Nature of vibrational eigenmodes in topologically disordered solids

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We use a local projectional analysis method to investigate the effect of topological disorder on the vibrational dynamics in a model glass simulated by molecular dynamics. Evidence is presented that the vibrational eigenmodes in the glass are generically related to the corresponding eigenmodes of its crystalline counterpart via disorder-induced level-repelling and hybridization effects. It is argued that the effect of topological disorder in the glass on the dynamical matrix can be simulated by introducing positional disorder in a crystalline counterpart.

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I. INTRODUCTION

Vibrational properties of disordered materials is one of the current lively topics of modern condensed-matter physics. Features of the disordered vibrational spectrum, such as the boson peak, the Ioffe-Regel crossover and vibrational localization are being investigated (see e.g.1–7). Disorder in condensed matter can essentially be classified into two basic types: (i) lattice disorder in crystals, e.g. in substitutional alloys, and (ii) topological disorder in, e.g., glasses. Crystalline structures with lattice disorder can be studied analytically and considerable progress has been achieved in understanding their vibrational behaviour.8–10 Structures with topological disorder have mainly been investigated numerically but in some aspects analytically.11–13 Different possible ways of describing the origin and nature of vibrational modes, particularly in the low-frequency range, in such disordered systems are still under debate.

The vibrational properties of crystals with lattice disorder can be successfully treated mainly because of the existence of a well-defined reference structure, viz. the same crystalline structure but without disorder. The choice of a reference structure for a topologically disordered material is less obvious. In some cases, such a choice can be based on the existence of similar local order in both the topologically disordered structure and its crystalline prototype. For example, α-cristobalite is found to be a good crystalline counterpart for vitreous silica.14–16 The existence of similar structural elements in both crystalline and glassy systems is expected to lead to similar vibrational dynamics. Similarities in the vibrational dynamics within these structural units can be revealed by comparing vibrational eigenmodes of the topologically disordered structure with those of its crystalline counterpart. Such a comparison can elucidate the nature of the disordered vibrational eigenmodes and their possible generic connection to crystalline modes. Basically, it can be expected that vibrational eigenmodes in topologically disordered structures have approximately the same nature as in crystals with lattice disorder.14–16 Namely, they can be regarded as strongly hybridized crystalline eigenmodes, shifted in frequency due to disorder-induced level-repelling effects. In this paper, we present arguments, based on a numerical analysis of a representative topologically disordered structure, that this conjecture holds true, at least, for the structure considered.

The existence of a crystalline counterpart allows a comparison to be made between a glass and its reference structure and conclusions to be drawn about the effect of topological disorder on the spectrum of vibrational excitations. Moreover, it is possible to compare positional, e.g. quenched thermally-induced, disorder in a crystal with topological disorder in the corresponding glass in terms of their respective influence on the vibrational properties. In a computer simulation, positional disorder can be introduced into a crystalline structure by heating it in the course of a molecular-dynamics run up to a certain temperature below the melting point. Then the dynamical matrix of an instantaneous configuration corresponding to this temperature can be calculated and analyzed. Thus it is possible to mimic the main features of the vibrational dynamics of topologically disordered systems by the vibrational dynamics of their positionally disordered crystalline counterparts. In the following, we demonstrate that this can be done, at least for some representative structures.

The rest of the paper is arranged as following. The local projectional analysis method is developed and described in Sec. II. The model is described in Sec. III. Results and conclusions are given in Sec. IV and V, respectively.
II. LOCAL PROJECTIONAL ANALYSIS

A comparison of the vibrational dynamics in a glass and its counterpart crystalline phase can be based on a local projectional analysis. Of course, the equilibrium atomic arrangements in the glass and the crystal are essentially different, and thus we cannot expand the eigenmodes in a disordered system in terms of the crystalline eigenmodes, as we can do for disordered lattices. Nevertheless, what we can do in such a situation is to compare the local atomic motions in topologically similar constituent structural units in different frequency ranges in the glass and the crystal.

