ORIGIN OF LIGHT SCATTERING FROM DISORDERED SYSTEMS

P. Benassi, W. Frizzera, M. Montagna and G. Viliani
Dipartimento di Fisica, Università di Trento
Povo, Trento, I-38050, Italy.

V. Mazzacurati
Dipartimento di Scienze e Tecnologie Biometriche e Biometria
Università dell’Aquila, Collemaggio, L’Aquila, I-67100, Italy.

G. Ruocco and G. Signorelli
Dipartimento di Fisica, Università dell’Aquila
Coppito, L’Aquila, I-67100, Italy.
ABSTRACT

Anelastic light scattering is computed numerically for model disordered systems (linear chains and 2-dimensional site and bond percolators), with and without electrical disorder. A detailed analysis of the vibrational modes and of their Raman activity evidences that two extreme mechanisms for scattering may be singled out. One of these resembles scattering from finite size systems, while the other mechanisms originates from spatial fluctuations of the polarizability and is such that modes in even small frequency intervals may have very different Raman activities. As a consequence, the average coupling coefficient $C(\omega)$ is the variance of a zero-average quantity. Our analysis shows that for both linear chains and 2-dimensional percolators the second mechanism dominates over the first, and therefore Raman scattering from disordered systems is essentially due to spatial fluctuations.
1. INTRODUCTION

Inelastic light scattering is a powerful tool to obtain information on the vibrational properties of solids. In the case of crystals, the existence of translational symmetry produces a selection rule which links the pseudo-momentum of the created or annihilated phonon, $\mathbf{k}$, to the momentum $\mathbf{q}$ exchanged between the photon and the system, i.e. $\mathbf{k} = \mathbf{q}$; as a consequence acoustic phonons produce Brillouin scattering, while optical phonons at the centre of the Brillouin zone ($k \ll \pi/a$ where $a$ is the interatomic distance) produce Raman scattering. The reason why phonons with $\mathbf{k} \neq \mathbf{q}$ do not scatter, is that in this case the electromagnetic waves produced by the polarizability modulations that are induced at different sites of the crystal by the modes, interfere in an exactly destructive way.

In disordered solids the situation is quite different. Brillouin scattering still produces a sharp and intense peak as in crystals. In fact, the wavelength of the light $\lambda \sim 5000\,\text{Å}$ is much greater than the correlation length of disorder so that the vibrational modes with such wavelength are much the same as phonons in crystals, i.e. plane waves which propagate in the disordered medium with a well defined $\mathbf{k}$. So the rule $\mathbf{k} = \mathbf{q}$ is still valid. In the case of Brillouin scattering it is customary to speak about ”coherent” scattering because the polarizability changes induced by the vibrations with $\mathbf{k} = \mathbf{q}$ scatter the light inelastically in a coherent way, which means that the electric field amplitudes scattered by atoms placed at different sites of the crystal by the modes, interfere in an exactly destructive way. In the following we will assume $q \simeq 0$ and we will not consider the Brillouin scattering any more in this paper.

Besides the Brillouin peaks, a continuum spectrum is present; this is due to the absence of spatial periodicity. In fact, the spatial fluctuations of the acousto optical constants prevent the above mentioned completely destructive interference among the scattered waves; this produces inelastic scattering which originates from the spatial variations of the electric polarizability (disorder-induced light scattering, DILS) and which has a continuum spectrum ranging from $\omega = 0$ up to some hundred wavenumbers. This is often called ”incoherent” scattering, though in our opinion this terminology can produce confusion.

In the harmonic approximation the DILS scattered intensity $I(\omega, T)$ can be put in the form \cite{1}:

$$I(\omega) \propto |n(\omega) + 1|C(\omega)\rho(\omega)/\omega$$

where $n(\omega, T)$ is the Bose-Einstein population factor, $\rho(\omega)$ is the density of vibrational states and $C(\omega)$ is the average light-vibration coupling coefficient for the modes whose frequency lies between $\omega$ and $\omega + \delta\omega$:

$$C_{\alpha\beta}(\omega) = \frac{\sum_p C_{\alpha\beta}(p)\delta(\omega - \omega_p)}{\sum_p \delta(\omega - \omega_p)}$$  \hspace{1cm} (1)
where

