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Heat capacity of mesoscopically disordered superconductors with emphasis on MgB_2

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Abstract

The temperature dependence of the electronic heat capacity $C(T)$ was calculated for a mesoscopically disordered s-wave superconductor treated as a spatial ensemble of domains with continuously varying superconducting properties. The domains are assumed to have sizes $L > \xi$, where ξ is the coherence length. Each domain is characterized by a certain critical temperature T_{c0} in the range $[0, T_c]$. The averaging over a broad superconducting gap distribution leads to $\langle C(T)\rangle \propto T^2$ for low T, whereas the specific heat anomaly at T_c is substantially smeared. For narrow gap distributions there exists an intermediate-T range, where the curve $\langle C(T)\rangle$ can be well approximated by an exponential Bardeen–Cooper–Schrieffer-like dependence with the effective gap smaller than the weak-coupling value. The results are applicable in the general case of inhomogeneous superconductors including, e.g., electron-doped and hole-doped cuprates. The $C(T)$ data for MgB_2, where multiple gaps are observed, are discussed in more detail.

1. Introduction

The unexpected discovery of the relatively high-temperature (high-T) superconductor MgB_2 with a critical temperature $T_c \approx 40$ K [1] has cast considerable doubt on the validity of the notion that high T_cs are exclusive to substances with a spin-fluctuation-driven Cooper pairing and, consequently, with a predominantly $d_{x^2-y^2}$-wave symmetry of the superconducting order parameter. Indeed, an obvious absence of magnetic ions, a considerable isotopic effect [2], and Bardeen–Cooper–Schrieffer-like (BCS-like) coherent peaks in the optical conductivity [3] and spin–lattice relaxation [4] are indicative of the conventional s-wave pairing in MgB_2. As for the electron–phonon background of the superconductivity, it also seems highly probable, although a fairly exotic multiple-gap scenario is needed to reconcile the available data (see, e.g., the [5]). It is very remarkable that the multiple-gap conventional Cooper pairing is found directly in a number of point-contact, tunnelling, and Raman measurements [6]. It is even more important that the distribution of gaps may be rather broad [7] and spatially resolved [8], although much controversy exists over the number and widths of gaps in the electron density of states (DOS).

In actual fact, the order parameter symmetry for MgB_2 is not unambiguously determined. Some muon spin-rotation [9] and optical [3] measurements demonstrate that the low-T asymptotics of the magnetic field penetration depth $\lambda(T)$ is a power-law one. This was interpreted as either an unconventional superconductivity or at least a highly anisotropic s-wave pairing. On the other hand, other muon spin-relaxation [10] and microwave surface impedance [11] investigations show either the conventional exponential low-T behaviour of $\lambda(T)$, although with a reduced gap value in comparison with the BCS result [11], or a dependence governed by the weighted combination of two exponential terms with different gap parameters [10]. Measurements of $\lambda(T)$ with the help of the same radio-frequency technique both for sintered pellets and thin films of MgB_2 clearly indicated that the degree of disorder is at least one of the key factors affecting the low-T behaviour [12]. That is, $\lambda(T) \propto T^2$ for pellets and $\lambda(T)$ is exponential with a reduced value of the energy gap for films.

Thermodynamic measurements might be especially important in determining the low-T symmetry-based superconducting properties of MgB_2, because the minority phases or grain boundaries do not affect the results substantially, in contrast to, e.g., transport phenomena. The electronic heat capacity ($C(T)$) behaviour near T_c is also of great importance for elucidating the nature of the superconductivity here. And, indeed, there were a lot of specific heat investigations for MgB_2 performed by various groups [13].

