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Application of fracton theory to nanocrystalline iron

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Abstract. The dimensionality of harmonic modes in nanocrystalline iron is determined by a nonlinear method, and the result is analysed in terms of the Debye model and a fracton model. The negative of the exponent of the recoiless fraction, F, is calculated as a function of temperature from data in the literature. It is found that the temperature variation of F cannot be reproduced by the Debye model for two- or three-dimensional lattices, but can be reasonably well fit by a fracton model. Using the fracton model, it is found that the density of states scales with modal frequency and has a scaling exponent of 1.22 ± 0.13. This suggests that the modes have a dimensionality, $\overline{\overline{d}}$, of 2.22 ± 0.13. It is concluded that the value of $\overline{\overline{d}}$ may indicate the presence of fractons and a fractal atomic structure in nanocrystalline iron.

1. Introduction

The geometric language of fractals (Mandelbrot 1988) can be used to describe the characteristics of certain real-life objects as well as those of some geometric shapes. Fractals are self-similar or self-affine patterns, formed by quantities that vary exponentially with resolution or a length scale (Aharony 1996). Non-random fractals, such as the Sierpiński gasket, are self-similar across all magnifications (Mandelbrot 1988), but do not occur in nature. Instead, natural objects may contain patterns that are statistically self-similar or self-affine over a limited range of magnifications (Kjems 1996). Such patterns are called random fractals. Even though the median scaling range of random fractal patterns is just 1.3 orders of magnitude (Avnir *et al* 1998), they are of interest due to their abundance in natural objects (Biham *et al* 1998). The properties of random fractals are consistent with the distributions of mass observed in silica aerogels (Courtens and Vacher 1988), smoke–grain aggregates (Forrest and Witten 1979) and iron–nickel alloys (Li *et al* 1997).

Fractals have one or more properties of the form *property* $\propto$ *linear scale* ^{x} (Aharony 1996), where x is a non-integer, referred to as a scaling exponent. For example, mass (m) scales with length (L) as $m \propto L^{\overline{d}}$. For a material whose mass is distributed in a fractal pattern, $\overline{d}$ lies in the range 1–3 (Courtens and Vacher 1988), and is called the fractal dimensionality. The value of $\overline{d}$ provides a measure of roughness (Mandelbrot 1988) by indicating the extent to which the mass occupies an embedding Euclidean dimension (d) of one, two or three. Similarly, the density of states (DOS) of harmonic vibrational modes ($\rho(\omega)$) scales with modal frequency (ω) as $\rho(\omega) \propto \omega^{\overline{\overline{d}}-1}$ (Orbach 1989). In materials thought to contain modes that are localized on fractal atomic structures, $\overline{\overline{d}}$ is referred to as the fracton dimensionality and the modes are called fractons (Alexander and Orbach 1982). The fracton dimensionality represents the dimensionality of harmonic vibrations in a solid and characterizes the effect of a fractal atomic structure on the degrees of freedom (DOF) of its oscillators. The value of $\overline{\overline{d}}$ can lie in the range

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1–3 for a material with a fracton DOS (Courtens and Vacher 1988), but must equal 2 or 3 for two- or three-dimensional Debye lattices respectively (Debye 1912).

It is interesting to determine whether the Mössbauer spectra of compacted ultrafine iron grains contain evidence of fractons. Forrest and Witten (1979) found that chain-like aggregates of iron grains, 8 nm in diameter, have a fractal dimensionality of 1.51 ± 0.05. The detection of behaviour consistent with that of fractons would provide additional evidence for the presence of fractal structures in iron. Mössbauer spectroscopy is a suitable technique for the detection of fractons. The spectral area depends on the recoiless fraction (f) of γ-ray transitions between nuclear levels, which depends on the DOS (Wertheim 1968) and its scaling exponent, $\overline{\overline{d}} - 1$. Shrivastava (1986) predicted that the Debye–Waller factor will be reduced in fractal materials compared with well-crystallized solids. Shrivastava and Misra (1988) examined the recoiless fraction of frozen deoxygenated myoglobin in terms of a fracton DOS, and deduced that it represented an effective (fracton) dimensionality of 1.40 ± 0.07.

