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Abstract. The coupling of atomic tunneling states with vibration modes in amorphous materials is considered on the basis of dangling-bond and void-like structure model proposed by the authors. The theoretical investigation shows the atomic tunneling state has a strong influence on its nearby vibration modes. As a result, this mode will either be softened or become unstable causing a new tunneling mode. A hierarchy of constrained double-well potentials is then obtained. The recurrence formulae of the hierarchy are derived; the termination emerging naturally. The possible relation between the hierarchical constrained dynamics and the 'anomalous' relaxation in condensed matter is discussed.

1. Introduction

The anomalous thermal and acoustic properties of amorphous materials at low temperatures still remain obscure to theoretical researchers. Recent explanations of these properties are based on a phenomenological model of two-level systems (TLS) put forward by Phillips (1972) and Anderson et al (1972); it is assumed that in amorphous solids some localised low-energy elementary excitations exist, which result from the atomic tunneling states. However, the relationship between the double-well potential (DWP) in which the tunneling process occurs and local structural mechanisms is unclear. Several microscopic models have been proposed so far, e.g. the free-volume model of Cohen and Grest (1980), the layer-like model of Phillips (1981), the critical-potential model of Klinger et al (1982), the non-ideal Frenkel–Kontorova model of Geszti (1986) and the chaotic-configuration model of Schilling (1984), but they can only explain part of the anomalous low-temperature properties. In fact, numerous experimental findings (Phillips 1985) indicate that a strong correlation exists between the anomalous low-temperature properties.

The present authors have proposed a dangling-bond and void-like structure (DVS) model (Wu and Chen 1986) which attributes the microscopic mechanism of the appearance of the DWP to two structural characteristics of amorphous materials: dangling-bonds and void-like structural defects. By applying the path integral theory (Chen et al 1986), the analytic form of the effective potential which the atom in the void experiences is obtained, and simultaneously the critical condition when the DWP arises is given and the

behaviours of the DWPs and TLSs in the vicinity of the critical point are also investigated. However, in our previous discussions, the atom in the void can only move in an adiabatic effective potential and the coupling of the atomic tunneling states with its environment has not been considered directly. In order to fit the model to the real circumstances in amorphous materials, in this paper the coupling effect is led directly into the Hamiltonian. Therefore, two problems ought to be considered: one is the influence of local vibration modes on the atomic tunneling process in the void, the other is the influence of the atomic tunneling states on the behaviours of surrounding atoms. The former has been discussed indirectly by the authors (Chen and Wu 1986), the latter will be discussed in this paper.

We start from a void in a one-dimensional chaotic chain of atoms. Using the analytic form of the effective potential solved by the DVS model and moreover adopting a special data fitting method, we find the following results.

(i) There exists a set of low-frequency vibration modes associated with a DWP, and this set of modes causes the fluctuations of the shape of the DWP. This result is in agreement with the findings of Buchenau *et al* (1984, 1986) from their neutron scattering measurements.

(ii) The existence of the DWP in which the atom in the void moves induces a hierarchical, constrained sequence of DWPs. This result not only provides plentiful low-energy elementary excitations for explaining the anomalous low-temperature properties of amorphous materials but also introduces a microscopic model and constraining mechanisms to the principles outlined by Palmer *et al* (1984) for interpreting the 'anomalous' relaxation in glasses.

(iii) The effective range of the coupling, i.e. the termination of the hierarchical constrained DWPs, emerges naturally from the DVS model. This result accords with the demands suggested by Fleurov (1988) for the hierarchical scale of DWPs. In addition, the termination also meets the requirements of the fracton model proposed by Alexander *et al* (1983).

Recently, two types of DWPs—soft and rigid—were investigated under a unified approach by Fleurov *et al* (1985) and Fleurov and Trakhtenberg (1986). Soft DWPs are mainly responsible for the anomalous low-temperature properties, whereas rigid ones determine the ultrasonic attenuation and other kinetic properties of amorphous materials at higher temperatures. In this paper, both types arise naturally and their characteristics can be discussed analytically. The numerical result agrees well with the one obtained by Fleurov *et al* (1985) using the quantum chemical computation method; moreover, our result can describe the behaviours in the vicinity of the critical point consistently.

The above introduction shows that DVS model can provide a unified foundation for the theoretical explanation of the anomalous thermal, ultrasonic, optical and electronic (Wu and Chen 1986) properties in amorphous materials, and may make a connection between the models proposed to explain the anomalous low-temperature thermal properties and the anomalous Debye relaxation. In § 2, we start from the Hamiltonian considering the influence of neighbouring atoms to get the effective potential which can be easily treated theoretically. In § 3 the appearance of a low-frequency vibration mode and a DWP of higher rank is analysed on the basis of the DVS model. The recurrence formulae and the termination are derived in § 4. A discussion and concluding remarks are given in § 5.
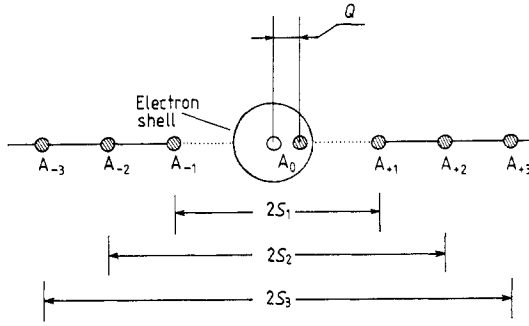
Figure 1. Schematic of a one-dimensional DVS model. Atom A_0 is located in a void and is displaced from its centre. S_1 is the radius of the void, $2S_2$ is the distance of randomly located second-nearest-neighbour atoms and $2S_3$ is that of third-nearest-neighbour atoms.

