

## A new approach for the analytic computation of the instantaneous normal modes spectrum

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2000 J. Phys.: Condens. Matter 12 6295

(<http://iopscience.iop.org/0953-8984/12/29/301>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.221

The article was downloaded on 16/05/2010 at 05:22

Please note that [terms and conditions apply](#).

## A new approach for the analytic computation of the instantaneous normal modes spectrum

Andrea Cavagna<sup>†§</sup>, Irene Giardina<sup>†||</sup> and Giorgio Parisi<sup>‡</sup>

<sup>†</sup> Theoretical Physics, University of Oxford, 1 Keble Road, Oxford OX1 3NP, UK

<sup>‡</sup> Dipartimento di Fisica, Università di Roma La Sapienza and INFN Sezione di Roma I, P.le Aldo Moro 5, 00185 Roma, Italy

E-mail: andrea@a13.ph.man.ac.uk, giardinia@spht.saclay.cea.fr and giorgio.parisi@roma1.infn.it

Received 15 February 2000

**Abstract.** In the context of the instantaneous normal mode approach, the spectrum of the Hessian of Hamiltonian is a key quantity to describe a liquids behaviour. The determination of the spectrum represents a major task for theoretical studies, and has recently been addressed in various works. In this work a new approach for the analytic computation of the Hessian spectrum is presented. The one-dimensional case for a system of particles interacting via a purely repulsive potential at low density is analysed in detail and, also in the localized sector, the spectrum is computed exactly. Finally the possible extensions of the method are discussed, together with a comparison with different approaches to the problem.

### 1. Introduction

In this work we describe an approach we have recently developed to compute analytically the so-called instantaneous normal mode (INM) spectrum of a liquid system [1]. That is, more precisely, the density of eigenvalues of the Hessian of the Hamiltonian, averaged over the equilibrium distribution.

The general frame where our computation acquires particular meaning, and where a great number of recent analyses have been performed, is the INM approach [2]. The main idea of this approach is that liquids are ‘solid-like’ at short times and that liquids’ dynamics thus correspond to vibrations about some equilibrium positions with periodic jumps into new local minima [3].

In this context, a crucial quantity is the typical spectrum of the Hessian of the Hamiltonian, which describes the structure of the energy landscape around the typical configurations. This is the INM spectrum and the related eigenvectors are the so-called INMs.

Recently there have been many attempts to quantitatively relate the knowledge of the INM spectrum with measurable quantities, such as the velocity–velocity correlation function, more complicated self-correlation functions, and even diffusion properties [2]. The main idea is to generalize the standard harmonic analysis used for solids, taking into account the

§ Current address: Department of Physics and Astronomy, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK.

|| Current address: Service de Physique Theorique CEA-Saclay, Orme des Merisiers, 91191 Gif sur Yvette Cedex, France.

finite hopping rate from one local minimum to another [2, 4]. For many systems simulations have shown that the predictions given by the INM analysis are in very good agreement with the results obtained directly from molecular dynamics. Simulations also show that the INM spectrum of a liquid system always displays both positive and negative eigenvalues (i.e. real and imaginary frequencies), not only in the liquid, but also in the supercooled phase as the glass transition is approached. While the interpretation of the positive modes seems straightforward (they represent harmonic vibrations in well defined wells), this is not the case for the negative modes. The contributions to these modes come from regions of negative curvature in the phase space sampled by the system at equilibrium. Indeed the system does not remain forever in a local minimum of the energy surface, but jumps into new regions over certain time scales, in this way exploring an enormously complicated landscape.

Unfortunately, it is not possible to distinguish, in a simple way, the modes related to real barriers (which can be used to give an estimate of the hopping rates, see [2]) from those related to anharmonic deformations of the local landscape. Recently it has been proposed [5] that a crucial difference exists between localized and extended negative modes: the localized modes involve a finite number of particles and can be associated with local barriers hopping; the extended modes involve an extensive number of particles and can be associated with structural rearrangements. Thus these two kinds of modes should be related to different diffusional processes. Simulations seem to confirm this view for some fragile models: in [5] it is shown that the number of extended negative modes goes to zero as a particular temperature is approached and that this temperature is the same as the temperature  $T_c$  where the mode coupling theory (MCT) would predict a dynamical transition [6]. The temperature  $T_c$  represents an important reference value for fragile glasses: above  $T_c$  MCT successfully predicts the observed dynamical behaviour of the system, and the viscosity has a non-Arrhenius temperature dependence; below  $T_c$  MCT breaks down and the viscosity behaviour becomes Arrhenius-like. It is commonly believed that  $T_c$  coincides with the crossover temperature postulated long ago by Goldstein [7], below which hopping processes become relevant. The result of [5] shows that, for the particular system studied there,  $T_c$  can be found via spectrum analysis, looking for the temperature where the extended negative modes disappear. It is not clear how general this phenomenon is, but, at least, it indicates that the localization properties of the INM represent a useful tool, and, moreover, can be used to give an estimate of the temperature  $T_c$ .

