmonic potentials with locally varying parameters. However, neither the tunnelling model nor its extension gives any justification for the existence of these soft localized modes.

A clue to the origin of the soft localized modes in glasses is given by recent numerical work on liquids and clusters at elevated temperatures (Hahn and Matzke 1984, 1987, Matzke and Hahn 1988, Seeley and Keyes 1989, Adams and Stratt 1990, Beck and Marchioro 1990, Xu and Stratt 1990). These investigations show a fraction of unstable modes with negative eigenvalues and imaginary frequencies in the normal-mode analysis of frozen-in configurations. The results suggest a connection between the anomalous behaviour of glasses and instabilities in the liquid state.

The present paper begins by a quantitative formulation of the concept of a localized soft mode in §2. The localization is expressed in terms of the eigenvector of the soft mode. It is shown that it can be connected to the fourth-order term of the potential in the mode displacement. Section 3 reviews recent experimental and theoretical work on the soft modes in glasses. As will be seen, these soft modes are localized to about 100 participating atoms. This result has been obtained in two different ways. The first is based on a consideration of the cross-over frequency between tunnelling states and soft vibrations. The cross-over frequency was extracted from specific heat data at low temperatures (Buchenau, Galperin, Gurevich and Schober 1991). The second and more

direct way was to calculate the number of participating atoms for the soft modes of a model glass of soft spheres (Laird and Schober 1991).

Section 4 considers the undercooled liquid just above the glass transition temperature T_g , where these soft modes are formed and frozen in. At this temperature, the atomic diffusion is slow enough to define long-time equilibrium positions of the atoms, from which in turn a dynamical matrix and vibrational eigenmodes can be derived. Using strong simplifications, the equilibrium condition is transformed into a stability equation for the soft modes. The section discusses the implications of that stability equation for the soft modes and their localization. The results are compared with those obtained at low temperatures.

The final section § 5 gives a summary of the results of the preceding sections on the soft modes in the glassy and in the liquid state.

§ 2. FUNDAMENTALS

Let us consider the glass or the undercooled liquid as a giant molecule, for which a dynamical matrix may be defined. Then one can, at least in principle, calculate normal modes with eigenvalues and eigenvectors. The localization of a mode can be defined in terms of its eigenvector. We denote the component of the mode k for the atom i in direction α by $e_{i\alpha}^k$. The eigenvector is normalized:

$$\sum_{i=1}^N \sum_{\alpha=1}^3 (e_{i\alpha}^k)^2 = 1. \quad (1)$$

Here N is the number of atoms in the glass or the undercooled liquid. One of the possible ways to define the number N_k of atoms partaking in the mode k is

$$N_k = \left(\sum_{i=1}^N [(e_{i1}^k)^2 + (e_{i2}^k)^2 + (e_{i3}^k)^2]^2 \right)^{-1}. \quad (2)$$

This definition is often used in numerical work (Bell 1976) and will be also used here. Since it contains the fourth powers of the atomic eigenvector components, it can be related to the fourth-order term in the potential. This quartic term is especially important for the stabilization of the soft modes with their very small or even negative harmonic term. Of course, in a solid such a quartic term always exists. Following the convention of the soft-potential model, it is expressed in terms of a length a of the order of the interatomic distance, at which the fourth order of the mode potential becomes as high as the harmonic term for an unsoftened mode at a crystal-like frequency ω_c . Since the effect of the correlation of the atomic motion on the magnitude of the fourth-order term can be absorbed into the value of a , the fourth-order term may be assumed to depend only on single-atom motions:

$$V_4(u_i) = \frac{Ma^2\omega_c^2}{2} \left(\frac{u_i}{a} \right)^4, \quad (3)$$

where M is the atomic mass (for simplicity assumed to be equal for all atoms) and u_i is the amplitude of atom i .

A different way to define the localization of the mode is via the largest atomic displacement in the mode and an effective mass which connects the kinetic energy of the mode to that displacement (Buchenau *et al.* 1991). Although that definition tends to give smaller numbers of participating atoms than that introduced here (Laird and Schober 1991), it is essentially equivalent and can be similarly connected to the fourth-order term of the potential.

Let η_k be the fraction of the crystal-like restoring force for mode k . Then the harmonic term in the mode potential is $\eta_k \omega_c^2 A_k^2/2$, where A_k is the amplitude of the mode k . One can define a typical atomic displacement x_k of the atoms in the mode k by

$$x_k = \frac{A_k}{(N_k M a^2)^{1/2}}. \quad (4)$$

The potential of the mode in this coordinate is

$$V_k(x_k) = \varepsilon_k (\eta_k x_k^2 + x_k^4), \quad (5)$$

with $\varepsilon_k = N_k M a^2 \omega_c^2/2$.

