

broadening of the electron-density distribution affects the small residual overlap so strongly that a maximum can appear before melting sets a limit on the thermal amplitudes. From then on the electron density in the free volume declines and the lifetime rises.

It remains to discuss briefly  $\tau_B$  and  $\tau_{C'}$ . In Sec. 3 we were led to identify the component  $B$  with the annihilation of positrons which did not form Ps, because of the magnitude of  $\tau_B$  and the small variations of  $\tau_B$  and  $i_B$  with temperature. Even the slight trends with temperature discernable in Figs. 2 and 4, if significant, would be consistent with this interpretation for reasons similar to those advanced for the changes in  $\tau_A$ , if consideration is given to the differences in the affinities between a molecule and a positron on one hand and a positronium atom on the other. The small values

of  $\tau_{C'}$  are comparable to the lifetime of  $\text{Ps}^-$  and reflect the high pickoff rates in the ordered domains of the lattice<sup>6</sup>; indeed the values of  $\tau_A$  as plotted in Fig. 8 extrapolate to just such short lifetimes as  $v^*(T) = 1 + v_f^*(T)$  approaches the tight-packing (t.p.) value  $1 + v_{t.p.}^*$ .

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## Molecular Distribution and Elasticity in Crystals

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The well-known fluctuation-compressibility theorem for fluids is generalized to crystalline solids. It is pointed out that in anisotropic media, fluctuating local strains induce long-range pair correlations. The resulting anomalous contributions to the theorem's pair-correlation-function integral are related to certain components of an elastic tensor for the crystal. Detailed knowledge of particle-singlet and pair equilibrium distributions alone thus provides constraints on, but does not wholly determine, the solid's elastic constants.

### I. INTRODUCTION

ONE of the central results in the statistical-mechanical theory of classical fluids is the density-fluctuation theorem<sup>1</sup> relating the isothermal compressibility  $\kappa$  to an integral of the difference between the pair distribution function,  $\rho^{(2)}(r_{12})$ , and the product of singlet distribution functions,  $\rho^{(1)}(\mathbf{r}_1)\rho^{(1)}(\mathbf{r}_2) = \rho^2$ :

$$\rho^2 kT\kappa = \rho + \int d\mathbf{r}_{12} [\rho^{(2)}(r_{12}) - \rho^2], \quad (1)$$

$$\kappa = (1/\rho)(\partial\rho/\partial p)_T, \quad \rho = N/V.$$

The local density-fluctuation integrand refers to an infinite isotropic and homogeneous system, which normally ensures integrability. One of the persistent interests in relation (1) concerns critical phenomena; for the liquid-vapor transition the anomalous behavior of  $\kappa$  at the corresponding critical point is thus related to a long-range nonintegrable tail of the integrand, and hence to critical opalescence.<sup>2</sup>

<sup>1</sup> L. S. Ornstein and F. Zernike, *Proc. Akad. Sci. (Amsterdam)* **17**, 793 (1914); J. Yvon, *Fluctuations en Densité*, *Actualités Scientifique et Industrielles*, No. 542 (Hermann et Cie., Paris, 1937).

<sup>2</sup> M. E. Fisher, *J. Math. Phys.* **5**, 944 (1964).

The isothermal compressibility  $\kappa$  measures the elastic response of a fluid to an isotropic stress. Indeed, this quantity is essentially the single elastic constant that fluids possess. It is the aim of this article to examine the modification of Eq. (1) required by crystalline substances where there may be up to 18 independent elastic constants.

An interesting and evidently characteristic feature of solids that emerges from the analysis below is that the density fluctuation integrand corresponding to  $\rho^{(2)}(r_{12}) - \rho^2$  possesses long-range character, decaying to zero as the inverse distance cubed. This behavior (unlike the exponentially damped behavior expected for fluids under normal circumstances) has nothing to do with critical phenomena, but arises from local stress fluctuations. The long-range crystalline-phase "tail" requires careful handling, and indeed holds the key to proper modification of Eq. (1) in the presence of direction-dependent elastic properties.