The main features of the data for $C(T)$ are:

(i) small values of the phase transition anomaly $\Delta C = C_s - C_n$ at T_c [13–16] in comparison to the BCS case [17], when the ratio $\mu = \Delta C/[\gamma_S(T_c)T_c]$ is equal to $\mu_{BCS} = 12/[7\zeta(3)]$; and

(ii) deviations from the asymptotic BCS behaviour at $T \ll T_c$:

$$C_{BCS}^{asympt}(T) = N(0)\left(\frac{2\pi\Delta_0^5}{T^3}\right)^{1/2} \exp\left(-\frac{\Delta_0}{T}\right). \tag{1}$$

Here γ_S is a Sommerfeld constant, the subscripts s and n correspond to the superconducting and normal states, respectively, $N(0)$ is the electron DOS at the Fermi level, Δ_0 is the energy gap value at $T = 0$, $k_B = \hbar = 1$. The deviations from equation (1) may be twofold: power-law-like $\propto T^2$ [14] and of the form $\propto \exp(-\frac{A}{T})$ [16, 18], where the constant A is much less than $\frac{\pi}{\gamma}T_c \approx 1.76T_c$, as it should be for a weak-coupling superconductor [17]; $\gamma = 1.78\ldots$ is the Euler constant. Thus, the raw specific heat data do not give definite answers to the problems of the order parameter symmetry and the underlying mechanisms of superconductivity.

In this article, on the basis of the experimentally proven *distribution* of energy gaps, we show that *both* main features of $C_s(T)$ observed for MgB_2 can be explained by the conventional s-wave superconductivity, so these data can be easily reconciled with other observations [3, 4]. The approach adopted, being an extension of the earlier one [19], is phenomenological because the origin of the gap distribution is not known precisely. However, in accordance with the tunnelling data [8], the gap distribution is considered to occur *in real space* rather than in $\boldsymbol{k}$-space, as was suggested, e.g., in [13, 14]. The theoretical description of such spatially disordered superconductors depends on the ratio between the characteristic superconducting domain size L and the coherence length ξ [20]. If $L > \xi$, superconducting properties are determined by local values of the order parameter Δ. Our approach corresponds to these so-called large-scale inhomogeneities, whereas the small-scale inhomogeneities are matched by the reverse inequality $L < \xi$ [21, 22]. The quantity ξ is T-dependent and tends to infinity at T_c. Hence, in the close vicinity of T_c, strictly speaking, all inhomogeneities become small-scale ones and a divergent correction proportional to $(\frac{T_c}{T} - 1)^{-1/2}$ appears in the expression for $C(T)$ [21]. Nevertheless, it can be easily shown that for conventional superconductors including MgB_2, the relevant T-range is very small, so its influence on the phase transition smearing is negligible. Moreover, it has been disclosed recently that the correction for three-dimensional superconductors is actually finite [22]. Therefore we can identify ξ with the T-independent coherence length, dependent on the Pippard coherence length $\xi_0 \approx \frac{\hbar v_F}{\pi\Delta}$ and the mean free path l [17]. Here v_F is the Fermi velocity. For MgB_2, which can be considered a clean superconductor [23, 24], the quantities of interest are $\xi \approx \xi_0 \ll l \approx 600$ Å, although there is a significant scatter of ξ_0, inferred from different experiments and for *different kinds* of sample [6, 25], so we may estimate this quantity as lying in the range from 25 to 120 Å. This dispersion of ξ_0 qualitatively correlates with the broad spectra of gaps in tunnel and point-contact spectra [6–8, 13].

So far, our reasoning has ignored the possible influence of the strong-coupling effects on $C_s(T)$ in MgB_2. At the same time, recent de Haas–van Alphen measurements [26] revealed strong electron–phonon renormalization of the effective masses, differing for various Fermi surface sheets. The last property is in agreement with the band-structure calculations [27]. This circumstance does not require, however, an obligatory accounting for strong-coupling effects while studying $C_s(T)$. In fact, the asymptotic low-T dependence of $C_s(T)$ in the framework of the Eliashberg theory remains the same as in the weak-coupling limit, although its amplitude is reduced [28]. On the other hand, the anomaly ΔC near T_c *increases* in the strong-coupling regime [28, 29], the effect being opposite to what is observed for MgB_2 [13–16, 30]. This means that the effects to be accounted for formidably exceed the strong-coupling augmentation. Hence, hereafter we shall neglect the electron–phonon renormalization altogether.

