PHYSICS

A modification of the soft potential model of glasses

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Abstract. Replacement of the cubic term in the soft potential model of glasses with a linear one can be interpreted as an introduction of the internal pressure (coefficient of the linear term), additive to the applied external pressure. It makes the potential unique and removes the ambiguity, present in the conventional representation (different choices of the origin). Pressure-induced transformation of soft systems becomes in the new representation extremely simple – they evolve with the pressure along straight lines. The "sea-gull" distribution of the soft potential parameters transforms into a uniform one in these new variables.

Key words: glass, soft potential, pressure.

1. INTRODUCTION

Anomalous low-temperature thermal properties of glasses have been an active field of research since their discovery in the early 1970s [1]. The soft potential model (SPM) [2–6] has turned out to be a versatile method for capturing essential features of glasses: two-level systems, soft vibrations (the boson peak), and relaxation systems. In particular, effects of hydrostatic pressure on low-temperature glasses can be modelled in this way [7–9]. While the effect of pressure on crystalline solids is mainly expressed by increase in vibrational frequencies, the pressure effects in glasses are more diverse. Besides the increase in vibrational frequencies, transitions between different types of excitations (two-level systems and soft vibrations) can be induced by the applied pressure. The pressure can also affect barriers between the two equilibria in SPM and, as a result, influence the irreversible relaxation in glasses [9]. On the other hand, studies of these effects can yield valuable information on the local structure and dynamics in glasses – problems still gaining much attention. In what follows, we demonstrate that a slight modification of SPM, introduction of "internal pressure" as a linear term in the potential instead of the cubic one, enables one to handle both pressurized and unpressurized situations on a

similar footing and gives, in our opinion, a more physical interpretation of the parameters of the potential.

2. THE SOFT POTENTIAL

In what follows, the quantities with the subscript "0" refer to the situation with no external pressure applied.

The form of the soft potential is given by [2–4]

$$U(x) = U_0(x^4 + \xi_0 x^3 + \eta_0 x^2), \quad (1)$$

where the quantities ξ_0 and η_0 are random dimensionless parameters, subject to distribution to fit the experimental data. For the sake of simplicity, in what follows we take the energy factor $U_0 = 1$. Depending on the values of parameters ξ_0 and η_0, the potential Eq. (1) has either a single minimum $(\eta_0 > (9/32)\xi_0^2)$ or two minima $(\eta_0 < (9/32)\xi_0^2)$, separated by a barrier. In the (ξ_0, η_0)-plane these two types of potentials are separated by the parabola $(\eta_0 = (9/32)\xi_0^2)$ (Fig. 1a). Note that the presentation of Eq. (1) is not unique: potentials with

$$\eta_\pm = \frac{1}{16}\left[9\xi_0^2 - 32\eta_0 \mp 3\xi_0\sqrt{9\xi_0^2 - 32\eta_0}\right],$$

$$\xi_\pm = \frac{1}{2}\left[\pm\sqrt{9\xi_0^2 - 32\eta_0} - \xi_0\right] \quad (2)$$

differ from (1) only by the choice of the origin [5].

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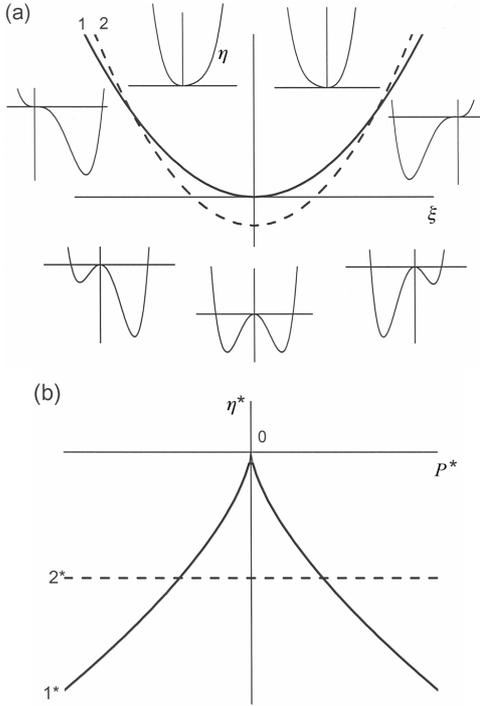
Fig. 1. The "phase space" of soft potentials in (ξ, η) (a) and (P^*, η^*) (b) parametrization. Above the separatrices 1 (a) and 1* (b) the single-well potentials are located. The parabolic curve 2 (see Eq. (12)) (a) and straight line 2* (b) represent the pressure evolution of a selected soft system.

