# Structural defects and soft localized modes in disordered systems

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The relation between localized low-frequency excitations, which are known from the theory of low-temperature anomalies in glasses, and structural defects, which play a decisive role in plastic deformation, has been studied. We examine the position dependence of the decay of the strain field, corresponding to a local mode, as a function of the anharmonicity of the system. The long-range strain field due to localized modes has been calculated, its properties have been analyzed, and the results have been compared to the data obtained independently by computer simulation. © *1996 American Institute of Physics.* [S1063-7761(96)02107-5]

## 1. INTRODUCTION

It is well known that low-temperature anomalies in glasses can be interpreted only assuming molecular mobility that does not exist in crystals. It is natural to relate this mobility to the presence of structural defects in a disordered medium, generating both tunneling modes and soft localized vibrations.[1–4]

Presently there are reasons to suppose that these structural defects are also responsible for plastic deformation of glasses.[5,6]

Models of low-frequency local mobility proposed in recent years can be efficiently applied to the analysis of electronic and thermodynamic properties of glasses, but are not adequate for describing their plastic properties. The reason is that interaction between structural defects is important in this case, and in order to take this interaction into account, the defect model must be consistent with elastic-continuum theory.

This reconcilliation of the two models is described in the present paper, which suggests a continuum description of a structural defect. Calculations of soft local vibrations based on this model are given.

In Sec. 2 we discuss the feasibility of a localized mode due to local nonuniformity of elastic and density properties. Section 3 is dedicated to a qualitative analysis of the role of anharmonicity in generating soft localized modes. In effect, the region where anharmonicity arises is a structural defect, for which model is given in Sec. 4. Finally, in Sec. 5 we discuss the soft localized mode as a small perturbation of an equilibrium configuration due to a structural defect.

## 2. VIBRATIONS OF LINEAR ELASTIC CONTINUUM WITH A POINT-LIKE SINGULARITY OF ELASTIC MODULI

The simplest model of a defect in glass can be described in terms of a local nonuniformity of elastic parameters and/or density. In the theory of elastic continua, it can be defined as a point-like singularity; to be specific, let us suppose that all the parameters of the continuum have delta-singularities at the point $R=0$:

$$\mu=\mu_0+\frac{m}{4\pi R^2}\delta(R),$$

$$\lambda=\lambda_0+\frac{l}{4\pi R^2}\delta(R), \qquad (1)$$

$$\rho=\rho_0+\frac{r}{4\pi R^2}\delta(R).$$

Here $\rho$ is the continuum density, and $m$, $l$, and $r$ are the amplitudes of the singularities of the Lamé coefficients $\mu$ and $\lambda$, and of the density, respectively.

Let us consider the feasibility of localized waves caused by this singularity. The boundary condition for such waves is their decay at infinity, hence the elasticity equations can be reduced to the d'Alembert equations in which transverse and longitudinal waves are separated.[7] For simplicity, we only consider centrally symmetrical longitudinal waves. In this case, the strain field is characterized by a potential described by the scalar d'Alembert equation:

$$\mathbf{U}=\nabla\Phi,\quad \Phi_{tt}-c_1^2\Delta\Phi=0,$$
$$c_1^2=\frac{\lambda+2\mu}{\rho},\quad \Delta=\frac{\partial^2}{\partial R^2}+\frac{2}{R}\frac{\partial}{\partial R}. \qquad (2)$$

In this equation $\mathbf{U}$ is the elastic displacement vector and $\Phi$ is the scalar potential.

Given Eqs. (1) and (2) and the central symmetry of vibrations, the equation for vibrations due to a structural defect can be reduced to

$$\Phi_{tt}-c_{10}^2\Delta\Phi=\frac{g}{4\pi R^2}\delta(R)\Delta\Phi, \qquad (3)$$

where

$$c_{10}^2=\frac{\lambda_0+2\mu_0}{\rho_0},\quad g=c_{10}^2\left(\frac{l+2m}{\lambda_0+2\mu_0}-\frac{r}{\rho_0}\right).$$

The terms containing first derivatives of the delta function and scalar potential are omitted in Eq. (3) because the solution is presumed to be centrally symmetrical and the perturbation of elastic moduli point-like.