This study uses a fracton theory to interpret the temperature dependence of f in nanocrystalline iron. In section 2.1, an equation is derived for the fracton density of states (DOS) which, in section 2.2, is used to develop an expression for f. Section 3 describes how the resulting fracton model of f was used to interpret data obtained from a previous study (Herr *et al* 1987) of nanocrystalline iron, a material thought to consist of nanometre-scale crystallites separated by atomically disordered grain boundaries. Later parts of this paper contain the results (section 4), discussion (section 5) and concluding remarks (section 6).

2. Theory

2.1. Density of states

The Debye model (Debye 1912) assumes that normal modes of vibration occur in a continuous, isotropic medium as standing waves. It considers the modal frequency, ω, to lie in the range $0 \leqslant \omega \leqslant \omega_D$, where $\omega_D = k_B\theta_D/\overline{h}$. Here, ω_D is the Debye frequency, k_B is Boltzman's constant, $\overline{h}$ is Planck's constant, and θ_D is the Debye temperature of the material. In an isotropic lattice, the total number of modes equals $\mathrm{d}N$, the number of vibrational degrees of freedom (d) multiplied by the number of oscillators (N) (Debye 1912). Consider, for example, a two-dimensional lattice in which the effective spring constant is zero in the direction out of the plane and identical for two orthogonal directions within the plane. The oscillators have two vibrational DOF, and the Debye model predicts a total of $2N$ modes. For a well-crystallized material, whose oscillators are free to vibrate in three spatial dimensions, the Debye model predicts $3N$ modes. Since the normalized Debye DOS (Debye 1912) has the form $\rho(\omega) \propto \omega^{d-1}$, it varies in proportion to ω for Euclidean surfaces and ω^2 for well-crystallized material (Courtens and Vacher 1988). For lattices with three vibrational DOF, the normalized Debye DOS is (Wertheim 1968)

$$\rho(\omega) = \frac{9N\omega^2}{\omega_D^3}. \tag{1}$$

Equivalent expressions may be obtained for fractons to describe the total number of modes and the fracton DOS. For fractons, as well as phonons, the total number of modes equals N multiplied by the number of vibrational DOF. Thus, the total number of fracton modes, counted in terms of fracton dimensionality (Alexander and Orbach 1982), is $\overline{\overline{d}}N$. The fracton DOS includes a minimum vibrational frequency (ω_C) whose value is related to the length of the longest vibrating segment in the fractal structure. The normalized fracton DOS may therefore

be derived from the normalized Debye DOS, and expressed as

$$\rho(\omega) = \begin{cases} 0 & 0 < \omega < \omega_C \\ \dfrac{\bar{\bar{d}}^2 N \omega^{\bar{\bar{d}}-1}}{\omega_{fD}^{\bar{\bar{d}}} - \omega_C^{\bar{\bar{d}}}} & \omega_C \leqslant \omega \leqslant \omega_{fD}. \end{cases} \tag{2}$$

In analogy to the Debye model (Debye 1912), $\omega_C = k_B\theta_C/\bar{h}$ and $\omega_{fD} = k_B\theta_{fD}/\bar{h}$. The parameters θ_C and θ_{fD} are, respectively, the minimum and maximum temperatures at which a fracton may be excited.

2.2. Calculation of the recoiless fraction in terms of a fracton density of states

In this section, an expression for f is derived for a material with a fractal atomic structure and fracton DOS. The derivation has essentially the same form as that of Wertheim (1968). However, equation (2) replaces the Debye DOS in the development of expressions for the mean square displacement of vibrating atoms, $\langle r^2\rangle$, and then for f.

In a material with a fracton DOS, the modal frequencies lie in the range ω_C–ω_{fD} as shown by equation (2), and Wertheim's expression (Wertheim, 1968) for $\langle r^2\rangle$ may be modified to

$$\langle r^2\rangle = \frac{\bar{h}}{NM}\int_{\omega_C}^{\omega_{fD}} \left[\frac{1}{2} + \frac{1}{\exp(\bar{h}\omega/k_BT) - 1}\right]\frac{\rho(\omega)}{\omega}\mathrm{d}\omega. \tag{3}$$

