Elastic Constant Inhomogeneity and the Broadening of the Dynamic Structure Factor in One-Dimensional Disordered Systems

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The dynamical structure factor of a model disordered system consisting of spring-disordered harmonic linear chains is studied both analytically and numerically. The width of the acoustic excitation peak is observed to grow quadratically with the exchanged momentum Q; such behavior is shown to be due to spatial fluctuations of the local wavelength of the collective vibrational modes, which in turn is produced by the inhomogeneity of the interparticle elastic constants. The extent to which this mechanism can explain the Q^2-dependent broadening recently observed in many glasses both experimentally and numerically is discussed.

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The study of the nature of collective atomic excitations in disordered solids at wavelengths λ approaching the interparticle separation a, $a/\lambda \approx 0.1$–0.5, has received renewed interest in the last few years thanks to new experimental tools and to improved numerical techniques. On the experimental side, the dynamics of the collective excitations is often investigated via the dynamic structure factor $S(Q, \omega)$, i.e., the space-time Fourier transform of the particle density correlation function. The study of $S(Q, \omega)$ in the region of mesoscopic exchanged momentum ($Q = 1$–$10\ \mathrm{nm}^{-1}$) has become recently possible in disordered systems thanks to the development of the inelastic x-ray scattering (IXS) technique [1], by which many glasses [2] and liquids [3] have been studied. Although specific *quantitative* differences exist among different systems, all the investigated glasses show some common features that can be summarized as follows: (i) there exist propagating acousticlike excitations up to $Qa \approx 3$; (ii) the slope of the Q-ω dispersion relation in the $Q \to 0$ limit extrapolates to the macroscopic sound velocity; (iii) the broadening of the excitation peaks in $S(Q, \omega)$ follows a Q^2 law. These general features of $S(Q, \omega)$ have been confirmed by numerical calculations in different glasses, using both standard molecular dynamics simulations [4], and the normal mode analysis in the harmonic approximation [5].

The Q^2 dependence of the excitation broadening in the $Q = 1$–$10\ \mathrm{nm}^{-1}$ region has not yet received a theoretical explanation. Such dependence is the same as predicted by hydrodynamics, but this coincidence is only accidental. Indeed, in the IXS experiments, the broadening is found to be temperature independent and the Q^2 law is numerically found also in *harmonic* glass models; these results indicate that the origin of this behavior should be found in *structural* rather than *dynamical* properties, i.e., it should be associated with the atomic disorder in the glass and not with dissipative phenomena such as anharmonicity or relaxation processes. In order to get insight into the physical properties that may be at the origin of this dependence, we consider here harmonic systems, leaving apart all the difficulties due to dynamical processes such as, for example, anharmonicity. Moreover, in order to obtain simple analytical results, we also concentrate on one-dimensional (1D) systems, specifically on spring-disordered linear chains. These systems, as we will see, show the same broadening law as their 2D and 3D counterparts. We find that in disordered linear chains the Q^2 broadening of $S(Q, \omega)$ can be related to the spatial fluctuations of the elastic constants.

The dynamics of 1D disordered lattices has been thoroughly investigated in the past, and a recent overview on this subject can be found in [6]. One-dimensional systems often show peculiar characteristics; more specifically it is well known [7] that, at variance with $d > 1$ cases, in a (infinite) disordered linear chain all vibrational modes are localized and localization by itself contributes to the broadening of the inelastic peaks of $S(Q, \omega)$. However, as we will show, this effect can be disentangled. Our model consists of a linear chain of N particles ($i = 1, \ldots, N$) of mass M placed a distance a apart from each other ($x_i = ia$), and joined by next-neighbor springs (K_i) randomly chosen from a flat distribution [$\mathcal{P}(K)$] with extrema $K \pm \Delta K$ (mean $\mu_k = K$ and standard deviation $\sigma_k = \Delta K/\sqrt{3}$). The dynamics of the system can be expressed in terms of its eigenvalues (ω_p) and eigenvectors [$e_p(i)$], $p = 1, \ldots, N$ being the mode label. The high-temperature dynamic structure factor is expressed as

$$S(Q, \omega) = \frac{K_B T}{M} \frac{Q^2}{\omega^2} E(Q, \omega), \tag{1}$$

with

$$E(Q, \omega) = \sum_p |\tilde{e}_p(Q)|^2 \delta(\omega - \omega_p), \qquad \tilde{e}_p(Q) = N^{-1/2} \sum_i \exp(iQx_i) e_p(i). \tag{2}$$

