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# Thermodynamics of superconductors with charge-density waves

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**Abstract**

Equations for the temperature- ($T$-) dependent superconducting ($\Delta(T)$) and dielectric ($\Sigma(T)$) order parameters are solved self-consistently in the partial dielectric gapping model of Bilbro and McMillan for superconductors with charge-density waves (CDWs). It is shown that for the close enough structural phase transition temperature, $T_s$, and superconducting one, $T_c$, with $T_s > T_c$, $\Sigma$ below $T_c$ may become smaller than $\Delta$. The electronic heat capacity $C(T)$ is calculated. It is shown that the discontinuity $\Delta C$ at $T = T_c$ is always smaller than the Bardeen–Cooper–Schrieffer value. The effect is detectable over a wide range of the model parameters. Experimental implications for CDW superconductors, such as A15 compounds, high-$T_c$ cuprates, and $MgB_2$, are suggested and discussed.

## 1. Introduction

The superconducting transition due to the small role of the fluctuation effects near the critical temperature $T_c$ for almost all objects is a classical example of a mean-field second-kind transition [1]. The standard weak-coupling Bardeen–Cooper–Schrieffer (BCS) microscopic theory predicts, in particular, a universal discontinuity $\Delta C = C_s - C_n$ of the electronic heat capacity $C(T)$ at $T_c$, namely,

$$\frac{\Delta C}{\gamma_S T_c} = \frac{12}{7\zeta(3)} \approx 1.43. \qquad (1)$$

Here the subscripts s and n denote the superconducting and normal states, respectively, $T$ is temperature, $\zeta(x)$ is the Riemann zeta function, and $\gamma_S$ is the Sommerfeld constant pertinent to the normal phase, which can be expressed in terms of the electron density of states (DOS) per spin on the Fermi surface (FS) $N(0)$: $\gamma_S = 2\pi^2 N(0)/3$.

Of course, the weak-coupling theory is applicable only to conventional low-$T_c$ metals and alloys. Strong-coupling effects, as is well known [2–4], may substantially enhance $\Delta C$ over the BCS value. This remains true for any boson field giving rise to the Cooper pairing. On the other hand, there are many factors that may influence the heat capacity jump. For example, it is worth noting that the Van Hove singularity of the electron spectrum leads [5] (see also the discussion in [6, 7]) to the $\Delta C$ increase in comparison with the conventional case when the primordial electronic DOS is considered structureless.

All the aforesaid concerns only the conventional s-wave superconductivity, whatever its specific mechanism or strength. The unconventional order parameter symmetry may influence thermodynamic characteristics conspicuously, although the underlying mean-field picture remains the same as for conventional superconductors [8]. On the other hand, the more exotic approaches, such as a bipolaronic one [9], revoke the second-kind transition itself, so the very discussion of the specific heat jump is out of the question.

Now let us turn our attention to the experiment. The well-justified strong-coupling increase of $\Delta C$ was undoubtedly observed in many materials [10] and is not the subject of our investigation presented here. However, there are quite different cases of a discontinuity *decrease* for superconductors with high and moderately high temperature when one would expect the usual behaviour. In particular, in the first publications [11, 12] on $C(T)$ measurements for the ceramics $BaPb_{1-x}Bi_xO_3$ with $x = 0.25$ and $T_c = 12$–13 K, the conclusion that the jump $\Delta C$ was absent was drawn. Only very sensitive measurements [13] and a special treatment [14] proved the discontinuity to exist in these solid solutions. $C(T)$ studies for a member, $Ba_{0.6}K_{0.4}BiO_3$, of the related oxide family with the maximal $T_c \approx 30$ K also show either a total absence of the anomaly [15] or a 60% reduction [16] in comparison with the discontinuity calculated on the basis of the BCS theory from the upper critical magnetic field ($H_{c2}(T)$) data [17]. On the other hand, a recent heat-pulse investigation of $Ba_{1-x}K_xBiO_3$ with $x = 0.40$ and 0.47 in magnetic fields [18] did reveal BCS-size jumps near $T_c$. Since the precise determination of the ratio $\Delta C/\gamma_S T_c$ is hampered by the quite ambiguous isolation of the electronic contribution to $C_s(T)$, the controversy still persists. An anomalously small $\Delta C/\gamma_S T_c \approx 0.6$ was observed [19] for $Li_{1.16}Ti_{1.84}O_4$ with $T_c^{\mathrm{onset}} \approx 9$ K, with a FS partially reduced by the composition variation relative to the parent compound $LiTi_2O_4$ with $T_c^{\mathrm{onset}} \approx 12.6$ K. Experiments on the Laves phases $HfV_2$ and $ZrV_2$ [20] demonstrated that the ratio $\Delta C/\gamma_S T_c$ is considerably smaller than the BCS value (1).