An alternative way of representing the potential (1) is the replacement of the cubic term by a linear one [6]. With the change of variable $x \to z = x - t$ and taking $t = -(1/4)\xi_0$, we get

$$U(z) = z^4 + \eta_0^* z^2 + P_0^* z + \Delta U_0^*, \tag{3}$$

where

$$\eta_0^* = \eta_0 - \frac{3}{8}\xi_0^2,$$

$$P_0^* = \frac{1}{2}\xi_0\left(\frac{1}{4}\xi_0^2 - \eta_0\right), \tag{4}$$

$$\Delta U_0^* = \frac{1}{2^4}\xi_0^2\left(\eta_0 - \frac{3}{2^4}\xi_0^2\right).$$

In what follows, the quantities with the asterisk (*) correspond to such a linearized form of the soft potential. The "linear" form of the potential (3) makes it unique and removes the ambiguity present in the conventional (ξ_0, η_0)-representation (different choices of the origin, Eq. (2)). The separatrice between the double-well and single-well potentials in the (P_0^*, η_0^*)-plane is given by

$$\eta_0^* = -\frac{3}{2}\left|P_0^*\right|^{2/3}, \tag{5}$$

with the single-well potentials located above this curve (see Fig. 1b). Notice that only single-well potentials with $\eta_0^* < 0$ can be transformed into double-well ones with pressure.

In order to match the experimental observation, a distribution of the soft potential parameters of the "seagull" type is usually adopted:

$$F(\xi_0, \eta_0) \propto \left|\eta_0\right|. \tag{6}$$

Using (4), it is easy to show that the distribution for the variables P_0^* and η_0^* becomes uniform (constant):

$$F^*(P_0^*, \eta_0^*)$$

$$= F[\xi_0(P_0^*, \eta_0^*), \eta_0(P_0^*, \eta_0^*)]\left|\frac{\partial(P_0^*, \eta_0^*)}{\partial(\xi_0, \eta_0)}\right|^{-1} \propto \text{const.} \tag{7}$$

3. PRESSURE EFFECTS

The applied hydrostatic pressure P is usually described as an additional linear term in the soft potential [7–9]:

$$U(x) = x^4 + \xi_0 x^3 + \eta_0 x^2 + Px. \tag{8}$$

In Eq. (8) pressure is introduced as a dimensionless quantity; in physical units the pressure $P = 1$ is of the order of the Young modulus. With a proper choice of the origin, the potential can always be brought to a form where there is no linear term. For this one must take $x \to y = x - s$. For s we get the following equation:

$$s^3 + \frac{3}{4}\xi_0 s^2 + \frac{1}{2}\eta_0 s + \frac{1}{4}P = 0. \tag{9}$$

Then

$$U(y) = y^4 + \xi(P)y^3 + \eta(P)y^2 + \Delta U. \tag{10}$$

This is essentially Eq. (1) where the coefficients, however, acquire now pressure dependence:

$$\xi(P) = 4s + \xi_0,$$

$$\eta(P) = 6s^2 + 3\xi_0 s + \eta_0, \tag{11}$$

$$\Delta U(P) = s^4 + \xi_0 s^3 + \eta_0 s^2 + Ps,$$

and s is the solution of Eq. (9). Note that in this section the quantities without the subscript "0" are, in general, pressure-dependent. In Figs 2 and 3 the dependences of s, $\xi(P)$, $\eta(P)$ and the points $(\xi(P), \eta(P))$ on the hydrostatic pressure P are presented. Note that the points $(\xi(P), \eta(P))$ lie on the parabola

$$\eta(P) = \frac{3}{8}\xi^2(P) + \left(\eta_0 - \frac{3}{8}\xi_0^2\right). \tag{12}$$

(a)