The fracton DOS in equation (2) is substituted in equation (3) to obtain

$$\langle r^2\rangle = \frac{\bar{\bar{d}}^2\bar{h}}{M(\omega_{fD}^{\bar{\bar{d}}} - \omega_C^{\bar{\bar{d}}})}\int_{\omega_C}^{\omega_{fD}} \left[\frac{1}{2} + \frac{1}{\exp(\bar{h}\omega/k_BT) - 1}\right]\omega^{\bar{\bar{d}}-2}\,\mathrm{d}\omega. \tag{4}$$

After integrating the first term under the integral sign, equation (4) becomes

$$\langle r^2\rangle = \frac{\bar{\bar{d}}^2\bar{h}}{2(\bar{\bar{d}}-1)M(\omega_{fD}^{\bar{\bar{d}}} - \omega_C^{\bar{\bar{d}}})}\left[\omega_{fD}^{\bar{\bar{d}}-1} - \omega_C^{\bar{\bar{d}}-1} + 2(\bar{\bar{d}}-1)\int_{\omega_C}^{\omega_{fD}} \frac{\omega^{\bar{\bar{d}}-2}}{\exp(\bar{h}\omega/k_BT) - 1}\,\mathrm{d}\omega\right]. \tag{5}$$

To clarify the integral in equation (5), the expressions $\omega_{fD} = k_B\theta_{fD}/\bar{h}$, $\omega_C = k_B\theta_C/\bar{h}$ and $u = \bar{h}\omega/k_BT$ were substituted in equation (5), and appropriate adjustments were made to the limits of integration. Thus

$$\langle r^2\rangle = \frac{\bar{\bar{d}}^2\bar{h}^2}{2(\bar{\bar{d}}-1)Mk_B(\theta_{fD}^{\bar{\bar{d}}} - \theta_C^{\bar{\bar{d}}})}\left[\theta_{fD}^{\bar{\bar{d}}-1} - \theta_C^{\bar{\bar{d}}-1} + 2(\bar{\bar{d}}-1)T^{\bar{\bar{d}}-1}\int_{\theta_C/T}^{\theta_{fD}/T} \frac{u^{\bar{\bar{d}}-2}}{\mathrm{e}^u - 1}\,\mathrm{d}u\right]. \tag{6}$$

The recoiless fraction is given by (Wertheim 1968)

$$f = \exp\left(-\left(\frac{E_\gamma}{\bar{h}c}\right)^2\frac{\langle r^2\rangle}{3}\right) \tag{7}$$

where E_γ, the energy of the emitted γ-ray, equals 3.19×10^{-22} J for the 14.4 keV transition in ^{57}Fe. When $E_R = E_\gamma^2/(2Mc^2)$ and equation (6) are substituted into equation (7), the expression for f has the form

$$f = \exp\left\{-\frac{\bar{\bar{d}}^2 E_R(\theta_{fD}^{\bar{\bar{d}}-1} - \theta_C^{\bar{\bar{d}}-1})}{3(\bar{\bar{d}}-1)k_B(\theta_{fD}^{\bar{\bar{d}}} - \theta_C^{\bar{\bar{d}}})}\left[1 + \frac{2(\bar{\bar{d}}-1)T^{\bar{\bar{d}}-1}}{\theta_{fD}^{\bar{\bar{d}}-1} - \theta_C^{\bar{\bar{d}}-1}}\int_{\theta_C/T}^{\theta_{fD}/T} \frac{u^{\bar{\bar{d}}-2}}{\mathrm{e}^u - 1}\mathrm{d}u\right]\right\}. \tag{8}$$

The negative of the exponent in equation (8), F, is given by

$$F = \frac{\bar{\bar{d}}^2 E_R(\theta_{fD}^{\bar{\bar{d}}-1} - \theta_C^{\bar{\bar{d}}-1})}{3(\bar{\bar{d}}-1)k_B(\theta_{fD}^{\bar{\bar{d}}} - \theta_C^{\bar{\bar{d}}})} \left[1 + \frac{2(\bar{\bar{d}}-1)T^{\bar{\bar{d}}-1}}{\theta_{fD}^{\bar{\bar{d}}-1} - \theta_C^{\bar{\bar{d}}-1}} \int_{\theta_C/T}^{\theta_{fD}/T} \frac{u^{\bar{\bar{d}}-2}}{e^u - 1} du\right]. \quad (9)$$

