Proceedings of the 9th International Conference on Hopping and Related Phenomena, Shefayim, Israel, Sept. 2001, published in
phys. stat. sol(b), 230, 31 (2002)

Anharmonic Interactions and the Low-Temperature Thermal Anomalies in Glasses

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Abstract

We study the low-temperature behaviour of an elastic continuum with spatially fluctuating elastic constants and a cubic anharmonic coupling. By functional integral quantization and techniques used previously for treating an electron gas in the presence of disorder and interactions, we obtain an effective action which describes the density fluctuations of the system. By means of a saddle-point approximation we derive a self-consistent set of equations for the density fluctuation propagator $Q(\omega, T)$ where ω is the frequency and T is the temperature. From the low-frequency solutions of these equations we are able to obtain a mean free path $\ell \propto \omega^{-1}\mathrm{Im}\{Q(\omega, T)^{1/2}\}$. This leads roughly to a T^2 law for the thermal conductivity. We also obtain a specific heat which varies linearly with temperature at low temperatures. At higher temperatures the quantity $C(T)/T^3$ exhibits a peak ("boson peak"). The former features are due to the anharmonic interactions, whereas the latter is mainly due to the disorder in the harmonic elastic constants.

To find a theoretical explanation of the low-frequency vibrational properties of glasses and their related low-temperature thermal properties is a long-standing problem [1]. At very low temperatures (around and below 1K) the specific heat $C(T)$ varies (almost) linearly with temperature T[2], and the thermal conductivity [3] $\kappa(T)$ obeys a $\kappa \propto T^s$ law with $s \approx 2$. At higher temperatures (around 10 K) $\kappa(T)$ shows a characteristic plateau, and the quantity $C(T)/T^3$ has a peak which corresponds to a peak in the density of states $g(\omega)$, divided by ω^2 ("boson peak"). The anomalies below ~ 1 K are traditionally discussed in terms of the tunneling model [4] in which the acoustic waves are assumed to be scattered inelastically by local bistable defects (two-level systems, TLS) with a broad distribution of resonance energies. The features around 10 K have been discussed in terms of a generalization of this model (soft-potential model) [5]
Although many of the observed experimental findings including the absorption and dispersion of ultrasound could be explained by the TLS model and its generalizations [5, 6], doubts where raised [7] whether the presence of TLS centers could be the only reason for the observed anomalies. Indeed, for the anomalous specific heat a much more general explanation in terms of a spin-glass like model was proposed recently [8].

For the boson peak there exist a large number of alternative models [9]. To our opinion the most simple and obvious explanation is the one in terms of spatially fluctuating elastic constants [10, 11] which lead to elastic scattering of phonons by the frozen disorder [12].

In the present contribution we generalize our previous ideas concerning the boson peak anomaly to include an anharmonic interaction. By field theoretic methods, borrowed from the theory of electrons in disordered systems [13, 14, 15, 16], we show that the low-temperature anomalies can be explained as a result of the inelastic mode-mode coupling of acoustic phonons induced by the combined effect of disorder and anharmonicity.

In order to demonstrate the main features of our proposed scenario we consider a simplified model of an elastic continuum which supports only longitudinal vibrations[1] (i. e. $\nabla \times \mathbf{u} = 0$, where $\mathbf{u}(\mathbf{r}, t)$ are the displacements), which is described by a classical Lagrangian density of the form

$$\mathcal{L}(\mathbf{r}, t) = \mathcal{T}(\mathbf{r}, t) - \mathcal{V}(\mathbf{r}, t) = \frac{m_0}{2}\left([\partial_t \mathbf{u}(\mathbf{r}, t)]^2 \;-\; \bar{K}[\nabla \cdot \mathbf{u}(\mathbf{r}, t)]^2\right) \tag{1}$$