The local projectional analysis uses the information about atomic displacements contained in the vibrational eigenmodes \( \{ \mathbf{e}_{\xi,i} \} \) with eigenfrequencies \( \{ \omega_{\xi} \} \). Here the index \( i \) enumerates the atoms and the index \( \xi \) labels the eigenmodes. Let us introduce the normalized local displacement vector \( \{ \mathbf{u}_{\xi,j}^{(l)} \} \) for a structural element \( l \): \( \mathbf{u}_{\xi,j}^{(l)} = \mathbf{e}_{\xi,j} / |p_l(\omega_{\xi})|^{1/2} \), where \( j \) enumerates the atoms within the structural unit, and \( p_l(\omega_{\xi}) = \sum_j |\mathbf{e}_{\xi,j}|^2 \). In the case of a crystal, an eigenmode is identified by the wavevector \( \mathbf{k} \) and the dispersion branch \( \beta \). For a crystalline structural unit \( l \), a disordered structural unit \( m \), a crystalline eigenmode \( \mathbf{k}\beta \) and a disordered eigenmode \( \mu \), we can define the squared scalar product:

\[
A_{l,m}(\omega_{k\beta}, \omega_\mu) = |\mathbf{u}_{k\beta}^{(l)} \mathbf{u}_{\mu}^{(m)}|^2 ,
\]

where \( \mathbf{u}_{k\beta}^{(l)} , \mathbf{u}_{\mu}^{(m)} \) have \( 3N_u \) components, \( N_u \) being the number of atoms within the structural unit. This quantity depends on the mutual orientation of the crystalline and disordered structural units. We are interested in finding the crystalline eigenmode which most resembles a given disordered mode and, therefore, we choose the maximum value (among all possible mutual orientations preserving the symmetry) of the squared scalar product,

\[
\hat{A}_{l,m}(\omega_{k\beta}, \omega_\mu) = \max\{A_{l,m}(\omega_{k\beta}, \omega_\mu)\} .
\]

The next step is to average \( \hat{A}_{l,m}(\omega_{k\beta}, \omega_\mu) \) over all possible disordered and crystalline structural units, and thus calculate the averaged squared scalar product, \( \mathcal{A}(\omega_{k\beta}, \omega_\mu) \),

\[
\mathcal{A}(\omega_{k\beta}, \omega_\mu) = \frac{1}{P} \sum_{l,m} p_l(\omega_{k\beta})p_m(\omega_\mu)\hat{A}_{l,m}(\omega_{k\beta}, \omega_\mu) ,
\]

where the local normalization factors, \( p_l(\omega_\mu) \), take into account the weight of the corresponding structural unit in the entire mode and \( P = \sum_{l,m} p_l(\omega_{k\beta})p_m(\omega_\mu) \). The averaged scalar products \( \mathcal{A}(\omega_{k\beta}, \omega_\mu) \) can be reduced to a function of two continuous variables \( \mathcal{A}(\omega_{\text{cryst}}, \omega_{\text{dis}}) \) by an interpolation. In this case, \( \mathcal{A}(\omega_{\text{cryst}}, \omega_{\text{dis}}) \) quantifies the degree of local similarity between one disordered mode with a frequency about \( \omega_{\text{dis}} \) and a crystalline mode with a frequency about \( \omega_{\text{cryst}} \). One way to obtain \( \mathcal{A}(\omega_{\text{cryst}}, \omega_{\text{dis}}) \) from \( \mathcal{A}(\omega_{k\beta}, \omega_\mu) \), which was used in the present work, is to compile a histogram for a discrete set of equidistant values of \( \omega_{\text{cryst}} \) for an arbitrary set of \( \omega_\mu \equiv \omega_{\text{dis}} \), i.e. for each \( \omega_{\text{dis}} \),

\[
\mathcal{A}(\omega_{\text{cryst}}, \omega_{\text{dis}}) = \frac{1}{N(\omega_{\text{cryst}})} \sum_{k\beta'} \mathcal{A}(\omega_{k\beta'}, \omega_\mu) .
\]

Here \( k\beta' \) assume values for which \( \omega_{\text{cryst}} \leq \omega_{k\beta'} < \omega_{\text{cryst}} + d\omega \), where \( d\omega \) is the width of a histogram bin, and \( N(\omega_{\text{cryst}}) \) is the number of crystalline states in this spectral interval of width \( d\omega \).

III. STRUCTURAL MODEL

As a representative example of a topologically disordered model structure, we consider a single-component glass with predominantly icosahedral order (the IC glass) constructed by means of molecular-dynamics simulation with the use of a pair-wise interatomic potential \( V \) (all quantities used in this paper are expressed in Lennard-Jones reduced units \( \text{[21,22]} \) for more detail). The \( 3N \times 3N \)-dynamical matrix (the number of particles is \( N = 16000 \)) for a glassy minimum-energy configuration has been calculated and diagonalized, yielding all vibrational eigenvectors, \( \{ \mathbf{e}_{\mu,i} \} \), and eigenfrequencies, \( \{ \omega_\mu \} \) for this system. A good crystalline counterpart for the IC glass is the \( \sigma \) phase, a Frank-Kasper crystal \[23,24\].