A more familiar expression, the negative of the exponent (F_D) in the Debye–Waller factor for a three-dimensional lattice, may be recovered from equation (9) by replacing θ_{fD} by θ_D, θ_C by zero, and $\bar{\bar{d}}$ by three:

$$F_D = \frac{3E_R}{2k_B\theta_D}\left[1 + 4\left(\frac{T}{\theta_D}\right)^2 \int_0^{\theta_D/T} \frac{u}{e^u - 1} du\right]. \quad (10)$$

There are two main differences between the Debye (1912) model and the fracton model for the DOS. The former has modes with a minimum temperature of zero and requires d to equal two and three for two- and three-dimensional lattices respectively; the latter has modes with a non-zero minimum temperature and only requires $\bar{\bar{d}}$ to lie in the range 1–3.

Equation (9) expresses the relationship between F and T for a fracton DOS. In a previous study (Shrivastava and Misra 1988), it was assumed that the relationship was approximately $F \propto T^{\bar{\bar{d}}-1}$ in the temperature region for which the integral in equation (9) is nearly constant. In such circumstances, the gradient of a double-logarithmic plot of F as a function of T equals $\bar{\bar{d}} - 1$, and was used to calculate $\bar{\bar{d}}$ (Shrivastava and Misra 1988). However, it is preferable to use the whole of equation (9) to interpret the temperature dependence of F: due to the effect of the logarithmic function, the temperature-independent term in equation (9) may not be neglected. In this study, the full expression in equation (9) was used to interpret $f(T)$ in terms of $\bar{\bar{d}}$ for nanocrystalline iron.

3. Calculations

Numerical results were obtained by applying the theory developed in section 2 to the data of Herr *et al* (1987). Herr *et al* (1987) collected Mössbauer spectra of a specimen of nanocrystalline iron, made by compacting 6 nm iron grains produced by inert-gas condensation. Originally, the data were reported in the form of $f(T)/f(10)$, and $\theta_D = 345$ K. To check this value of θ_D, the Debye formula in equation (10) with $\theta_D = 345$ K was fitted to the data. The validity of this low value of θ_D is discussed in section 5. To obtain $f(T)$, each value of $f(T)/f(10)$ was multiplied by the value of $f(10)$, approximately 0.904, obtained by using $\theta_D = 345$ K. The value of $\ln F$, where $F = -\ln f$, was plotted as a function of $\ln T$. The calculations were repeated using $\theta_D = 467$ K to estimate the uncertainty in the final numerical result.

To simplify the fracton model, values were estimated for θ_C and θ_{fD} in equation (9). The value of θ_C was obtained from the relationship

$$\theta_C = \frac{h\upsilon}{2k_B x} \quad (11)$$

where h is Planck's constant, x is the mean grain diameter and υ is the wave velocity. It is assumed that the least energetic fracton modes have a wavelength of $2x$, corresponding to the presence of nodes at the grain boundaries, and that υ is the same as that of sound waves in similar, well-crystallized material. The value of θ_{fD} was approximated as θ_D; it is expected to be at least as large as θ_D since fracton modes are excited at higher temperatures than phonon modes (Courtens and Vacher 1988).