This formulation in terms of an energy ε_k and a dimensionless coordinate x_k corresponds to the usual convention of the soft-potential model (Karpov *et al.* 1983). In general, one would have to add a small first- or third-order term to describe the asymmetry of the potential. These terms are important if one wants to determine the distributions of tunnelling states and soft vibrations in detail but need not be considered here.

The cross-over from soft vibrations to tunnelling states occurs as η_k goes from positive to negative values. Let us assume that we can describe all the modes in the neighbourhood of that cross-over by a common participation number N_s . Let us denote the corresponding value of ε_k by ε_0 . Then

$$\varepsilon_0 = \frac{N_s M a^2 \omega_c^2}{2}. \quad (6)$$

The cross-over frequency between soft vibrations and tunnelling states observed in experiment should correspond roughly to the energy spacing of the lowest levels of the purely quartic potential with $\eta_k = 0$. These spacings are of the order of the energy W with

$$W = \left(\frac{\hbar^2}{2N_s M a^2} \right)^{2/3} \varepsilon_0^{1/3}. \quad (7)$$

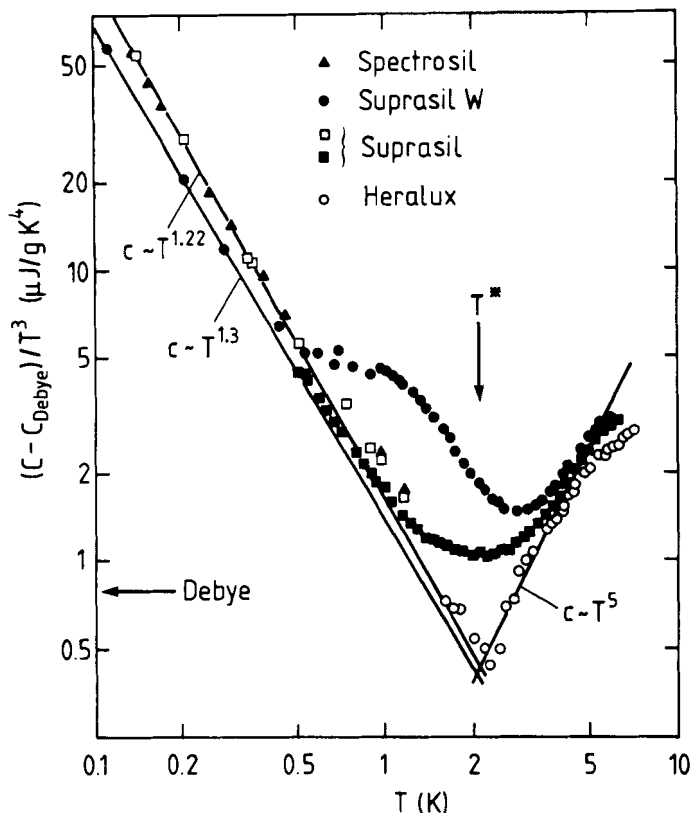
With eqns. (6) and (7) it is possible to calculate $N_s a^2$, if one knows W and ω_c from experimental data. Having $N_s a^2$, one can then estimate N_s , since a should be of the order of the interatomic distances. This will be one of the ways to estimate the localization of the soft modes in glasses described in the next section.

§ 3. LOCALIZATION OF THE SOFT VIBRATIONAL MODES IN GLASSES

This section reviews recent work on the localization of the soft vibrations in glasses. Let us begin with the estimate of the number of partaking atoms from the low-temperature specific heat and from neutron data based on the soft-potential model (Buchenau *et al.* 1991).

The low-temperature specific heat c_p of glasses is dominated by additional excitations which coexist with the sound waves. Figures 1 and 2 show this for two well studied glasses, vitreous silica and amorphous selenium respectively. The data are plotted as c_p/T^3 against T on a double-logarithmic scale. The Debye contribution of the sound waves is subtracted in order to show only the additional excitations. The measurements were done by different workers on different samples and do in fact differ appreciably. Nevertheless, they all show a c_p essentially linear in T at the lowest temperatures. This part is satisfactorily described in terms of the tunnelling model. Also, all these different measurements show a marked increase above 1 K. The