For the local projectional analysis of the type described above, we have chosen two \( Z14 \) Frank-Kasper polyhedra (point group \( D_{6h} \), interpenetrating along the six-fold symmetry axes. This structural unit \( Z14_{32} \), comprised of 22 atoms, represents a short segment of the -72° disclination line \[23,24\]. Within the unit cell of the \( \sigma \) phase, there are 16 interior atoms (centers of \( Z14 \) polyhedra) of eight partially overlapping \( Z14_{32} \) units. In the 16000-atom sample of the simulated IC glass, 236 such units (some of them partially overlapping) covering 20% of all atoms have been identified. These atoms, together with their nearest neighbors, represent 52% of all atoms.

IV. RESULTS

A. Level-repelling effect

Fig. 3 shows the calculated \( \mathcal{A}(\omega_{\text{cryst}}, \omega_{\text{dis}}) \) for disordered eigenmodes from the low-, middle- and high-frequency parts of the spectrum (a set of crystalline eigenmodes uniformly covering the first Brillouin zone with \( 10^3 \) points in \( \mathbf{k} \)-space was used). As seen from this figure, \( \mathcal{A}(\omega_{\text{cryst}}, \omega_{\text{dis}}) \), as a function of \( \omega_{\text{cryst}} \), represents a broad distribution centered around \( \omega_{\text{cryst}} = \omega_{\text{dis}} \) the frequency of the crystalline spectrum for which the crystalline modes have the greatest overlap with the modes of the disordered system. This suggests that a disordered mode with a frequency about \( \omega_{\text{dis}} \) is generically related to crystalline modes about \( \omega_{\text{cryst}} \); in other words,
the first moment of this function:

$$\omega^{(1)}_{\text{cryst}}(\omega_{\text{dis}}) = \int \omega A(\omega, \omega_{\text{dis}}) d\omega / \int A(\omega, \omega_{\text{dis}}) d\omega .$$  

(5)

In Fig. 2(b), $\omega^{(1)}_{\text{cryst}}$, calculated both by the direct method and by subtracting a background from $A(\omega_{\text{cryst}}, \omega_{\text{dis}})$, is plotted versus $\omega_{\text{dis}}$.

From Fig. 2, it is evident that the peak-shaped function, $A(\omega_{\text{cryst}}, \omega_{\text{dis}})$, becomes increasingly broader with increasing frequency, $\omega_{\text{cryst}}$, meaning that there is an increasingly weak correlation in character between modes in the disordered system and specific similar modes in the crystal. Such a large broadening of the peaks at high frequencies has also been found in lattice model [31] subject to large force-constant disorder and, in this sense, it is not surprising and reflects the large degree of disorder in the IC-glass. A similar effect is known for vibrational plane waves (analog of crystalline eigenmodes) propagating in glass. The distribution of the weights of different disordered eigenmodes contributing to a propagating plane wave becomes increasingly broad with increasing plane-wave frequency (the strong-scattering regime). The shape of this distribution can even approach the shape of the entire frequency spectrum of a glass. However, at low frequencies, and specifically in the boson-peak region, $\omega_{\text{BP}} \simeq 2.5$, the peak width of $A(\omega_{\text{cryst}}, \omega_{\text{dis}})$ is rather narrow, indicating that there is quite a strong correlation between the two types of modes there (this is analogous to the weak-scattering regime for plane waves).

The deviation of the curve $\omega^{(1)}_{\text{cryst}}(\omega_{\text{dis}})$ from the straight-line bisector, $\omega^{(1)}_{\text{cryst}} = \omega_{\text{dis}}$, reflects the level-repelling effect. It is clearly seen that the disordered modes from the low-frequency region are pushed down in frequency as compared to the related crystalline frequency, while the disordered modes from the high-frequency part of the spectrum are shifted upwards with respect to $\omega^{(1)}_{\text{cryst}}$.