2. Theory

Let us examine a T-independent configuration of mesoscopic domains, with each domain having the following properties:

(i) at $T = 0$, it is described by a certain superconducting order parameter $\Delta_0 \leqslant \Delta_0^{\max}$;
(ii) up to a relevant critical temperature $T_{c0}(\Delta_0) = \frac{\gamma}{\pi}\Delta_0$, it behaves like an isotropic BCS superconductor, i.e. the superconducting order parameter $\Delta(T)$ is the Mühlschlegel function $\Delta(T) = \Delta_{BCS}(\Delta_0, T)$ [17]; and the electronic specific heat is characterized in this interval by the function $C_s(\Delta, T)$;
(iii) at $T > T_{c0}$, it transforms into the normal state, and the relevant property is [17]

$$C_n(T) = \frac{\pi^2}{3} N(0) T. \tag{2}$$

At the same time, the values of Δ_0 scatter for various domains. The current carriers move freely across domains and inside each domain acquire appropriate properties. The picture adopted is especially suitable for superconductors with small coherence lengths ξ_0 [13].

For simplicity, we restrict ourselves to the situation when the whole sample above T_c is electronically homogeneous, i.e. is characterized by a common approximately constant $N(0)$ value. Therefore, we completely neglect possible electron wavefunction mismatches or formation of potential barriers at the domain boundaries. Such an assumption is justified for an analysis of bulk thermodynamic properties or, e.g., the magnetic field penetration depth, but may be invalid for transport phenomena, which fall beyond the scope of this publication. Below T_{c0} for a given mesoscopic domain, a corresponding isotropic gap appears on the Fermi surface. The microscopic background of the assumed scatter in the T_{c0}s may be diverse but ultimately manifests itself as a variation either of the electron–phonon interaction magnitude or of local values of the Coulomb pseudopotential.

In the framework of our phenomenological approach, the superconductivity (if any) inside a chosen domain is described by the relevant parameters Δ_0 and T_{c0}. They are bounded from above by $\Delta_0^{\max}$ and T_c, respectively. These Δ_0s may or may not group around a certain crowding value Δ_0^*, depending on the sample texture. The existence of two such possibilities is in accordance with the varied data for MgB_2 [6–8, 13, 31]. The specific gap distribution is described by the function $f_0(\Delta_0)$.

Thus, for all T in the interval $[0, T_c]$, where $T_c = \max T_{c0}$, the superconducting sample consists of superconducting (s) and nonsuperconducting (n) grains more or less homogeneously distributed over the sample volume.

The measured $C_s(T)$ is an averaged sum of contributions from the two phases:

$$\langle C(T)\rangle = \langle C_n(T)\rangle + \langle C_s(T)\rangle, \tag{3}$$

which depends on the distribution function $f(\Delta, T)$ of superconducting domains, and on the fraction $\rho_n(T)$ of the normal phase [19]:

$$\langle C_n(T)\rangle = C_n(T)\rho_n(T), \tag{4}$$

$$\langle C_s(T)\rangle = \int_0^{\Delta^{\max}(T)} C_s(\Delta, T) f(\Delta, T)\, d\Delta. \tag{5}$$

Here $\Delta^{\max}(T) = \Delta_{BCS}(\Delta_0^{\max}, T)$ and $f(\Delta, T)$ is a result of the thermal evolution of the initial (at $T = 0$) distribution function $f_0(\Delta_0)$. It is convenient to normalize all temperatures by T_c and all energy parameters by $\Delta_0^{\max}$: $t = T/T_c$, $\delta = \Delta/\Delta_0^{\max}$ with relevant indices retained, and to consider $C_s(T)$ and $C_n(T)$ together with their averaged counterparts, normalized by the $C_n(T_c)$ value, i.e., $c_{s,n}(t) = C_{s,n}(T)/C_n(T_c)$. Then one can easily find that for each domain, characterized by the parameter δ_0 at $t = 0$, the dimensionless heat capacity is either

$$c_n(t) = t, \qquad t > \delta_0 \tag{6}$$

or

$$c_s(t) = \delta_0 c_{BCS}\left(\frac{t}{\delta_0}\right), \qquad t < \delta_0, \tag{7}$$

where $c_{BCS}(x)$ is a well-known normalized heat capacity function for a standard BCS superconductor [17]. For a surmised domain ensemble, a distribution function $f(\Delta, T)$ for finite T is defined by the formula

$$f(\Delta, T)\, d\Delta = f_0(\Delta_0)\, d\Delta_0. \tag{8}$$

Then the dimensionless heat capacity takes the form

$$\langle c_s(t)\rangle = \int_t^1 c_{BCS}\left(\frac{t}{\delta_0}\right) f_0(\delta_0)\delta_0\, d\delta_0. \tag{9}$$

