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A qualitative study of the instability of fractal aggregates

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Abstract. The instability of fractal aggregates is discussed qualitatively using the self-consistent harmonic approximation. The density of elastic vibrational states originating from phonon and elastic fracton modes is used to calculate the mean square displacement of lattice constituents, and the transition temperature is estimated for fractal aggregates with different dimensionalities of elastic fractons.

The instability of fractal aggregates is an interesting problem. Some important discussions of the thermal stability for some special fractal systems (DLA-like, etc) at finite temperatures have been made [1, 2]. In this paper, from a new viewpoint and approach which are based on the self-consistent harmonic approximation (SCHA) [3], we shall carry out a further study on the instability problem of a general fractal system with a nearest-neighbour interaction potential. Our discussions concern especially the occurrence of the instability, the cases of different fracton dimensionalities, the cases of general mixed-type fractals and the fluctuation of zero temperature, all of which have not been discussed by others as far as we know. Accordingly, some new and general conclusions have been obtained.

As is well known, the mean-square displacement (MSD) $\langle u^2\rangle = \Sigma_l \langle u_l^2\rangle/N$ is an important quantity related closely to elastic vibrational problems and the stability of long-range order (LRO) in the lattice. For an ordinary Debye crystal, the LRO of the lattice is broken down as the MSD $\langle u^2\rangle$ reaches a critical value [3, 4]. Similarly, the MSD $\langle u^2\rangle_{\mathrm{f}}$ will increase monotonically with increasing temperature for a given fractal lattice, and it could be proposed that the order of the fractal aggregates is broken down as the threshold u_{c}^2 is reached and the corresponding temperature at the onset is called the instability temperature. Since there is no order on the fractal lattice at least as $\langle u^2\rangle_{\mathrm{f}}$ reaches about a^2 (a is the lattice constant), the instability viewpoint for fractal aggregates is visible from direct physical considerations. Now, let us try to demonstrate the occurrence of the instability of fractal aggregates with the SCHA [3] and other appropriate approximations. It is reasonable to write the interaction potential for many fractal aggregates as

$$V_{ll'} = V(|\boldsymbol{R}_l - \boldsymbol{R}_{l'}|) \tag{1}$$

where (ll') denotes nearest neighbours and $\boldsymbol{R}_l$ is the position vector of site l. By making use of the SCHA, we can obtain a general Born potential with temperature-dependent coefficients. In three Euclidean dimensions, this is as follows [3, 5]:

$$V_{ll'} = \alpha_{ll'}(\boldsymbol{u}_l - \boldsymbol{u}_{l'})^2_{\parallel} + \beta_{ll'}(\boldsymbol{u}_l - \boldsymbol{u}_{l'})^2_{\perp} \tag{2a}$$

with

$$\alpha_{ll'} = \int \mathrm{d}^3q\, \alpha(q) \exp[\mathrm{i}(\boldsymbol{R}_l^0 - \boldsymbol{R}_{l'}^0)\cdot \boldsymbol{q}] \exp\left(-\frac{q^2}{6}\langle(\boldsymbol{u}_l - \boldsymbol{u}_{l'})^2\rangle\right) \tag{2b}$$

$$\beta_{ll'} = \int \mathrm{d}^3q\, \beta(q) \exp[\mathrm{i}(\boldsymbol{R}_l^0 - \boldsymbol{R}_{l'}^0)\cdot \boldsymbol{q}] \exp\left(-\frac{q^2}{6}\langle(\boldsymbol{u}_l - \boldsymbol{u}_{l'})^2\rangle\right) \tag{2c}$$

where $\boldsymbol{R}_l^0$ is the original position vector of the site l and $\boldsymbol{u}_l$ is the displacement vector of the site l. $(\boldsymbol{u}_l - \boldsymbol{u}_{l'})_{\parallel}$ is the relative displacement of the site l' in the direction parallel to the bond $\boldsymbol{R}_l^0 - \boldsymbol{R}_{l'}$, and $(\boldsymbol{u}_l - \boldsymbol{u}_{l'})_{\perp}$ is the relative displacement in the perpendicular direction. For simplicity, the isotropic case is considered in detail, i.e. $\alpha_{ll'} = \beta_{ll'} = \alpha$. Considering that the density of states (DOS) for elastic vibrational modes on fractals takes the form: $\rho_{\mathrm{fr}}(\omega) \propto \omega^{\tilde{d}-1}$ [6, 7], where $\tilde{d}$ is called the fracton dimensionality, we have for the DOS of pure fractals