(b)
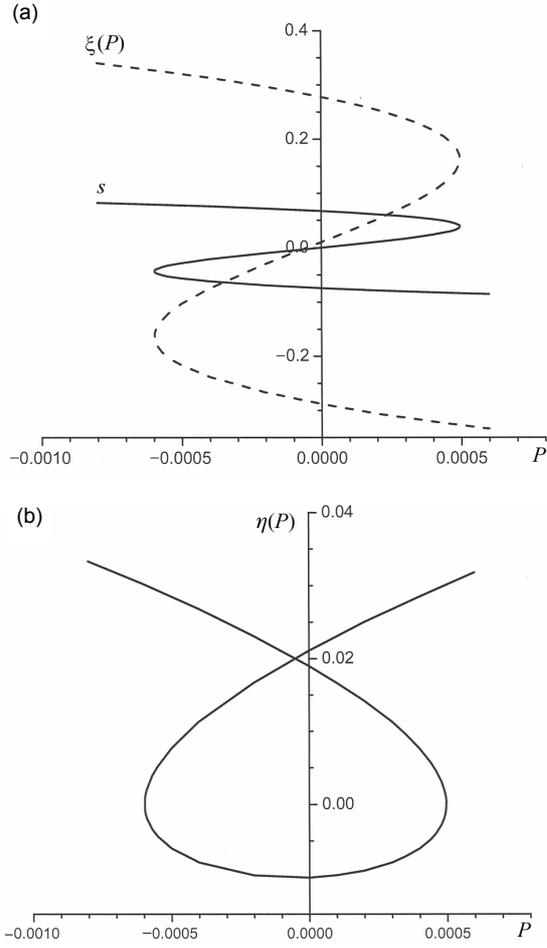
Fig. 2. The dependence of s, $\xi(P)$ (a), and $\eta(P)$ (b) on the hydrostatic pressure P for initial parameters $\xi_0 = \eta_0 = 0.01$ (see Eqs (9), (11)).
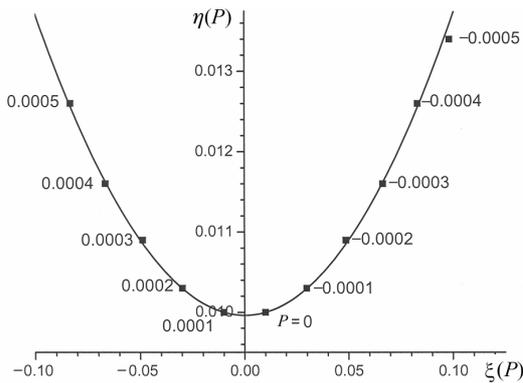
Fig. 3. The dependence of the points $(\xi(P), \eta(P))$ on the hydrostatic pressure P for initial parameters $\xi_0 = \eta_0 = 0.01$ (see Eqs (9), (11)).

The cubic term can be removed from the "pressurized" potential (8), (10) as well. This is reached by transformation $x \to z = x - t$ with $t = -(1/4)\xi_0$. The result is
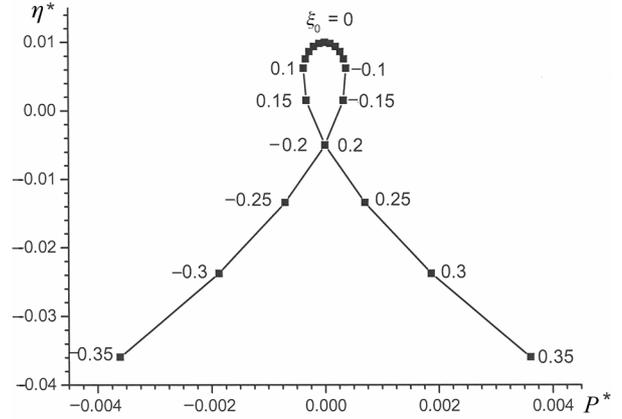
Fig. 4. The dependence of the points (P^*, η^*) on the initial parameter ξ_0 for the initial parameter $\eta_0 = 0.01$ and the hydrostatic pressure $P = 0$ (see Eq. (4)).