Fig. 1



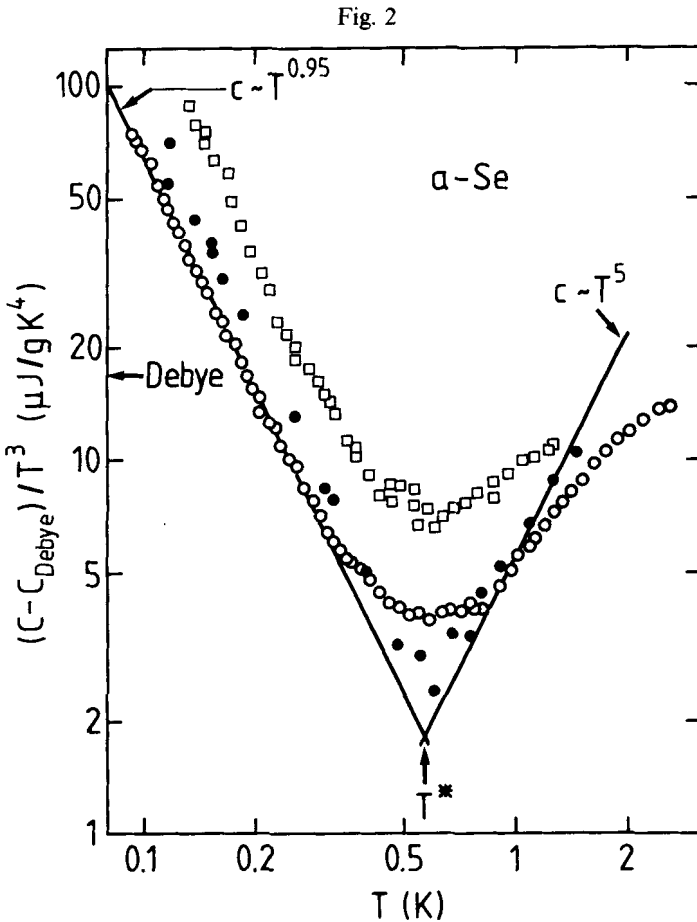
Specific heat c_p of vitreous silica shown as c_p/T^3 against temperature T on a double-logarithmic scale. The Debye contribution is subtracted. (From Buchenau *et al.* (1991).)

temperature T^* at the minimum of c_p/T^3 is a quantitative measure for the cross-over between the tunnelling states and these higher-frequency excitations.

The nature of the higher-frequency excitations has been investigated by spectroscopic methods, in particular by neutron scattering (for a review see Egelstaff (1989) and Buchenau (1989b)). The temperature dependence of the signals shows them to be harmonic vibrations which become anharmonic as one approaches from above the cross-over frequency to the tunnelling states corresponding to T^* . This finding supports the interpretation in terms of the soft-potential model described above. A detailed study gives the quantitative relation

$$W = 1.6k_B T^* \quad (8)$$

Figures 3 and 4 show the vibrational density of states determined from neutron scattering data for vitreous silica (Buchenau *et al.* 1986) and amorphous selenium (Phillips *et al.* 1989) respectively. The data show a broad first maximum of the vibrational density of states at 3 THz for vitreous silica and at 1.3 THz for amorphous selenium. These maxima are only insubstantially lower than the lowest maxima in the density of states of the corresponding crystals and will be identified with the frequency



Specific heat c_p of amorphous selenium shown as c_p/T^3 against temperature T on a double-logarithmic scale. The Debye contribution is subtracted. (From Buchenau *et al.* (1991).)

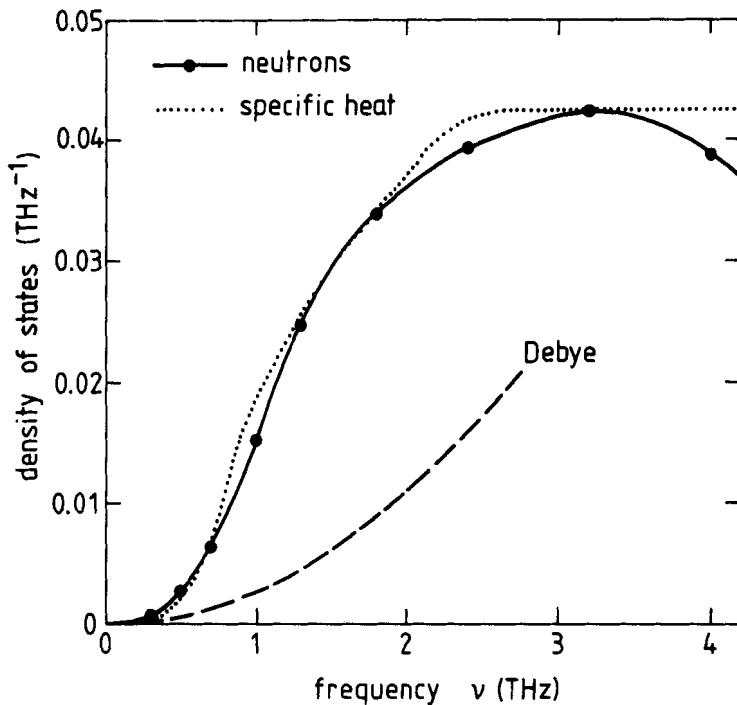
ω_c . They are about two orders of magnitude higher than the cross-over frequency corresponding to the temperatures T^* extracted from the specific heat data at low temperatures.