With the change of variables

$$\Psi=\Phi/\sqrt{R},$$

we transform Eq. (3) to the form

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$$\Psi_{tt}-c_{10}^2\Delta_{1/2}\Psi=\frac{g}{4\pi R^2}\,\delta(R)\Delta_{1/2}\Psi, \tag{4}$$

where

$$\Delta_{1/2}=\frac{\partial^2}{\partial R^2}+\frac{1}{R}\frac{\partial}{\partial R}-\frac{1}{4R^2}. \tag{5}$$

We solve the equation containing the Laplacian in the form of Eq. (5) using the Hankel transform of order 1/2:

$$\mathscr{H}(\Psi_0)=\int_0^\infty \Psi_0 R J_{1/2}(sR)\sqrt{sR}\,dR, \tag{6}$$

where

$$J_{1/2}(z)=\sqrt{\frac{2}{\pi}}\frac{\sin z}{\sqrt{z}},$$

and $s$ is the transform parameter. After substituting $\Psi=\Psi_0\exp(-wt)$ into Eq. (4) and using the Hankel transform, we obtain

$$\mathscr{H}w^2+c_{10}^2s^2\mathscr{H}=\sqrt{\frac{2}{\pi}}\frac{sgA_0}{4\pi}, \tag{7}$$

where

$$\Delta_{1/2}\Psi\to A_0R,\quad R\to 0. \tag{8}$$

Thus we have

$$\mathscr{H}=\sqrt{\frac{2}{\pi}}\frac{gA_0}{4\pi}\frac{s}{w^2+s^2c_{10}^2}. \tag{9}$$

Finally, the inverse transform yields

$$\Psi=e^{-wt}\frac{gA_0c_{10}^2}{4\pi}\exp\left(-\frac{wR}{c_{10}}\right). \tag{10}$$

Given a specific model of the defect kernel, the parameter $w$ can be derived from Eqs. (8) and (10). In other words, if we assume that Eq. (8) is valid at a small distance $R=\kappa$, the parameter $w$ can be derived from the equation

$$R=\frac{gc_{10}^2}{4\pi}\left(\frac{\partial^2}{\partial R^2}+\frac{1}{R}\frac{\partial}{\partial R}-\frac{1}{4R^2}\right)e^{-wR/c_{10}},\quad R=\kappa. \tag{11}$$

In some range of parameters, this equation has complex roots $w=d\pm i\omega$ with a negative real part, which implies the existence of a localized vibrational mode. As expected, characteristic vibrations of the combined defect continuum system decay exponentially with distance. Excitation of such vibrations can be described in terms of linear resonance, i.e., there is an external-field frequency $\omega$ at which these vibrations have maximum amplitude. This shape of the vibrational spectrum is typical of crystals with interstitial defects.[7] Similar results, namely exponential decay with time and distance, were obtained in solving for the characteristic vibrations of an infinite elastic medium with a spherical void.[9]

The strain field potential described by Eq. (10) decays exponentially with distance. In other words, the mode due to a defect caused by a point-like singularity in an elastic continuum is highly localized. Highly localized high-frequency modes are well known in a wide range of amorphous systems, and the present approach to these defects can be applied to the elastic strain field generated by such vibrations. Linear vibrations in the vicinity of a screw dislocation in a crystal also turn out to be highly localized.[8]

Higher harmonics and transverse waves can also be analyzed using this technique. This analysis is not given in the paper because it is very cumbersome. It is obvious, however, that the basic qualitative conclusions about the nature of these vibrational modes (such as exponential decay and strong localization) also apply to more complex vibrations.

Can a model based on such vibrations adequately describe the low-frequency localized excitations responsible for the anomalous thermodynamic properties of glasses at low temperatures? The answer should probably be negative. First, according to our present understanding of thermodynamic anomalies,[10] parameters characterizing phonon absorption are universal for a wide range of glasses (essentially all glasses except metallic ones). This universality can be theoretically interpreted only by assuming a strong interaction among localized vibrational modes. The strong interaction demands that the elastic strain field as a function of distance be described by a power law ($R^{-2}$, see Ref. 10) rather than by an exponential one, i.e., the vibrational mode must be localized weakly rather than strongly. Second, all computer simulations[11] and physical experiments indicate that the localization region of a soft mode is notably larger (three to four coordination spheres) than that of a high-frequency mode, and spectral bands of the two types of excitation are separated, since strongly localized high-frequency modes occur on the high-energy edge of the phonon spectrum.