For high-$T_c$ oxides, one would even more confidently expect a substantial increase of $\Delta C$ to be caused by the strong-coupling renormalization. Instead, usually, jumps for different substances are smaller than the BCS values. For example, for $La_{2-x}Ba_xCuO_{4-y}$, adiabatic calorimetry usually does not reveal any noticeable $\Delta C$, although differential methods find a specific heat anomaly [21]. For $La_{2-x}Sr_xCuO_4$ the specific heat anomaly becomes comparable with the conventional BCS theory predictions only above $x = \frac{1}{8}$ [22]. The jumps for $YBa_2Cu_3O_{7-y}$ as well as Tl- and Bi-based oxide families are also reduced and smeared in comparison with the conventional ones [21, 23, 24]. In particular, the discontinuity $\Delta C$ decreases steeply for underdoped compositions of $YBa_2Cu_3O_{7-y}$ below the critical hole concentration per Cu atom $p_c \approx 0.19$ [25].

Our view is that this phenomenon is caused by the development at a certain temperature $T_s$ of a structural phase transition accompanied by the appearance of a static charge-density wave (CDW) described by the dielectric order parameter $\Sigma$ on the nested segments of the FS [26–29]. The possibility of this was demonstrated by us in the framework of the so-called partially gapped model [30] with a strong mixing of states from nested and non-nested FS sections. Our predictions were obtained on the basis of calculations made for the most common situation, when $T_s \gg T_c$. This assumption permitted us to not consider the problem self-consistently, i.e. to disregard the negative feedback below $T_c$ of the superconducting order parameter $\Delta$ onto the dielectric one $\Sigma$. However, adopting these approximations, we eliminated from consideration some A15 superconductors [31] and some cuprates [28, 32, 33] for which the two instabilities have almost the same strength, with the result that $T_s$ exceeds its superconducting counterpart $T_c$ only slightly. In this paper, the thermodynamical properties of partially gapped CDW superconductors are treated self-consistently, which made it possible to extend the range of calculation applicability to arbitrary ratios $T_s/T_c > 1$. It turned out quite unexpectedly that, to some extent, the self-consistency simplified the calculations and made the results clearer. The qualitative conclusions of [26, 27] concerning the reduction of the anomaly $\Delta C$ at $T_c$ caused by CDW gapping are confirmed. On the other hand, the magnitude of the effect is estimated for the first time. It is shown that for reasonable FS gapped fractions, the normalized jump $\Delta C/\gamma_S T_c$ may be several times lower than the BCS value (1). This result agrees qualitatively with observations for high-$T_c$ oxides. In both limits of a very small CDW-gapped FS area and almost complete dielectrification, the BCS value is restored. It should be noted that we assume mean-field behaviour of both $\Delta$ and $\Sigma$. A fluctuating nature of the latter in the absence of a true long-range order was suggested in [34].