For all these reasons, the analytic determination of the INM spectrum represents a vital task for any theoretical study of liquids. Indeed, there have been in the last years various attempts to perform such a computation and some important steps in this direction have been performed in [8, 9]. In the following we will try to outline the standard procedure to compute analytically the INM spectrum and we will briefly mention what are the main assumptions and strategies adopted in these previous works [8, 9]. This will help us in introducing our new approach, commenting on what are the differences in perspective and procedure, and what are the objectives we propose to address.

## 2. General procedure

Let us consider a system of  $N$  interacting particles with the Hamiltonian

$$H = \sum_{k>1}^N V(r_{kl}) \quad (1)$$

where  $V(r)$  is a two-body potential. The Hessian matrix  $\mathbf{A}$  is defined by,  $A_{kl}^{\mu\nu} = \partial_k^\mu \partial_l^\nu H$ , with  $\mu, \nu = 1, \dots, d$ , being  $d$  the dimension of the space. The general form of  $\mathbf{A}$  with respect to

particle indices is

$$A_{kl} = -J_{kl} + \delta_{kl} \sum_i^N J_{ki} \tag{2}$$

where  $J_{kl} = J(\vec{r}_{kl}) = V''(r_{kl})\hat{r}_{kl}\hat{r}_{kl} + (V'(r_{kl})/r_{kl})(1 - \hat{r}_{kl}\hat{r}_{kl})$ ,  $\hat{r}_{kl}$  being the versor along the inter-particle  $k, l$  distance. The diagonal term of  $\mathbf{A}$  is a consequence of the translational invariance of the system, which requires  $\sum_k^n A_{kl} = 0$ .

The standard procedure to compute the INM spectrum is to consider the well known relation between the density of eigenvalues of the matrix  $\mathbf{A}$  and the corresponding resolvent operator  $\mathbf{G}(\lambda) = (\lambda\mathbf{1} - \mathbf{A})^{-1}$ :

$$D_A(\lambda) = \lim_{\epsilon \rightarrow 0} -\frac{1}{N\pi} \text{Im Tr} \mathbf{G}(\lambda + i\epsilon). \tag{3}$$

This formula provides the spectrum of a single matrix  $\mathbf{A}$ , that is of the Hessian matrix evaluated in a particular particles configuration. To obtain the INM spectrum  $D(\lambda)$ , one has to average (3) over the Boltzmann equilibrium distribution. In this way the computation of the INM spectrum is reduced to the computation of the average diagonal element of the resolvent matrix  $\langle G_{ii}(\lambda + i\epsilon) \rangle$ . The standard procedure is at this point to use an integral representation for the resolvent:

$$\langle G_{ii}(\lambda + i\epsilon) \rangle = \frac{1}{Z} \int d\phi_1 \dots d\phi_N \phi_i^2 \exp \left[ -\frac{1}{2} \phi(\lambda - \mathbf{A} + i\epsilon)\phi \right].$$

In this way some new variables, the internal fields  $\phi_i$ , enter in the computation, beside the space coordinates  $r_i$  included in the explicit expression of the Hessian  $\mathbf{A}$ . A further step consists in adopting a replica trick to bring the normalization factor  $Z$  at the numerator:  $\frac{1}{Z} = \lim_{n \rightarrow 0} Z^{n-1}$ . In this way we finally get

$$\langle G_{ii}(\lambda - i\epsilon) \rangle = \lim_{n \rightarrow 0} \int d\vec{\phi}_i (\phi_i^1)^2 \Omega(\vec{\phi}_i) \tag{4}$$

where now the  $\vec{\phi}_i$  are vectors in a  $n$ -dimensional space. The function  $\Omega$  is given by

$$\Omega(\vec{\phi}_i) = \int d\vec{\phi}_1 \dots d\vec{\phi}_N \langle e^{-\frac{1}{2} \vec{\phi}(\lambda - \mathbf{A} + i\epsilon)\vec{\phi}} \rangle \tag{5}$$

where the integration is performed over all, but the  $i$ , internal fields.