### II. DEDUCTION

DeGennes<sup>3</sup> has remarked that Eq. (1) represents for fluids the long-wavelength limit of a more general

<sup>3</sup> P. G. deGennes, *Nuovo Cimento* **9**, Suppl. 1, 240 (1958).

susceptibility relation for behavior of the system toward a weak externally applied sinusoidal potential. Under present circumstances, it will prove convenient to adopt the same point of view, passing ultimately to infinite wavelength to recover the desired generalization of Eq. (1).

$N$  molecules,<sup>4</sup> contained in a box  $V$  with periodic boundary conditions, will be presumed at a temperature  $T$  such that a crystalline phase obtains.  $\Phi(\mathbf{r}_1 \cdots \mathbf{r}_N)$  will denote the total potential energy of the system of molecules at  $\mathbf{r}_1 \cdots \mathbf{r}_N$ , and it consists of three parts: (1) the intermolecular interaction  $\Phi_1$ , whose precise character is unimportant for present considerations<sup>5</sup>; (2) an external potential  $\Phi_2$  preserving any desired crystal orientation<sup>6</sup>; (3) a second external potential  $\epsilon\Phi_3$ , whose coupling constant  $\epsilon$  may be varied at will for the purpose of observing the system's response. We then take

$$\Phi_3(\mathbf{r}_1 \cdots \mathbf{r}_N) = \sum_{i=1}^N \sin(\mathbf{k} \cdot \mathbf{r}_i), \quad (2)$$

where  $\mathbf{k}$  is a vector of the reciprocal lattice generated by

$$\rho^{(n)}(\mathbf{r}_1 \cdots \mathbf{r}_n, \epsilon) = \frac{N(N-1) \cdots (N-n+1) \int \cdots \int \exp[-\beta\Phi] d\mathbf{r}_{n+1} \cdots d\mathbf{r}_N}{\int \cdots \int \exp[-\beta\Phi] d\mathbf{r}_1 \cdots d\mathbf{r}_N}. \quad (5)$$

Then applying an  $\epsilon$  derivative to Eq. (4),

$$(\partial F(\epsilon)/\partial \epsilon)_{\beta, V} = \int \sin(\mathbf{k} \cdot \mathbf{r}_1) \rho^{(1)}(\mathbf{r}_1, \epsilon) d\mathbf{r}_1. \quad (6)$$

When  $\epsilon=0$ ,  $\rho^{(1)}(\mathbf{r}_1, \epsilon=0)$  exhibits only the fundamental lattice periodicity, which will have no Fourier components in common with the slowly varying external potential; consequently  $F'(0)$  vanishes. A second  $\epsilon$  derivative yields:

$$\begin{aligned} (\partial^2 F(\epsilon)/\partial \epsilon^2)_{\beta, V} &= \int \sin(\mathbf{k} \cdot \mathbf{r}_1) [\partial \rho^{(1)}(\mathbf{r}_1, \epsilon)/\partial \epsilon] d\mathbf{r}_1 = -\beta \int \sin^2(\mathbf{k} \cdot \mathbf{r}_1) \rho^{(1)}(\mathbf{r}_1, \epsilon) d\mathbf{r}_1 \\ &\quad - \beta \int \int \sin(\mathbf{k} \cdot \mathbf{r}_1) \sin(\mathbf{k} \cdot \mathbf{r}_2) [\rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2, \epsilon) - \rho^{(1)}(\mathbf{r}_1, \epsilon) \rho^{(1)}(\mathbf{r}_2, \epsilon)] d\mathbf{r}_1 d\mathbf{r}_2. \quad (7) \end{aligned}$$

In the  $\epsilon=0$  limit in this last relation, the  $\mathbf{r}_1$  integrals over  $V$  (at fixed  $\mathbf{r}_{12}$ ) may be performed to yield the following<sup>7</sup>:

$$\begin{aligned} (\partial^2 F(0)/\partial \epsilon^2)_{\beta, V} &= -(\beta V/2) \left\{ \rho + \int \cos(\mathbf{k} \cdot \mathbf{r}_{12}) \right. \\ &\quad \left. \times \langle \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2) - \rho^{(1)}(\mathbf{r}_1) \rho^{(1)}(\mathbf{r}_2) \rangle_{\omega} d\mathbf{r}_{12} \right\}, \quad (8) \\ \rho &= \langle \rho^{(1)}(\mathbf{r}_1) \rangle_{\omega} = N/V. \end{aligned}$$

<sup>4</sup> For simplicity, we suppress internal degrees of freedom.