## 2. Theory and numerical results

### 2.1. Superconducting and dielectric parameters

The Dyson–Gor'kov equations for the normal ($\mathcal{G}_{ij}$) and anomalous ($\mathcal{F}_{ij}$) temperature Green functions in the case of coupled superconducting ($\Delta_{im}^{\alpha\gamma}$) and dielectric ($\Sigma_{im}^{\alpha\gamma}$) matrix order parameters are well known [28] and are presented below for the sake of completeness:

$$[\mathrm{i}\omega_n - \xi_i(\boldsymbol{p})]\mathcal{G}_{ij}^{\alpha\beta}(\boldsymbol{p};\omega_n) - \sum_{m\gamma}\Sigma_{im}^{\alpha\gamma}\mathcal{G}_{mj}^{\gamma\beta}(\boldsymbol{p};\omega_n) + \sum_{m\gamma}\Delta_{im}^{\alpha\gamma}\mathcal{F}_{mj}^{\dagger\gamma\beta}(\boldsymbol{p};\omega_n) = \delta_{ij}\delta_{\alpha\beta}, \tag{2}$$

$$[\mathrm{i}\omega_n + \xi_i(\boldsymbol{p})]\mathcal{F}_{ij}^{\dagger\alpha\beta}(\boldsymbol{p};\omega_n) + \sum_{m\gamma}\Sigma_{im}^{\dagger\alpha\gamma}\mathcal{F}_{mj}^{\dagger\gamma\beta}(\boldsymbol{p};\omega_n) - \sum_{m\gamma}\Delta_{im}^{\dagger\alpha\gamma}(\boldsymbol{p})\mathcal{G}_{mj}^{\gamma\beta}(\boldsymbol{p};\omega_n) = 0. \tag{3}$$

Here $\omega_n = (2n+1)\pi T$, $n = 0, \pm 1, \pm 2, \ldots$, $T$ is the temperature, $\hbar = k_B = 1$, Greek superscripts correspond to electron spin projections, and italic subscripts describe the natural split of the FS into degenerate (nested, d) and non-degenerate (non-nested, nd) sections. For the former, the following condition necessary for dielectric gapping holds:

$$\xi_1(\boldsymbol{p}) = -\xi_2(\boldsymbol{p}+\boldsymbol{Q}), \tag{4}$$

where $\boldsymbol{Q}$ is the CDW vector. This equation binds the electron and hole bands $\xi_{1,2}(\boldsymbol{p})$ for the excitonic insulator [35, 36] and different parts of the one-dimensional self-congruent band in the Peierls insulator case [37]. At the same time, the rest of the FS remains undistorted and is described by the electron spectrum branch $\xi_3(\boldsymbol{p})$. Such an approach was suggested long ago by Bilbro and McMillan [30]. We solve equations (2) and (3), adopting the strong-mixing approximation for states from different FS sections. This means the appearance of a single superconducting order parameter for d and nd FS sections. The spin-singlet structure (s-wave superconductivity and CDWs) of the matrix normal ($\Sigma_{ij}^{\alpha\beta} = \Sigma\delta_{\alpha\beta}$) and anomalous ($\Delta_{ij}^{\alpha\beta} = \boldsymbol{I}^{\alpha\beta}\Delta$) self-energy parts (where $(\boldsymbol{I}^{\alpha\beta})^2 = -\delta_{\alpha\beta}$) in the weak-coupling limit are also taken into account. The self-consistency equations for the order parameters $\Sigma$ and $\Delta$ in accordance with the fundamentals [1, 36] can be expressed in the following form (more details on the subject can be found in [27, 28]):

$$1 = V_{BCS}N(0)[\mu I(D) + (1-\mu)I(\Delta)], \tag{5}$$

$$1 = V_{CDW}N(0)\mu I(D), \tag{6}$$

where

$$I(x) = \int_0^{\Omega} \frac{d\xi}{\sqrt{\xi^2 + x^2}} \tanh \frac{\sqrt{\xi^2 + x^2}}{2T}. \tag{7}$$