Of course the difficult task is to compute the function  $\Omega$ . In [8, 9], more or less explicitly, the authors assumed a Gaussian shape for  $\Omega(\vec{\phi})$ , and then computed self-consistently its variance using a sort of generalized liquid theory (we will come back to this point later). Thus, they assumed a quite rough approximation for the general  $\vec{\phi}$  dependence of  $\Omega$ , but accurately took into account the many-particles contribution terms included in the definition (5). Our approach has been precisely the opposite. As you will shortly see, we did not assume any *a priori* form for  $\Omega(\vec{\phi})$ , but we disregarded many-particles contribution terms. As a result, we have an approach which is much simpler to deal with, but still allows for non-trivial spectral properties. In [8, 9], excellent results were obtained for the density of eigenvalues, but, on the other hand, the computational procedure was too complicated to reasonably look at the localization properties of the eigenfunctions. Moreover, a Gaussian shape for  $\Omega$  is too simple to detect non-trivial localization properties. On the contrary, our simpler approach is more suitable for an eventual study of localization.

### 3. Our approach

We consider a liquid system at low density, that is precisely in the physical context where we can expect the many-particles correlations not to be important. At low density the most particles will be very far from one other and we can imagine that, picking out at random a couple of particles  $i$  and  $j$ , the interaction  $V(r_{ij})$  between them will be very low, practically zero. This will be true also for the Hessian element  $A_{ij}$  between the two particles, since the Hessian is much more short ranged than the potential itself. The whole matrix  $\mathbf{A}$  will therefore have the most of its elements equal to zero and very few greater than zero: it will be, in other terms, a *diluted* matrix. In this context, the main idea underlying our approach is to model the matrix  $\mathbf{A}$  as a random diluted matrix and to use all the techniques developed from random matrix ensembles to compute the INM spectrum.

As a first step in this program, we must find out what is the probability distribution of the matrix  $\mathbf{A}$ , or, which is the same, of the matrix  $\mathbf{J}$ . This distribution is naturally induced by the equilibrium probability over the positions of the particles via expression (2). Consistent with our low-density approximation, we can assume that the probability distribution  $P[\mathbf{J}]$  is factorized into the individual probabilities of the particles pairs. In this way the elements of  $\mathbf{J}$  (but not of  $\mathbf{A}$ ) are independently distributed, i.e.

$$P[\mathbf{J}] \equiv \prod_{k>l}^N p(J_{kl}). \quad (6)$$

It is clear that with (6) we are disregarding three-particles correlations. Thus, (6) becomes a reasonable assumption when three-particles correlations are not important, for example at low densities.

Once we have assumed this factorized form for  $P[\mathbf{J}]$ , we can express the pair probability  $p(J_{kl})$  as

$$\begin{aligned} p(J_{kl}) &= \int dr_1 \dots dr_N e^{-\beta H} \delta(J_{kl} - J(r_{kl})) \\ &= \frac{\rho}{N} \int dr_k dr_l g^{(2)}(r_{kl}) \delta(J_{kl} - J(r_{kl})) \end{aligned} \quad (7)$$

where  $g^{(2)}(r)$  is the two-particles correlation function and  $\rho$  is the average density. At this point there are various ways to exploit equation (7). One can, for example, insert in (7) the numerical values obtained for  $g^{(2)}(r)$  by numerical simulations, or by some classical liquid theory approaches. Alternatively, and this is the simplest possibility, we can adopt a low-density expansion for the two-points correlation function. In this way,  $g^{(2)}(r) = \exp(-\beta V(r))$  at the first order of the virial expansion. This approximation is consistent with our previous assumption (6) where we disregarded three-particles correlations, and, as we shall see, it enables us to perform completely analytically the calculations.

In the general frame we have described, we finally have the Hessian matrix  $\mathbf{A}$  and its distribution, as given by (7). This is enough to start a random matrix computation. However, the algebra is still very complicated: the matrices involved ( $\mathbf{A}$  and  $\mathbf{J}$ ) are actually tensors with particle and space coordinates indices. Before dealing with this general case, we decided to test our method under the simplest possible conditions, that is when the particles live in one dimension. This case is conceptually analogous to the three-dimensional case, with the advantage of a simpler algebra.