<sup>5</sup> In particular, there is no requirement of pairwise additivity, or harmonicity.

<sup>6</sup> Since  $N$  and  $V$  are arbitrarily large, and since at most only the five orientational and translational degrees of freedom for the crystal as a whole (each with average energy  $kT$ ) need to be frozen, the interaction energy *density* of the crystal with  $\Phi_2$  may be made arbitrarily small.  $\Phi_2$  presumably would have periodic minima at the equilibrium crystal sites, though like  $\Phi_1$  its precise form is immaterial.

<sup>7</sup> Here and in the following, the  $\epsilon$  dependence of the  $\rho^{(n)}$  will be suppressed, since always  $\epsilon=0$ .

$V$ . Thus

$$\Phi(\mathbf{r}_1 \cdots \mathbf{r}_N, \epsilon) = \Phi_1(\mathbf{r}_1 \cdots \mathbf{r}_N) + \Phi_2(\mathbf{r}_1 \cdots \mathbf{r}_N) + \epsilon\Phi_3(\mathbf{r}_1 \cdots \mathbf{r}_N). \quad (3)$$

The  $\epsilon$ -dependent free energy  $F$  for the system is provided by the canonical partition function:

$$\begin{aligned} \exp[-\beta F(\epsilon)] &= (\lambda_T^{3N} N!)^{-1} \int \cdots \int \exp[-\beta\Phi(1 \cdots N, \epsilon)] \\ &\quad \times d\mathbf{r}_1 \cdots d\mathbf{r}_N, \quad (4) \end{aligned}$$

where  $\beta = (kT)^{-1}$ , and  $\lambda_T$  is the mean thermal deBroglie wavelength. The behavior of  $F(\epsilon)$  for small  $\epsilon$ , specifically the second  $\epsilon$  derivative  $F''(0)$  will be computed in two ways, by direct differentiation of (4), and by means of macroscopic elasticity theory. In order for the latter to be applicable,  $|\mathbf{k}|$  must necessarily be small.

The  $n$ -particle molecular distribution functions are defined by:

Here, the operation denoted by  $\langle \cdots \rangle_{\omega}$  is an average over positions  $\mathbf{r}_1$  in a unit cell  $\omega$  ( $\mathbf{r}_{12}$  fixed).

The alternative route in calculating  $F''(0)$ , by means of linear elasticity theory, requires an expression for the stress tensor  $\boldsymbol{\sigma}$ . There will be an isotropic contribution  $\boldsymbol{\sigma}^{(0)}$  due to the hydrostatic pressure in the system, plus an extra contribution resulting from the external force field, so we write:

$$\boldsymbol{\sigma}(\mathbf{r}) = \boldsymbol{\sigma}^{(0)}(\mathbf{r}) + \epsilon \boldsymbol{\sigma}^{(1)}(\mathbf{r}), \quad (9)$$

$$\boldsymbol{\sigma}^{(0)} = -p \mathbf{1}.$$

It suffices in the present calculation to regard the force per unit volume on the crystal due to  $\Phi_3$  in the coarse-grained sense of the unit cell average  $\langle \cdots \rangle_{\omega}$ . Then since the divergence of  $\boldsymbol{\sigma}^{(1)}$  is related to this body force in the

standard manner,<sup>8</sup>

$$\sigma^{(1)}(\mathbf{r}) = -\rho \sin(\mathbf{k} \cdot \mathbf{r}) \mathbf{e}\mathbf{e}, \quad (10)$$

where  $\mathbf{e} = \mathbf{k}/|\mathbf{k}|$  is the unit vector in the direction of  $\mathbf{k}$ . From the displacement vector  $\mathbf{u}(\mathbf{r})$  one constructs the strain tensor, with components

$$u_{ij} = \frac{1}{2}(\partial u_i/\partial x_j + \partial u_j/\partial x_i). \quad (11)$$

Then the stress and strain tensors are related to one another by means of the elastic modulus tensor of rank four,  $\lambda$ :<sup>9</sup>

$$\epsilon \sigma_{ij}^{(1)} = \lambda_{ijkl} u_{lm}; \quad (12)$$

here, the repeated-index summation convention applies.