These considerations lead us to conclude that soft vibrations cannot be described using the linear approach. This will obviously be so even if we abandon the model of a point-like perturbation of an elastic continuum.

This means that the anharmonicity of interatomic interaction must be included in the model of localized vibrations, and the effect of the anharmonicity on the vibrational spectrum must be considered.

## 3. VIBRATIONS OF ELASTIC CONTINUUM WITH A STRUCTURAL DEFECT WITH ANHARMONICITY OF INTERATOMIC INTERACTION TAKEN INTO ACCOUNT

Anharmonicity is included in the model of longitudinal waves, which was invoked in the linear approximation in the previous section. Let us consider Eq. (2) for the elastic strain potential supplemented with terms describing a cubic nonlinearity[12]:

$$\Phi_{tt}-c_1^2\left(\Delta\Phi+B\Phi\frac{\partial\Phi}{\partial R}\right)=0. \tag{12}$$

We consider small oscillations in the system described by Eq. (12) in the presence of a permanent structural defect. To this end, we express the solution for the potential $\Phi$ as a sum of the field generated by the structural defect and a small time-dependent component (as in the previous section, only solutions with central symmetry are discussed):

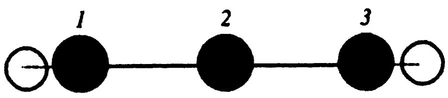


FIG. 1. One-dimensional three-atom model.

$$\Phi=\frac{D}{R}+f(R,t),\quad f\ll\Phi. \tag{13}$$

Substituting this expression into Eq. (12) and retaining only terms linear in $f$, we obtain

$$f_{tt}-c_1^2\frac{\partial^2 f}{\partial R^2}-\frac{2+BD}{R}\frac{\partial}{\partial R}+\frac{BDf}{R^2}=-\frac{BD^2}{R^3}. \tag{14}$$

The substitution $f=f_0\exp(i\omega t)$ turns Eq. (14) into a Bessel equation with a nonvanishing right-hand side. By using an approach similar to that of the previous section, i.e., assuming point-like singularities in elastic moduli and density, we can prove that the equation has localized solutions with the following properties.[8]

(1) If

$$3-2\sqrt{2}<DB<3+2\sqrt{2}, \tag{15}$$

acquires the equation can be the standard Bessel form with an imaginary exponent $\nu$. In this case, it can be solved using the $K$-transform, and its solutions can be expressed in terms of Bessel functions of the first kind, which decay as a power of the distance.

2) If $DB$ does not fall in the interval defined by Eq. (15), solutions similar to those discussed in the previous section can be expressed in terms of modified Bessel functions of the third kind (Besse or Macdonald functions), which decrease exponentially with distance.

Thus, power-law decay of the localized vibration amplitude with distance (weak localization) is a property of a nonlinear elastic continuum with a structural defect only over a certain range of the volume dilation around the defect and the anharmonicity parameter. The anharmonicity factor $B$ can be estimated to be

$$B\sim\frac{\alpha_3}{a_0(\lambda+2\mu)},$$

where $\alpha_3$ is the coefficient of the cubic term in the interatomic potential (the interatomic distance $a_0$ is included because this factor must have the same units as the elastic modulus.

In the case of positive dilation ($D<0$), (15) is satisfied if and only if the anharmonicity of the interatomic potential is negative, i.e., $\alpha_3<0$. If the dilation volume is negative, (15) is satisfied if the coefficient of the cubic term of the interatomic potential is positive, although the absolute value of this factor turns out to be comparable to the elastic moduli of the material, i.e., to the linear rigidity of this potential.