Why do we describe this phenomenon as a level-repelling effect? One can argue alternatively that disordered systems are “softer” than their crystalline counterparts, and this results in a smaller sound velocity and in excess low-frequency eigenmodes which have nothing to do with the level-repelling effect. In the high-frequency range, the extra modes could be due to vibrations in local structural blocks characterized by relatively large (due to statistical fluctuations) spring constants which are also not related to the level-repelling effect. These arguments are not based on particular assumptions about the topology of the disordered system and therefore should be valid for disordered lattices as well. However, we have demonstrated the relevance of the level-repelling effects in describing vibrational spectra of disordered lattices with force-constant disorder [31]. This has been done within the mean-field approximation (homogeneous disorder), ignoring the role of statistical fluctuations in the force-constant distributions. We have explained the appearance of the low-frequency excess modes (with the VDOS...
\( \propto \omega^2 \) in terms of the well-known effect of the shift of the low-frequency part of the spectrum of quasi-particles to lower frequencies due to level-repelling effects. The mean-field approach is very accurate in the low-frequency regime. This demonstrates the relative unimportance of local soft regions (soft configurations) characterized by relatively small force constants which are due to statistical fluctuations. These regions, of course, exist but the low-frequency eigenmodes are not localized at them due to the delocalized properties of the vibrational states spectrum around zero frequency. We have also examined and confirmed this by means of multifractal analysis (to be published elsewhere). This is in contrast to the high-frequency part of the VDOS where the mean-field approach describes well only the main features and tendencies in the spectrum, such as the shift to higher frequencies (again due to level-repelling effects) but not the band tails containing localized states where statistical fluctuations of the force constants are very important.

Thus, bearing in mind the importance of level-repelling effects in determining the spectrum of disordered lattices, we conjecture that a similar effect can also be important in topologically disordered structures. By this we mean level-repelling effects relative to the spectra of corresponding crystalline counterparts of disordered systems which are caused by positional and topological disorder. The crystalline counterparts are characterized by the same local order as in the corresponding disordered structures. In topologically disordered systems, the atoms in local structural units are displaced from their ideal crystalline positions and the values of the force-constants are distributed, giving a situation resembling lattice models with force-constant disorder. However, the force constants of bonds emanating from a given central atom can be correlated with each other (see also Ref. 27). This situation is in contrast to lattice models with uncorrelated force constants. The strength and role of these correlations in the vibrational spectrum are not presently known. Another distinctive feature of glassy systems is the presence of topological disorder, the role of which in vibrational spectra is not yet established. In our current analysis, we try to find out to what extent the "lattice ideology" (including the level-repelling effect) is applicable to glassy models.

### B. The boson peak

Fig. 2(b) can be used to establish the nature of disordered modes in different frequency ranges. The low-frequency regime is of particular interest for vibrations in glasses, because of the so-called boson peak (BP) which occurs in the reduced VDOS, \( g_{\text{dis}}(\omega)/\omega^2 \), [see Figs. 2(a) and 3] and in the temperature dependence of the (vibrational) heat capacity \( C_v \), normalized by the Debye \( T^3 \) dependence [the solid line in Fig. 4].

The vibrational contribution to the heat capacity per particle is given by

\[
C_v(T) = \frac{1}{N} \sum_{j=1}^{3N} \left( \frac{\hbar \omega_j}{T} \right)^2 \frac{\exp(\hbar \omega_j/T)}{[\exp(\hbar \omega_j/T) - 1]^2},
\]

(6)

Since all quantities in this paper are expressed in Lennard-Jones reduced units, we use the value of \( \hbar \approx 0.02959 \) referred to argon. The Debye approximation of the above equation is given by

\[
C_v^D(T) = \frac{12 \pi^4}{5} \left( \frac{T}{\theta_D} \right)^3,
\]

(7)

where \( \theta_D = \hbar \omega_D = 2\pi \hbar (9N/4\pi V)^{1/3} c_D \) is the Debye temperature. The Debye velocity of sound \( c_D \) is defined as \( c_D = (1/c^3_t + 2/c^3_t)^{-1/3} \), where \( c_t \) and \( c_l \) are the
longitudinal and transverse sound velocities respectively, which can be estimated from the slopes of the acoustic dispersion branches. The Debye velocities for the IC glass and the σ phase are $c_{\text{dis}}^{\text{IC}} \approx 3.43$ and $c_{\text{dis}}^{\text{cryst}} \approx 4.74$, respectively.