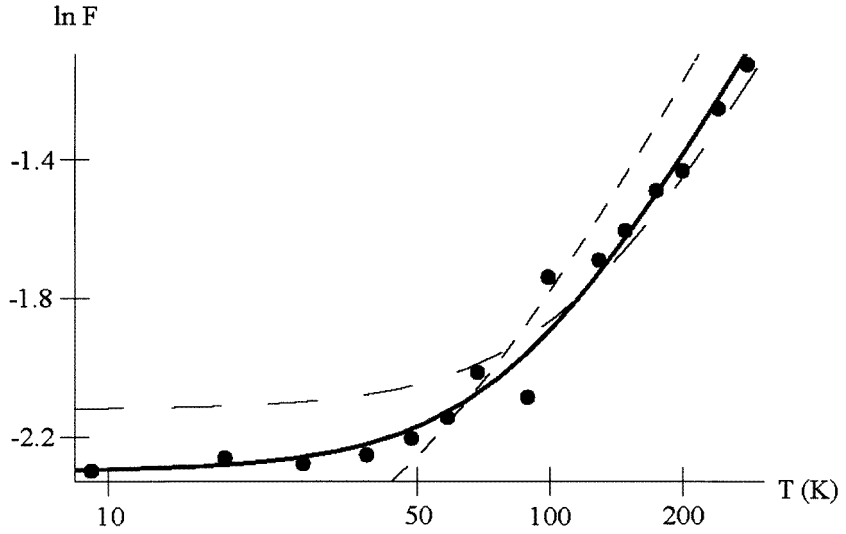

Figure 1. A plot of the data from Herr *et al* (1987) and curves representing the Debye model for a lattice of dimension d. The solid curve represents the fit obtained with $\theta_D = 345$ K and $d = 3$. The broken curves were obtained using the Debye model in conjunction with an additive constant, with (short dashes) $\theta_D = 467$ K, $d = 2$ and (long dashes) $\theta_D = 467$ K, $d = 3$.

In the method of Shrivastava and Misra (1988), it was assumed that the gradient of a double-logarithmic plot of F as a function of T was equal to $\overline{\overline{d}} - 1$, in the temperature region for which the integral in equation (9) is approximately constant. The temperature dependence of the integral term was examined here using `Mathematica` (Wolfram 1996). On the basis of the `Mathematica` findings, the applicable temperature range was determined to be 28–68 K, and $\overline{\overline{d}}$ was calculated from a linear fit to the data in this temperature region.

To evaluate $\overline{\overline{d}}$ by a nonlinear method, `Mathematica` (Wolfram 1996) was used to fit equation (9) to the entire set of data at temperatures above θ_C. The model was also fitted to the data obtained using the second value of $f(10)$, approximately 0.928, which corresponded to $\theta_D = 467$ K. The relationship between $\overline{\overline{d}}$ and θ_C was determined by selecting different values of θ_C, then fitting equation (9) to the data. The potential effect of a grain size distribution was determined from the relationship between $\overline{\overline{d}}$ and θ_C.

4. Numerical results

As expected, the Debye model does not fit the data. When θ_D is set to 467 K a poor fit is obtained using either the two- or three-dimensional Debye model (figure 1) whether or not an additive constant is included. As pointed out by Herr *et al* (1987), the three-dimensional Debye model only fits the data when θ_D is permitted to have an anomalously low value of 345 K (figure 1). The effect on the fit of including a non-zero lower cut-off temperature was found to be relatively insignificant compared with the value of d.

In order to apply the fracton models, values were estimated for θ_C and θ_{fD}. In iron grains, assuming $\upsilon = 5960$ ms^{-1} (*CRC Handbook of Chemistry and Physics* 1996–97) and a mean grain diameter of 6 nm, the maximum fracton wavelength is approximately 12 nm. Thus, equation (11) yields $\theta_C = 24$ K. The value of θ_{fD} was assumed to be approximately equal to

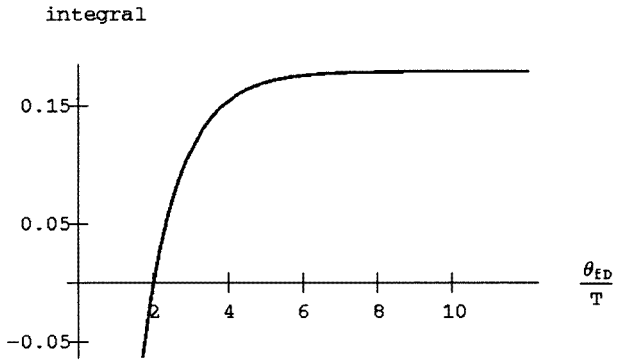

Figure 2. Plot of the integral $\int_2^{\theta_{fD}/T} \frac{u^{\bar{\bar{d}}-2}}{e^u-1}\mathrm{d}u$ as a function of θ_{fD}/T for $\bar{\bar{d}} = 2.2$.