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The eigenvectors and eigenvalues are the solutions of the secular problem, $\sum_j D_{ij} e_p(j) = \omega_p^2 e_p(i)$, where D_{ij} is the dynamical matrix. The basic properties of $S(Q,\omega)$ are contained in $E(Q,\omega)$, the squared space Fourier transform of the eigenvectors of modes with frequency ω_p "close" to ω. A typical eigenvector obtained from the diagonalization of the dynamical matrix is reported in Fig. 1. As is well known (see, for example, [8]), there exists a rather well defined wavelength but, with respect to the sine function expected in the case of an ordered system, three main differences can be noted: (i) the peak height is not constant, i.e., there exists an envelope which is localized in space; (ii) the wavelength λ (i.e., the distance between two next nearest nodes) is not constant, but is rather a space fluctuating quantity; its statistical distribution is reported in inset (a) of Fig. 1; finally, (iii) by analyzing in detail the eigenvector between two successive nodes [see insets (b) and (c)], some deviations from the simple sine law can be evidenced. In principle, all these three characteristics could contribute to the broadening of the inelastic peaks in $S(Q,\omega)$. In the following, we will show that the broadening is mainly due to fluctuations of the local wavelength and (in 1D systems) to localization. Effect (iii) has minor consequences, and its signature can be found only in the low-ω tail of $S(Q,\omega)$.

Let us first focus on effect (ii), i.e., the effect of spatial wavelength fluctuations. To show how these fluctuations can by themselves produce the Q^2 broadening, we build up a fictitious, model eigenmode, which possesses only characteristic (ii), i.e., a sequence of alternating positive and negative semiperiods of a sine function, each having a slightly different wavelength. If h_m ($m = 1, 2, \ldots$) are the positions of the nodes, we define a local wavelength $\lambda_m = h_m - h_{m-2}$ and compute the Fourier transform of such model eigenvector, i.e., $\tilde{e}(Q) \propto \sum_m \int_{h_{m-2}}^{h_m} dx \exp(iQx) \sin[2\pi/\lambda_m(x - h_{m-2})]$. This is then averaged over different realizations, assuming a Gaussian distribution of wavelengths centered at λ and with variance σ_λ^2. The calculation of $E(Q) = N^{-1}\langle|\tilde{e}(Q)|^2\rangle$ is straightforward but long; the resulting $E(Q)$, which is peaked at $Q = 2\pi/\lambda$, is quasi-Lorentzian near the peak itself and its half width at half maximum (HWHM), $\Gamma_F^{(Q)}$, is given by

$$\frac{\Gamma_F^{(Q)}}{Q} = \pi \frac{\sigma_\lambda^2}{\lambda^2}. \tag{3}$$

Equation (3) is merely a mathematical result; we need now to establish a relationship between σ_λ and the characteristics of disorder. In an ordered chain, ω and λ are related by $\lambda = \frac{2\pi}{\omega} a\sqrt{K/M}$. In the disordered chain, the fluctuations of K produce fluctuations of λ and for the local wavelength we will have

$$\lambda_m^{\mathrm{loc}} = \frac{2\pi}{\omega} a\sqrt{K_m^{\mathrm{eff}}/M}, \tag{4}$$

where K_m^{eff} is the effective elastic constant obtained by averaging the individual spring constants K_i that are found between h_{m-2} and h_m: $(K_m^{\mathrm{eff}})^{-1} = \sum_{i\in m} P_i K_i^{-1} / \sum_{i\in m} P_i$. Here, the P_i's are weights that take into account the fact that springs near the nodes are more effective (because highly stretched) than those near the antinodes (not stretched) in determining K_m^{eff}.