\[ C_{\alpha \beta}(p) = \left| \sum_{ij} \sum_{\gamma} \frac{\partial \pi_{i \beta}}{\partial u_{j \gamma}} \left[ \frac{e_{\gamma}(j|p)}{\sqrt{m_j}} - \frac{e_{\gamma}(i|p)}{\sqrt{m_i}} \right] \right|^2 = |B_{\alpha \beta}(p)|^2 \]  

which defines the quantity \( B_{\alpha \beta}(p) \) whose utility will become clear later. Note that since \( B_{\alpha \beta}(p) \) is a linear function of the atomic displacements \( e(j|p)/\sqrt{m_j} \) produced by mode \( p \), and since the sign of the \( e(j|p) \)'s is arbitrary, the only physically relevant quantity is \( |B_{\alpha \beta}(p)| \).

For low frequency acoustic modes (i.e. those whose wavelength is greater than the correlation length of disorder) what is expected is \( C(\omega) \propto \omega^2 \) \[2\], which has been observed experimentally in some systems \[3\]. At higher frequency the situation is more complicated.

The DILS Raman scattering is due to a spatially disordered distribution of the polarizabilities of the scattering units (ions, atoms or molecules). In infinite, mechanically and electrically ordered systems the scattering intensity vanishes. In disordered systems the spatial polarizability fluctuations produce a finite scattering intensity: these fluctuations can be due to mechanical disorder alone, to electrical disorder alone, or to both. The situation is somewhat different in finite systems, whose edges can be considered as regions where the polarizability varies from the value of the scattering units to that of vacuum, so that even fully ordered finite systems scatter.

Even though scattering arises in all cases from polarizability variations, it may be useful to define two extreme types of scattering. (i) Scattering that is produced by spatial polarizability fluctuations, in the sense that different sites have different polarizabilities. This kind of scattering is present both in mechanically ordered \[3\] and disordered solids. (ii) "Finite-size" scattering, i.e. the only contribution to scattering present in fully ordered finite systems; this is due to edge effects.

As we will see, even in mechanically disordered systems some of the scattering intensity can be ascribed to mechanisms very similar to edge effects. We will also see that these two scattering mechanisms have different characteristics which can be used to define them \textit{a posteriori}.

In the present paper we will study the relative importance of these two mechanisms; this depends in principle on the scattering system and on the frequency range. We will investigate two simple model systems: linear chains and two-dimensional site- and bond-percolators: the statistical method set up for linear chains will be utilized to study the percolators, which, at threshold, are fractal structures.

Raman scattering from supposedly fractal systems was studied in a number of recent experimental \[5\] \[6\] \[7\] papers. The experimental results seem to imply that the Raman coupling coefficient follows a power law \( C(\omega) \propto \omega^x \). Much numerical work \[8\] \[9\] \[10\] was devoted to determine \( x \) in percolators, while theoretical efforts \[11\] \[12\] \[13\] were devoted to derive scaling laws for \( C(\omega) \) in...
terms of the mechanical parameters (like the fractal dimension $D$, the spectral dimension $d$, and others) \[1\] \[2\]. In the next sections, we will show that for the systems under consideration the DILS mechanism is by far the dominant one.

The paper is organized as follows: in section 2 we present the results relative to linear chains with electrical and/or mechanical disorder. In section 3 we report the results on percolators, while the final section is devoted to discussion and conclusions.

2. LINEAR CHAIN

In order to better illustrate the scattering mechanisms proposed in the Introduction, we will apply the previous formalism to various situations relative to the linear chain.

Consider an ordered, infinite ($N \to \infty$) linear chain with equal masses and equilibrium positions $x^j = a_j$, $j = 1,...,N$, connected by equal springs. In this case, disregarding Brillouin scattering, and omitting the polarization subscripts, equation (2) becomes:

$$C(p) = \left| \sum_{ij} A_{ij} [e(j|p) - e(i|p)] \right|^2$$

(3)

where $A_{ij} = \partial \pi^i / \partial u^j$. The explicit form of $A_{ij}$ depends on the scattering mechanism used (Dipole-Induced-Dipole (DID), Bond Polarizability (BP), etc) but in any case it is an odd function, $A_{ij} = -A_{ji}$.