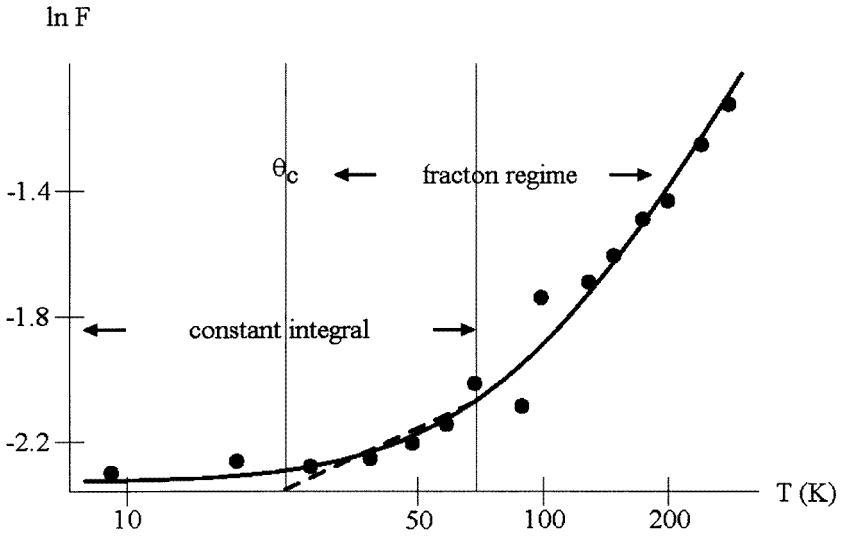

Figure 3. Fitted temperature dependence of F. Dots: data from Herr *et al* (1987). Solid curve: fracton model with an added constant of 0.41. Dashed line: linear fit between 24–69 K ($3.2 \leqslant \ln T \leqslant 4.2$). Vertical lines indicate the bounds of the region in which the integral in equation (9) is approximately equal to 0.18 for $\bar{\bar{d}} = 2.2$, and the lower limit of the fracton regime.

467 K, the value of θ_D in well-crystallized iron.

When using the linear method of Shrivastava and Misra (1988) to calculate $\bar{\bar{d}}$, it was found that the value of $\bar{\bar{d}}$ affects the range of temperatures over which the integral in equation (9) is approximately constant. For $\bar{\bar{d}} = 1.3$, the integral is found to be approximately constant for $\frac{\theta_{fD}}{T} > 5$, as assumed previously (Shrivastava and Misra 1988); when $\bar{\bar{d}} = 2.2$ (figure 2) the integral is approximately constant for $\frac{\theta_{fD}}{T} > 6$. The constant value of the integral is approximately 0.07 for $\bar{\bar{d}} = 1.3$ and 0.18 for $\bar{\bar{d}} = 2.2$. However, in this study, the value of $\bar{\bar{d}}$ barely affects the set of data to be fitted. Upon deleting the datum at $T = 89$ K ($\ln T = 4.488$), which is inconsistent with the other data (figure 3), the applicable temperature range turns out to be the same for values of $\bar{\bar{d}}$ in a range of at least 1.3–2.2. Thus, the data set contains the

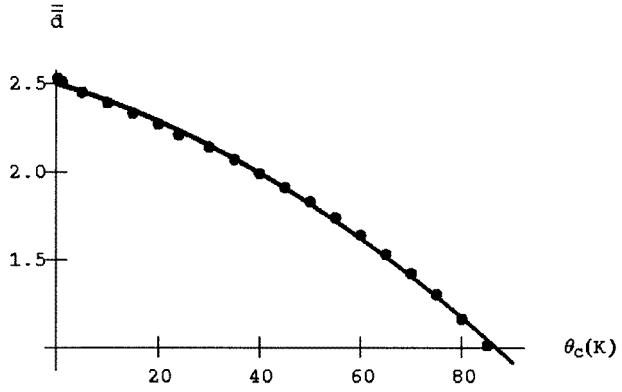

Figure 4. Relationship between $\overline{\overline{d}}$ and θ_C for nanocrystalline iron. The solid curve is a guide to the eye.

same five data over a reasonably broad range of $\overline{\overline{d}}$ values, and is assumed to be suitable for the linear method of calculating $\overline{\overline{d}}$.