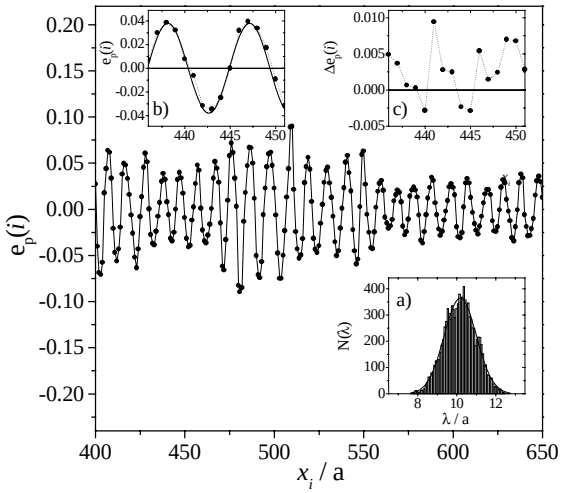

FIG. 1. Portion of the eigenvector of the mode at $\omega/\sqrt{K/M} = 0.571$ of a chain with $N = 1000$ particles and $\Delta K/K = 0.6$ reported as a function of the particle coordinate. In the insets, the most relevant features of the eigenvectors are emphasized: (i) Wavelength fluctuations [the distribution of λ_m calculated from 100 realizations of disorder is reported in inset (a) together with the Gaussian fit, $\bar{\lambda}/a = 10.18$, $\sigma_\lambda/a = 0.827$]; and (ii) deviation from sine curve [a blowup of the eigenvector (dots) is reported in the inset (b)] together with the best local sine approximation (full line). In the inset (c) it is reported the residual of the eigenvector after the subtraction of the full line in the inset (b).

The validity of Eq. (4), which is derived for the fictitious mode, has been tested numerically by diagonalizing the dynamical matrix of many realizations of disordered linear chains. In Fig. 2 we show the correlation between the diagonalized λ and λ^{loc} obtained from the analysis of the eigenvectors of the modes at $\omega/\sqrt{K/M} \approx 0.031$ of 1000 disordered chains with $\Delta K/K = 0.3$, for different choices of the weights P_j. As can be seen, the correlations are satisfactory, indicating the validity of the assumption that λ_m^{loc} is determined by an averaged local spring constant. Moreover, we observe that the highest correlation is found for weights proportional to the square of the local strain. From the previous relations, it is easily deduced that

$$\frac{\sigma_\lambda}{\lambda} = \frac{1}{2}\frac{\sigma_{K^{\mathrm{eff}}}}{K^{\mathrm{eff}}} = \frac{1}{2}\sqrt{\frac{\alpha}{n}}\frac{\sigma_k}{\mu_k}, \tag{5}$$

where $n = \langle n_m\rangle = \lambda/a$ and $\alpha = \langle P^2\rangle/\langle P\rangle^2$. In the following, we will choose P proportional to the square of the

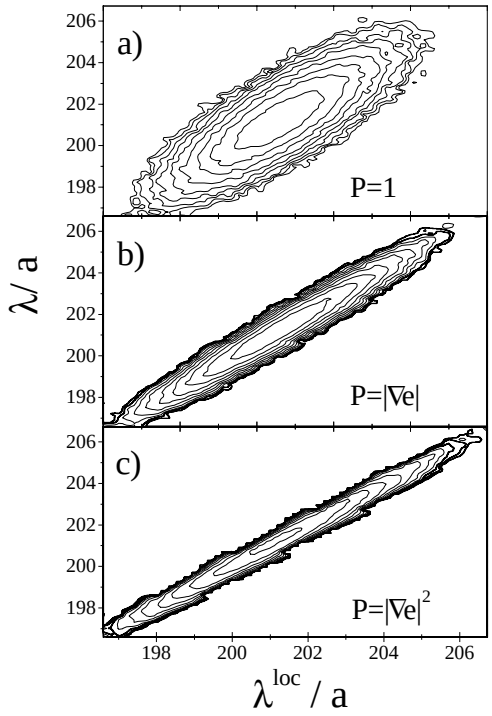

FIG. 2. Contour plot of the numerical joint distribution function $\mathcal{P}(\lambda, \lambda^{\mathrm{loc}})$ obtained by the analysis of the modes at $\omega/\sqrt{K/M} \approx 0.031$. The contour plot has been obtained averaging 1000 realization of disordered chains ($\Delta K/K = 0.3$) with $N = 1000$. The analysis has been performed with different choices for the weight: (a) $P = 1$; (b) $P = |\nabla e_p(i)|$; and (c) $P = |\nabla e_p(i)|^2$. The $\mathcal{P}$ scale is logarithmic and the ith line indicates the level at $10^{-i/4}\mathcal{P}_{\max}$.

strain and, therefore, $\alpha = 3/2$. By substituting Eq. (5) into (3), we obtain

$$\Gamma_F^{(Q)} = a\,\frac{\sigma_k^2}{\mu_k^2}\,\frac{\alpha}{8}\,Q^2, \tag{6}$$

which reproduces the Q^2 behavior. In summary, we have shown here that the spatial fluctuations of the "effective" (i.e., averaged over one wavelength) elastic constant, by themselves produce a quasi-Lorentzian, broadened $E(Q, \omega)$. These fluctuations become smaller and smaller as the wavelength (i.e., as the number of involved springs) increases and, in particular, from basic statistics it is $\propto \lambda^{-1/2}$. This result leads to a Q^2 dependence of the Brillouin linewidth at all Q values.