essentially the potential (3), where the coefficients P_0^* and ΔU_0^* are replaced by values depending on the applied pressure P:

$$P^*(P) = P_0^* + P,$$
$$\Delta U^*(P) = \Delta U_0^* - (1/4)\xi_0 P, \tag{13}$$

yielding

$$U(z) = z^4 + \eta^* z^2 + P^*(P)z + \Delta U^*(P). \tag{14}$$

The value of $\eta^* \equiv \eta_0^*$ remains, however, independent of pressure and is determined by the "unpressurized" values ξ_0 and η_0 only. In Fig. 4 the dependence of the points (P^*, η^*) on the initial parameter ξ_0 is presented. Notice that the coefficient of the linear term $P^*(P)$ now sums up from two contributions: external pressure P and the quantity P_0^*, with the value given by (4). Hence it is reasonable to interpret the latter quantity as an "internal pressure", additive to the applied external pressure and subject to variations from location to location. Pressure-induced transformation of soft systems becomes in the (P^*, η^*)-representation extremely simple – they evolve with the pressure along straight lines in the (P^*, η^*)-plane, parallel to the P^*-axis. It then immediately follows from (6), (7) that the distribution $F(P^*, \eta^*)$ remains invariant under the changing pressure.

4. CONCLUSIONS

We have demonstrated that the replacement of the cubic term in the soft potential model with a linear one can be interpreted as an introduction of the internal pressure, additive to the applied external pressure. It also makes the potential unique and removes the ambiguity present

in the conventional (ξ, η)-representation (different choices of the origin). Pressure-induced transformation of soft systems becomes extremely simple in the (P^*, η^*)-representation – they evolve with the pressure along straight lines in the (P^*, η^*)-plane. In a further paper we are going to calculate the distribution of the internal pressure proceeding from the postulated (ξ, η)-statistics of glasses in order to compare it to some microscopic elastic models of glasses [10].

ACKNOWLEDGEMENT

REFERENCES

1. Zeller, R. C. and Pohl, R. O. Thermal conductivity and specific heat of noncrystalline solids. *Phys. Rev. B*, 1971, **4**, 2029–2041.

2. Karpov, V. G., Klinger, M. I., and Ignat′ev, F. N. Theory of low-temperature anomalies in thermal properties of amorphic structures. *Sov. Phys. JETP*, 1983, **57**, 439–448.

3. Il′in, M. A., Karpov, V. G., and Parshin, D. A. Parameters of soft atomic potentials in glasses. *Sov. Phys. JETP*, 1987, **65**, 165–168.

4. Parshin, D. A. Model of soft potentials and universal properties of glasses. *Phys. Solid State*, 1994, **36**, 991–1024.

5. Karpov, V. G. and Parshin, D. A. Heat-conductivity of glasses at temperatures below the debye temperature. *Sov. Phys. JETP*, 1985, **61**, 1308–1317.

6. Buchenau, U., Galperin, Yu. M., Gurevich, V. L., and Schober, H. R. Anharmonic potentials and vibrational localization in glasses. *Phys. Rev. B*, 1991, **43**, 5039–5045.

7. Karpov, V. G. and Grimsditch, M. Pressure-induced transformation in glasses. *Phys. Rev. B*, 1993, **48**, 6941–6948.

8. Hizhnyakov, V., Laisaar, A., Kikas, J., Kuznetsov, An., Palm, V., and Suisalu, A. Transformation of soft localized modes in glasses under pressure. *Phys. Rev. B*, 2000, **62**, 11296–11299.

9. Kikas, J., Suisalu, A., Kuznetsov, An., Laisaar, A., Takahashi, J., and Hizhnyakov, V. Pressure effects on relaxation in a polymer glass: a persistent spectral hole burning study. *Opt. Spectrosc.*, 2005, **98**, 675–680.

10. Alexander, S. Amorphous solids: their structure, lattice dynamics and elasticity. *Phys. Rep.*, 1998, **296**, 65–236.

Klaaside pehme potentsiaali mudeli modifitseerimine

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Kuupliikme asendamine lineaarsega klaaside pehme potentsiaali mudelis on interpreteeritav siserõhuna (lineaar-liikme võrdtegur), mis liitub välisele süsteemile rakendatud rõhule. See kõrvaldab traditsioonilisele esitusele omase mitteühesuse (erinevad koordinaadi nullpunkti valikuvõimalused). Pehmete süsteemide rõhuline transformatsioon muutub uues esituses äärmiselt lihtsaks: rõhu muutudes liiguvad need mööda sirgeid. Pehmete süsteemide statistiline jaotus transformeerub uutes muutujates ühtlaseks jaotuseks.