The relationship between features of the phonon spectrum due to a structural defect and the specific microstructure of the center of this defect is a very difficult problem. In

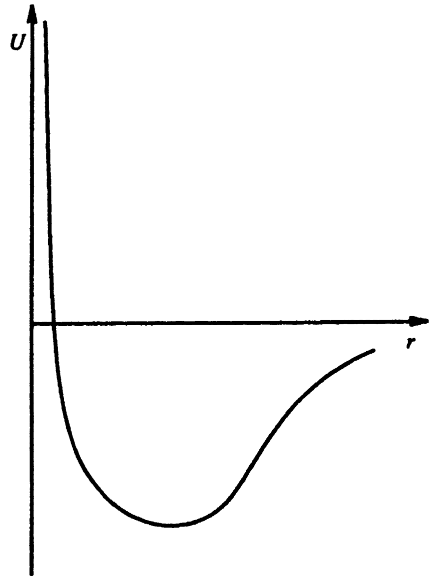


FIG. 2. Typical shape of two-atom interaction potential. The presence of an inflection point enables us to introduce an effective two-well potential.

order to get some idea of this relationship, we consider the simplified three-atom model shown in Fig. 1. Suppose that the atoms interact with each other through the potential given in Fig. 2. In this case, the effective potential acting on the atom 2 varies with interatomic distance as shown in Fig. 3.

This potential obviously undergoes a transition between states with one and two equilibrium positions. Let us focus our attention on the region around this transition. If the potential has two wells, its second derivative at the center is negative. The transition between the states with one and two wells leads to a sign change in the quadratic term of the potential expansion around the center. This means that from continuity considerations, at least one of the two relations

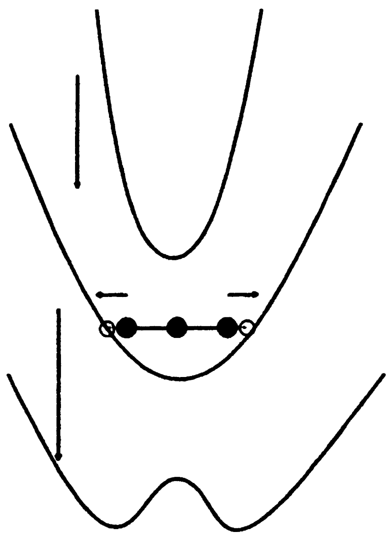

FIG. 3. Effective potential acting on the middle atom in the three-atom model at different distances between atoms.

between the coefficients of the linear and nonlinear terms in the potential expansion mentioned above must be satisfied near the transition, resulting in weak localization of the defect phonon mode.

It is clear that far from this transition (near the minimum of the pairwise interaction potential), the condition of weak localization cannot be satisfied because the positive anharmonicity factor is small compared to that of linear rigidity. Therefore the necessary condition of weak localization in this model is that the potential parameters be in the vicinity of the transition between the states with one and two wells.

From the viewpoint of real glasses, this conclusion is very approximate. Really, the proposed model implies that the anharmonicity is constant throughout the continuum, whereas in real materials the transformation can take place only in one or more neighboring bonds of a defect kernel. Note, however, that since the elastic stress rapidly drops with the distance from the defect, the nonlinear interaction takes place mostly in the neighborhood of the kernel. This suggests that the above conclusion also applies to realistic systems with strong local nonlinearity.

Thus, using the above reasoning and the terminology of Ref. 13, we may assert that there are states with "softened" one-well and two-well potentials in which weak (power-law) localization of vibrations is possible. According to the independent conclusions reported in Refs. 3 and 13, these states are responsible for the anomalous thermal conductivity and heat capacity of glasses at temperatures near absolute zero.

On the other hand, it is clear that the microstructure of such states is very different from that of the bulk material (this depends on proximity to the transition point). Therefore we may conclude that generation and evolution of structural defects under consideration leads to irreversible modifications in the microstructure (topology) of the amorphous system. Thus there is every reason to suppose that such defects may also control the process of plastic deformation in an amorphous system.

The approximate estimate of the anharmonicity effect given above does not yield an expression for the long-range strain field due to a localized vibrational mode that can be compared to computer simulations.[11,14] But it follows from the above discussion that the oscillation amplitude drops as a power of the distance. For a more detailed description of the elastic strain field due to a soft localized vibrational mode, we have to consider a model of a static point defect.