The free-energy density  $f^{(d)}$  of elastic deformation, resulting from the sinusoidal  $\Phi_3$ , is a quadratic form in the strain components<sup>9,10</sup>:

$$f^{(d)}(\mathbf{r}, \epsilon) = -\frac{1}{2} \lambda_{ijkl} u_{ij} u_{lm}. \quad (13)$$

In order to re-express  $f^{(d)}$  in terms of stress-tensor components, we introduce a tensor  $\Lambda$  conjugate to  $\lambda$  by means of the relation:

$$\lambda_{ijkl} \Lambda_{lmnp} = \delta_{in} \delta_{jp}. \quad (14)$$

Then we obtain the following total deformation free energy:

$$\int f^{(d)}(\mathbf{r}, \epsilon) d\mathbf{r} = -\frac{1}{2} \epsilon^2 \int \Lambda_{ijkl} \sigma_{ij}^{(1)}(\mathbf{r}) \sigma_{lm}^{(1)}(\mathbf{r}) d\mathbf{r}. \quad (15)$$

Since the components of  $\Lambda$  are  $\mathbf{r}$ -independent, Eq. (15) may trivially be integrated after insertion of expression (10) for  $\sigma^{(1)}$ . The implied second free-energy derivative is thereupon calculated to be:

$$(\partial^2 F(0)/\partial \epsilon^2)_{\beta, \nu} = -\frac{1}{2} V \rho^2 \mathbf{e}\mathbf{e} : \Lambda : \mathbf{e}\mathbf{e}. \quad (16)$$

Now we may equate the two expressions (8) and (16), if in the former the magnitude of  $\mathbf{k}$  passes to zero<sup>11</sup>:

$$\rho^2 k T \mathbf{e}\mathbf{e} : \Lambda : \mathbf{e}\mathbf{e} = \rho + \lim_{k \rightarrow 0} \int \cos(\mathbf{k} \cdot \mathbf{r}_{12}) \times \langle \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2) - \rho^{(1)}(\mathbf{r}_1) \rho^{(1)}(\mathbf{r}_2) \rangle_{\omega} d\mathbf{r}_{12}. \quad (17)$$

In homogeneous fluids (barring critical phenomena), the difference  $\rho^{(2)}(12) - \rho^{(1)}(1)\rho^{(1)}(2)$  decays to zero fast enough that  $\cos(\mathbf{k} \cdot \mathbf{r}_{12})$  may automatically be replaced by unity in Eq. (17), so that Eq. (1) is recovered, since also

$$\Lambda = \kappa \mathbf{1} \quad (18)$$

for fluids. In the case of crystals, though, the situation requires more careful analysis, for the left member must

<sup>8</sup> L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1959), p. 5.

<sup>9</sup> Reference 8, p. 37.

<sup>10</sup> The deformation free energy is usually quoted with positive sign. The free energy relevant to partition function (4) however, differs by a Legendre transformation which induces sign change, and which reflects the tendency of the system to deform in the sinusoidal external potential so as to lower its free energy.

<sup>11</sup> Implicit here is passage first to infinite system size (the  $k$  vectors become dense), then  $|\mathbf{k}| \rightarrow 0$  at fixed  $\mathbf{e}$ .

generally display dependence on the direction of  $\mathbf{e}$  as a result of crystal anisotropy, whereas treatment of the right member as for fluids would invariably yield an isotropic answer.