Here $\mathcal{T}$ and $\mathcal{V}$ are the kinetic and potential energy densities, $m_0 = M\rho_0$ is the mass density, M the atomic mass, ρ_0 the number density. $\bar{K} = c^2 = (\lambda + 2\mu)$, is the elastic constant composed of the Lamé constants λ, μ, and c is the sound velocity. The elastic constant $\bar{K}$ is now assumed both to have a spatial variation due to the structural disorder of the material as well as a space-and-time variation due to an anharmonic coupling to the density fluctuations $\Delta\rho(\mathbf{r}, t)/\rho_0 = -\nabla \cdot \mathbf{u}(\mathbf{r}, t)$:

$$\bar{K}(\mathbf{r}, t) = [K_0 + \Delta K(\mathbf{r})][1 - 2g\nabla \cdot \mathbf{u}(\mathbf{r}, t)] \tag{2}$$

$\Delta K(\mathbf{r})$ is the static spatial fluctuation of $\bar{K}$ which is supposed to have a Gaussian distribution of the form

$$P[\Delta K] = P_0 \exp\{-\frac{1}{2\gamma}\int \mathrm{d}^3\mathbf{r}[\Delta K(\mathbf{r})]^2\} \tag{3}$$

where $\gamma = \overline{\Delta K^2}$ (the variance of ΔK) is the parameter which measures the strength of the disorder. g is the Grüneisen parameter defined by

$$g = \mathrm{d}\ln c/\mathrm{d}\ln\rho|_{\rho=\rho_0} = \frac{1}{2}\mathrm{d}\ln\bar{K}/\mathrm{d}\ln\rho\Big|_{\rho=\rho_0} \tag{4}$$

The quantum partition function can now be written as [18]

$$Z = \mathrm{Tr}\{e^{\hat{\mathcal{H}}/k_BT}\} = \int \mathcal{D}[\mathbf{u}]\, e^{\frac{1}{\hbar}S[\mathbf{u}(\mathbf{r}, \tau = it)]} = \int \mathcal{D}[\mathbf{u}]\, e^{\frac{1}{\hbar}\int_0^\beta \mathrm{d}\tau \int \mathrm{d}^3\mathbf{r}\mathcal{L}(\mathbf{r}, \tau = it)} \tag{5}$$

S is the Euklidean (imaginary-time) action and $\beta = \hbar/k_BT$.

From now on we use dimensionless units such that lengths are measured in units of the inverse of the Debye wavenumber $k_D = [6\pi^2\rho_0]^{1/3}$, times in units of the inverse Debye frequency $\omega_D = ck_D$,

[1] The calculation including the transverse degrees of freedom is straightforward [17] but cumbersome and will be carried out in a later paper. However – as we believe – the main features of the full theory will be similar to those described by the present simplified version

energies in units of $\hbar\omega_D$, and temperatures in units of the Debye temperature $\Theta_D = \hbar\omega_D/k_B$. In these units the dimensionless mass density is given by $\tilde{m}_0 = m_0 c/\hbar k_D^4 = Mc^2/6\pi^2\hbar\omega_D$, and for the dimensionless average force constant we have $\tilde{K}_0 = 1$.

In order to be able to perform the disorder average from the outset we make use of the replica trick: Averages over physical variables are carried out with weights $e^{\frac{1}{\hbar}S[\mathbf{u}_\alpha(\mathbf{r},\tau)]}$ without denominators Z for $\bar{n}$ replicas (labelled by the index α), and we have to let $\bar{n} \to 0$ at the end of the calculation.

Integrating out the disorder produces an effective quartic interaction which is taken apart by the Fadeev-Popov ghost field method [16], introducing the matrix fields $Q_{\alpha\alpha'}(\mathbf{r},\tau,\tau') \equiv \frac{\tilde{m}_0}{2}\nabla\cdot\mathbf{u}_\alpha(\mathbf{r},\tau)\nabla\cdot\mathbf{u}_{\alpha'}(\mathbf{r},\tau')$ and the "ghost fields" $\Lambda_{\alpha\alpha'}(\mathbf{r},\tau,\tau')$. The latter turns out to play the role of a self ernergy and the average of the former is the density-density propagator, taken at $\mathbf{r}' \to \mathbf{r}$. Integrating out the original fields in the standard way [13, 14, 15, 16, 17] yields the following effective action