In Fig. 4, the peak in the curve of $C_\nu/T^3$ versus $T$ is due to states in the boson-peak range of frequencies in the vibrational spectrum. Indeed, if we cut the low-frequency part of the spectrum, including the boson peak, then the peak in $C_\nu/T^3$ is significantly suppressed (see the dash-dotted line in Fig. 4). The heat capacity of the IC-glass can be easily compared with that of the σ-phase crystal. It can be seen from Fig. 4 that the plot of $C_\nu/T^3$ for the σ-phase crystal [the circles in Fig. 4] tends to the Debye limit (the dotted line) at low temperatures. The increase above this level at higher $T$ is due to the influence of van Hove singularities in the crystalline VDOS. This peak for the σ-phase crystal closely resembles the peak for the IC-glass, when the modes in the boson-peak region of the VDOS are omitted from the calculations. However, the peak in the total curve of $C_\nu/T^3$ for the IC glass is significantly shifted to lower frequencies, and also approaches a higher value of the Debye limit (the dashed line in Fig. 4) because of a lower value of the sound velocity.

Following the arrows in Figs. 2(a)-(c), it appears that the vibrational states in the boson-peak region mainly correspond to crystalline states in the vicinity of the lowest van Hove singularity in the σ phase. Although this method of analysis is unavoidably approximate, it does seem to confirm the physical picture for the origin of the boson peak previously found in the f.c.c. force-constant disordered lattice and in models of disordered silicon.

Assuming such a scenario for the boson-peak origin, we can compare its position, $\omega_{\text{BP}} \approx 2.5$, with the position of the lowest van Hove singularity, $\omega_{\text{VH}} \approx 4$, and thus conclude that $\omega_{\text{VH}} - \omega_{\text{BP}} \approx \omega_{\text{VH}}$, which is an indication of strong disorder. In this model, level repelling of low-frequency optic modes to even lower frequencies signifies contributions to the excess mode density in the boson-peak region (see below).

### C. Branch-hybridization effects

The level-repelling effect in glass is accompanied by another general effect, namely strong disorder-induced hybridization of crystalline eigenmodes. In this effect, many crystalline eigenmodes contribute to a particular disordered eigenmode (cf. the broad distributions of $A(\omega_{\text{cryst}}, \omega_{\text{dis}})$ in Fig. 3). Disorder-induced hybridization also involves hybridization between different crystalline vibrational branches. A quantitative characteristic of such hybridization can be defined via the branch hybridization parameter, $w_\beta(\omega_\mu)$:

$$w_\beta(\omega_\mu) = \sum_k A(\omega_{k\beta}, \omega_\mu) / \sum_{k\beta} A(\omega_{k\beta}, \omega_\mu).$$

Knowing $w_\beta(\omega_\mu)$, we can say which crystalline branch mainly contributes to a particular disordered eigenmode. In particular, this is of interest for the low-frequency regime, where only the acoustic branches exist in the crystal, but where we could also expect an admixture of low-lying optic branches in a glass due to hybridization effects. The results of a calculation of the hybridization parameter for acoustic, $w_{\text{ac}} = \sum_{\beta=1}^3 w_\beta$, and optic, $w_{\text{opt}} = \sum_{\beta=4}^{3N_{\text{u.c.}}} w_\beta$, branches are presented in Fig. 5 (with $N_{\text{u.c.}}$ being the number of atoms in the crystalline unit cell; $N_{\text{u.c.}} = 30$ for the σ phase). Here we have averaged the hybridization parameter $w_\beta$ over all acoustic and all optic branches in order to demonstrate the relative role of these types (acoustic or optic) vibrations in disordered modes. As seen from Fig. 5, even for the lowest-frequency disordered modes, the contribution of optic crystalline modes is dominant [$w_{\text{opt}}(\omega_{\text{dis}} \approx 1) \approx 0.8$] which indicates very strong hybridization effects with the dominant number of optic modes. Precisely because of such strong hybridization, it is not possible to distinguish between the contributions of acoustic branches of different polarizations (transverse or longitudinal). The branches of different polarization are distinct for small wavevectors only, but this part of the dispersion is suppressed by the dominant contribution from all other wave vectors for which the polarization is not well defined. It is also seen in Fig. 5 that the contribution of the acoustic branches is maximum at low frequencies and then monotonically decays with increasing frequency (whereas the opposite behavior is, obviously, found for optic modes), thus indicating an enhancement of the hybridization with optic branches. Such behaviour is qualitatively different from the plateau behaviour found at low frequencies in the f.c.c. disordered lattice, where only acoustic modes exist, and is due to the existence of the extensive optic spectrum in the crystalline counterpart of the IC glass.
D. Distribution of the matrix elements in the dynamical matrix