#### 4. The one-dimensional case

In one dimension the explicit expression of the matrix  $\mathbf{J}$  is much simpler, since only the longitudinal part of (2) survives, and we have  $J(r) = V''(r)$ . To obtain the distribution  $p(J_{kl})$  we have to solve equation (7). In the light of the greatest simplicity we consider a soft-spheres potential  $V(r) = 1/r^m$ . Thus, from equation (7) we have,

$$p(J) \sim \frac{1}{N} \frac{e^{-\hat{\beta} J^b}}{J^{1+c}} \equiv \frac{1}{N} q(J) \quad (8)$$

with  $\hat{\beta} = \beta[m(m+1)]^{1/(m+2)}$ ,  $b = m/(m+2)$  and  $c = 1/(m+2)$ . (From now on we indicate with  $J$  an individual element of the matrix  $\mathbf{J}$ .) For realistic values of  $m$  (typically  $m = 12$ ) the parameter  $b$  is very close to one. Therefore, we will directly set  $b = 1$  in  $p(J)$  in order to simplify our calculation. We will show in the discussion of our results that the actual spectrum is very weakly dependent on this approximation.

As it stands, the distribution  $p(J)$  is not normalizable, but we can regularize it in the following way. Let us put an IR cut-off  $\vec{r}$ , by setting  $V(r) = 0$  for  $r > \vec{r}$ , and let  $\eta = V(\vec{r})$ . In this way we obtain a regularized form of the pair probability:

$$p_\eta(J) = \delta(J) + \frac{1}{N} \left( q(J) \theta(J - \eta) - \delta(J) \int_\eta^\infty dJ' q(J') \right) \quad (9)$$

where  $q(J)$  is defined in equation (8). A few comments about equation (9):

- the distribution  $p_\eta(J)$  is *diluted* as we expected, since the probability of finding an element of the matrix  $\mathbf{J}$  equal to zero is of order one, while the probability of finding one element larger than  $\eta$  is of order  $1/N$ ;
- $p_\eta(J)$  explicitly depends on the value  $\eta$  of the cut-off. However, if we consider a generic function  $f(J)$  and compute its average over  $p_\eta(J)$ , we get:  $\langle f(J) \rangle \equiv \int_\eta^\infty dJ q(J) [f(J) - f(0)]$ . Thus, if the function  $f$  is differentiable in zero, its average value has a well defined limit when  $\eta \rightarrow 0$ . This means that in our computation, after averaging over the Hessian distribution, we can safely take the limit  $\eta \rightarrow 0$ , recovering the original problem without cut-off.

At this point we finally have a well defined random matrix problem: we have an ensemble of matrices  $\mathbf{A}$ , and their distribution, as given by equation (9). We can therefore try to apply random matrix techniques to compute the density of eigenvalues  $D(\lambda)$ .

Before going on with the computation, we would like to add some more general remarks on the one-dimensional case.

First of all, we note that the low-density approximation we made when disregarding three-particles correlations in (6) is, at fixed density and temperature, less appropriate the lower the dimension. For  $d = 1$  the geometrical constraints on the particles are much stronger and, indeed, the true Hessian matrix has a band structure. For this reason our analysis has to be regarded more as a training example for the three-dimensional case rather than a predictive computation for a real one-dimensional system (our one-dimensional treatment is very similar to what, for an electronic band structure problem, would be an *s*-band computation [10]).

Second, in the specific example we are dealing with we have chosen a soft-sphere potential  $V(r)$ . It is easy to see that in this case the Hessian  $\mathbf{A}$  is a positive defined matrix which therefore has a positive defined density of eigenvalues. This is not the case for a three-dimensional system, where the role of the negative modes is an important issue. However, the qualitative shape of the spectrum is actually very similar to that found, for example, in simulations on three-dimensional systems [11]. Moreover, the eigenfunctions exhibit non-trivial localization properties in the tails, thus providing a good context when testing a localization analysis

procedure. Finally, to avoid confusion in the future, we note once again that the matrix  $J$  is *not* a nearest-neighbours matrix, but a diluted matrix. This is why, even in one dimension, we find both localized and extended states (and not only localized states [12]).

Let us now proceed with the computation. The procedure is very similar to that outlined in the previous section for the standard computation of the INM spectrum: one has to relate  $D(\lambda)$  to the resolvent operator  $\mathbf{G}(\lambda)$ , and the diagonal element of the resolvent to the one-particle function  $\Omega$ . The main difference is that now all the averages involved in the definition (5) of  $\Omega$  are not averages over the Boltzmann distribution, but over the distribution  $p_\eta(J)$ .

The crucial point is, of course, the computation of  $\Omega(\vec{\phi}_i)$ . This can be performed in different ways, generalizing some random matrix computations [13] to the particular case of the distribution  $p_\eta(J)$  [1]. In our case it is convenient to write  $\Omega$  as (the particle index  $i$  is, from now on, understood)  $\Omega(\vec{\phi}) = e^{-\frac{1}{2}\lambda\phi^2 + g(\vec{\phi})}$  and look for a self-consistent equation for the exponent  $g(\vec{\phi})$ . We note that  $g(\vec{\phi})$  measures how much the function  $\Omega$  is Gaussian: a non-quadratic shape of  $g$  implies a non-Gaussian  $\Omega$ .