## 4. MODEL OF A STATIC STRUCTURAL DEFECT

From the viewpoint of the theory of a continuum with Volterra defects, an arbitrary point defect can be described as a set of cuts, splices, and inserts in an infinite elastic continuum in a local region. The distinctive feature of all combinations of such elements is that the elastic displacement field decays as $R^{-2}$, and accordingly the stress field decays as $R^{-3}$. This stress distribution is typical of vacancies.

The presence of a "defect ring"[5] implies anisotropy of the local elastic field caused by the break of one interatomic bond (the alignment of this bond defines a singular direction in the elastic field generated by a defect). It is reasonable to suppose that the system is axially symmetric about this direction, which means that all other sources of anisotropy have a weaker effect on the elastic field than one broken bond.

Writing the elastic equilibrium equations for an isotropic continuum in spherical coordinates,

$$\mu\left[\nabla^2 U_R-\frac{2}{R^2}\left(U_R+\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(U_\theta\sin\theta)+\frac{1}{\sin\theta}\frac{\partial U_\phi}{\partial\phi}\right)\right]+(\lambda+\mu)\frac{\partial\mathscr{D}}{\partial R}=0,$$

$$\mu\left[\nabla^2 U_\theta+\frac{2}{R^2}\left(\frac{\partial U_R}{\partial\theta}-\frac{U_\theta}{2\sin^2\theta}-\frac{\cos\theta}{\sin^2\theta}\frac{\partial U_\phi}{\partial\Phi}\right)\right]+(\lambda+\mu)\frac{1}{R}\frac{\partial\mathscr{D}}{\partial\theta}=0, \quad (16)$$

$$\mu\left[\nabla^2 U_\phi+\frac{2}{R^2\sin\theta}\left(\frac{\partial U_R}{\partial\phi}+\cot\theta\frac{\partial U_\theta}{\partial\phi}-\frac{U_\phi}{2\sin\theta}\right)\right]+\frac{\lambda+\mu}{R\sin\theta}\frac{\partial\mathscr{D}}{\partial R}=0,$$

where

$$\nabla^2=\frac{1}{R^2}\frac{\partial}{\partial R}\left(R^2\frac{\partial}{\partial R}\right)+\frac{1}{R^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)+\frac{1}{R^2\sin\theta}\frac{\partial^2}{\partial\phi^2},$$

$$\mathscr{D}=\frac{1}{R^2}\frac{\partial}{\partial R}(R^2U_R)+\frac{1}{R\sin\theta}\frac{\partial}{\partial\theta}(\sin\theta U_\theta)+\frac{1}{R\sin\theta}\frac{\partial U_\phi}{\partial\phi},$$

assuming that all components of the displacement field are proportional to $R^{-2}$, and taking the polar axis ($\theta=0$) as a symmetry axis of the strain field due to a structural defect (in this case, all its components are obviously independent of $\phi$), we obtain the following singular solution:

$$U_R=\frac{A}{R^2}(C+\cos 2\theta),\qquad U_\phi=0, \quad (17)$$

$$U_\theta=-\frac{A}{R^2}\gamma\sin 2\theta.$$

In this equation $\gamma=2\mu/(3\lambda+5\mu)$. The parameters $A$ and $C$ are determined by the microscopic structure of the defect core. The parameter $C$ determines the contribution of dilation to the defect strain field. In fact, if we take the limit $A\rightarrow 0$ in Eq. (17) and assume that $AC=\text{const}$, we obtain the field generated by a conventional vacancy or a dilation center.

Another approach to the strain field generated by a point defect can be based on the concept of one broken interatomic bond. From the viewpoint of the theory of elasticity, this approach is equivalent to a pair of equal forces, directed oppositely, and applied to two points on a straight line aligned with the forces in the continuum. The resulting field can easily be calculated and is called in the literature a field generated by a pair of forces with zero moment.[7] This field is described by the following equation:

$$U_i=-Fh\frac{\lambda+\mu}{8\pi\mu(\lambda+2\mu)}\frac{\partial}{\partial x_j}\left(\frac{x_i x_j}{R^3}+\frac{\lambda+3\mu}{\lambda+\mu}\frac{\delta_{ij}}{R}\right) \quad (18)$$