In order to check the validity of Eq. (6), we performed a numerical calculation of $E(Q, \omega)$. At selected values of ω, and for different values of ΔK, we calculated the eigenvectors of 50 different realizations of a disordered chain composed of 20 000 atoms, by using the Dyson-Schmidt (DS) method [9]. Using Eq. (2), the functions $E(Q, \omega)$ are then calculated as a function of Q and fitted by a Lorentzian; representative examples are reported in Fig. 3 for the indicated values of ω and $\Delta K/K = 0.6$. The HWHM Γ from the fitting are reported in the inset of Fig. 3, together with the prediction of Eq. (6) (dashed line), which reproduces

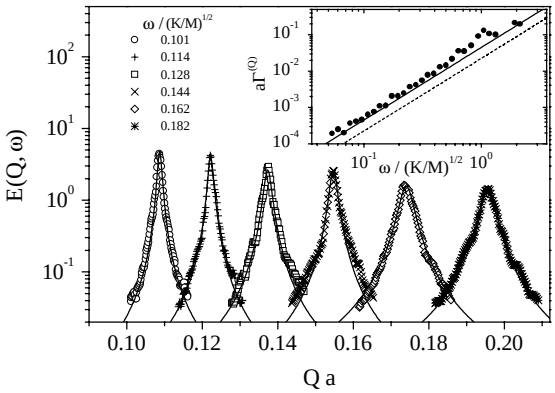

FIG. 3. Examples of $E(Q, \omega)$ vs Q at the indicated ω values calculated with the Dyson-Schmidt method [9] averaged over 50 realizations of disordered linear chains ($\Delta K/K = 0.6$) of length $N = 20\,000$. The full lines are the best Lorentzian fits. Inset: ω dependence of the HWHM of the Lorentzian fit to the $E(Q, \omega)$ reported in the figure (full dots), compared with the predictions from elastic constant fluctuations including (full line) or not including (dashed line) the localization effects.

correctly the Q^2 behavior but is about a factor 2 too small. This discrepancy is due to the omission of localization effects which, as mentioned before, are certainly effective in 1D systems. In fact, in these systems the eigenvectors have an exponential envelope in the tails, with a decay length L, and this contributes a term $\Gamma_L^{(Q)} = 1/L$ to the linewidth of $E(Q, \omega)$ [6]. Since localization also gives rise to a Lorentzian line shape, we expect a total broadening given by $\Gamma_{\mathrm{tot}}^{(Q)} = \Gamma_L^{(Q)} + \Gamma_F^{(Q)}$. The values of L were determined numerically by fitting the tails of the eigenvectors produced by the DS technique; as expected [6] we found $L \propto \omega^{-2}$, i.e., $\Gamma_L^{(Q)} \propto Q^2$ in this case also. The full line in the inset of Fig. 3 represents the total HWHM $\Gamma_{\mathrm{tot}}^{(Q)}$ and is in quantitative agreement with the widths of $E(Q, \omega)$ determined numerically. It is worth remarking that the effect of localization is, to a large extent, peculiar to 1D systems and is not relevant to 2D and 3D disordered systems [5].

The extension of the present results to 2D and 3D cases is not straightforward. Indeed, it has been recently numerically found [10] that the Q dependence of the Brillouin widths in spring-disordered 3D lattices is much richer than the simple Q^2 law found here for linear chains. Their $\Gamma^{(Q)}$ is proportional to Q^2 only at high Q, and shows a deviation towards a Q^4 behavior in the $Q \to 0$ limit. A similar behavior of the Brillouin linewidth has also been recently inferred for *topologically* disordered glasses [11], where $\Gamma^{(Q)}$ follows the Q^2 law at high Q and tends to a higher Q dependence at small Q. The exponent found at low Q in Ref. [11], however, is not as large as 4. The fact that (contrary to the 1D case where $\Gamma^{(Q)} \propto Q^2$ holds in any case) in 3D systems $\Gamma^{(Q)}$ tends to grow faster than Q^2 at long wavelength can be rationalized as follows. In $d > 1$, the effective elastic constant is no longer determined by averaging the springs along a direction parallel to the