We will not enter into the details of how to obtain the self-consistent equation (the interested reader is referred to [1]), but simply give the result:

$$g(\vec{\phi}) = \int d\vec{\phi}' e^{-\frac{1}{2}\lambda\vec{\phi}'^2 + g(\vec{\phi}')} \int dJ q(J) [e^{-\frac{1}{2}J(\vec{\phi} - \vec{\phi}')^2} - 1]. \quad (10)$$

This equation has still to be averaged over the distribution  $q(J)$ . This can be achieved exactly with a few algebraic tricks, and finally we get

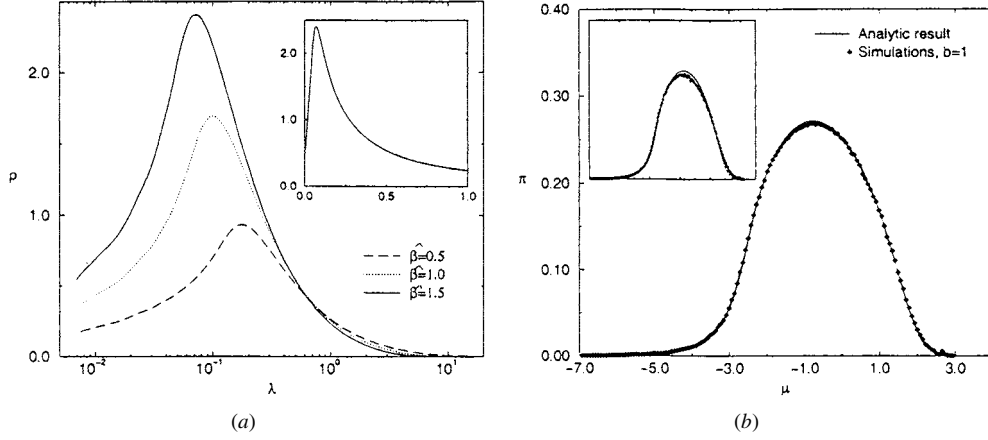
$$\hat{g}(x) = g(e^{i\pi/4}x) = K_1(x) - x \int_0^\infty dy K_2(x, y) \exp\left(i\frac{\lambda}{2}y^2 + \hat{g}(y)\right) \quad (11)$$

where  $x = |\vec{\phi}|$ , and  $K_1(x)$  and  $K_2(x, y)$  are expressed in an analytic form [1].

First of all, it is possible to check analytically that asymptotically  $g(x) \sim x^{2c}$ , thus proving that  $\Omega$  is definitely *not* a Gaussian function. In addition, we have been able to numerically solve the equation for  $g(x)$  without any further approximation. Indeed, (11) has the form of a fixed-point equation and can be solved numerically by iteration, discretizing the function  $\hat{g}$  and the kernel  $K$  on a lattice.

Once one has obtained  $g$  for a given value of  $\lambda$ , it is possible to compute the spectrum, using backward relations between  $\Omega$  and the resolvent, and finally between the resolvent and  $D(\lambda)$  (see previous sections). The results are shown in figure 1(a), where we have plotted the INM spectrum  $D$  as a function of  $\lambda$ , for  $m = 12$ . The spectrum has positive support and it depends on the scaled inverse temperature  $\hat{\beta}$  in the expected way: for low temperatures (high  $\hat{\beta}$ ) the collisions among particles are weaker, so that the spectrum is peaked on lower values of the eigenvalues. On the other hand, the tail for large  $\lambda$  is larger at higher temperatures. The behaviour in the right of the tail is of the form  $D(\lambda) \sim e^{-\alpha\lambda}$  where  $\alpha$  is an increasing function of  $\beta$ .

As previously stated, this one-dimensional case represents for us, first of all, a way to test our analytic procedure. A crucial task is therefore to check whether the result we have found is correct. To this aim we performed extensive numerical simulations. Once we have drawn a matrix  $\mathbf{J}$  with probability (9), we build  $\mathbf{A}$  and diagonalize it numerically. Since the spectrum has huge tails for large eigenvalues, it is convenient, in order to compare simulations with analytic results, to consider the probability distribution  $\pi$  of  $\mu \equiv \ln \lambda$ , that is  $\pi(\mu) = D(e^\mu) e^\mu$ . In figure 1(b) we plot  $\pi(\mu)$  as obtained from the analytic form of  $D(\lambda)$ , together with that obtained from numerical simulations. The two curves are in excellent agreement confirming the validity of our result. In addition, we show, in the insert of figure 1(b), the numerical spectrum obtained with the original value of  $b = m/(m+2)$ . The result justifies the sensibility of the approximation  $b \sim 1$ .