Introducing a new variable $z = t/\delta_0$ and expanding the function $f_0(t/z)$ into a series, we arrive at the proper low-t asymptotics:

$$\langle c_s(t \to 0)\rangle = t^2 \int_0^1 \frac{dz}{z^3} f_0(0) c_{BCS}(z) \approx 2.45 f_0(0) t^2. \tag{10}$$

The t-dependence of the next term in the expansion for $\langle c_s(t)\rangle$ can be estimated in the limit $t \to 0$ by substitution of the normalized expression (1) for $c_{BCS}(z)$. It turns out that this expression decreases as $O[t^{5/2}\exp(-\frac{\pi}{\gamma t})]$.

Now, in the same low-T region let us take a look at the contribution $\langle c_n(t)\rangle$ of the continuously expanding normal phase. At any T, all domains with $\Delta_0 < \frac{\pi}{\gamma}T$ (i.e. $\delta_0 < t$) are nonsuperconducting, with the total normal phase fraction being

$$\rho_n(t) = \rho_n(0) + \int_0^t f_0(\delta_0)\,d\delta_0. \tag{11}$$

For simplicity, below we restrict ourselves to the case when all domains at $t = 0$ are superconducting, i.e. $\rho_n(0) = 0$. A generalization to the case $\rho_n(0) \neq 0$ is obvious: at each temperature there exists an additional contribution from the normal phase. Then the function $f_0(\delta_0)$ should be normalized by $1 - \rho_n(0)$, and all averaging-driven effects would accordingly decrease. Moreover, if $\rho_n(0) \neq 0$, the observed heat capacity $\langle c(t)\rangle$ must include an extra linear contribution $\rho_n(0)t$ in the true superconducting state exhibiting the Meissner effect.

As for the second term in the equation (11), the approximation of $f_0(\delta_0)$ by its limiting value $f_0(0)$ demonstrates that the main temperature-dependent contribution to $\rho_n(t)$ is *linear* in t. Since $c_n(t)$ is also a linear function of t, the apparent contribution $\langle c_n(t)\rangle$ of the normal phase to the resulting specific heat $\langle c(t)\rangle$ is quadratic in t for small t, similarly to $\langle c_s(t)\rangle$. Thus, in the suggested model of the disordered superconductor with a broad continuous spatial distribution of domains, possessing different T_cs, normal and superconducting contributions to thermodynamical quantities are *functionally indistinguishable* from each other.

The analysis of equation (9) shows that it is a phase redistribution (superconducting versus nonsuperconducting) in the ensemble rather than the presence of domains with infinitesimally small δs which necessarily leads to the $\langle c_s(t)\rangle$ power-law low-t asymptotics. From the mathematical point of view this circumstance is reflected by the variability of the lower limits of the integral in equation (9). And to obtain a power-law asymptotics persisting down to $T = 0$, it is essential for the distribution function $f_0(\delta_0)$ to extend down to $\delta_0 = 0$. Otherwise, in the vicinity of the zero temperature the $\langle c_s(t)\rangle$ exponential dependence will be recovered.

Moreover, if there exists a certain $\delta_0^{\min}$ such that $f_0(\delta_0) = 0$ for all $\delta_0 \leqslant \delta_0^{\min}$, then for $t < \delta_0^{\min}$ the 'effective' lower limit of the integral (9) becomes constant. Then, taking into account the exponential behaviour (1) it is easy to show that ultimately the integral (9) at $t \to 0$ is a sum of two terms proportional to $\exp(-\frac{\pi}{\gamma t})$ and $\exp(-\frac{\pi}{\gamma t}\delta_0^{\min})$, notwithstanding the $f_0(\delta_0)$ profile in the interval $\delta_0^{\min} \leqslant \delta_0 \leqslant 1$. The latter will define the pre-exponential factors in these two terms. This result is an alternative to the explanation of some experimental data for MgB_2 in the framework of the well-known two-gap model extensively developed by other investigators [5, 32, 33].