$$\rho_{\mathrm{fr}}(\omega) = \alpha_{\mathrm{s}}^{-\tilde{d}/2}\rho_{\mathrm{fr}}^0(\omega) \tag{3}$$

where $\alpha_{\mathrm{s}} = \alpha/\{\int \mathrm{d}^3q\, \alpha(q) \exp[\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{R}_l^0 - \boldsymbol{R}_{l'}^0)]\}$, and $\rho_{\mathrm{fr}}(\omega)$ and $\rho_{\mathrm{fr}}^0(\omega)$ are DOSs corresponding to self-consistency and non-self-consistency, respectively. From equation (3), we can also easily obtain

$$\langle u^2\rangle_{\mathrm{fr}} = \alpha_{\mathrm{s}}^{-\tilde{d}/2}\langle u^2\rangle_{\mathrm{fr}}^0 \tag{4}$$

where $\langle u^2\rangle_{\mathrm{fr}}$ and $\langle u^2\rangle_{\mathrm{fr}}^0$ are the MSDs for pure fractals corresponding to self-consistency and non-self-consistency, respectively. Because $\langle u^2\rangle_{\mathrm{fr}}$ is much less than a^2 before the instability occurs (this will be demonstrated later), we neglect the specific dependence of various terms in the integral and the cross terms in the MSDs [3]. Now, for the bulk of fractal aggregates, we can simply write

$$\alpha_{\mathrm{s}} \simeq \exp(-2\lambda\langle u^2\rangle_{\mathrm{fr}}) \tag{5}$$

where a microscopic expression for λ can in principle be estimated: $\lambda \sim \pi^2/a^2$ [3]. In the above treatment, we also made use of the rough approximation within which the specific term in the exponent is replaced by the average value $\langle u^2\rangle_{\mathrm{fr}} = \Sigma_l\langle u_l^2\rangle/N$. This is reasonable because of the qualitative nature of our discussion, and the influence of the errors does not cause changes in the qualitative conclusion about the occurrence of the instability of the lattice. Solving equations (5) and (4), we find that a threshold exists: $u_{\mathrm{c}}^2 = 1/\lambda\tilde{d}e$. Beyond the threshold, equation (5) has no solution. This means that the order of original fractal structure is broken down and the so-called instability occurs. Moreover, for mixed fractal aggregates, in which the total size and the fractal characteristic length are defined as L and ξ, respectively, the threshold is estimated to be between $1/3\lambda e$ and $1/\lambda\tilde{d}e$. Similarly, the above results can be obtained for a centre-force potential, except that $\tilde{d}$ is different. It is also worth pointing out that the main qualitative conclusions can be generalised to the case $\alpha \neq \beta$. In other words, the order of fractal aggregates is broken down as the MSD $\langle u^2\rangle_{\mathrm{f}}^0$ reaches a certain value. Thus the equation for describing the transition temperature is established as

$$u_{\mathrm{c}}^2 = \langle u^2\rangle_{\mathrm{f}}^0(T_{\mathrm{c}}). \tag{6}$$

On the basis of equation (6), now let us discuss the instability problem in detail. As we know, the usual Debye-type DOS changes to the elastic fracton DOS for length scales less than the characteristic length ξ, corresponding to frequencies greater than a crossover frequency which scales as $\omega_{co} \sim \omega_{fD}\xi^{-(2+\sigma_e)/2}$. Here ω_{fD} is called the elastic fracton Debye frequency, which is the upper limit on the frequency region of fractons, and σ_e satisfies the relation $\tilde{d} = 2\bar{d}/(2 + \sigma_e)$ ($\bar{d}$ is the Hausdorff dimensionality of fractals). After some analysis, the numbers of fracton and phonon modes are given by