2. Solution of the effective potential

A one-dimensional DVS model is shown in figure 1. Inside a void, atom A_0 interacts with two dangling-bonds, left and right. S_1 is the radius of the void (half of the distance from A_{-1} to A_{+1}). $2S_2$ is the distance of randomly located neighbouring atoms beyond the void (A_{-2} to A_{+2}). $2S_3$ is the distance beyond the above ones (A_{-3} to A_{+3}), and so on.

The Hamiltonian of the DVS model we use is

$$H_{\mathrm{DVS}} = \sum_{i,\sigma} \varepsilon_i N_{i\sigma} + \varepsilon_0 \sum_d N_{0d} + [\tfrac{1}{2}m\dot{Q}^2 + \alpha Q + m\Omega^2 Q^2 + m\Omega^2(S_1 - S_0)^2]$$

$$+ \sum_{i,\sigma,d} (V_{0d,i\sigma} a_{0d}^+ a_{i\sigma} + \mathrm{HC}) - M(a_{0s}^+ a_{0p} + a_{0p}^+ a_{0s})Q. \qquad (1)$$

The first term of the Hamiltonian expresses the electronic energy of dangling-bonds, $i = L, R$ corresponding to two dangling-bonds, $\sigma = \uparrow, \downarrow$ to spins and $N_{i\sigma} = a_{i\sigma}^+ a_{i\sigma}$, where $a_{i\sigma}^+, a_{i\sigma}$ are the electronic creation and annihilation operators of the dangling-bonds respectively. The second term expresses the electronic energy of the electron shell of atom A_0; we assume that the electronic states of the shell can be divided into two parts, symmetric ($d = s$) and antisymmetric ($d = p$), and $N_{0d} = a_{0d}^+ a_{0d}$, where a_{0d}^+, a_{0d} are the electronic creation and annihilation operators of the shell. The third term is the vibration energy of the ion which has considered the influence of its neighbouring atoms, A_{-1} and A_{+1}. The fourth and fifth terms are the coupling terms of the electron shell of atom A_0 to the electrons of the dangling-bonds and to its ion.

By using the treatment of the Hamann (1970) path integral theory and integrating all electronic degrees of freedom in (1), we can obtain the adiabatic effective potential of atom A_0. Except for the interaction term of atom A_0 with its neighbouring atoms, (1) is completely similar to the corresponding Hamiltonian we used previously (Chen *et al* 1987); nevertheless, the tiny difference has no influence on integrating over all electronic degrees of freedom. In the same treatment as before, the total partition function of DVS model can be written as
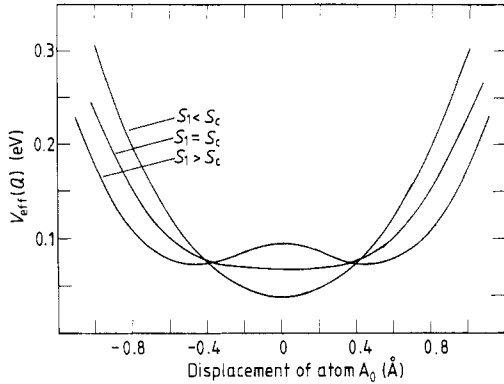
$$Z_{\mathrm{DVS}} = Z_{\mathrm{el}} Z_{\mathrm{vib}} \qquad (2)$$

where Z_{el} is the partition function of electrons, Z_{vib} is the vibrational partition function of atom A_0. So
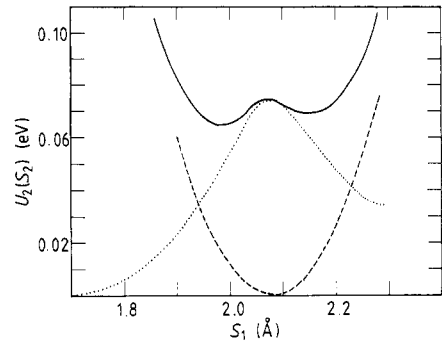
$$Z_{\mathrm{vib}} = \exp\left\{-\frac{1}{\hbar}\int_0^{\beta\hbar} \mathrm{d}\tau\, [T_{\mathrm{eff}}(\tau) + V_{\mathrm{eff}}(\tau)]\right\} \qquad (3)$$

where $\beta = 1/k_B T$, and T_{eff} and V_{eff} are the effective kinetic energy and the effective potential, respectively. Therefore we get

$$V_{\mathrm{eff}}(Q, S_1) = \alpha Q + m\Omega^2 Q^2 + m\Omega^2(S_1 - S_0)^2$$
$$+ (2MQ/\pi)\tan^{-1}(-MQ/\Sigma) + (\Sigma/\pi)\ln(1 + M^2Q^2/\Sigma^2) \qquad (4)$$

Figure 2. How the shape of the effective potential $V_{eff}(Q)$ of g-SiO$_2$ changes with respect to S_1, showing the transition from a single-well potential to a double-well potential.