3. Numerical results

In addition to the low-T asymptotics the overall T-dependence of the heat capacity C up to T_c is also of considerable interest. Especially important is tracing the smearing of the anomaly ΔC by the *same* effect of disorder which leads to the transformation of the intrinsic exponential low-T behaviour of $C_s(T)$ into a power-law one. These objectives were met by numerical calculations.

For this purpose, a Gaussian model distribution function $f_0^G(\delta_0)$ was used:

$$f_0^G(\delta_0) \propto \exp\left[-\frac{(\delta_0 - \delta_0^*)^2}{2d^2}\right]. \tag{12}$$

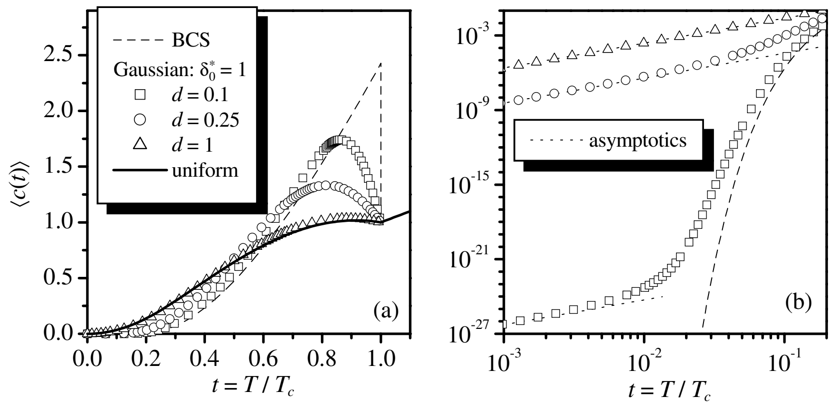

Figure 1. (a) Temperature dependences of the normalized total electronic heat capacity $\langle c(t)\rangle$ in comparison with the BCS dependence of the superconducting phase fraction. Gaussian distributions with $\delta_0^* = 1$. (b) Low-temperature portions of the relevant curves on a log–log scale together with their t^2-asymptotics.

The parameter δ_0^* designates the peak position, which may vary from 0 to 1. By changing the parameter d we control the dispersion of the domain superconducting properties. Nevertheless, for any d the function $f_0^{\mathrm{G}}(\delta_0)$ does not vanish in the limit $\delta_0 = 0$ and its Taylor series begins with a constant as the main term. Only for highly improbable distribution functions, when simultaneously $f_0(\delta_0)$ extends to $\delta_0 = 0$ and matches the condition $f_0(\delta_0 = 0) = 0$, may the Taylor series begin with the next term resulting in the asymptotics $C_{\mathrm{s}}(T) \propto T^3$.

In figure 1 the dependences $\langle c(t)\rangle$ are depicted in panel (a) for $\delta_0^* = 1$ and different dispersion values d. A substantial spreading of the anomaly ΔC readily seen in figure 1 seems quite natural in view of the results for $\mathrm{MgB_2}$ [13–16]. However, the concomitant superposition of various domain contributions distorts the whole of curves $C_{\mathrm{s}}(T)$ and $C(T)$, which is much less trivial. This very superposition leads for low T to the power-law behaviour, the asymptotics of which was analysed above. The low-T parts of the curves $\langle c(t)\rangle$ are displayed on the log–log scale in panel (b). Dotted straight lines correspond to the pertinent T^2-asymptotics for each curve. It is clear that the validity range of the asymptotics extends with the increase of d. Although intervals where the T^2-approximation holds good exist for any d, for small d it is merely of academic interest, because both temperatures and heat capacities become too tiny to be experimentally significant. On the other hand, for higher T in this case the averaged dependences $\langle c(t)\rangle$ lie rather close to the exponential curve inherent to the BCS theory (the dashed curve). Such transitional parts of the dependences $\langle c(t)\rangle$ describe well the exponential low-T behaviour for some samples of $\mathrm{MgB_2}$ [16, 18] with smaller exponents than in the BCS case.