Here $V_{BCS}$ and $V_{CDW}$ are contact interactions responsible for superconductivity and CDW gapping, respectively,

$$D(T) = [\Delta^2(T) + \Sigma^2(T)]^{1/2}, \tag{8}$$

and $\mu$ is the parameter of the FS dielectrization (we use this term in some places to avoid confusion with the term for the superconducting gapping), so $N_d(0) = \mu N(0)$ and $N_{nd}(0) = (1 - \mu)N(0)$ are the DOSs on the nested and non-nested FS sections, respectively. The upper limit $\Omega$ in equation (7) is the relevant cut-off frequency, which is assumed to take the same value for the two interactions. If the cut-offs $\Omega_{BCS}$ and $\Omega_{CDW}$ are considered different, the correction arising, $\log(\Omega_{CDW}/\Omega_{BCS})$, is logarithmically small [30] and does not alter qualitatively the results presented below. Only in the case of almost complete electron spectrum dielectric gapping ($\mu \to 1$) does the difference between $\Omega_{BCS}$ and $\Omega_{CDW}$ become important for the phase coexistence problem [38]. This situation is, however, of no relevance for substances with detectable superconductivity, since $T_c$ tends to zero for $\mu \to 1$.

The physical meaning of the quantity $D$ is a combined gap appearing on the nested FS sections, whereas the order parameter $\Delta$ defines the gap on the rest of the FS. Here we confine ourselves to the case $\Sigma > 0$ since the sign does not affect the thermodynamic properties (see the relevant discussion in [28]).

Introducing the primordial order parameters $\Delta_0 = 2\Omega \exp[-1/V_{BCS}N(0)]$ and $\Sigma_0 = 2\Omega \exp[-1/V_{CDW}N_d(0)]$, we can rewrite the equation set (5) and (6) in the equivalent form

$$I_M(\Delta, T, \Delta(0)) = 0, \tag{9}$$

$$I_M(D, T, \Sigma_0) = 0, \tag{10}$$

where

$$I_M(\Delta, T, A) = \int_0^{\infty} d\xi \left( \frac{1}{\sqrt{\xi^2 + \Delta^2}} \tanh \frac{\sqrt{\xi^2 + \Delta^2}}{2T} - \frac{1}{\sqrt{\xi^2 + A^2}} \right) \tag{11}$$

is the standard Mühlschlegel integral and

$$\Delta(0) = (\Delta_0 \Sigma_0^{-\mu})^{\frac{1}{1-\mu}}. \tag{12}$$

Equations (9) and (10) mean that both gaps have the BCS form $G(T) = \Delta_{BCS}(G(T = 0), T)$, namely:

(i) $\Delta(T) = \Delta_{BCS}(\Delta(0), T)$, i.e. the actual value of the superconducting gap of the CDW superconductor at $T = 0$ is $\Delta(0)$ rather than $\Delta_0$, and the actual superconducting critical temperature is $T_c = \frac{\gamma}{\pi}\Delta(0)$; and
(ii) $D(T) = \Delta_{BCS}(\Sigma_0, T)$, which determines $T_s = \frac{\gamma}{\pi}\Sigma_0$.

Here $\gamma = 1.7810\ldots$ is the Euler constant.

From equation (8) we obtain that, at $T = 0$,

$$\Delta^2(0) + \Sigma^2(0) = \Sigma_0^2. \tag{13}$$

Replacing $\Delta(0)$ by its value (12), we arrive at the conclusion that in the model adopted, two order parameters coexist only if $\Delta_0 < \Sigma_0$. Then, according to equation (12), $\Delta(0) < \Delta_0$; i.e. the appearance of the CDW, if it occurs, always *inhibits* superconductivity. And *vice versa*, according to equation (8), for $T < T_c$, $\Sigma(T) < \Delta_{BCS}(\Sigma_0, T)$; i.e. superconductivity depresses dielectrization.