$$S_{eff}[Q,\Lambda] = -\frac{1}{2}\mathrm{Tr}\{\log[\hat{A}_0 + \hat{1} + \hat{\Lambda}]\} + \mathrm{Tr}\{\hat{\Lambda}\hat{Q}\} + \frac{\gamma}{2}\mathrm{Tr}\{\hat{Q}^2\} + \frac{4\gamma g^2}{\tilde{m}_0}\mathrm{Tr}\{\hat{Q}^3\} + \Delta S[Q,\Lambda] \tag{6}$$

where the operators appearing in (6) are defined by

$$\begin{aligned} <\mathbf{r}\tau|\hat{Q}|\mathbf{r}'\tau'>_{\alpha\alpha'} &= Q_{\alpha\alpha'}(\mathbf{r},\tau,\tau')\delta(\mathbf{r}-\mathbf{r}')\delta_{\alpha\alpha'} \\ <\mathbf{r}\tau|\hat{\Lambda}|\mathbf{r}'\tau'>_{\alpha\alpha'} &= \Lambda_{\alpha\alpha'}(\mathbf{r},\tau,\tau')\delta(\mathbf{r}-\mathbf{r}')\delta_{\alpha\alpha'} \\ <\mathbf{k}n|\hat{A}_0|\mathbf{k}'n'>_{\alpha\alpha'} &= (\omega_n^2/k^2)\,\delta_{\mathbf{k}\mathbf{k}'}\delta_{nn'}\delta_{\alpha\alpha'} \end{aligned} \tag{7}$$

Here n are integers which run from $-\infty$ to $+\infty$ and are related to the Matsubara frequencies by $\omega_n = 2\pi n k_B T$. ΔS is a contribution, which arises from odd powers in $\nabla\mathbf{u}$ and vanishes in the saddle-point approximation introduced below.

Varying the matrix fields Q and Λ yields saddle-point equations which can be solved by replica-diagonal and space independent functions $Q_{\alpha\alpha'}(\mathbf{r},\tau,\tau') = Q(\tau-\tau')\delta_{\alpha\alpha'}$ and $\Lambda_{\alpha\alpha'}(\mathbf{r},\tau,\tau') = \Lambda(\tau-\tau')\delta_{\alpha\alpha'}$. They obey self-consistent equations, which can be written in terms of the Fourier coefficients $Q_n = Q(z=i\omega_n) = \int_0^\beta \mathrm{d}\tilde{\tau}e^{-i\omega_n\tilde{\tau}}Q(\tilde{\tau})$ and $\Lambda_n = \Lambda(z=i\omega_n)$ with $\tilde{\tau} = \tau-\tau'$ and

$$Q(\tilde{\tau}) = \lim_{\mathbf{r}\to\mathbf{r}'}\frac{\tilde{m}_0}{2}\overline{<\nabla\cdot\mathbf{u}(\mathbf{r},\tilde{\tau}+\tau')\nabla\cdot\mathbf{u}(\mathbf{r}',\tau')>} \tag{8}$$

$\overline{\ldots}$ denotes a configuational average, $<\ldots>$ a thermal one. The self consistent saddle-point equations are

$$\begin{aligned} Q_n &= \frac{1}{2}\sum_{|\mathbf{k}|<k_D}\frac{k^2}{-(i\omega_n)^2 + k^2(1+\Lambda_n)} \\ \Lambda_n &= -\left(\gamma Q_n + \frac{12\gamma g^2}{\tilde{m}_0}Q_{2,n}\right) \end{aligned} \tag{9}$$

Here

$$Q_{2,n} = Q_2(z=i\omega_n) = T\sum_\nu Q_\nu Q_{n-\nu}$$

$$= \int_0^\beta \mathrm{d}\tilde{\tau} e^{-i\omega_n \tilde{\tau}} Q(\tilde{\tau})^2 \tag{10}$$

The quantity

$$G(k,z) = [-z^2 + k^2(1+\Lambda_n)]^{-1} \tag{11}$$

is the averaged displacement Green's function (propagator), and $Q(k,z) = k^2 G(k,z)$ is the density fluctuation propagator (also called response function or dynamical susceptibility). The density of states is given by $g(\omega) = -(\pi/2)\omega \mathrm{Im}\{G(z\!=\!\omega\!+\!i\epsilon)\}$ with $G(z) = \sum_k G(k,z)$.