With the values of W and ω_c obtained in this way, one calculates $N_s a^2 = 162 \text{ \AA}^2$ for vitreous silica and $N_s a^2 = 289 \text{ \AA}^2$ for amorphous selenium. Similar data for vitreous boron trioxide give $N_s a^2 = 392 \text{ \AA}^2$. Since a should be of the order of 1 \AA , one must reckon with an order of magnitude of 100 for the number of N_s of atoms participating in these soft vibrational and tunnelling modes.

For comparison with the results in the undercooled liquid described in § 4, we note that the corresponding values of the energy ϵ_0 are 55, 89 and 45 eV for vitreous silica, amorphous selenium and vitreous boron trioxide respectively.

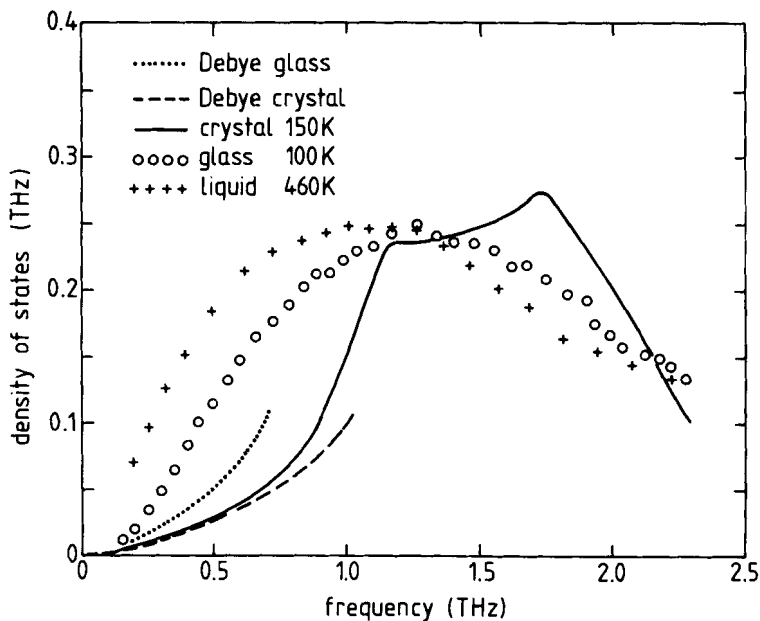
The chain of arguments leading to the result of 100 participating atoms given above is relatively long and involves several assumptions, the most important being the validity of the soft-potential model. Therefore the conclusion would seem all the better for a direct proof. Such a direct proof has been given recently, at least for the soft

Fig. 3

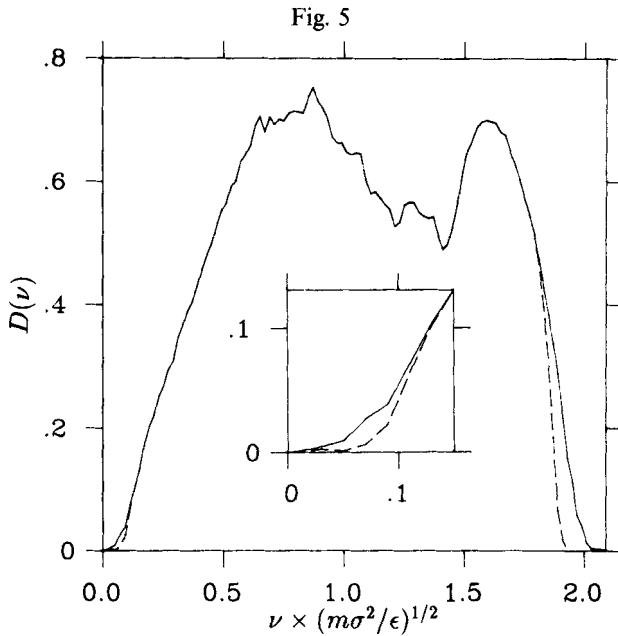


Vibrational density of states of vitreous silica determined from neutron scattering (—●—) and from specific heat data (...). (From Buchenau *et al.* (1986).)

Fig. 4



Vibrational density of states of amorphous and crystalline selenium determined from neutron scattering data at different temperature. (From Phillips *et al.* (1989).)