$$N_{fr} = nA_p(L/\xi)^3\{A_f(\xi/a)^{\bar{d}} - [1 - (\xi/L)^3]\} \tag{7a}$$

$$N_{ph} = nA_p(L/\xi)^3(1 - \xi/L) \tag{7b}$$

where A_p and A_f are proportionality constants depending on the structure of the fractal aggregates and n is the number of vibrational degrees of freedom. According to the normalisation condition, we have

$$\rho^0_{fr}(\omega) = N_{fr}\tilde{d}\omega^{\tilde{d}-1}/(\omega^{\tilde{d}}_{fD} - \omega^{\tilde{d}}_{co}) \tag{8a}$$

$$\rho^0_{ph}(\omega) = N_{ph}3\omega^2/(\omega^3_{co} - \omega^3_0) \tag{8b}$$

where $\omega_0 \sim \omega_{fD}(\xi/a)^{-\sigma_e/2}(L/a)^{-1}$. By using equations (8), the MSD can be calculated, i.e.

$$\langle u^2\rangle^0_f = u^2_{fr} + u^2_{ph} \tag{9}$$

with

$$\begin{aligned} u^2_{fr} &= u^2_0\frac{N_{fr}}{N}\frac{\tilde{d}}{3n\omega^{\tilde{d}-1}_{fD}[1-(\xi/a)^{-\bar{d}}]}\left(\frac{\omega^{\tilde{d}-1}_{fD} - \omega^{\tilde{d}-1}_{co}}{2(\tilde{d}-1)} + \int_{\omega_{co}}^{\omega_{fD}}\frac{\omega^{\tilde{d}-2}\,d\omega}{\exp(\hbar\omega/kT) - 1}\right) \\ u^2_{ph} &= \frac{u^2_0}{A_p}\left(\frac{\omega_{co}}{\omega_{fD}}\right)^{\tilde{d}-1}\left[\frac{1}{4}\left(1 - \frac{\xi^2}{L^2}\right) + \frac{1}{\omega^2_{co}}\int_{\omega_0}^{\omega_{co}}\frac{\omega\,d\omega}{\exp(\hbar\omega/kT) - 1}\right] \end{aligned} \tag{10}$$

where $u^2_0 = 3n\hbar/m\omega_{fD}$ and N is the total number of sites. It is convenient to introduce the new parameters $T_f = T_{Dc}u^2_c/u^2_{Dc}$ and $y_f = u^2_c/u^2_0$, where u^2_{Dc} and T_{Dc} are the critical MSD and the melting transition temperature of a Debye crystal with the same material parameters as those of the fractals. In the ordinary case, we have $kT_{Dc} \gg \hbar\omega_D$; this implies the inequality $y_f \gg 1$. In the following, we shall calculate the instability temperature of fractal ordering in different cases.

(i) When $\tilde{d} > 2$, for pure fractal aggregates ($\xi = L$), from equations (6), (9) and (10), we have

$$\frac{\tilde{d}}{6(\tilde{d}-1)}\left[1 - \left(\frac{\omega_{co}}{\omega_{fD}}\right)^{\tilde{d}-1}\right] + \frac{\tilde{d}}{3\omega^{\tilde{d}-1}_{fD}}\int_{\omega_{co}}^{\omega_{fD}}\frac{\omega^{\tilde{d}-2}\,d\omega}{\exp(-\hbar\omega/kT) - 1} = y_f. \tag{11}$$

From equation (11), it is known that $kT_c \gg \hbar\omega_{co}$. On comparison with the second term, the first term on the left-hand side can be neglected. By evaluating the second term approximately, the instability temperature is given by

$$T_c \sim [3(\tilde{d} - 2)/\tilde{d}]\, T_f. \tag{12}$$

Since the transition point of fractal ordering is independent of the size, the LRO may exist below T_c in this case, just as for a Debye lattice. For mixed fractal aggregates, the same result can be obtained in a similar way. Accordingly, we may view T_c as the melting point of a fractal lattice.