Figure 3. Effective void potential for g-SiO$_2$ versus void radius S_1 (after Liu *et al* (1988)). Dotted line, $V_{eff}(S_1, Q_{min})$; broken line, interaction energy of the atom in the void with its neighbouring atoms; full curve, the sum of the two lines, being the effective potential which the vibrational mode of the void meets, $U_2(S_2)$.

where Σ is the resonant width of the electron shell. The equilibrium position of atom A_0 is determined by

$$\partial V_{eff}(S_1, Q)/\partial Q|_{Q=Q_{min}} = \alpha + 2m\Omega^2 Q_{min} - (2M/\pi) \tan^{-1}(-MQ_{min}/\Sigma) = 0. \tag{5}$$

For small asymmetry ($\alpha \simeq 0$), we have

$$m\Omega^2 Q_{min} = (M/\pi) \tan^{-1}(-MQ_{min}/\Sigma). \tag{6}$$

The above equation tells us that the parameter

$$y = M^2/\pi m\Omega^2 \Sigma \tag{7}$$

determines the shape of V_{eff}. For $y \leqslant y_0 = 1$, it is a single-well potential while, for $y > y_0$, it is a DWP. The resonant width Σ is a function of the radius of the void S_1, and in general can be expressed as (Chen *et al* 1987)

$$\Sigma = \Sigma_c(S_c/S_1)^{2k} \tag{8}$$

where S_c is the critical radius, satisfying $y(S_1 = S_c) = y_0 = 1$. In other words, for $S_1 \leqslant S_c$, V_{eff} is a single-well potential, while for $S_1 > S_c$, it is a DWP. In figure 2, the change of the effective potential from a single-well form to a double-well one with increasing S_1 is demonstrated. The calculation is for g-SiO$_2$. The parameters according to our previous paper (Liu *et al* 1988) are: $m = 2.66 \times 10^{-26}$ kg, $\Omega = 3 \times 10^{12}$ Hz (which is the dominant frequency from Buchanau *et al* (1984)), $\Sigma_c = 0.5$ eV which is the order of magnitude of the electrons band width in glasses, $S_0 = 1.7$ Å from Fleurov *et al* (1985), and $k = 6$ for the Lennard–Jones potential considered.

From $V_{eff}(Q, S_1)$, the potential of the cluster (A_{-1}, A_0, A_{+1}) i.e. the effective potential of the void, as a function of its radius S_1 can also be obtained. We assume that atom A_0 is always in its equilibrium position located so as to minimise the total potential. This is demonstrated in figure 3 by the dotted curve. The numerical result is for g–SiO$_2$ and agrees with the ones obtained by Fleurov *et al* (1985). The curve $V_{eff}(S_1, Q_{min})$ corresponds to a harmonic potential for small deviation of S_1 from its crystalline value S_0, then becomes anharmonised and passes over a maximum in the critical region.
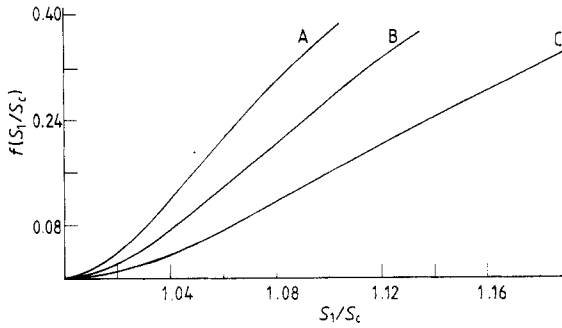
Figure 4. The function $f(S_1/S_c)$: curve A, $k = 8$; curve B, $k = 6$; curve C, $k = 4$. We find that it can be divided into three parts. The data is for g-SiO$_2$.

This property is very important for the following discussions of the appearance of the hierarchical, constrained DWPs. So it is necessary to find an easily tractable form of $V_{eff}(S_1, Q_{min})$.

Combining (7) and (8) we get

$$M^2 = \pi m^2 \Omega^2 \Sigma_c. \tag{9}$$

Substituting (8) into (6), we see that for a certain s_1, the equilibrium position of atom A_0 can be determined. But (6) is a transcendental equation which cannot be solved analytically, and what is more, expansion cannot give a reliable result for large deviations from the point about which the expansion is made. For this reason, a special data fitting method is applied. First of all, two dimensionless parameters are introduced

$$Q' = \sqrt{\pi m \Omega^2 / \Sigma_c} \, Q_{min} \qquad C = (S_c/S_1)^{2k}. \tag{10}$$

Thus (6) can be rewritten as

$$Q' = \tan^{-1}(Q'/C). \tag{11}$$

Using (9) and (11), $V_{eff}(S_1, Q_{min})$ can be expressed as

$$V_{eff}(S_1, Q_{min}) = -\Sigma_c f(c) + m\Omega^2(S_1 - S_0)^2 \tag{12}$$

where

$$f(c) = Q'/\pi - (c/\pi) \ln(1 + Q'^2/C^2). \tag{13}$$

We find that (13) is only a pure mathematical transcendental function and the functional relation between C and f has nothing to do with any particular material parameters, where Q' is determined by (11) for a certain value of C. Furthermore, as $C = (S_c/S_1)^{2k}$, apart from the parameter k, the functional relation between S_1/S_c and f is also independent of material parameters. In figure 4, a series of the curves $f_k(S_1/S_c)$ are demonstrated for different values of k ($k = 4, 6, 8$). By analysing these curves, we find that though for different materials k takes different values, the shape of the curves on the whole remaining the same. Thus the curves can be divided into three parts: a straight line lying on the axis of abscissa from S_0/S_c to 1, then a quadratic parabola with its vertex on (1,0) followed by a straight line in contact with it, which can be expressed as