By virtue of the intermolecular forces operative in crystals, the displacement of a given particle from its nominal lattice position can induce a local dilatation, an even function of the displacement, which should distort the lattice sufficiently far from the given particle according to the requirements of macroscopic elasticity. The displaced particle then plays a local role similar to that of a lattice point defect, and we may then draw upon the elastic continuum theory of lattice defects<sup>12</sup> as a heuristic guide in further analysis of identity (17). Especially revealing in this regard is Eshelby's result concerning the dilatation (i.e., density change) about a defect in a cubic medium ( $|\mathbf{r}| > 0$ ):<sup>13</sup>

$$\nabla \cdot \mathbf{u}(\mathbf{r}) = \text{const} \times (x^4 + y^4 + z^4 - \frac{3}{5} r^4) / r^7, \quad (19)$$

when the crystal is aligned along the coordinate axes. Thus one observes local density changes that fall to zero radially as the inverse distance cubed, but with an angular dependence which averages to zero over the sphere. By implication the same will be true in other crystal types, but with differing angular variations consistent with the various symmetries.

For sufficiently large  $\mathbf{r}_{12}$ , then, our integrand  $\langle \rho^{(2)}(12) - \rho^{(1)}(1)\rho^{(1)}(2) \rangle_{\omega}$  for crystals should decay to zero also as  $r_{12}^{-3}$ , with vanishing angular average. Therefore we add and subtract under the integral of Eq. (17) the spherical average (denoted by subscript  $s$ ) of the quantity:

$$\begin{aligned} & \langle \rho^{(2)}(12) - \rho^{(1)}(1)\rho^{(1)}(2) \rangle_{\omega} \\ &= \langle \rho^{(2)}(12) - \rho^{(1)}(1)\rho^{(1)}(2) \rangle_{\omega, s} \\ & \quad + [\langle \rho^{(2)}(12) - \rho^{(1)}(1)\rho^{(1)}(2) \rangle_{\omega} \\ & \quad - \langle \rho^{(2)}(12) - \rho^{(1)}(1)\rho^{(1)}(2) \rangle_{\omega, s}]. \quad (20) \end{aligned}$$

Then since sphericalization removes the long-ranged tail, leaving an integrable function of  $r_{12}$ , the  $|\mathbf{k}| \rightarrow 0$  limit for the first term is automatic:

$$\begin{aligned} & \rho^2 \kappa T \mathbf{e}\mathbf{e} : \Lambda : \mathbf{e}\mathbf{e} \\ &= \rho + \int \langle \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2) - \rho^{(1)}(\mathbf{r}_1) \rho^{(1)}(\mathbf{r}_2) \rangle_{\omega, s} d\mathbf{r}_{12} \\ & \quad + \lim_{k \rightarrow 0} \int \cos(\mathbf{k} \cdot \mathbf{r}_{12}) [\langle \dots \rangle_{\omega} - \langle \dots \rangle_{\omega, s}] d\mathbf{r}_{12}. \quad (21) \end{aligned}$$

In order to evaluate the remaining limit in the last term of Eq. (21), the two integrand factors will next be expanded in spherical harmonics. First,

$$\cos(\mathbf{k} \cdot \mathbf{r}_{12}) = \sum_{n=0}^{\infty} (-1)^n (4n+1) P_{2n}(\cos\theta) j_{2n}(kr_{12}), \quad (22)$$

<sup>12</sup> J. D. Eshelby, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 3, p. 79.

<sup>13</sup> J. D. Eshelby, *Acta Met.* **3**, 491 (1955).

where  $\theta$  is the angle between  $\mathbf{e}$  and  $\mathbf{r}_{12}$ , and  $j_{2n}$  is the spherical Bessel function:

$$j_{2n}(x) = (\pi/2x)^{1/2} J_{2n+1/2}(x). \quad (23)$$

The analogous expansion of the square-bracketed factor in (21) will have the following form:

$$\begin{aligned} & [\langle \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2) - \rho^{(1)}(\mathbf{r}_1)\rho^{(1)}(\mathbf{r}_2) \rangle_\omega \\ & - \langle \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2) - \rho^{(1)}(\mathbf{r}_1)\rho^{(1)}(\mathbf{r}_2) \rangle_{\omega, \mathbf{e}}] \\ & = \sum_{l=1}^{\infty} \sum_{m=-l}^l h_{lm}(r_{12}) P_l^m(\cos\theta) \exp(im\varphi), \quad (24) \end{aligned}$$

in which the  $h_{lm}$  are suitable functions only of scalar pair separation.