It is easy to see that $B(p) = 0$ due to cancellation effects which involve all the scatterers. In the case of a finite chain, on the contrary, $B(p) \neq 0$ due to the absence of complete cancellation for the scatterers at the edges. It is clear that ordered systems do not scatter, and it is only the breakdown of the translational symmetry of the polarizability which makes finite ordered systems slightly Raman active.

Another interesting case is that of electrical disorder in a mechanically ordered system; the coupling coefficient was derived in \[4\]; here we study the distribution $B(p)$ for one dimensional systems. Let us consider a linear chain of identical masses linked by identical next-neighbour springs of rest length $a$, and let us assume that the scattering mechanism is next-neighbour DID (ND) \[8\] (extension to three dimensions and full DID (FD) is straightforward). The electrical disorder is produced by randomly assigning to each mass a different isotropic, point-like polarizability $\alpha_i$. In this case

$$A_{(i-j)} = \begin{cases} \alpha_i \alpha_j T^{(3)}(a) & \text{if } i=j+1 \\ -\alpha_i \alpha_j T^{(3)}(a) & \text{if } i=j-1 \\ 0 & \text{otherwise} \end{cases}$$
where
\[ T^{(3)}_{\alpha\beta\gamma}(\vec{r}) = -\nabla_\alpha \nabla_\beta \nabla_\gamma (1/|\vec{r}|) \] (4)

It is easy to obtain, with straightforward but lengthy algebra, the following expression:
\[ B(p) = \frac{2}{\sqrt{N}} T^{(3)}(a)[e^{ik_p a} - 1] \left[ (1 + e^{-ik_p a})(\alpha) F + G \right] \] (5)

where
\[ F = \sum_j \delta \alpha_j e^{ik_p a_j} \] (6)
\[ G = \sum_j \delta \alpha_j \delta \alpha_{j+1} e^{ik_p a_j} \] (7)

For a large number of masses with polarizability \( \alpha \), and for many independent statistical replicas, the statistical distribution of the quantity \( F \) is a gaussian centered at \( \langle F \rangle = 0 \) and with a variance \( \sigma^2_F \equiv \langle F^2 \rangle = N \langle (\delta \alpha)^2 \rangle \). The distribution of \( G \) is also a zero-centered gaussian with \( \sigma^2_G = \langle \sigma^2_F \rangle/N \). Therefore the distribution of \( B(p) \), which is the convolution of two gaussians, is again a zero centered gaussian with a variance \( \sigma^2_B \) given by:
\[ \sigma^2_B = \frac{2 T^{(3)}(a)}{N} \left[ (1 + e^{-ik_p a})(\alpha) \sigma^2_F + \sigma^2_G \right]^2 \]
\[ \sigma^2_B = \frac{9}{4N} \left( \frac{2^8}{a} \right) \left[ \langle (\alpha)^2 \rangle (\delta \alpha)^2 \sin^2(k_p a) + \langle (\delta \alpha)^2 \rangle^2 \sin^2(k_p a/2) \right] \] (8)
in agreement with eqs. (22), (31), (35) and (36) of reference [4].

From this analysis it is clear that the origin of DILS is in fluctuations: in fact, \( C(p) \) is just the variance of the zero-average quantity \( B(p) \).

We consider next a linear chain with \( m_1 = 1 \) and \( m_2 = 2 \) randomly distributed with equal probability and linked by identical springs of elastic constant \( K = 1 \), with periodic boundary conditions. The maximum frequency is \( \omega_{\text{max}} = 2 \sqrt{K/\mu} \) where \( \mu \) is the lighter mass, and in the following \( \omega \) will be given in units of \( \omega_{\text{max}} \). In the infinite chain all modes would be localized [4], but in a finite chain the localization length of the lowest energy modes may be larger than the chain length, so that these modes appear not to be localized. Roughly speaking, there are two kinds of modes:

- acoustic-like modes;
- very localized modes.

We assign the polarizabilities \( \alpha_1, \alpha_2 \) to the masses \( m_1 \) and \( m_2 \) respectively. Vibrational eigenvalues and eigenvectors were obtained by diagonalizing the dynamical matrix of many replicas of the system. In Fig. 1(a) we report \( D(|B|) \),
i.e. the distribution of the $|B(p)|$ relative to the 2 modes with lowest frequency of 2000 chains containing 150 masses each, with $\alpha_1 = 0$ $\alpha_2 = 2$. $C(\omega)$ may be then obtained from equation (1) by averaging on the modes with energy $\omega \pm \delta \omega$ which are produced by the different realizations.