$$
\begin{aligned}
f(S_1/S_c) &= 0 & S_0/S_c &\leq S_1/S_c < 1 \\
f(S_1/S_c) &= d(S_1/S_c - 1)^2 & 1 &\leq S_1/S_c < \lambda \\
f(S_1/S_c) &= -a + bS_1/S_c & S_1/S_c &\geq \lambda
\end{aligned}
\tag{14}
$$
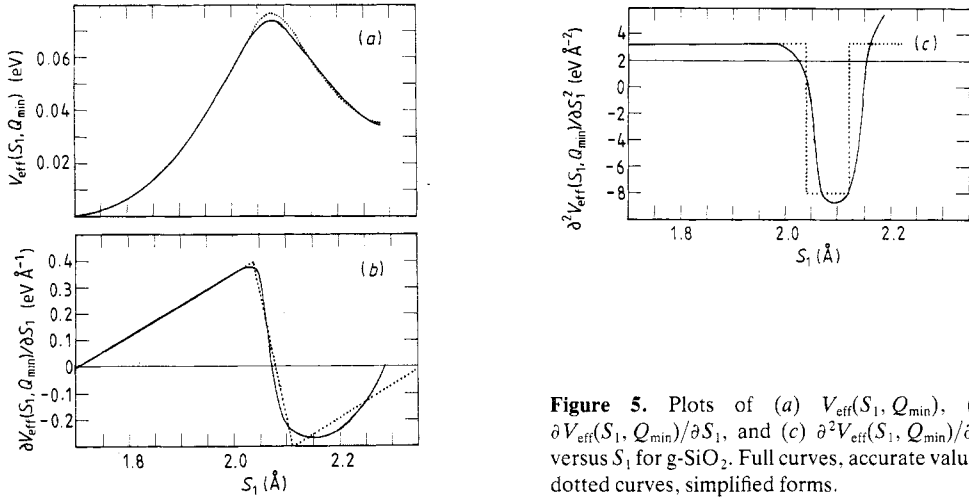
Figure 5. Plots of (a) $V_{eff}(S_1, Q_{min})$, (b) $\partial V_{eff}(S_1, Q_{min})/\partial S_1$, and (c) $\partial^2 V_{eff}(S_1, Q_{min})/\partial S^2$ versus S_1 for g-SiO$_2$. Full curves, accurate values; dotted curves, simplified forms.

where a, b, d, λ are dimensionless parameters depending on the values of k, e.g. for Lennard-Jones potential $k = 6$, we get $a = 13.6$, $b = 13.2$, $d = 45.8$, $\lambda = 1.04$.

By introducing

$$\sigma(\chi) = \begin{cases} 1 & \text{for} \quad \chi \leqslant 0 \\ 0 & \text{for} \quad \chi > 0 \end{cases} \tag{15}$$

$V_{eff}(S_1, Q_{min})$ can be expressed approximately as

$$V_{eff}(S_1, Q_{min}) = m\Omega^2(S_1 - S_0)^2 - \Sigma_c\, d(S_1/S_c - 1)^2\sigma[(S_1/S_c - 1)(S_1/S_c - \lambda)]$$

$$+ \Sigma_c(a - b\, S_1/S_c)\, \sigma[\lambda - S_1/S_c]. \tag{16}$$

From the above consideration, the effective potential of the void as a function of its radius has been simplified to a semi-analytic form which can be easily treated in the following discussions. In figure 5, the full curves represent the accurate values of $V_{eff}(S_1, Q_{min})$, its derivative and its second derivative for g–SiO$_2$, while the dotted curves are their simplified forms. We see that this kind of simplification is acceptable.

3. The appearance of low-frequency vibration modes and the instability of critical DWP

According to the result of the above considerations, the shape of the effective potential of atom A_0 depends on the value of S_1. As the neighbouring atoms A_{-1} and A_{+1} vibrate in the vicinity of their equilibrium positions, the shape of V_{eff} will change considerably. This indicates that the tunneling process and the vibration of neighbouring atoms are closely related and have interaction with one another. In our previous paper (Chen and Wu 1986), the temperature related TLS was introduced by considering the influence of lattice vibrations on the atomic tunneling process. In this section the influence of the atomic tunneling states on local vibration modes will be discussed.

In order to deal with this problem, a more extended cluster of atoms is considered, containing the neighbouring atoms beyond the void (A_{-2} and A_{+2}). In this cluster the

atoms of the dangling-bonds (A_{-1} and A_{+1}) are not fixed but free to oscillate. As was shown in figure 1, we have three variables to describe the cluster, S_2, S_1 and Q, one of which (keeping S_2 fixed) simulates a static disorder. The effective potential of the whole cluster can be expressed as

$$U_2(S_2, S_1, Q) = V_{\text{eff}}(S_1, Q) + V(l_{-2} - l_{-1}) + V(l_{+2} - l_{+1}) \tag{17}$$

where l_i represents the deviation of atom A_i from its equilibrium position, while $V(l_{-2} - l_{-1})$ and $V(l_{+2} - l_{+1})$ represent the interaction energy between two atoms. In the harmonic approximation

$$V(r) = \tfrac{1}{2}\varepsilon r^2 \tag{18}$$

where ε corresponds to the elastic coefficient. In the symmetric approximation

$$|l_{+2} - l_{+1}| = |l_{-2} - l_{-1}| = |S_2 - S_1 - S_0|. \tag{19}$$

As explained in § 2, considering the influence of the outside atoms, the atom in the void is always in its equilibrium position, so

$$U_2(S_2, S_1) = U_2(S_2, S_1, Q_{\min}) = V_{\text{eff}}(S_1, Q_{\min}) + \varepsilon(S_2 - S_1 - S_0)^2. \tag{20}$$