Insertion of (22) and (24) into the last term of Eq. (21) leads to the following form for this quantity:

$$\lim_{k \rightarrow 0} \sum_{n=1}^{\infty} \int j_{2n}(kr_{12}) h_{10}(r_{12}) d\mathbf{r}_{12}. \quad (25)$$

Near the origin

$$j_{2n}(x) \sim 2^{2n} (2n)! x^{2n} / (4n)!, \quad (26)$$

so that the only parts of the functions  $h_{10}$  which are relevant to the  $k \rightarrow 0$  limit in integrals (25) are those for large argument. In view of the earlier discussion therefore, we shall asymptotically represent the  $h_{10}$  thus:

$$h_{10}(r_{12}) \sim h_l / r_{12}^3 \quad (r_{12} \rightarrow \infty), \quad (27)$$

where the set of numerical constants  $h_l$  of course depend upon the relative orientation of crystal principal directions, and  $\mathbf{e}$ . Consequently the expression (25) may be simplified somewhat:

$$\lim_{k \rightarrow 0} 4\pi \sum_{n=1}^{\infty} h_{2n} \int_0^{\infty} dr j_{2n}(kr) / r. \quad (28)$$

Equality (23) now converts the integrals appearing in (28) to standard forms.<sup>14</sup> The crystalline phase modification of theorem (1) then may be finally written down:

$\rho^2 \kappa T \mathbf{e} : \mathbf{A} : \mathbf{e} \mathbf{e}$

$$\begin{aligned} & = \rho + \int \langle \rho^{(2)}(\mathbf{r}_1, \mathbf{r}_2) - \rho^{(1)}(\mathbf{r}_1)\rho^{(1)}(\mathbf{r}_2) \rangle_{\omega, \mathbf{e}} d\mathbf{r}_{12} \\ & + \sum_{n=1}^{\infty} (-1)^n \pi^{3/2} \Gamma(n) h_{2n}(\mathbf{e}) / \Gamma(n + \frac{3}{2}). \quad (29) \end{aligned}$$

<sup>14</sup> I. M. Ryslik and I. S. Gradshteyn, *Tables of Series, Products and Integrals* (Veb Deutscher Verlag der Wissenschaften, Berlin, 1963), p. 231, formula 1.

The last set of terms provides the necessary orientation dependence, by virtue of  $\mathbf{e}$  dependence of the  $h_{2n}$ .

### III. COMMENTS

A few short comments regarding the extended theorem (29) are in order. First, it must be acknowledged that the argument leading to the universality of inverse-cube asymptotes for all crystal classes, as shown in (27), is far from complete. The relevant solutions for dilatations in continuum models for lattice defects are not generally available in all these classes, and indeed the result (19) obtained in Ref. 13 represents only a low-order perturbation result for the cubic case. However, it is clear from the foregoing analysis that asymptotes which fall off more or less slowly than  $r_{12}^{-3}$ , respectively, would fail to give directional dependence for  $F''(0)$ , or to give any convergent result at all. *A posteriori*, then, the postulate gains credence, but it would be desirable to augment the currently available relevant solutions in continuum defect theory.

On account of the special nature of the scalar contraction  $\mathbf{e} \mathbf{e} : \mathbf{A} : \mathbf{e} \mathbf{e}$ , it is generally not possible to obtain the entire set of up to 18 independent elastic constants from the right member of Eq. (29). Roughly, this derives from the restriction, when one is committed to strict equilibrium conditions (as we were above by operating on the canonical partition function), to use only of conservative external force fields. However, the observation of the entire set of elastic constants for a crystal will generally require use of shear stress, which is not derivable from an external potential. The remainder set of constants left undetermined by Eq. (29) therefore could only be obtained in a rigorous statistical mechanical theory by calculation of time-dependent system responses, unless the system model had inherent characteristics preventing unbounded energy absorption from the shear field at zero driving frequency.<sup>15</sup>

Finally, we remark that Eq. (29) retains full validity in the quantum-mechanical regime, if the  $\rho^{(n)}$  are interpreted as diagonal elements of reduced density matrices in direct space representation.

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<sup>15</sup> This would be the case for cross-linked polymers, or rigid spheres sufficiently highly compressed to prevent plastic flow.