In order to test if $D(|B|)$ is a gaussian, we computed its first 10 moments:

$$M_n = \int dBD(|B|)|B|^n$$

and compared them with the corresponding ones, $M_n^G$, of the gaussian having the same zero and second moment. The ratios $M_n/M_n^G$ are reported in Table I together with the $\chi^2$ test values. We find that the gaussian distribution is a very good approximation for all acoustic-like modes (i.e. for $\omega < 0.5$), so that the introduction of mechanical disorder does not change the results we obtained for electrical disorder alone. For some frequency intervals for $\omega > 0.5$ important departures from gaussian shape are observed which show up in the appearance of peaks at well defined $|B|$ values (see Fig. 1(b)).

By choosing narrower frequency intervals on which the averages are taken it is possible to enhance the peaks (see Fig. 1(c)). The frequencies around which Fig. 1(a) and Fig. 1(b-c) were computed are indicated by the arrows in Fig. 2, where $C(\omega)$ is shown. The modes which produce the peaks at $|B| \neq 0$ are those where two neighboring light masses are surrounded by two long sequences of heavy masses, and oscillate in counter phase with $\omega \approx 0.9$, while the heavy masses are practically stationary. These modes scatter by a mechanism analogous to that of finite size systems: they cannot produce exact cancellation of the scattering amplitude.

The present analysis evidences therefore that there are two extreme mechanisms for Raman activity in disordered linear chains: DILS and finite-size scattering. The former produces a distribution $D(|B|)$ which is a gaussian centered at zero, while the latter produces peaks. In any case we have seen that the DILS mechanism is the largely dominant one when many masses are involved in the mode. Only in very narrow and high frequency ranges one can find modes localized on few atoms which give finite-size scattering.

### 3. SCATTERING FROM 2-DIMENSIONAL PERCOLATING STRUCTURES

The same analysis as in the previous section was performed on 2-dimensional site and bond percolators at percolation threshold, containing identical masses with identical bare polarizability. In the systems, each mass is bound to its nearest neighbours, the connecting springs are identical and scalar elasticity is assumed. Periodic boundary conditions are imposed and the eigenvalues are normalized to the maximum frequency of a full square lattice $\omega_{max} = 2\sqrt{2K/\mu}$. The Raman coupling coefficient was computed assuming that the scattering
mechanism is full DID, nearest neighbour DID, and Bond Polarizability (BP) \(^{[13]}\).

In two dimensions equation (2) for FD becomes \(^{[8]}\):

\[
C_{\alpha\beta}(p) = \left| 2\alpha^2 \sum_{ij} \sum_{\gamma} T_{\alpha\beta\gamma}^{(3)}(p) [e_{\gamma}(j|p) - e_{\gamma}(i|p)] \right|^2 = |B_{\alpha\beta}(p)|^2 \quad (9)
\]

To calculate the depolarized Raman scattering, \(B_{xy}(p)\) (or \(B_{yx}(p)\)) are used, while for the polarized scattering \(B_{xx}(p)\), \(B_{xy}(p)\) and \(B_{yy}(p)\) are used. We have computed separately the statistical distributions for the two groups of \(B\)'s (\(B_{\alpha\alpha}\) or \(B_{\alpha\beta}\) with \(\alpha \neq \beta\)). In Fig. 3 (a-c) we report the \(B_{\alpha\alpha}\) distributions relative to the modes in three frequency intervals (0.08-0.12, 0.37-0.43, 0.57-0.63, respectively) for 450 replicas of a 20×20 site percolator. For the two lowest-frequency intervals the distributions are very well fitted by gaussians, as shown in Table I. For the frequency range \(0.57 < \omega < 0.63\), important deviations from the gaussian shape are observed near the origin, and are quantified in Table I. In particular, there is an excess in the first channel, showing that many modes are little Raman active, which means that cancellation effects are very relevant. By expanding the abscissa (Fig. 4) it comes out that most of the excess modes are confined to a very small range of \(|B|\) values, of the order of \(\approx 10^{-5}\) times the value of \(\sigma\) of the distribution of Fig. 3 (c).