For a fixed S_2, the equilbrium position of S_1 is determined by

$$\partial U_2(S_2, S_1)/\partial S_1 = \partial V_{\text{eff}}(S_1, Q_{\min})/\partial S_1 - 2\varepsilon(S_2 - S_1 - S_0) = 0. \tag{21}$$

Solving equation (21) and at the same time considering that

$$\partial^2 U_2(S_2, S_1)/\partial S_1^2 = \partial^2 V_{\text{eff}}(S_1, Q_{\min})/\partial S_1^2 + 2\varepsilon \tag{22}$$

we obtain the results below. If the condition

$$S_2 \leqslant S_{2c_1} = (1 + m\Omega^2/\varepsilon)[(2d + b)/2d]\,S_c + (1 - m\Omega^2/\varepsilon)S_0 - b\Sigma_c/2\varepsilon S_c \tag{23}$$

holds, the equation (21) has one solution corresponding to a minimum of the potential (20), namely

$$S_{1S} = [(m\Omega^2 - \varepsilon)S_0 + \varepsilon S_2]/(\varepsilon + m\Omega^2). \tag{24}$$

If the condition

$$S_2 \geqslant S_{2c_2} = (1 + m\Omega^2/\varepsilon)S_c + (1 - m\Omega^2/\varepsilon)S_0 \tag{25}$$

holds, equation (21) has one solution corresponding to a minimum of the potential (20), too

$$S_{1l} = [(m\Omega^2 - \varepsilon)S_0 + \varepsilon S_2 + b\Sigma_c/2S_c]/(\varepsilon + m\Omega^2). \tag{26}$$

The most significant thing is that if the condition

$$S_{2c_1} < S_2 < S_{2c_2} \tag{27}$$

holds, equation (21) has three solutions

$$S_{1m_1} = [(m\Omega^2 - \varepsilon)S_0 + \varepsilon S_2]/(\varepsilon + m\Omega^2)$$

$$S_{1m_2} = [(m\Omega^2 - \varepsilon)S_0 + \varepsilon S_2 - \Sigma_c d/S_c]/(\varepsilon + m\Omega^2 - \Sigma_c d/S_c^2) \tag{28}$$

$$S_{1m_3} = [(m\Omega^2 - \varepsilon)S_0 + \varepsilon S_2 + b\Sigma_c/2S_c]/(\varepsilon + m\Omega^2)$$

where S_{1m_1}, S_{1m_3} correspond to two minima of the potential (20), while S_{1m_2} corresponds to the maximum.

The above result can be explained as follows. Considering the influence of the vibrations of atoms A_{-2} and A_{+2}, S_1 (the only degree of freedom of the void) can no longer be fixed, but oscillates in a potential determined by the coupling effect. The frequency squared of this vibration mode is

$$\Omega_{S_1}^2(S_2) = (1/2m) \cdot \partial^2 U_2(S_2, S_1)/\partial S_1^2 = (1/2m)[\partial^2 V_{\text{eff}}(S_1, Q_{\min})/\partial S_1^2 + 2\varepsilon]. \tag{29}$$

When S_2 is small enough, i.e. $S_2 \leqslant S_{2c_1}$, the vibration of S_1 sits in a single-well potential and

$$\Omega_{S_1}^2(S_2 \leqslant S_{2c_1}) = (1/m)(m\Omega^2 + \varepsilon). \tag{30}$$

In this case the vibration of Q can prove to be in a single-well potential as well. So it is just like the situation in crystalline form of the material. With increasing S_2, the local tension increases greatly, and hence the total potential of the cluster although atom A_0 is in its equilibrium position located to minimise the total potential. At the same time, the vibration mode of S_1 becomes softer and softer, i.e. $\Omega_{S_1}^2(S_2)$ decreases constantly. However, since the interaction energy per atom in non-crystalline solids is close to that in their crystalline counterparts, neither the local tension nor the total potential can increase any further until S_2 exceeds a certain value, at which point a local 'phase transition' occurs. This indicates that the system changes locally from elastic to plastic with partial relaxation of the local tension and decrease of the total potential. The same result can be recognised more clearly by

$$\Omega_{S_1}^2(S_{2c_1} < S_2 < S_{2c_2}) = (1/m)(m\Omega^2 + \varepsilon - d\Sigma_c/S_c^2) \leqslant 0. \tag{31}$$

(The equality holds at the critical point.) The negative frequency squared implies breakdown of the single-well potential, and hence an establishment of a DWP; as a result, the vibration of S_1 meets a DWP instead of a single-well potential. A new equilibrium is then set up when S_1 sinks into one of the two newly created wells. The first and third formulae in (28) correspond to the positions of both wells, while the second corresponds to the barrier between them. The newly created DWP is then induced by the atomic tunneling states occurring in the primary DWP of atom A_0 and it can be regarded as a DWP of higher hierarchical rank. If S_2 enlarges further, i.e. $S_2 \geqslant S_{2c_2}$, S_1 will come back to a single-well potential once more. In this case, the interaction between the dangling-bonds and their outside neighbouring atoms is far beyond the elastic region, so the harmonic approximation becomes invalid; nevertheless, such a large value of S_2 is rarely seen even in amorphous materials. So this case is not worth discussing further. In figure 6, the potential where S_1 vibrates is demonstrated for different values of S_2, showing a change from a single-well potential to a DWP and in reverse order.