Different results were obtained from the linear and nonlinear fits (figure 3). Using the linear method to fit the fracton model, the result was $\overline{\overline{d}} = 1.27 \pm 0.14$. When the full fracton model in equation (9) was fitted to the set of data at temperatures above 24 K, an additive constant of 0.41 was required. The result was $\overline{\overline{d}} = 2.22 \pm 0.13$. Using data calculated from the second value of $f(10)$, which had been obtained by assuming $\theta_D = 467$ K, the result was $\overline{\overline{d}} = 2.37 \pm 0.16$. The uncertainties were assumed to be twice as large as the standard deviations.

It was found that the calculation of $\overline{\overline{d}}$ depends on the value to which θ_C is set in equation (9). For example, values of θ_C between 70 K and 10 K correspond to values of $\overline{\overline{d}}$ between 1.4–2.4, respectively (figure 4).

5. Discussion

The value of $\overline{\overline{d}} = 2.22 \pm 0.13$, obtained from the nonlinear fit, appears to be more reliable than the result obtained from the linear fit. The nonlinear method involves fitting the full model given by equation (9), in conjunction with an additive constant of 0.41, to the entire set of data; two different estimates of $f(10)$ led to results that were the same within the experimental uncertainties. In contrast, the linear method neglects the temperature-independent term in equation (9) and is applied to a smaller range of data.

Since the value of $\overline{\overline{d}}$ differs from that expected for a three-dimensional Debye lattice, the result supports the contention of Herr *et al* (1987) that grain boundaries in nanocrystalline material are probably not Debye solids. The Debye model assumes the presence of a continuous, isotropic medium, but the structure of nanocrystalline matter is neither continuous nor isotropic. The small grain size means that 10–50% of the volume fraction consists of atomically disordered grain boundaries (Schaefer *et al* 1988), which inhibit the formation of extended modes.

Nanocrystalline iron is unlikely to be a Debye solid (Herr *et al* 1987) not only because its structure is incompatible with the Debye model, but because the Debye model leads to an uncharacteristically low value of θ_D. If the Debye model was somehow applicable to

nanocrystalline iron, the work of Niemantsverdriet *et al* (1984), who found that α-FeOOH and α-Fe_2O_3 microcrystals have the same θ_D as well-crystallized material, suggests that θ_D ought to be the same as in well-crystallized iron. Erroneously low values of θ_D can be obtained for microcrystals in calculations based on $f(T)$, when the recoiless fraction is reduced by temperature-dependent grain vibrations as well as normal mode vibrations (Niemantsverdriet *et al* 1984). In the future, it would be useful to compare a grain-vibration model with the fracton model. The necessity of including a constant with equation (9) suggests that grain-vibration effects may occur in addition to fracton behaviour.

A preliminary statistical analysis confirms that the value of $\overline{\overline{d}}$ is significantly different to that expected from the Debye model for a three-dimensional lattice. In ultrafine (3–200 nm) iron grains produced by inert-gas condensation, the grain size follows a log-normal distribution; the geometric standard deviation of the distribution is between 1.36–1.60 (Granqvist and Buhrman 1976). Thus, approximately 95% of the grains have diameters between 2–15 nm, which, respectively, correspond to minimum fracton temperatures of 70 K and 10 K. The results in figure 4 suggest that even if the grains had an average diameter as large as 15 nm, $\overline{\overline{d}}$ would be approximately 2.4, rather than 3. The smaller the grain, the larger the minimum fracton temperature, and the greater the difference between $\overline{\overline{d}}$ and the value expected for well-crystallized material (figure 4). This difference is consistent with the presence of a larger volume fraction of atomically disordered material at the surfaces of small grains or in the grain boundaries of finely grained material, than in well-crystallized material.

6. Concluding remarks

Since the Debye model does not reproduce $f(T)$ with the use of the correct value of θ_D, a different explanation of the data is warranted. The fracton model is applicable in this context, as it is able to reproduce the data with physically realistic parameter values and assumptions. The result of $\overline{\overline{d}} = 2.22 \pm 0.13$ is consistent with the poorly crystalline structure of nanocrystalline iron, as it is significantly less than the value of 3 expected for a three-dimensional Debye lattice. The detection of behaviour consistent with that of fractons provides additional evidence for the presence of fractal structures in iron. Therefore, this paper has shown that fractal theory may be suitable for the characterization of atomic structure in agglomerates of ultrafine iron grains.

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