Let us now first discuss equations (9) in the absence of the anharmonic Q_2 term (g=0). Without this term they constitute the self-consistent born approximation (SCBA) for force-constant disorder, which has been derived previously for mass disorder in ref. [17] by similar techniques. On the other hand, it can be shown that the well-known coherent-potential approximation (CPA) [19] reduces to the SCBA in the regime of weak disorder ($\gamma/K_0^2 < 1$). By means of the CPA (compared with a numerical simulation) it has been demonstrated in ref. [11] that the boson peak is a natural consequence of disorder in a harmonic solid with fluctuating elastic constants. Therefore it is not surprising that within the harmonic limit of the present model the SCBA also predicts a boson peak both in the quantity $g(\omega)/\omega^2$ and in $C(T)/T^3$ calculated from this $g(\omega)$. In fig. 1 we show $C(T)/T^3$ calculated for the harmonic model in SCBA for three values of γ. For $\gamma > \gamma_{crit} = 0.5$ the model becomes instable as in the harmonic models considered in [11].

Let us now turn to a discussion of the self-consistent equations (9) in the presence of the anharmonic terms. As in the harmonic case we use the analytic functions $Q(z), Q_2(z)$ and $\Lambda(z)$ in the real frequency domain $z\!=\!\omega\!+\!i\epsilon$, i. e. we have $Q(z) = Q'(\omega) + iQ''(\omega)$ with the spectral representation $Q(z) = (1/\pi)\int_{-\infty}^{+\infty} \mathrm{d}\bar{\omega} Q''(\bar{\omega})/(\bar{\omega} - z)$. The dynamical susceptibility can be re-written as

$$Q(z) = \frac{1}{2[1+\Lambda(z)]}\left(1 + z^2 G(z)\right) \tag{12}$$

We see that in the low-frequency regime (which we are interested in) the $z^2G(z)$ term is negligible. In the real frequency domain we can use the fluctuation-dissipation theorem [20] to relate $Q''(\omega)$ to the van-Hove correlation function $S(t) = \lim_{\mathbf{r}\to\mathbf{r}'} \frac{\tilde{m}_0}{2} \overline{< \nabla\cdot\mathbf{u}(\mathbf{r}, t+t_0)\nabla\cdot\mathbf{u}(\mathbf{r}', t_0) >}$ and its Fourier transform $S(\omega) = \int_{-\infty}^{+\infty} \mathrm{d}\, t\, e^{i\omega t}\, S(t)$ in the following way

$$\begin{aligned} Q''(\omega) &= \frac{1}{2}\left(1 - e^{-\beta\omega}\right) S(\omega) \equiv f(\beta\omega)\, S(\omega) \\ Q_2''(\omega) &= f(\beta\omega)\, S_2(\omega) \end{aligned} \tag{13}$$

with $S_2\omega) = \int_{-\infty}^{+\infty} \mathrm{d}t\, e^{i\omega t}\, S^2(t)$.

We have now a simplified set of equations which bears strong similarity to the set of equations studied in the context of glass transition singularities [12, 21]. The detailed analytic and numerical solution of the full and simplified set of equations will be studied in a future publication [22]. For the present discussion we only note that in the low-frequency regime the frequency dependence of $S(\omega)$

is very weak, and the temperature dependence enters via the thermal factors $f(\beta\omega)$. In order to be able to calculate the thermal conductivity we remind ourselves that the mean free path $\ell(\omega)$ can be calculated from the imaginary part of the inverse of the complex sound velocity $c(z) = \sqrt{1+\Lambda(z)}$:

$$\frac{1}{\ell(\omega)} = 2\omega \mathrm{Im}\{[1+\Lambda(z)]^{-1/2} \tag{14}$$

Using (12) this becomes for small ω:

$$\begin{aligned} \frac{1}{\ell(\omega)} &= 2\omega\sqrt{2}\mathrm{Im}\{[Q(z)]^{1/2} \\ &= 2\omega\left[|Q(z)| - Q'(\omega)\right]^{1/2} \end{aligned} \tag{15}$$

The function $Q(z)$ can be written as

$$Q(z) = \frac{1}{\pi}\int_{-\infty}^{+\infty} \mathrm{d}x f(x) \frac{S(x/\beta)}{x-\beta z} \tag{16}$$