**Figure 1.** (a) The INM spectrum  $D$  as a function of  $\lambda$  for different values of the scaled temperature  $\hat{\beta}$ ,  $m = 12$  and  $\epsilon = 0$ . The plot is in log-linear scale. The insert shows  $D(\lambda)$  for  $\hat{\beta} = 1.5$  in linear-linear scale. The spectrum vanishes at  $\lambda = 0$ . (b) Comparison of numerical simulations and analytic solution. We plot here the probability distribution  $\pi(\mu)$ , with  $\mu = \ln \lambda$ . Both the curves correspond to a probability distribution with  $b = 1$ ,  $N = 600$ ,  $\eta = 10^{-4}$ ,  $\hat{\beta} = 1$  and  $m = 12$ . In the insert, on the same scale, we compare the analytic result for  $b = 1$  with the simulations performed with  $b = m/(m + 2)$ .

In the introduction we mentioned that an important issue in the context of the INM calculations is the analysis of the localization properties of the negative modes. As we said previously, even if in this one-dimensional case the spectrum is positive defined, the tails exhibit non-trivial localization properties. From a numerical point of view an important quantity in order to investigate localization properties is the average inverse participation ratio  $Y(\lambda_\alpha) = \sum_{i=1}^N (w_\alpha^i)^2$ , where  $\alpha = 1 \dots N$  is the eigenvalue index and  $w_\alpha^i = |\langle \alpha | i \rangle|^2$  is the weight of site  $i$  in the eigenfunction  $|\lambda_\alpha\rangle$ . In figure 2 we plot  $Y$  as a function of  $\lambda$ , as obtained via numerical diagonalization. It is clear from the figure that there are two localization edges, separating a central region of extended eigenvalues, from the tails where localized states are present. Note that for  $\lambda \rightarrow 0$  the inverse participation ratio goes to one and this corresponds to a single particle which happens to be nearly decoupled from the rest of the system. On the other hand, the localized states of the right of the tail correspond to pairs of very strongly interacting particles and this naturally leads to an inverse participation ratio equal to  $\frac{1}{2}$ .

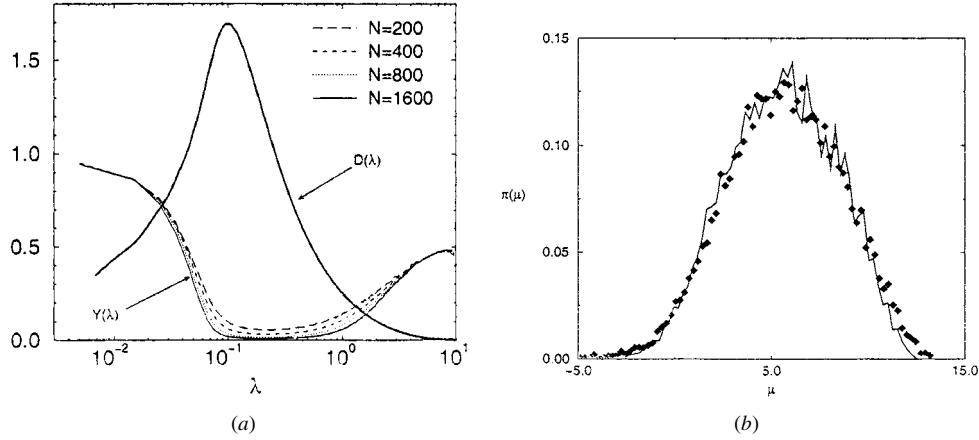
Our aim for the future is to find, analytically, the two localization edges revealed by the numerical analysis. Our idea is to generalize to our case the methods used for the Bethe lattice [14] and we will address this problem in a future work [15].