For large d, when the Gaussian distribution function $f_0^{\mathrm{G}}(\delta_0)$ becomes almost uniform (such a random dense, although quasi-discrete, distribution of gaps was found in point-contact spectra [7]), the quadratic asymptotics is valid at least up to $t = 0.1$ (for the uniform distribution $f_0^{\mathrm{U}}(\delta_0) =$ constant, the relative error of the t^2-asymptotics is $\approx$0.6% at $t = 0.1$ and $\approx$5% at $t = 0.2$), which agrees with the measurements [14]. For intermediate d the experimental data in the relevant T-range may be satisfactorily represented by power-law curves $C(T) \propto T^n$ with $n \geqslant 2$.

One can draw another important conclusion from the numerical data shown in figure 1. A one-parameter fitting explains *both* the smearing of the heat capacity anomaly at T_{c} and the appearance of the power-law asymptotics. The latter reproduces the results appropriate to superconductors with order parameters of the $d_{x^2-y^2}$ wave [34] or extended s wave with uniaxial anisotropy [13, 14] symmetry. The patterns displayed in these figures explain well the experimental heat capacity dependences $C(T)$ for MgB_2, which demonstrate power-law behaviour for lowest attainable T [13, 14] or above the exponential low-T tail [18]. At the same time, the reduction of the anomaly ΔC at T_c with the increase of d, traced in figure 1(a), adequately describes the ΔC magnitudes inferred from the analysis of the observed total heat capacity of MgB_2, making allowance for crystal lattice and impurity components. That is, $\mu \approx 1.13$ [16], 0.82 [13, 14], 0.7 [15], so the experimental specific heat jump is substantially smaller than the BCS value μ_{BCS}.

4. Conclusions

The results obtained here are of a quite general nature and fit well the observed heat capacity dependences both for cuprates and for magnesium diboride. Our main assumption is the proposed large-scale ($L > \xi_0$) spatial inhomogeneities of Δ (and T_c). As for cuprates, the origin of those heterogeneities was discussed in our previous publications [19, 35]. On the other hand, in MgB_2 large enough inclusions (they influence the heat capacity!) of different phases or planar defects may be the most probable cause of the Δ spread. One could mention, e.g., observed MgB_4 grains and stacking faults [36] or nonstoichiometry modelled by $Mg_{1+\delta}B_2$ phases [37]. X-ray analysis shows that MgB_2 can be microscopically nonstoichiometric up to 5–10% [38]. However, we also cannot exclude the possibility of an electronic phase separation, since the substance concerned is on the verge of the electronic topological transition [39, 40].

It should be noted that a somewhat related idea of multi-gap superconductivity with gap diversity on different sheets of the Fermi surface, i.e. in the momentum space, was presented for MgB_2 [5, 6, 32, 41–43]. This is an extension of the well-known two-band superconductivity concept [44], which, in its turn, approximates the complex anisotropy of the electron spectrum. The expected observable results of the picture presented here and the two-band model differ in the sense that in the latter case there should be two different gap parameters connected by interband scattering matrix elements or gaps clustered into two groups [43]. On the other hand, our gap distribution should be quasi-continuous due to the proximity effect. Our point of view is directly supported by the point-contact observation of gap values in the interval between the so-called large and small gaps [7]. Although the relevant gap-maxima histogram corresponds to different contacts, it reflects properties of various contact areas of the same pellet. Another argument for the validity of the spatial gap distribution is the disappearance of the gap averaging for samples of better quality in the same experimental set-up [12]. Finally, a three-gap structure was also seen in the tunnel spectra [45].

To summarize, we presented a phenomenological model of the disordered s-wave superconductor with a random domain network possessing continuously varying superconducting properties. The spatially averaged electronic heat capacity $\langle C(T)\rangle$ is calculated. It is shown that its low-T asymptotics is a power-law one $\propto T^2$, whereas the anomaly ΔC at T_c is simultaneously smeared. These are just the features appropriate to the heat capacity of MgB_2 and cuprates.

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