The vibrational density of states calculated for a model glass of soft spheres for all modes (—) and for only the extended modes (---). The inset shows an enlargement of the low-frequency tail of these curves. (From Laird and Schober (1991).)

vibrations (Laird and Schober 1991). The result was obtained in numerical work in a model glass with a repulsive soft-sphere potential

$$V(r) = \frac{1}{r^6}. \quad (9)$$

Figure 5 shows the vibrational spectrum obtained for this model glass. Most of these modes are extended over the whole cell. At high frequencies, one finds some localized modes as observed already in earlier work (Nagel, Rahman and Grest 1984). What is new, however, is the observation of localized modes at the low-frequency end of the spectrum. Figure 6 shows the participation ratio $p = N_k/N$ as a function of the frequency. The low-frequency modes can be seen to be localized to about 100 participating atoms. The localization is restricted to the lowest portion of the spectrum.

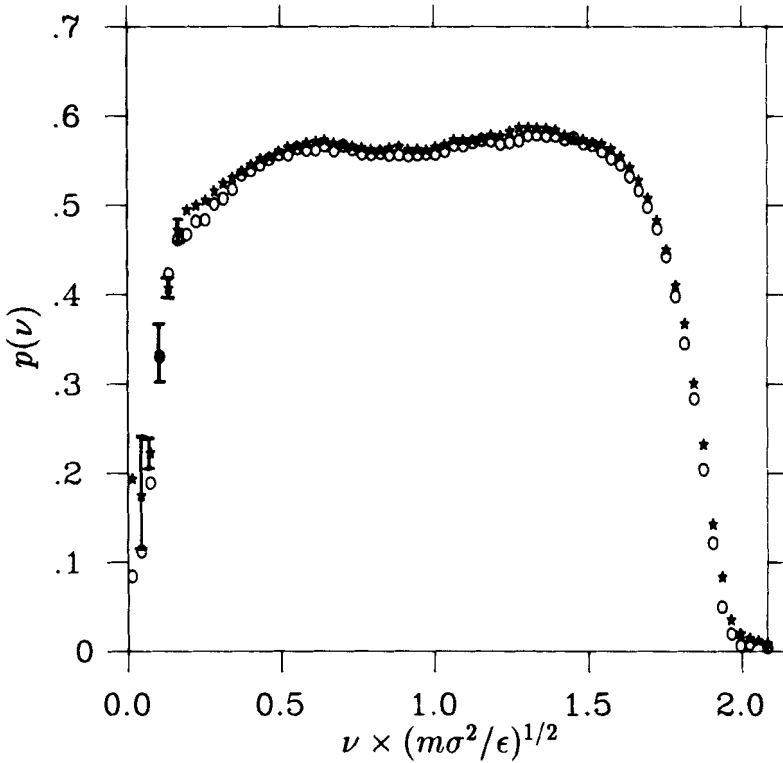
The finding is independent proof for a localization of the soft modes in glasses. It supports the interpretation of the specific heat and neutron data in terms of the soft-potential model described above which reaches the same conclusion.

§4. SOFT MODES IN THE UNDERCOOLED LIQUID

The present section intends to address the question of the thermodynamic stability of a soft mode in the undercooled liquid. Let us consider the undercooled liquid just above the calorimetric glass transition temperature T_g , where the time scale of the flow processes is many orders of magnitude longer than the vibrational time scale, even for a soft mode. Then one can define the long-time equilibrium positions of the N atoms by

$$\frac{\delta U}{\delta x_{ia}} + \sum_{k=1}^{3N} \frac{\delta F_k}{\delta x_{ia}} = 0, \quad (10)$$

Fig. 6



The participation ratio $p(\nu) = N_k/N$ as a function of frequency for a soft-sphere model glass with $N = 500$ (\star) and $N = 1000$ (\circ). (From Laird and Schober (1991).)

where $x_{i\alpha}$ is the displacement of atom i in direction α and U is the potential energy of the interacting atoms. The configurational free energy F_k of the vibrational eigenmode k is given by $F_k = -k_B T \ln Z_k$ with

$$Z_k = \int_{-\infty}^{\infty} dx_k \exp[-\beta V(x_k)], \quad (11)$$

where $\beta = 1/k_B T$ and $V(x_k)$ is the potential energy as a function of the normal mode displacement x_k as defined in § 2. Let us again denote the harmonic part of this potential by $\eta_k \epsilon_k x_k^2$ as in § 2. Neglecting the weak influence of the displacement on the fourth-order term, one gets