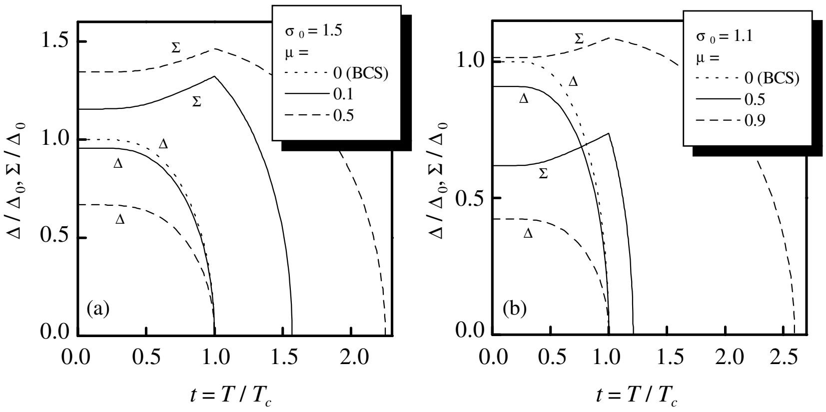


**Figure 1.** Temperature dependences of the superconducting ($\Delta$) and dielectric ($\Sigma$) order parameters for different values of the dimensionless parameters $\mu$ and $\sigma_0$ (see explanations in the text).

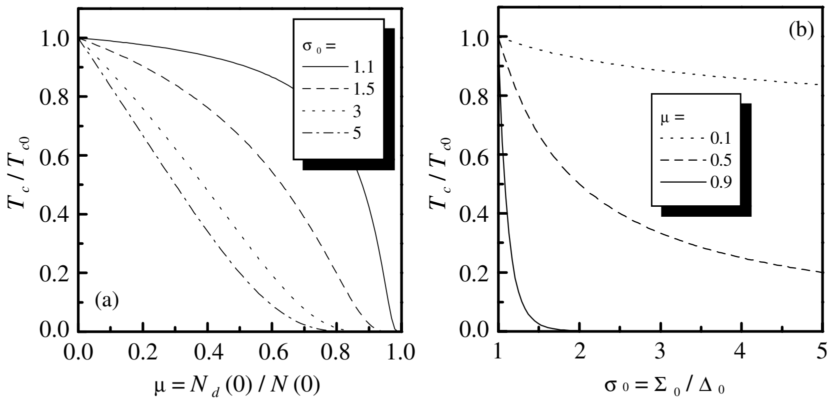


**Figure 2.** Dependences of the ratio $T_c/T_{c0}$, where $T_{c0} = \frac{\gamma}{\pi}\Delta_0$ is the primordial superconducting critical temperature, on $\mu$ (a) and $\sigma_0$ (b).

In figure 1 the $T$-dependences of the order parameters $\Delta$ and $\Sigma$ are shown for various parameters of the partially dielectrized superconductor. It can be easily inferred from the data shown in the figures that, in agreement with the foregoing, $\Delta(T)/\Delta(0)$ curves coincide with the Mühlschlegel one for any values of the dimensionless $\mu$ and $\sigma_0 \equiv \Sigma_0/\Delta_0$. The novel feature, which has been overlooked in previous investigations, is the possibility of such a strong suppression of $\Sigma$ for low enough $T$ that it becomes *smaller* than $\Delta$, although $T_s$ is *larger* than $T_c$ (see figure 1(b)). This intriguing situation can be realized for the parameter $\sigma_0$ close to unity. One should note that the gaps $\Delta$ and $D$ (the former coincides with the superconducting order parameter) are monotonic functions of $T$. But the dielectric order parameter $\Sigma$ is not.

The *magnitudes* of the $T_c$s and $\Delta(0)$s strongly depend on $\mu$ and $\sigma_0$, which is demonstrated in figure 2, although the simple BCS-like scaling between them is preserved. Now we would like to underline once more that all previous results obtained for the Bilbro–McMillan model of the CDW superconductor in the approximation $\Sigma_0 \gg \Delta_0$ are now verified by the exact self-consistent solution.