## 5. Comparison with other approaches

In order to better understand what are the differences between our approach and those of [8, 9], it is convenient to go back once again to the general procedure outlined in section 2, and, in particular, to expressions (4) and (5). A crucial observation made in [9] is that the function  $\Omega(\vec{\phi})$  is proportional to the one-particle correlation function of a generalized liquid system, whose coordinates are  $X_i = (r_i, \phi_i)$  and whose Hamiltonian is given by

$$-\beta H_{GEN} = \sum_i U(X_i) + \sum_{i < j} W(X_i, X_j) \quad (12)$$





**Figure 2.** (a) The inverse participation ratio  $Y$  as a function of the eigenvalue  $\lambda$  at different values of  $N$ .  $\eta = 10^{-4}$ ,  $\hat{\beta} = 1$  and  $m = 12$ . The thick curve is the spectrum  $D(\lambda)$  at the same values of the parameters. (b) Monte Carlo simulation for a three-dimensional soft-sphere mixture, at  $\Gamma = 0.2$ . The full curve represents the real INM spectrum. The points correspond to the spectrum obtained using a scrambled Hessian matrix.

where  $U(X_i) = -\frac{1}{2}\lambda(\vec{\phi}_i \cdot \vec{\phi}_i)$  and  $W(X_i, X_j) = -\beta V(r_{ij}) + A_{ij}(\vec{\phi}_i \cdot \vec{\phi}_j)$ . Indeed it is simple to check that

$$\Omega(\vec{\phi}) = \left\langle \frac{1}{N} \sum_i \delta(\vec{\phi} - \vec{\phi}_i) \right\rangle_{GEN} = s(X)/\rho$$

where  $s(X)$  is the one-particle correlation function of the generalized liquid system, and  $\rho$  is the average density of the original system. Given this analogy, it is possible to compute  $\Omega$  using the standard liquid theory [16] applied to this generalized liquid system. This is precisely what the authors of [9] did: they assumed a Gaussian shape for  $\Omega$ , and then computed the variance using a renormalized mean spherical approximation [16–18].

An alternative route is the following. We can use a hypernetted chain (HNC) approximation [16–18] to obtain the one-particle correlation function of the generalized liquid. The HNC equations can be deduced from a variational principle where the free energy of the system is written as a functional of the one-particle and two-particles correlation functions [17]. The variational free energy has the following form:

$$\begin{aligned} \beta F = & \frac{1}{2} \int dX dX' s(X)s(X')g^{(2)}(X, X') \log(g^{(2)}(X, X')) \\ & + \frac{1}{2} \int dX dX' s(X)s(X')[1 - g^{(2)}(X, X') + \beta W(X, X')g^{(2)}(X, X')] \\ & - \int dX s(X)U(X) + \int dX s(X)[\log(s(X)) - 1] \\ & - \frac{1}{N} \text{Tr} \left( \log(1 + sh) - sh + \frac{1}{2}(hshs) \right) \end{aligned} \quad (13)$$

where  $s(X)$  and  $g^{(2)}(X, X')$  are, respectively, the one-particle and the two-particles correlation functions,  $h(X, X') = g^{(2)}(X, X') - 1$  and in the trace term all the products are convolutions. Self-consistent equations for  $s(X)$  and  $g^{(2)}(X, X')$  are obtained variationally from (13). These equations are, in general, not easy to solve, but one can start facing them perturbatively. For example, we can look at a low-density situation where, in a first approximation, the trace

term will not contribute. In this case, the variational equation for the two-particles correlation function gives immediately  $g^{(2)}(X, X') = e^{-\beta W(X, X')}$ , which is nothing more than the first order of the virial expansion. The equation for the one-particle correlation function is, on the other hand, less trivial. In terms of  $\Omega(\vec{\phi}) = s(X)/\rho$  it gives

$$\log(\Omega(\vec{\phi})) + \frac{1}{2}\lambda\vec{\phi}^2 = \int d\vec{\phi}' \Omega(\vec{\phi}') \rho \left[ \int dr e^{-\beta V(r)} e^{\frac{1}{2}J(r)(\vec{\phi}-\vec{\phi}')^2} - N \right]. \quad (14)$$

The interesting fact about equation (14) is that it is exactly the same self-consistent equation that we get in our low-density approach presented in the previous section. To see this, we note that

$$\rho \int dr e^{-\beta V(r)} e^{\frac{1}{2}J(r)(\vec{\phi}-\vec{\phi}')^2} = \int dJ q(J) [e^{\frac{1}{2}J(\vec{\phi}-\vec{\phi}')^2} - 1] + N.$$

If we use now the previously-adopted notation  $\Omega(\vec{\phi}) = \exp(-\frac{1}{2}\lambda\vec{\phi}^2 + g(\vec{\phi}))$  and we insert it in (14) we finally get precisely equation (10) for  $g(\vec{\phi})$ .

This result shows that our simple low-density approach can be seen as the first-order approximation of a more complicated generalized HNC approach. We understand now in a deeper way what is the origin of the approximations we made and also what is the route we have to follow to improve our calculation. Indeed, we see that, if we want to go beyond the low-density approximation, we have to consider further terms in the trace appearing in (13) and consequently compute the self-consistent equations [15].