In order to gain more insight on the nature of these low activity modes, we tried to characterize their wavefunction by the localization length evaluated according to the Thouless definition:

\[
l_p = R_p^{-1/D}
\]

where \(D\) is the fractal dimension and \(R\) is the participation ratio:

\[
R_p = \frac{\sum_i |e(i|p)|^4}{(\sum_i |e(i|p)|^2)^2} \quad (10)
\]

In Fig. 5(a) we report the distribution of \(l\) for the \(\approx 10^4\) fractons with frequency \(0.57 < \omega < 0.63\), whose \(D(|B|)\) is shown in Fig. 3(c). As can be seen, in this frequency range fractons with very different localization lengths are present, in qualitative agreement with previous calculations \(^{[16]}\) \(^{[17]}\). If, from the modes which produce Fig. 3(c), we select those with \(|B| < 10^{-3}\) and consider the distribution of their localization lengths, we obtain Fig. 5(b). Most of these low-activity modes are localized on a few masses, and all have practically the same frequency (\(\omega \approx 0.61\)). Examples of this localized, low Raman active modes are in Fig. 6 (a-b) which shows that only few masses are involved in the mode (4 and 8 in Fig. 6(a) and 6(b) respectively), while all the other masses are practically stationary. In the same frequency range (\(\omega \approx 0.61\) however, there are many more modes (see Fig. 5(a)) involving many masses, as exemplified in Fig. 6(c).
In order to check if a correlation between Raman activity and localization length exists, we also studied the $D(|B|)$ relative to groups of modes having about the same $l$. Exception made for modes with $l < 3$ (which are those that produce the peak in $B = 0$ of Fig. 3(c)) all the others groups produce gaussian distributions for $D(|B|)$ with exactly the same variance and, therefore, the same $C(\omega)$. This result was to be expected as a consequence of DILS: in fact, within the bond polarizability model, $B(p)$ is the sum of $n$ fluctuating terms ($n \approx lD$ is the number of participating bonds), each having a mean squared amplitude $\approx 1/n$. This is true if, for a mass involved in mode $p$, $\langle (u_i - u_j)^2 \rangle \propto \langle u^2 \rangle$, because $\langle u^2 \rangle \propto 1/n$ for normalized modes. Thus in this case the quantity $\langle B^2 \rangle$ does not depend on $n$ or $l$, giving rise to the same $C(\omega)$ for modes with very different localization length.

Full DID in site percolators was studied as well. What we observe in this case is that the longer range of the FD interaction tends to smooth out the extremes of almost zero Raman activity observed above, so that it is less simple to single out the very localized modes, and the distributions look more like gaussians.

Bond percolators were also studied. $50 \times 50$ samples have been used to study the low frequency range (Fig. 7(a)). In this case and for FD, as Figs. 7 (b-c) show, there are some statistically significant departures (see Table I) from the gaussian shape at high frequencies, though the distributions still appear to be peaked at zero. This result was expected because in this case there may be disconnected pairs of nearest neighbours masses and since connected and disconnected nearest neighbours pairs give different DID contributions to $B$, a single gaussian distribution is unlikely to reproduce $D(|B|)$. On the contrary, BP produces gaussian-like distributions in this case also.

4. CONCLUSIONS

In the present work we studied electrically disordered linear chains with and without mechanical disorder, and two dimensional site and bond percolators without electrical disorder.

For the mechanically ordered linear chains, it was analytically shown that the distribution $D(|B|)$ obtained for each mode by randomly assigning the bare polarizability to the masses, is a gaussian centered at zero. Therefore, the Raman coupling coefficient of the modes is the variance of the distribution. By introducing mechanical disorder, no relevant difference is observed at low frequencies, while at higher frequencies $D(|B|)$ shows peaks centered either at zero or at finite values of $|B|$. The latter peaks are due to very localized, Raman active modes. Thus, this study singled out two scattering mechanisms: DILS (which is intrinsically due to spatial fluctuations) and finite-size scattering.

The same statistical analysis was performed on site and bond percolators, and what emerges is that the DILS mechanism dominates in all frequency ranges and for both scattering mechanisms, FD and BP.