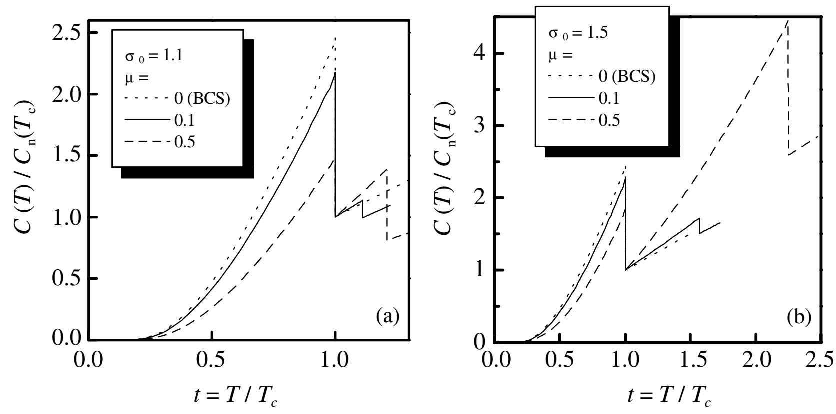


**Figure 3.** Temperature dependences of the normalized heat capacities $C$ for different values of $\mu$ and $\sigma_0$.

## *2.2. Electronic heat capacity*

The easiest way to calculate the electronic specific heat capacity $C(T)$ is to express it in terms of the quasiparticle dispersion relation, both for the normal and superconducting CDW states [1]. Two FS sections defined above contribute separately to this quantity. Since both gaps $\Delta$ and $D$ are BCS-like, it is possible to express each contribution through the well-known normalized heat capacity function of the BCS superconductor

$$c_{\mathrm{BCS}}\left(t = \frac{T}{T_c^{\mathrm{BCS}}}\right) = \frac{C_{\mathrm{BCS}}(T)}{C_{\mathrm{BCS}}(T = T_c^{\mathrm{BCS}} + 0)}. \tag{14}$$

The function $c_{\mathrm{BCS}}(t)$ increases at $t < 1$, has a jump (1) at $t = 1$, and equals $t$ for $t > 1$. Then the function to find can be expressed as

$$C(T) = N(0)\left[(1-\mu)T_c c_{\mathrm{BCS}}\left(\frac{T}{T_c}\right) + \mu T_s c_{\mathrm{BCS}}\left(\frac{T}{T_s}\right)\right]. \tag{15}$$

Since both CDW and superconducting gaps lead to the exponential decrease of relevant contributions to $C(T)$ for $T \to 0$, it would be difficult to distinguish between them below $T_c$. However, the very existence of the CDW gap should manifest itself above $T_c$ (i) in a non-linear dependence of $C(T)$ and (ii) in an additional jump $\Delta C$ at $T_s > T_c$.

The overall $C(T)$ curves normalized to the $C_n(T_c) = C(T = T_c + 0)$ value are shown in figure 3.

At the same time, the normalized discontinuity $\Delta C/C_n$ at $T_c$ may also serve as indirect evidence of the CDW gap on the FS, because in this case it is not at all a trivial BCS jump (1). (This change of behaviour was recognized long ago [26, 27]. But only the self-consistent approach used here allows us to give the answer explicitly for any value of the parameters appropriate to the partially dielectrically gapped superconductor.) It can be seen from figure 4(a), where the superconducting phase transition anomaly is shown as a function of $\mu$. The discontinuity is always smaller than the BCS value, $\approx$1.43, in agreement with previous qualitative considerations [26, 27]. At the same time, the BCS ratio is restored not only for $\mu = 0$, i.e. in the absence of the dielectrization, but also for $\mu \to 1$. In the former case it is clear because we are dealing with a conventional BCS superconductor. On the other hand, for large enough $\mu$, CDW gapping almost completely destroys superconductivity, so $T_c \ll T_s$. Therefore, in the relevant superconducting temperature range the contribution to the