If we believe that the frequency dependence of $S(\omega)$ is very weak, $Q(z)$ is essentially a function of βz so that we can state

$$\ell(\omega) \approx \tilde{f}(\beta\omega)/\omega \tag{17}$$

where $\tilde{f}(\beta\omega)$ is some function of $\beta\omega$. This is the same type of frequency and temperature dependence as that of the mean free path for inelastic scattering from two-level systems [4], which, inserted into the conventional formula for the thermal conductivity derived from the phonon Boltzmann equation

$$\kappa(T) = \int \mathrm{d}\omega g(\omega)\beta^2\omega^2 \frac{\exp\{\beta\omega\}}{(\exp\{\beta\omega\}-1)^2}\frac{1}{3}c_0^2\ell(\omega) \tag{18}$$

leads to

$$\kappa(T) \propto T^2 \tag{19}$$

In order to be able to discuss the specific heat we have to consider the average energy which can be easily calculated by adding a static source j to the Lagrangian: $\mathcal{L}(j) = \mathcal{T}(1+j) - \mathcal{V}(1-j)$, and we have

$$\begin{aligned} \overline{\mathcal{H}} &= \overline{\frac{1}{\beta}\frac{\mathrm{d}}{\mathrm{d}j}\ln Z(j)|_{j=0}} \\ &= \lim_{\bar{n}\to 0} \overline{Z(j)^{\bar{n}-1}\frac{1}{\beta}\frac{\mathrm{d}}{\mathrm{d}j}Z(j)|_{j=0}} \end{aligned} \tag{20}$$

Omitting the ΔS term the effective j dependent action is

$$S_{eff}(j) = -\frac{1}{2}\mathrm{Tr}\{\log[\hat{A}_0(1+j)+\hat{1}(1-j)+\hat{\Lambda}]\} + \mathrm{Tr}\{\hat{\Lambda}\hat{Q}\} + \left(\frac{\gamma}{2}\mathrm{Tr}\{\hat{Q}^2\} + \frac{4\gamma g^2}{\tilde{m}_0}\mathrm{Tr}\{\hat{Q}^3\}\right)(1-j)^2 \tag{21}$$

This gives in saddle-point approximation

$$\begin{aligned} \overline{\mathcal{H}} &= const. + T\sum_{n,\mathbf{k}} \frac{-\omega_n^2}{\omega_n^2 + k^2[1+\Lambda_n]} + \frac{4\gamma g^2 T^2}{\tilde{m}_0}\sum_{n,\nu} Q_n Q_\nu Q_{-n-\nu} \\ &= const. + \overline{\mathcal{H}_D(T)} + \overline{\Delta\mathcal{H}(T)} \end{aligned} \tag{22}$$

Here $\overline{\mathcal{H}_D(T)}$ is the usual Debye expression for the average energy[2] and $\overline{\Delta\mathcal{H}(T)}$ is the anomalous contribution which arises due to the anharmonic coupling. Since Q_n is an even function of the Matsubara frequencies $\omega_n \propto T$, we can state $\overline{\Delta\mathcal{H}(T)} = (4\gamma g^2 T^2/\tilde{m}_0)Q(z=0)^3 T^2 + O(T^4)$ and therefore

$$\lim_{T\to 0} C(T) = \frac{8\gamma g^2}{\tilde{m}_0} Q(0)^3 T \tag{23}$$

Numerical calculations to determine the entire temperature dependence of the specific heat using the full self-consistent equations (9) will be done shortly [22].

We conclude by emphasizing that our anharmonic contribution to the action arises from a *combination* of disorder and anharmonicity. If one of the ingredients of our theory, disorder (represented by the parameter γ) or anharmonicity (represented by g) vanish, the anomalies disappear.

We are grateful to the Deutsche Forschungsgemeinschaft and the University of Oregon for financial support. During the stay of W. S. and M. P. at the University of Oregon we profited from numerous illuminating discussions with D. Belitz. We would like to thank S. Hunklinger for encouragement and helpful discussions.