The fluctuation origin of Raman scattering in these systems is shown by the Gaussian shape of the distribution $D(|B|)$.
FIGURE CAPTIONS

Fig. 1. Distribution of the $|B|$ values relative to 2000 replicas of mechanically and electrically disordered chains containing 150 masses each. (a) $m_1 = 1$, $m_2 = 2$, $\alpha_1 = 0$ and $\alpha_2 = 2$, the first two modes $0.005 < \omega < 0.018$. (b) $m_1 = 1$, $m_2 = 2$, $\alpha_1 = 0.9$, $\alpha_2 = 1.1$, for all the eigenvalues falling in the frequency range $0.8994 < \omega < 0.9050$. (c) Same as (b), but for $0.899450 < \omega < 0.899475$. 

Fig. 2. $C(\omega)$ for electrically and mechanically disordered linear chains. The arrows indicate the frequencies for which the statistical analysis relative to figures 1(a-b) was performed.

Fig. 3. Distribution of the $|B_{\alpha\alpha}|$ values computed in the bond polarizability model for all modes of 450 replicas of a 20×20 site percolator. (a): $0.08 < \omega < 0.12$; (b): $0.37 < \omega < 0.43$; (c): $0.57 < \omega < 0.63$. The arrow in (c) shows the number of modes which are left in the first channel neglecting those with $|B_{xx}|$ and $|B_{yy}| < 10^{-3}$.

Fig. 4. Same as Fig. 3(c), but plotted for $|B_{\alpha\alpha}| < 10^{-3}$.

Fig. 5. (a) Distribution of the localization lengths of $\approx 10^4$ fractons with frequency $0.57 < \omega < 0.63$, i.e. the same which produce Fig. 3(c). (b) Same as (a), but considering only the fractons with $|B_{xx}|$ and $|B_{yy}| < 10^{-3}$.

Fig. 6. Modes of a 20 × 20 site percolator with $\omega \approx 0.61$. The localization length are: (a) $l \approx 2.1$, (b) $l \approx 3.0$, (c) $l \approx 7.5$.

Fig. 7. Distribution of $|B_{\alpha\alpha}|$ values for modes of a bond percolator (FD). (a) $50 \times 50$ percolator in the range $0.014 < \omega < 0.018$. (b) $20 \times 20$ percolator in the range $0.37 < \omega < 0.43$ (c) $20 \times 20$ percolator in the range $0.57 < \omega < 0.63$

TABLE CAPTION

Table I. Ratios between the first 10 moments, $M_n$, of the distribution $D(|B|)$ and the moments of a gaussian having the same zero and second moment of $D(|B|)$. The values of the reduced $\chi^2$ are also reported.
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Table 1:

| Figures | $\chi^2$ | 1(a) | 3(a) | 3(b) | 3(c) | 7(a) | 7(b) | 7(c) |
|---------|---------|------|------|------|------|------|------|------|
| $M_1/M_1^C$ | 1.08 | 1.32 | 1.04 | 3.87 | 1.18 | 7.84 | 11.8 |
| $M_2/M_2^C$ | 1.00 | 1.01 | 1.00 | 1.00 | 0.99 | 0.97 | 0.97 |
| $M_3/M_3^C$ | 1.00 | 1.00 | 1.00 | 1.00 | 0.99 | 1.07 | 1.07 |
| $M_4/M_4^C$ | 1.01 | 0.97 | 0.97 | 0.97 | 1.03 | 1.07 | 1.07 |
| $M_5/M_5^C$ | 1.02 | 0.94 | 1.00 | 1.00 | 1.06 | 1.19 | 1.18 |
| $M_6/M_6^C$ | 1.04 | 0.99 | 0.99 | 0.99 | 1.11 | 1.36 | 1.33 |
| $M_7/M_7^C$ | 1.06 | 0.89 | 0.99 | 0.99 | 1.16 | 1.58 | 1.53 |
| $M_8/M_8^C$ | 1.09 | 0.85 | 0.89 | 0.89 | 1.20 | 1.86 | 1.79 |
| $M_9/M_9^C$ | 1.13 | 0.70 | 0.72 | 0.72 | 1.25 | 1.86 | 1.79 |
| $M_{10}/M_{10}^C$ | 1.16 | 0.65 | 1.00 | 1.00 | 1.25 | 3.08 | 2.00 |