Both level-repelling and hybridization effects have demonstrated the strong influence of disorder on properties of disordered and crystalline modes, thus indicating a strong degree of disorder in the IC glass. A possible way to quantify the degree of disorder is to compare the distribution of the dynamical-matrix elements in the IC glass with that in the σ phase (see Fig. 6). The positive-definite dynamical matrix consists of $D \times D$ blocks ($D$ stands for the dimensionality, $D = 3$ in this case). The elements of the diagonal blocks are subject to sum-rule correlations with the elements of the off-diagonal blocks. Therefore, there are four distinct distributions of the matrix elements: diagonal and off-diagonal elements in diagonal and off-diagonal blocks. In Figs. 6(a)-(b), we have plotted the representative distributions, $\rho^{\text{diag}}$ and $\rho^{\text{off}}$, for the traces of the diagonal elements in diagonal and off-diagonal blocks, respectively. The trace of the off-diagonal block (taken with opposite sign) for atom $i$ interacting with atom $j$ can be associated with the effective force constant, $\kappa_{ij} = -\sum_\alpha D_{i\alpha,j\alpha}$, for interactions between these atoms. Similarly the trace of the diagonal block for atom $i$ describes the effective force constant, $\kappa_{ii} = \sum_\alpha D_{i\alpha,i\alpha}$, for interactions of this atom with all other atoms. Two conclusions can be made from a comparison of these distributions for the IC glass and the σ phase. First, the distributions for the glass can be imagined as being generically obtained from those for the crystalline broadening the corresponding $\delta$-functions. Second, this broadening is strong enough so that no traces of the individual $\delta$-functions remain; this is an indication of a strong degree of disorder.

A natural question is, can we actually reproduce the distributions for the glass from the crystalline ones? The simplest way to try to do this is to introduce thermal (positional) disorder in the σ phase by increasing the temperature, and analyzing the dynamical matrix corresponding to the instantaneous non-equilibrium state in which all the atoms are displaced about their crystalline equilibrium positions (positional disorder). We have indeed found that, for such a force-constant disordered case, the distribution of the dynamical matrix elements (cf. the dashed and solid lines in Fig. 6(a) and (b)) and the VDOS for the thermally (positionally)-distorted σ phase and the IC glass are similar. This implies that the main features of the vibrational spectrum in a topologically disordered system can be essentially reproduced by introducing positional disorder in its crystalline counterpart, at least for the metallic-type system studied here. The existence of topological disorder, as distinguished from positional disorder about equilibrium crystalline positions, does not seem to play a major role in determining the character of the vibrational modes, at least in this case. However, we have not directly addressed the situation where there are differences in the topological connections of atoms with constant force constants.

We have also found that the distribution of the matrix elements for atoms belonging to $Z14_2$ units involved in the present analysis is similar to the total distribution (see Fig. 6(a)). This confirms the representative character of the $Z14_2$ units for which we have performed the local projectional analysis.

V. CONCLUSIONS

To conclude, we have presented evidence that the vibrational modes in a topologically disordered glass are generically related to the eigenmodes in the corresponding crystalline counterpart via disorder-induced level-repelling and hybridization effects. In particular, the extra states in the low-frequency regime (boson peak) appear to correspond to crystalline states in the vicinity of the lowest van Hove singularities in the crystalline VDOS. We have proposed a way of defining the degree of disorder (weak or strong) by comparing the distribution of the dynamical matrix elements for a topologically disordered structure with its crystalline counterpart. It appears that the main features of the vibrational spec-
trum of a topologically disordered (metallic) glass may be reproduced by the vibrational dynamics of its positionally (thermally) distorted crystalline counterpart.