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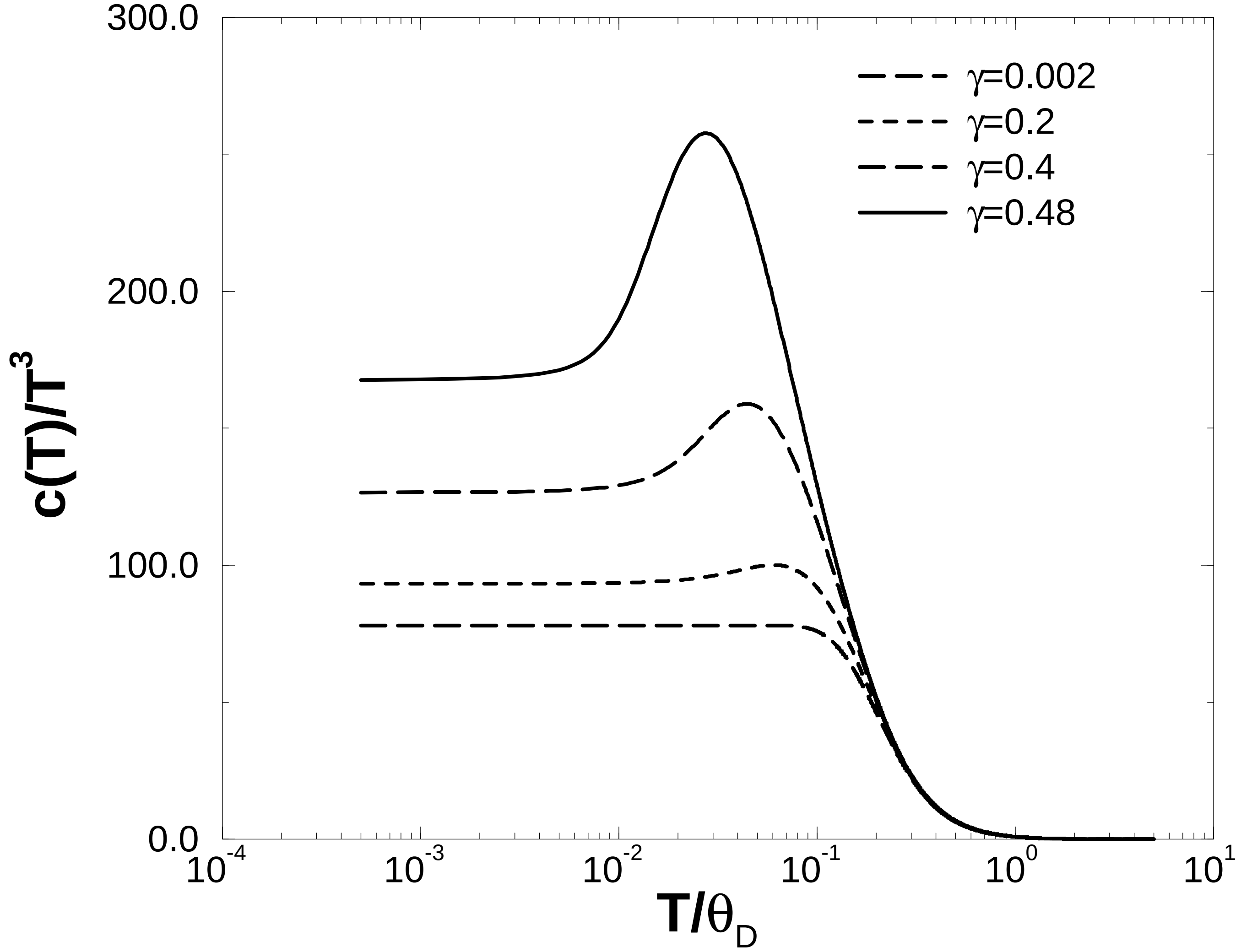
γ=0.002
γ=0.2
γ=0.4
γ=0.48
300.0
200.0
100.0
0.0
c(T)/T^3
10^-4
10^-3
10^-2
10^-1
10^0
10^1
T/θ_D

Figure Caption

Fig. 1: Reduced specific heat $C(T)/T^3$ against temperature, calculated in self-consistent Born approximation (g=0) for 4 values of the disorder parameter $\gamma = \overline{(\Delta K)^2}$.