As discussed above, with the increase of S_2, the frequency squared of the vibration of S_1 decreases considerably, and even passes zero to negative. The equilibrium becomes unstable and the system tends to find a new stable state causing the splitting of a single-well potential into a DWP. In this case S_1 can either shrink or elongate. When it shrinks to a subcritical region with $S_1 \leqslant S_c$, the primary DWP disappears. When it elongates to supercritical region, the primary DWP is no longer unstable and critical, but becomes rigid with higher barrier and smaller deformation. Therefore two different types of DWPs come out in the same model. The calculation of some characteristic parameters for g-SiO$_2$ will be given in a separate paper. Here we only conclude that when S_1 is in the vicinity of its critical point, the primary DWP is critical and soft; when $S_1 > \lambda S_c$, it is rigid.
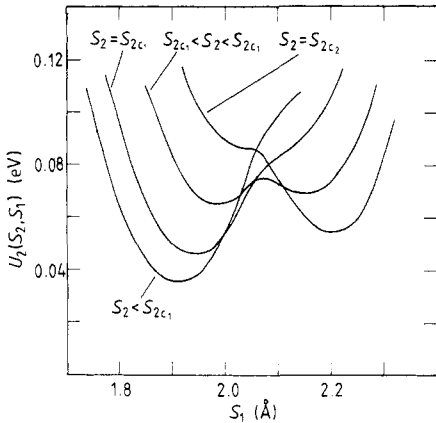
Figure 6. The changes induced in the shape of the DWP $U_2(S_2, S_1)$-versus-S_1 plot by varying S_2. Two critical points are shown; however, S_{2c_2} is by no means due to the breakdown of harmonic approximation for extremely large deformation. The data are for g-SiO$_2$.

Thus considering the interaction of the void with its neighbouring atoms, the vibration of atoms A_{+1} and A_{-1} can be treated as a vibration of a quasi-particle with the radius of the void being its degree of freedom. The quasi-particle sees an effective potential whose shape depends on the static formation parameters of surrounding atoms. At the same time, the primary DWP of the atom in the void is strongly influenced by the behaviour of the quasi-particle. So a strong constraint is clearly seen between the first two ranks of DWPs.

4. Hierarchy of DWPs and its termination

The above analyses inspire us to speculate as to whether there is any new DWP of higher rank and low-frequency vibration modes if we consider the system to be further and further extended. If this is true, can we continue the same consideration from rank to rank constructing an unlimited hierarchical ladder? These are the problems we treat in this section on the basis of the DVS model.

In one-dimensional DVS model, a more and more extended cluster of atoms is nothing but a cluster containing more and more neighbouring atoms from the outside. In the symmetric approximation, the characteristic degree of freedom specifying the shape and size of the cluster is its radius which can express the motion of the entire cluster, as was shown in figure 1.

The coupling of the vibrations of atoms A_{-2} and A_{+2} with the dynamical behaviours of the cluster consisting of atoms A_{-1}, A_0 and A_{+1} has been considered above. In the following, atoms A_{-2} and A_{+2} are involved in the extended cluster to consider its coupling with neighbouring atoms A_{-3} and A_{+3}. The vibrational degree of freedom of the extended cluster is its radius S_2, while S_3 remains fixed. As explained above, in this cluster, S_1 always minimises the total potential, i.e. the quasi-particle is in its equilibrium position. However, from figure 6, we see that the DWP in which S_1 moves is asymmetric, the relative depth of the two wells depending on the value of S_2. For convenience we assume that S_1 always sinks in a well having smaller value of S_1, i.e.

$$S_{1m} = [(m\Omega^2 - \varepsilon)S_0 + \varepsilon S_2]/(\varepsilon + m\Omega^2). \qquad (32)$$

In the same harmonic approximation of two neighbouring atoms, the total effective

potential of the extended cluster is

$$U_3(S_3, S_2) = U_3(S_3, S_2, S_{1m}) = U_2(S_2, S_{1m}) + \varepsilon(S_3 - S_2 - S_0)^2. \tag{33}$$

Substituting (32) into (33), we have

$$U_2(S_2, S_{1m}) = A_2 m\Omega^2(S_2 - 2S_0)^2 - d\Sigma_c(A_2 S_2/S_c - B_2 - 1)^2 \sigma[(S_2 - \alpha_2)(S_2 - \beta_2)]$$
$$+ \Sigma_c(a + bB_2 - bA_2 S_2/S_c)\sigma(\beta_2 - S_2) \tag{34}$$

where

$$A_2 = \varepsilon/(\varepsilon + m\Omega^2) \qquad B_2 = (\varepsilon - m\Omega^2)/(\varepsilon + m\Omega^2)$$
$$\alpha_2 = S_0 + S_c + (m\Omega^2/\varepsilon)(S_c - S_0) \tag{35}$$
$$\beta_2 = S_0 + \lambda S_c + (m\Omega^2/\varepsilon)(\lambda S_c - S_0).$$