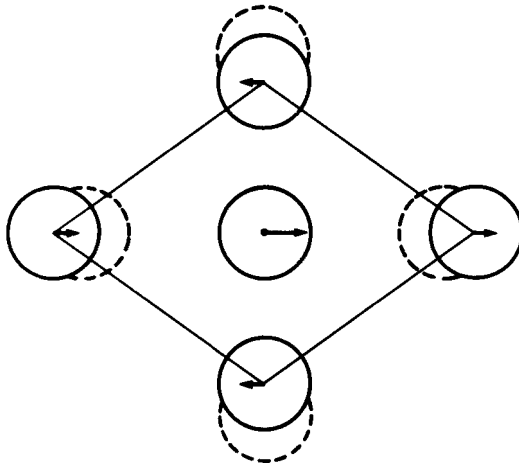
$$\frac{\delta F_k}{\delta x_{i\alpha}} = \frac{\delta F_k}{\delta \eta_k} \frac{\delta \eta_k}{\delta x_{i\alpha}} = \epsilon_k \langle x_k^2 \rangle \frac{\delta \eta_k}{\delta x_{i\alpha}}. \quad (12)$$

This entropic contribution to the force on a given atom would diverge at $\eta_k = 0$ for a purely harmonic mode potential. If one takes into account the quartic term (which in real solids always exists), it no longer diverges. Nevertheless, it still remains the dominating contribution to the entropic force. Its strength depends on the size of the quartic term. The consideration shows that the soft modes need a treatment which differs from that of the unsoftened crystal-like modes, for which the quartic term in the single-mode potential plays no important role.

If the internal strain energy is indeed balanced by the vibrational entropy, one will expect a marked increase in the number of soft modes in the liquid with increasing temperature. This must lead to a rapid increase in the mean square displacement of the vibrational motion above the glass transition. Such an increase, clearly distinguishable from the slow atomic diffusion through the liquid by its shorter time scale, has been in fact observed by neutron scattering in several different substances (Galli, Migliardo, Bellissent and Reichardt 1986, Fujara and Petry 1987, Frick, Richter, Petry and Buchenau 1988, Knaak, Mezei and Farago 1988, Phillips *et al.* 1989). This explanation, however, would have to compete with mode-coupling interpretations of the same phenomenon (Frick, Farago and Richter 1990). The mode-coupling scheme describes the glass transition in terms of dynamic freezing of the liquid (Bengtzelius, Götze and Sjölander 1984, Leutheusser 1984, Götze 1991). Its connection to the solid-like approach considered here is at present not clear.

Let us now turn to the practical question: how does one get a soft mode in a glass spending a minimum amount of potential energy? The destabilization of a stable structure by distortion requires potential energy. In general, that energy will grow with the spatial extent of the instability. This restricts the minimum-energy solution to the close neighbourhood of a single atom. On the other hand, allowing the neighbours to give way makes the destabilization easier. Thus the cheapest solution in energy will have a large amplitude on a central atom and smaller amplitudes on the neighbours. Its eigenvector will resemble those at the lowest maximum of the vibrational density of states of the corresponding crystal (transverse zone boundary modes in simple monatomic solids, tetrahedra rocking modes in quartz or octahedra rocking in perovskites, bond torsional motion in polymers, etc.) Thus the frequency scale ω_c defined above should indeed be chosen as the frequency of this lowest maximum in the crystalline density of states as done in § 3. The distortion needed to destabilize that mode locally is composed of extensions in the direction of the vibration of the central atom and compressions in the perpendicular directions, as shown schematically in fig. 7. For simple monatomic solids, the minimum energy ϵ_m required to form such a local instability has been estimated (Buchenau 1989a) to be of the order of a few times $k_B T_m$, where T_m is the melting temperature.

Fig. 7



Softening mechanism for the local soft mode (see text).

Let us consider such a minimum-energy soft mode. Clearly, that local soft mode need not be an eigenmode itself but can be described by a superposition of eigenmodes. We denote the amplitude of the soft mode by A_s and its (normalized) motional vector by \mathbf{s} . Then

$$\sum_{k=1}^{3N} (\mathbf{s} \cdot \mathbf{e}_k)^2 = 1. \quad (13)$$

Again, we define a fraction η_s to describe the harmonic term $\eta_s \omega_c^2 A_s^2 / 2$ of the local soft mode. From the decomposition of the local soft mode into eigenmodes it follows that

$$\eta_s = \sum_{k=1}^{3N} (\mathbf{s} \cdot \mathbf{e}_k)^2 \eta_k. \quad (14)$$

In order to be able to proceed, one has to make two simplifying assumptions. The first is to assume that the local distortion energy U_s depends only on η_s :

$$U_s = \varepsilon_m (1 - \eta_s)^2. \quad (15)$$

The eigenvector \mathbf{e} of mode k can be decomposed into a part $(\mathbf{s} \cdot \mathbf{e}_k)$ moving in phase with the eigenvector \mathbf{s} of the local soft mode and a rest which is orthogonal to it. The second simplifying assumption is to neglect the influence of the local distortion on that rest:

$$\frac{\delta \eta_k}{\delta \eta_s} = (\mathbf{s} \cdot \mathbf{e}_k)^2. \quad (16)$$

The approximation will be better the closer the motion of the mode k follows the soft-mode eigenvector locally.