## 6. Conclusion

We have presented a new approach for the analytic computation of the INM spectrum. At present we have successfully applied this approach to the case where particles live in one dimension and at low density. This simple case represents the ideal context in which we understand the potential of our method and outlines the procedure one has to follow in more general cases.

We expect this approach to give good results in realistic three-dimensional situations, where the geometrical constraints are much less important than in one dimension. A strong indication in this direction is provided in figure 2(b), where we plot the spectrum obtained from a Monte Carlo simulation for a soft-sphere mixture. The full curve is obtained considering, for each sampled configuration, the real Hessian matrix  $\mathbf{A}$  of the system. The points, on the other hand, are obtained considering a 'scrambled' Hessian matrix where, for each sampled configuration, a new  $\mathbf{J}$  matrix is built by mixing at random the elements  $J_{ij}$  of the real  $\mathbf{J}$ . This scrambling procedure destroys the three-particles correlations and is therefore equivalent to our approximation (6). The figure clearly shows that the scrambled approximation works very well for  $\Gamma = \rho\beta^{1/4} = 0.2$ . A more systematic analysis [15] with Monte Carlo simulations indicates that good results are also obtained at much higher values of  $\Gamma$ , suggesting that our analytic approach, even at the simplest first step where three-particles correlations are discarded, should describe quantitatively well the real spectral properties of a three-dimensional system over a wide range of temperatures and densities.

## References

- [1] Cavagna A, Giardina I and Parisi G 1999 *Phys. Rev. Lett.* **83** 108
- [2] See for example Keyes T 1997 *J. Chem. Phys. A* **101** 2921
- [3] Stillinger F H and Weber T A 1984 *J. Chem. Phys.* **80** 4434  
LaViolette R A and Stillinger F H 1985 *J. Chem. Phys.* **83** 4079

- Sastry S 1996 *Phys. Rev. Lett.* **76** 3738  
Sciortino F and Tartaglia P 1997 *Phys. Rev. Lett.* **78** 2385  
Schröder T B, Sastry S, Dyre J C and Glotzer S 1999 *Preprint cond-mat/9901271*
- [4] Zwanzig R 1983 *J. Chem. Phys.* **79** 4507  
[5] Bembenek S D and Laird B B 1996 *J. Chem. Phys.* **104** 5199  
[6] Gotze W 1989 *Liquids, Freezing and the Glass Transition* ed J P Hansen, D Levesque and J Zinn-Justin (Amsterdam: North Holland, Les Houches)  
[7] Goldstein M J 1969 *J. Chem. Phys.* **51** 3728  
[8] Wu T-M and Loring R F 1992 *J. Chem. Phys.* **97** 8568  
[9] Wan Y and Stratt R M 1994 *J. Chem. Phys.* **100** 5123  
[10] Xu B-C and Stratt R M 1990 *J. Chem. Phys.* **92** 1923  
Chen Z and Stratt R M 1991 *J. Chem. Phys.* **94** 1426  
[11] Sastry S private communication  
[12] Thouless D J 1974 *Phys. Rev.* **13** 93  
Kramer B and McKinnon 1993 *Rep. Prog. Phys.* **56** 287  
[13] Cizeau P and Bouchaud J P 1994 *Phys. Rev. E* **50** 1810  
Rodgers G J and Bray A J 1988 *Phys. Rev. B* **37** 3557  
Bray A J and Rodgers G J 1988 *Phys. Rev. B* **38** 11 461  
Biroli G and Monasson R 1999 *Preprint cond-mat/9902032*
- [14] Abou-Chacra R, Anderson P W and Thouless D J 1973 *J. Phys. C: Solid State Phys.* **5** 1734  
Kunz H 1983 *J. Physique* **44** L411  
Economou E N and Cohen M H 1972 *Phys. Rev. B* **5** 2931  
Mirlin A D and Fyodorov Y V 1991 *J. Phys. A: Math. Gen.* **24** 2273  
Mirlin A D and Fyodorov Y V 1991 *Nucl. Phys. B* **366** 507
- [15] Cavagna A, Giardina I and Parisi G in preparation  
[16] Hansen J P and McDonald I R 1986 *Theory of Simple Liquids* (London: Academic)  
[17] Morita T and Hiroike K 1961 *Prog. Theor. Phys.* **25** 537  
[18] Frisch H L and Lebowitz J L (eds) 1964 *The Equilibrium Theory of Classical Fluids* (New York: Benjamin)