propagation direction and extending for one wavelength, but one has also to average over orthogonal directions. For this reason, by increasing the wavelength, the number of springs which determine the effective spring increases faster than λ (which is the case in the 1D), the effective spring fluctuation decreases faster than $\lambda^{1/2}$, and, therefore, the Q dependence of $\Gamma^{(Q)}$ is faster than Q^2. This effect is more marked in the cases where the excitation wave fronts are planar and well defined, i.e., at long rather than at short wavelengths, and in disordered lattices rather than in topologically disordered systems.

In conclusion, we presented a mechanism that reproduces, in the 1D case, the Q^2 dependence of the broadening of the Brillouin peaks, which is observed experimentally at high Q ($Q = 1-10$ nm^{-1}) in a large variety of glasses. According to the present model, this broadening is a consequence of the spatial fluctuation of the effective (i.e., averaged over one wavelength) elastic constant. This fluctuation becomes smaller and smaller as the wavelength (i.e., as the number of involved springs) increases and, in particular, from basic statistics it is $\propto \lambda^{-1/2}$. This result leads to a Q^2 dependence of the Brillouin linewidth at all Q values.

[1] F. Sette, M. Krisch, C. Masciovecchio, G. Ruocco, and G. Monaco, Science **280**, 1550 (1998).

[2] C. Masciovecchio *et al.*, Phys. Rev. Lett. **76**, 3356 (1996); P. Benassi *et al.*, Phys. Rev. Lett. **77**, 3835 (1996); M. Foret *et al.*, Phys. Rev. Lett. **77**, 3831 (1996); A. Mermet *et al.*, Phys. Rev. Lett. **80**, 4205 (1998).

[3] F. Sette *et al.*, Phys. Rev. Lett. **75**, 850 (1995); G. Ruocco *et al.*, Nature (London) **379**, 521 (1996); F. Sette *et al.*, Phys. Rev. Lett. **77**, 83 (1996); C. Masciovecchio *et al.*, Phys. Rev. Lett. **80**, 544 (1998); G. Monaco *et al.*, Phys. Rev. Lett. **80**, 2161 (1998).

[4] D. Caprion, P. Jund, and R. Jullien, Phys. Rev. Lett. **77**, 675 (1996); S. N. Taraskin and S. R. Elliot, Phys. Rev. B **56**, 8605 (1997); J. Horbach, W. Kob, and K. Binder, Philos. Mag. B **77**, 297 (1998); M. C. C. Ribeiro, M. Wilson, and P. A. Madden, J. Chem. Phys. **108**, 9027 (1998); S. N. Taraskin and S. R. Elliot (to be published).

[5] J. L. Feldman, M. D. Kluge, P. B. Allen, and F. Wooten, Phys. Rev. B **48**, 12 589 (1993); H. R. Schober and C. Oligschleger, Phys. Rev. B **53**, 11 469 (1996); V. Mazzacurati, G. Ruocco, and M. Sampoli, Europhys. Lett. **34**, 681 (1996); R. Dell'Anna, G. Ruocco, M. Sampoli, and G. Viliani, Phys. Rev. Lett. **80**, 1236 (1998); J. L. Feldman, P. B. Allen, and S. R. Bickham (to be published).

[6] A. Crisanti, G. Paladin, and A. Vulpiani, *Products of Random Matrices in Statistical Physics* (Springer-Verlag, Berlin, 1992); J.-M. Luck, *Systemes Desordonnes Unidimensionels,* Alea-Saclay Vol. 2 (Commissariat à l'energie atomique, Gif-sur-Yvette, 1992).

[7] N. F. Mott and W. D. Twose, Adv. Phys. **10**, 107 (1961).

[8] K. Ishii, Suppl. Prog. Theor. Phys. **53**, 77 (1973).

[9] F. J. Dyson, Phys. Rev. **92**, 1331 (1953); H. Schmidt, Phys. Rev. **105**, 425 (1957).

[10] W. Schirmacher, G. Diezemann, and C. Ganter, Phys. Rev. Lett. **80**, 136 (1998).

[11] G. Ruocco *et al.* (to be published).