(ii) When $\tilde{d} = 2$, by using a similar evaluation procedure as above, the equation determining the instability temperature T_c is written as

$$(kT_c/\hbar\omega_{fD})\{(1 - \xi/L)/A_f + (\bar{d}/3)\ln(\xi/a) + \tfrac{2}{3}\ln(kT_c/\hbar\omega_{fD}) \times[1 - exp(-\hbar\omega_{fD}/kT_c)]\} = y_f. \tag{13}$$

The solution of this equation for T_c will be a decreasing function of ξ. Consequently, there is no LRO on this type of fractal aggregate. It can be seen easily that $T_c \sim [3/\bar{d}\ln(\xi/a)]\ T_f$ as ξ approaches infinity, just as in two-dimensional crystal lattice, which means there exists quasi-LRO on this type of fractal. Also, T_c is not very small so long as ξ is not much larger than the ordinary macroscopic scale (about $(10^8$–$10^9)a$). Here we should note that the results obtained in cases (i) and (ii) seem to be unable to be obtained in previous work [1, 2].

(iii) We now consider when $1 < \tilde{d} < 2$. Before continuing our discussion, we shall comment briefly on the fracton dimensions. According to equation (11), the necessary condition for which the equation of T_c has a real solution is

$$y_f - [\tilde{d}/6(\tilde{d} - 1)][1 - (\xi/a)^{-\bar{d}(\tilde{d}-1)/\tilde{d}}] \geqslant 0. \tag{14}$$

In order for the inequality to remain valid for any large ξ, including the case $T_c = 0$, we must have

$$\tilde{d} \geqslant (1 - 1/6y_f)^{-1} = d_c \simeq 1. \tag{15}$$

The same result still holds in the mixed case $\xi \neq L$. It also can be seen that the above result is unable to be derived in [1, 2]. As we discuss only the case in which $\tilde{d}$ is not close to unity, the second term on the left-hand side of equation (11) will play an important role. After some analysis, the instability temperature of fractal ordering is obtained approximately:

$$T_c \sim \hbar\omega_{fD}(\xi/a)^{-\bar{d}(2-\tilde{d})/\tilde{d}}[y_f - \tilde{d}/6(\tilde{d} - 1)]/k[(1 - \xi/L)/A_f + \bar{d}/3(2 - \tilde{d})]. \tag{16}$$

It is clear that neither LRO nor quasi-LRO exists on fractal aggregates in this case. In general, T_c will be extremely low due to the smallness of the factor $(\varepsilon/a)^{-\bar{d}(\tilde{d}-1)/\tilde{d}}$ except for when $\tilde{d}$ is relatively small or is very close to 2. This agrees with work in [2], which can give support for our work from another aspect.

(iv) Finally, we consider the case when $\tilde{d} \leqslant 1$. In the $d = 1$ case, because of the contribution from the first term on the left-hand side of equation (11), ξ cannot assume arbitrary large values even at absolute zero temperature. The upper limit of ξ is given by

$$\xi_{c(0)} = a\exp(6y_f/\bar{d}). \tag{17}$$

Thus, in a macroscopic fractal system, the linear size of which is about $10^8 a$, the conditions $\xi \ll \xi_c(0)$ and $\hbar\omega_{co} \ll kT_c$ can be satisfied simultaneously. After calculation, we obtain

$$T_c \sim \hbar\omega_{fD}(\xi/a)^{-\bar{d}}[y_f - (\bar{d}/6)\ln(\xi/a)]/k[(1 - \xi/L)/A_f + \tfrac{1}{3}]. \tag{18}$$

Consequently, since T_c decreases seriously with increasing ξ, the fractal order cannot be maintained for a system at ordinary low temperatures. In the $d \leqslant 1$ case, the conclusion is similar to the above case except that T_c decreases much more rapidly than earlier.

In conclusion, we have developed an approximate theory for the instability of fractal order. Moreover, several new important and interesting results have been obtained.

(i) In the $\tilde{d} > 2$ case, the onset temperature T_c for breaking of fractal order is independent of the characteristic length ξ. Thus, physically, T_c may be viewed as the melting temperature of fractal aggregates, just as for a three-dimensional Debye lattice.

(ii) In the $\tilde{d} = 2$ case, because of the asymptotically logarithmic dependence of T_c on ξ, there is only quasi-LRO, and fractal aggregates on a macroscopic scale (about $(10^8\text{–}10^9)a$) may exist at low temperatures.

(iii) For $\tilde{d} \leqslant 1$, even at zero temperature, the characteristic length ξ cannot be large.

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