Rephrasing eqn. (10) in terms of the variables η_s yields with eqns. (13)–(16)

$$\sum_{k=1}^{3N} \left(\omega_c^2 \langle A_k^2 \rangle - 2\varepsilon_m (1 - \eta_k) \right) (\mathbf{s} \cdot \mathbf{e}_k)^2 = 0. \quad (17)$$

Note that this stability equation for a local soft spot near $\eta_s = 0$ describes a metastable situation. The minimum of the local free energy lies around $\eta_s = 1$. Thus the other extremum near $\eta_s = 0$ must be maximum of the free energy. However, since only the local part of the free energy was considered, such a soft spot can be easily stabilized by the surroundings. The stabilization does not require any forces, because the soft spot is locally metastable. Thus the undercooled liquid would be stress free, although strongly strained at the soft spots. In this interpretation, the stresses found at low temperatures in glasses (Srolovitz, Vitek and Egami 1983) arise with the cooling because the balancing entropic forces disappear.

If eqn. (17) is valid for all soft spots,

$$\frac{\delta F_k}{\delta \eta_k} = \frac{\omega_c^2 \langle A_k^2 \rangle}{2} = 2\varepsilon_m (1 - \eta_k) \quad (18)$$

for each soft mode. This is the thermodynamical stability relation for the soft modes, which will be evaluated in the remainder of this section.

In the limiting case of a dominating harmonic term, the solution of the mode stability eqn. (18) is simple. In that case, ω_c^2 drops out and one obtains a quadratic equation for η_k with solutions

$$\eta_{1,2} = \frac{1}{2} (1 - 1/\beta \varepsilon_m)^{1/2}. \quad (19)$$

Obviously the scheme is only applicable if $k_B T < \varepsilon_m$. Then, one can have either a weakly softened mode at η_1 or a strongly softened mode at η_2 . The result is a simple parallel of more elaborate considerations on dynamic stability in the self-consistent harmonic theory, which have been taken to indicate a first-order phase transition at the stability limit (Choquard 1967).

With the definitions of § 2 the mode stability equation reads

$$\varepsilon_k \langle x_k^2 \rangle = 2\varepsilon_m(1 - \eta_k). \quad (20)$$

Note that ε_k scales with the number N_k of atoms participating in the mode. As long as the harmonic term dominates, this number cancels. This is no longer true for $\eta_k < \eta_2$. For instance in the simple case $\eta_k = 0$, only the quartic part of the potential remains. Then

$$\langle x_k^2 \rangle = \int_{-\infty}^{\infty} \frac{dx_k x_k^2 \exp(-\beta\varepsilon_k x_k^4)}{Z_k}. \quad (21)$$

This integral and the one in Z_k can be evaluated using the formula

$$\int_0^{\infty} dt \exp(-wt^n) t^{z-1} = \frac{\Gamma(z/n)}{nw^{z/n}}, \quad (22)$$

leading to

$$\beta\varepsilon_0 = 4 \left(\frac{\Gamma(\frac{1}{4})}{\Gamma(\frac{3}{4})} \right)^2 (\beta\varepsilon_m)^2 = 35.015(\beta\varepsilon_m)^2. \quad (23)$$

Here ε_0 denotes the value of ε_k at $\eta_k = 0$ as defined in § 2.

As noted above, $\beta\varepsilon_m$ is estimated to be about 3 at the melting temperature. Since the glass temperature T_g is usually found at about $0.6T_m$, $\beta\varepsilon_m$ should be around 5 at T_g , where the modes freeze in and are no longer free to change their degree of localization with changing temperature. Subsequent cooling from T_g to low temperatures should leave ε_0 essentially unchanged. Therefore at low temperatures an ε_0 of about three orders of magnitude higher than $k_B T_g$ is expected, that is of 50–100 eV, corresponding to about 100 atoms participating in the mode, as one indeed finds (see § 3). The quantity $\varepsilon_m/k_B T_g$ determined from the values of § 3 using eqn. (23) is 3.5 for silica, 10 for selenium and 5.3 for B_2O_3 , again of the expected order of magnitude.