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1. S. R. Elliott, *Physics of amorphous materials* (Longman, Harlow, 1990), 2nd ed.
2. W. Schirmacher, G. Diezemann, and C. Ganter, Phys. Rev. Lett. 81, 136 (1998).
3. T. S. Grigera, V. Martin-Mayor, G. Parisi, and P. Verrocchio, Phys. Rev. Lett. 87, 085502 (2001).
4. V. Martin-Mayor, M. Mezard, G. Parisi, and P. Verrocchio, J. Chem. Phys. 114, 8068 (2001).
5. S. N. Taraskin and S. R. Elliott, Phys. Rev. B 61, 12017 (2000).
6. S. N. Taraskin and S. R. Elliott, Phys. Rev. B 61, 12031 (2000).
7. S. N. Taraskin, Y. L. Loh, G. Natarajan, and S. R. Elliott, Phys. Rev. Lett. 86, 1255 (2001).
8. J. M. Ziman, *Models of disorder: the theoretical physics of homogeneously disordered systems* (Cambridge University Press, Cambridge, 1979).
9. V. Martin-Mayor, G. Parisi, and P. Verrocchio, Phys. Rev. E 62, 2373 (2000).
10. T.-M. Wu and R. F. Loring, J. Chem. Phys. 97, 8568 (1992).
11. Y. Wan and R. M. Stratt, J. Chem. Phys. 100, 5123 (1994).
12. G. Biroli and R. Monasson, J. Phys. A 32, L255 (2000).
13. R. Monasson, Eur. Phys. J. B 12, 555 (1999).
14. T. S. Grigera, V. Martin-Mayor, G. Parisi, and P. Verrocchio, cond-mat/0110129 (2001).
15. A. Barrat and M. Weigt, Eur. Phys. J. B 13, 547 (2000).
16. S. N. Taraskin and S. R. Elliott, Phys. Rev. B 56, 8605 (1997).
17. Y. Ding, T. Nanba, and Y. Miura, Phys. Rev. B 58, 14279 (1998).
18. S. N. Taraskin and S. R. Elliott, Phys. Rev. B 59, 8572 (1999).
19. M. Dzugutov, Phys. Rev. A 46, R2984 (1992).
20. M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids* (Clarendon Press, Oxford, 1987).
21. S. I. Sindyankin, S. N. Taraskin, M. Dzugutov, and S. R. Elliott, Phys. Rev. B 63, 184301 (2001).
22. S. I. Sindyankin, S. N. Taraskin, M. Dzugutov, and S. R. Elliott, Phys. Rev. B 62, 3223 (2000).
23. J. D. Harrop, S. N. Taraskin, S. I. Sindyankin, S. R. Elliott, and M. Dzugutov, J. Non-Cryst. Solids, to be published (2001).
24. R. Mosseri and J.-F. Sadoc, *Geometrical Frustration* (Cambridge University Press, 1999).
25. H. Ehrenreich and L. Schwarts, Solid State Phys. 31, 149 (1976).
26. S. John, H. Sompolinsky, and M. J. Stephens, Phys. Rev. B 27, 5592 (1983).
27. M. Mezard, G. Parisi, and A. Zee, Nucl. Phys. B 559, 689 (1999).
28. A. A. Maradudin, E. W. Montroll, G. H. Weiss, and I. P. Ipatova, *Theory of lattice dynamics in the harmonic approximation*, vol. Supplement 3 of *Solid State Physics* (Academic Press, New York, 1971), 2nd ed.
29. F. Finkemeier and W. von Niessen, Phys. Rev. B 63, 235204 (2001).
30. S. N. Taraskin and S. R. Elliott, J. Phys.: Cond. Matt. 13, 235204 (2001).
31. There is a non-zero background effect (due to the non-orthogonality of the local parts of $u_{k,\beta,j}^{(1)}$ and $u_{\mu,j}^{(m)}$) in the calculation of $\omega_{\text{cryst}}(k)$ which is clearly seen (solid circles in Fig. A) and is well defined for $\omega_\mu \equiv \omega_{\text{dis}} \lesssim 10$. For each value of $\omega_{\text{dis}}$, the background level can be estimated as the mean value of $\overline{\omega_{\text{cryst}}(k)}$ for $\omega_{\text{cryst}} > \omega_0$, where $\omega_0$ is an arbitrary cut-off frequency. The background can then be subtracted from $\overline{\omega_{\text{cryst}}(k)}$ and the corresponding renormalized value of first moment, $\omega_{\text{cryst}}^{(1)}$, can be calculated while assuming that $\overline{\omega_{\text{criys}} > \omega_0, \omega_{\text{dis}}}} = 0$. The sensitivity of this method can be assessed by changing the value of $\omega_0$. Fig. B(b) shows that the low-$\omega_{\text{dis}}$ data are the least sensitive with respect to the definition of the background level.