(no summation over the repeated index is assumed). In this equation $F$ is the applied force and $h$ is the distance between application points (usually the limit $h\rightarrow 0$ at $Fh=\text{const}$ is considered). By comparing Eqs. (17) and (18), we can find the relation between the parameter $A$ and parameters of the defect core:

$$A=Fh\frac{3\lambda+5\mu}{16\pi\mu(\lambda+2\mu)}. \quad (19)$$

## 5. SMALL PERTURBATION OF A STATIC DEFECT AS A LOCALIZED MODE

It is known that the displacement field in a dynamic problem can be expressed as a sum of potential and solenoidal components:

$$\mathbf{U}=\nabla\Phi+\text{curl }\mathbf{W}, \quad (20)$$

which, in turn, are determined by the scalar and vector d'Alembert equations:

$$\frac{\partial^2\mathbf{W}}{\partial t^2}=c_1^2\nabla^2\mathbf{W}, \quad \frac{\partial^2\Phi}{\partial t^2}=c_2^2\nabla^2\Phi, \quad (21)$$

where

$$c_1^2=\frac{\mu}{\rho}, \quad c_2^2=\frac{\lambda+2\mu}{\rho}, \quad \text{div }\mathbf{W}=0.$$

The problem is to find the profile of the localized vibrational mode due to an anisotropic structural defect whose strain field is determined by Eq. (17). It is natural to suppose that the dynamic strain field has the same symmetry as the steady-state defect field. Then it is easy to obtain expressions for the scalar and vector potentials:

$$\Phi=G_1\sqrt{R}Z_{5/2}(k_2R)(3\cos 2\theta+1)e^{i\omega t},$$
$$W_\phi=G_2\sqrt{R}Z_{5/2}(k_1R)\sin 2\theta e^{i\omega t}, \quad (22)$$
$$W_R=W_\theta=0, \quad k_1=\frac{\omega}{c_1}, \quad k_2=\frac{\omega}{c_2}.$$

In this equation

$$Z_{5/2}(x)=\sqrt{\frac{2x^3}{\pi}}\left(\frac{3\cos x}{x^4}+\frac{\sin x}{x^3}-\frac{\cos x}{x^2}\right) \quad (23)$$

is the spherical Bessel function of order 5/2.

It follows from Eq. (22) that the singularity in the dynamic mode is described by the function $R^{-4}$, which contradicts the accepted model of a steady-state defect. A solution which describes a microscopic model of a "breather" in a defect structure in terms of the theory of elasticity and is due to a pair of equal and oppositely directed forces periodic in time should, obviously, have a singularity at the origin similar to that of the steady-state solution. This condition is satisfied if the following constraint is imposed on the parameters of the scalar and vector potentials:

$$D=3B\alpha, \quad \alpha=(k_1/k_2)^{5/2}.$$

Given this condition, we finally obtain the following expressions for the displacement field of the soft mode generated by a structural defect:

$$U_R=B\left[\frac{d}{dR}\left(\sqrt{\frac{1}{R}}Z_{5/2}(k_2R)\right)+3\alpha\sqrt{\frac{1}{R^3}}Z_{5/2}(k_1R)\right]\times(3\cos 2\theta+1)e^{i\omega t},$$
$$U_\phi=0, \quad (24)$$
$$U_\theta=B\left[-\frac{3\alpha}{R}\frac{d}{dR}\left(\sqrt{\frac{1}{R}}Z_{5/2}(k_1R)\right)-6\sqrt{\frac{1}{R^3}}Z_{5/2}(k_2R)\right]\sin 2\theta e^{i\omega t}.$$

These expressions yield an estimate of the localization radius of the studied mode equal to the position of the first zero of the radial displacement component (this radius is, naturally, a function of the frequency). Then we can compare the localization radius derived from our model to the results of computer simulations and calculations of the spectrum of a disordered system of identical spheres.[11] According to these data, low-frequency localized modes exist in a frequency band of $(0.05–0.1)\omega_{\max}$, where $\omega_{\max}$ is the optical phonon frequency in the system. In these calculations, the localization region of the excitation contains about three or four coordination spheres, which is in good agreement with the estimate derived from Eq. (24).

Our results lead us to the conclusion that the linear theory of elasticity applied to the calculation of the profile of a localized vibrational mode yields fairly accurate results even for the region near the defect kernel. Therefore its application to the analysis of static defects and their interaction is justified.

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