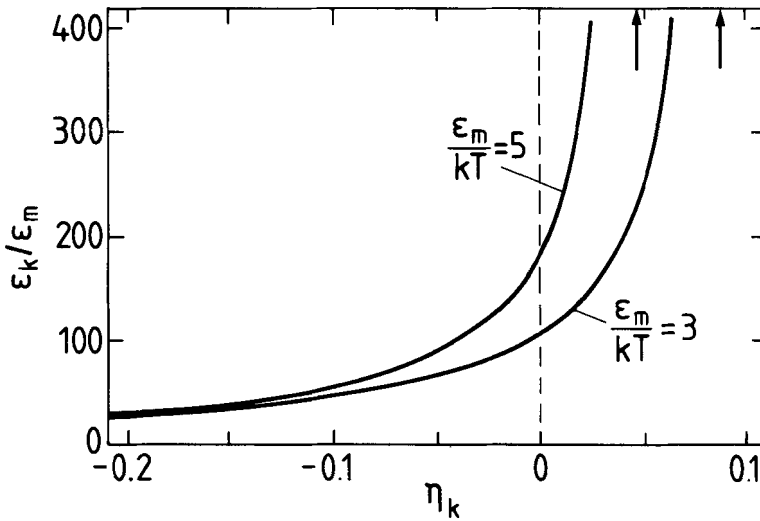
For the numerical treatment of eqn. (20) in the general case, one sets $u = (\beta\varepsilon_k)^{1/4} x_k$ and $\eta' = (\beta\varepsilon_k)^{1/2} \eta_k$. Then the mode stability eqn. (20) takes the form

$$(\beta\varepsilon_k)^{1/2} = \frac{\beta\varepsilon_m}{\langle u^2 \rangle} \pm \left[\left(\frac{\beta\varepsilon_m}{\langle u^2 \rangle} \right)^2 - \frac{2\beta\varepsilon_m \eta'}{\langle u^2 \rangle} \right]^{1/2}. \quad (24)$$

The negative sign corresponds to localized solutions above η_1 and the positive sign corresponds to localized solutions below η_2 . This equation can be used to calculate ε_k for a given η' and a given temperature. Since in the case of a dominating harmonic term $\langle u^2 \rangle = 1/2\eta'$, it is easy to see that with increasing η' the value of η_k approaches η_2 as ε_k goes to infinity. The general solution of eqn. (20) calculated in this way is shown in fig. 8 for two different temperatures which according to our estimates should be close to the melting and glass transition temperatures respectively. The number of atoms is seen to increase with the increasing harmonic term until it diverges at η_2 .

Why do we get such a close connection between anharmonicity and localization? The answer is simple: extended modes will never be able to develop a large displacement, because the small thermal energy is shared by many atoms. Thus the

Fig. 8



Dependence of the energy ε_k (a measure for the number of atoms participating in the mode) in units of the destabilization energy ε_m on the harmonic restoring force constant η_k . Solutions calculated for the cases $\varepsilon_m/k_B T = 3$ (near T_m) and 5 (near T_g). The arrows mark the η_2 values at which the two curves diverge respectively.

mode experiences the anharmonicity of the potential only via the combined action of all the other modes, a situation which is well known from the treatment of anharmonic effects in crystals (Choquard 1967). This is different for a localized mode, which samples the anharmonicity of the single-mode potential to an extent which becomes more pronounced as the mode becomes more localized. For this reason, only the product $N_k a^2$, which combines the number of participating atoms with the anharmonicity, is found in the equations.

§5. SUMMARY

In this paper, we have first discussed the tunnelling and soft vibrational modes in glasses. They could be shown to be localized to about 100 participating atoms in two different ways. The first was an interpretation of low-temperature specific heat and neutron scattering data in terms of the soft-potential model, which assumes a common origin of tunnelling and soft vibrations. In this model, a soft mode could be either a tunnelling or a soft vibrational mode, depending on small local variations in the parameters of the mode potential. For a small negative harmonic term, one gets a tunnelling mode and, for a small positive harmonic term, one gets a soft vibration. The model connects the average number of atoms participating in a soft mode to the change in the temperature dependence of the excess specific heat around 1 K from a linear to a more strongly rising behaviour.

The second and more direct way to determine the number of participating atoms was a numerical calculation in a glass of soft spheres (Laird and Schober 1991).

The good general agreement of the numbers obtained in these two independent ways supports the hypothesis of a common origin of tunnelling states and soft vibrations as postulated by the soft-potential model. Thus 20 years after the discovery of the low-temperature anomalies of glasses we begin to formulate a microscopic description of these modes.

On the basis of these findings, a first attempt was undertaken to understand the formation of the soft modes in the undercooled liquid. A long-time stability condition has been formulated by including a vibrational entropy term into the free energy of a quasi-stationary atomic configuration of a highly viscous liquid. In a strongly simplified picture, the stability condition could be transformed into a stability equation for the soft modes. This equation links the number of participating atoms to the small positive or negative harmonic term in the mode potential. The parameter of this model is the formation energy ε_m for a local instability. Assuming values of a few times $k_B T_g$ for this formation energy, one again gets about 100 atoms participating in a single soft mode.

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