We find that (34) has a form similar to that of $V_{eff}(S_1, Q_{min})$. It is just because of the particular form of $V_{eff}(S_1, Q_{min})$ that the vibrational mode of S_1 is softened, and under certain conditions its potential splits into a DWP of the second rank. So we continue the same consideration in the following. When S_3 is small enough, it locates in a single-well potential with a frequency determined by

$$\Omega_{S_2}^2(S_3) = (1/2m) \cdot \partial^2 U_3(S_3, S_2)/\partial S_2^2 = (1/2m)[\partial^2 U_2(S_2, S_{1m})/\partial S_2^2 + 2\varepsilon]$$
$$= (1/m)(m\Omega^2 A_2 + \varepsilon). \tag{36}$$

With the increase of S_3, the frequency squared of S_2 turns out to be

$$\Omega_{S_2}^2(S_3) = (1/m)(m\Omega^2 A_2 + \varepsilon - d\Sigma_c A_2^2/S_c^2). \tag{37}$$

If the condition

$$\varepsilon < d\Sigma_c A_2^2/S_c^2 - m\Omega^2 A_2 \tag{38}$$

holds, (37) becomes negative, then the single-well potential splits into a DWP.

Therefore, by considering an extended system we can get not only a new low-frequency vibration mode, but a DWP of third rank as well. The key condition to guarantee the occurrence of a DWP of higher rank is that the frequency squared becomes negative, and this condition in turn depends on the radius of the extented cluster. For a particular void in amorphous materials, such a hierarchy of DWPs cannot always happen. However, in our view, the structural defects such as voids exist everywhere with a distribution of $S_1, S_2, \ldots$, as $g(S_1), g(S_2), \ldots$. So for some of the voids, such hierarchies can really happen.

By continuing the same derivation, we obtain a hierarchy of DWPs and a large number of modes of low-frequency vibration. For convenience, we define

$$U_1(S_1, Q_{min}) = V_{eff}(S_1, Q_{min}). \tag{39}$$

The recurrence formulae of the total effective potential on the nth rank are then

$$U_n(S_n, S_{n-1m}) = A_n m\Omega^2(S_n - nS_0)^2 - d\Sigma_c(A_n S_n/S_c - B_n - 1)^2 \sigma[(S_n - \alpha_n)(S_n - \beta_n)]$$

$$+ \Sigma_c(a + bB_n - bA_n S_n/S_c)\sigma[\beta_n - S_n] \tag{40}$$

where

$$A_n = \varepsilon/(\varepsilon + (n-1)m\Omega^2) \qquad B_n = (n-1)(\varepsilon - m\Omega^2)/[\varepsilon + (n-1)m\Omega^2]$$

$$\alpha_n = (n-1)S_0 + S_c + [(n-1)m\Omega^2/\varepsilon](S_c - S_0) \tag{41}$$

$$\beta_n = (n-1)S_0 + \lambda S_c + [(n-1)m\Omega^2/\varepsilon](\lambda S_c - S_0).$$

When considering the influence of its neighbouring atoms, the total effective potential of the $(n+1)$th rank cluster is as

$$U_{n+1}(S_{n+1}, S_n) = U_n(S_n, S_{n-1m}) + \varepsilon(S_{n+1} - S_n - S_0)^2 \tag{42}$$

with the $(n-1)$th rank degree of freedom S_{n-1} to minimise it. When S_{n+1} is small enough, S_n locates in a single-well potential with a frequency determined by

$$\Omega^2_{S_n}(S_{n+1}) = (1/2m) \cdot \partial^2 U_{n+1}(S_{n+1}, S_n)/\partial S_n^2 = (1/2m)[\partial^2 U_n(S_n, S_{n-1m})/\partial S_n^2 + 2\varepsilon]$$

$$= (1/m)[A_n m\Omega^2 + \varepsilon]. \tag{43}$$

With increasing S_{n+1}, the squared frequency of S_n turns out to be

$$\Omega^2_{S_n}(S_{n+1}) = (1/m)(A_n m\Omega^2 - d\Sigma_c \cdot A_n^2/S_c^2 + \varepsilon). \tag{44}$$

If the condition

$$\varepsilon < \frac{d\Sigma_c A_n^2}{S_c^2} - A_n m\Omega^2 \tag{45}$$

holds, then (44) becomes negative, causing the splitting of a single-well potential into a DWP. The two wells are determined by

$$\partial U_{n+1}(S_{n+1}, S_n)/\partial S_n = \partial U_n(S_n, S_{n-1m})/\partial S_n - 2\varepsilon(S_{n+1} - S_n - S_0). \tag{46}$$
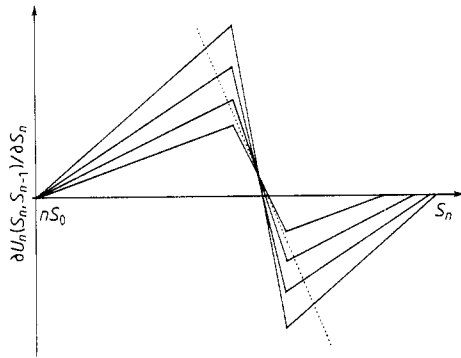
The solutions are

$$S_{nm_1} = \{[\varepsilon + (n-1)m\Omega^2]S_{n+1} - (\varepsilon - m\Omega^2)S_0\}/(\varepsilon + nm\Omega^2)$$

$$S_{nm_3} = \{[\varepsilon + (n-1)m\Omega^2]S_{n+1} - (\varepsilon - m\Omega^2)S_0 + b\Sigma_c/2S_c\}/(\varepsilon + nm\Omega^2). \tag{47}$$

The behaviours of the nth rank cluster determine the existence of the $(n-1)$th rank DWP and its rigidity.

Substituting (41) into (45), we find that with ascending rank, (45) cannot always hold, hence a termination emerges. The maximum rank is derived as

$$n \leqslant \sqrt{d\Sigma_c \varepsilon/m^2\Omega^4 S_c^2 + \tfrac{1}{4}} - (\varepsilon/m\Omega^2 - \tfrac{1}{2}). \tag{48}$$

Figure 7. Sketch of $\partial U_n(S_n, S_{n-1})/\partial S_n$ versus S_n. The set of full curves represents the first term in (46), while the dotted line represents the second term for a fixed S_{n+1}. The intersections of the full lines with the dotted line correspond to the solutions of (46).

The emergence of the termination can be seen more clearly as demonstrated in figure 7: a bunch of full lines represents the first term in (46), while the dotted line represents the second term. So the intersections of the full lines with the dotted line correspond to the solutions of equation (46). With ascending rank, the gradients of the full lines become less steep. Consequently the intersections reduce from three to one, this indicating that DWP can no longer exist. This result accords with the demands suggested by Fleurov (1988) for energy considerations of the hierarchy. The same idea of a characteristic length for the termination of elementary vibrational excitations was also introduced as an *ad hoc* assumption in a fracton approach proposed by Alexander *et al* (1983).

5. Discussion and concluding remarks

From (48), we see that the termination depends on the material features of amorphous solids. By defining

$$S_c/S_0 = \rho \tag{49}$$

we change (8) to

$$\Sigma_c/S_c^2 = \Sigma_0/\rho^{2k+2} S_0^2. \tag{50}$$

Substituting (50) into (48), we have

$$n \leqslant \sqrt{d\Sigma_c \varepsilon/m^2 \Omega^4 S_0^2 \rho^{2k+2} + \tfrac{1}{4}} - (\varepsilon/m\Omega^2 - \tfrac{1}{2}). \tag{51}$$

Among the parameters associated with the hierarchical termination, ρ and Ω are more important than m, Σ_c and S_0. It can be seen that the earlier the first-rank DWP appears, and the lower the dominant frequency of the system, the higher become the total rank of the hierarchical DWPs. Light atom on the chain and strong coupling of the electrons will also benefit the existence of the hierarchy.

It has been shown in (43) that with growing rank, the frequency of the additional vibration mode decreases, and it can be proved that the barrier height of the hierarchical DWPs decreases at the same time. A hierarchical scheme of DWPs with progressively decreasing characteristic parameters is then constructed. Mezard *et al* (1984) have

observed a hierarchical structure for distribution of pure states in configuration space. In this paper such a hierarchical structure has been established in real glassy systems, from which the non-exponential relaxation is observed.

It may be interesting to imagine this hierarchical structure to be a self-simulated one from rank to rank, and its fractal dimensionality $\bar{d}$ can then be determined by the fractal-like potential scheme. The above mentioned hierarchical termination sets the range of the elementary vibrational excitations. So using the method proposed by Alexander *et al* (1983), the vibrational density of states of the hierarchically dynamical modes can be determined by

$$N(\omega) = \bar{d}(L/S_0)^{\bar{d}}\omega^{\bar{d}-1}/\omega_{fD}^{\bar{d}} \tag{52}$$

where L is the interaction range determined by the hierarchical termination, and ω_{fD} is the fracton Debye frequency. Therefore, the physical nature of the hierarchical vibration modes is likely to be demonstrated in terms of the fracton approach. Detailed discussion of the determination of $\bar{d}$ and some physical properties associated are beyond the scope of the present paper and will be given in a separate one.

Besides the hierarchical scheme, our model also embodies constraints. The physical nature of the constraints is primarily the interaction between different ranks. When S_n locates in one particular well of the two, e.g. $S_n = S_{m_1}$, the only possible place for S_{n-1} to locate is the well of smaller value, and what is more, the behaviour of S_n is modulated by the detailed dynamics of S_{n+1}. So the relaxing process involves many sequential correlated activation steps, meanwhile, a wide range of relaxation times is generated. This result builds a complete connection to the phenomenological and illustrative class of models proposed by Palmer *et al* (1984). There are also some experimental implications on the constraints. The recent neutron-scattering measurements of Buchenau *et al* (1986) suggest that the DWPs and local vibration modes in vitreous silica have the same scattering vector and should be of the same origin. Strom (1986) deduced from his far-infrared absorption data of vitreous SiO_2 (Strom and Taylor 1977) that the coupling constant for an 'excess' absorption has a strong frequency dependence which indicates some degree of correlations between different ranks of vibrational modes.

Before giving conclusions, we want to make some remarks about the effective kinetic energy T_{eff}. T_{eff} is not local in time but involves the retarded interactions between the movements of atom A_0 from time τ' to τ. The retarded effect comes from the fact that the electrons are not in equilibrium with the atomic displacement, and it also shows the breakdown of the adiabatic approximation. We have proved that the retarded effect can be replaced by an effective mass (Chen and Wu 1989), namely $T_{eff} = \frac{1}{2}m_{eff}\dot{Q}^2$, where m_{eff} depends on temperature and tunneling path. The former has dominant influence at low temperatures.

In conclusion, by considering the coupling of the atomic tunneling states with local vibration modes, some interesting results are obtained based on the DVS model. First, we have found a hierarchy of constrained DWPs as well as plentiful low-frequency vibration modes. After that, the recurrence formula and the termination of the hierarchy are derived.

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