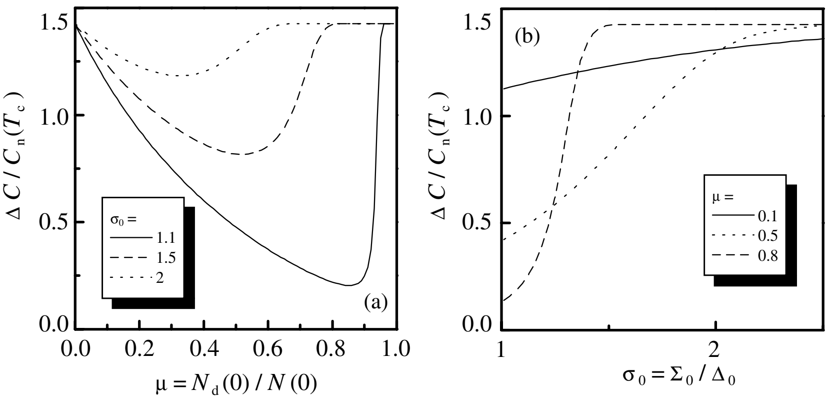


**Figure 4.** Dependences of the normalized heat capacity discontinuity $\Delta C$ at $T_c$ on $\mu$ (a) and $\sigma_0$ (b).

heat capacity from the d FS sections, governed by the gap $D \approx \Sigma$, becomes exponentially small. Another term, determined by the nd FS section, ensures the BCS limiting value of the normalized discontinuity.

Dependences of $\Delta C/C_n$ on $\sigma_0$ for various values of $\mu$ are depicted in figure 4(b). One sees that the effect is large for $\sigma_0$ close to unity, whereas the difference between 1.43 and $\Delta C/C_n$ goes to zero as $\sigma_0^{-2}$, verifying the asymptotical result obtained earlier [26].

It should be noted that the heat capacity calculation scheme adopted for partially dielectrized superconductors can be applied also to other types of order parameter symmetry.

## 3. Discussion

There are a lot of CDW superconductors which could be used to verify our theory [28, 33]. Unfortunately, as can be seen from the relevant tables in the cited reviews, for most of these substances the ratios $T_s/T_c$ are of the order of 10–100, so the predicted suppression of $\Sigma$ below the $\Delta$ level and the reduction of the ratio $\Delta C/C_n$ would be unobservable. $V_3Si$ with $T_c \approx 17$ K and $T_s \approx 21$ K remains the best candidate—as it was decades ago when Bilbro and McMillan suggested their model [30]. Investigation of other A15 compounds $Nb_3Sn$ ($T_c \approx 18$ K and $T_s \approx 43$ K) and $Nb_3Al_{0.75}Ge_{0.25}$ ($T_c \approx 20$ K and $T_s \approx 24$ K) can also remain on the agenda. The next low-$T_c$ candidate is much worse: 2H-$NbSe_2$ with $T_c \approx 7$ K and $T_s \approx 33.5$ K. Still, the possible suitability of the oxides $Li_{1.16}Ti_{1.84}O_4$ and $BaPb_{1-x}Bi_xO_3$ should not be ruled out. First, they clearly demonstrate a reduced $C(T)$ discontinuity and, second, relevant critical temperatures can be altered over a wide range by the control of the metal component and oxygen stoichiometry.

But high-$T_c$ oxides, notwithstanding the actual order parameter symmetry, should be considered as the most promising testing grounds for the partial-dielectrization concept and its specific consequences, such as the reduction of $\Delta C$. Since many measurements reveal structural and electronic peculiarities just above $T_c$ both for the $La_{2-x}M_xCuO_{4-y}$ (M = Sr or Ba) and $YBa_2Cu_3O_{7-y}$ families [28], which can be attributed to CDW gap formation, they may be intentionally doped to study correlations between the magnitude of $\Delta C$ and the CDW manifestations. Success of such examinations is highly likely, because the crucial role of CDWs in $T_c$-suppression for cuprates was clearly demonstrated in experiments [39].

Another possibility for checking the existence of CDW features in high-$T_c$ ceramics is applying a magnetic field $H$ to suppress the superconductivity in the substances concerned.

The revival of a reduced pseudogap below $T = T_c(H = 0)$, which we identify with $\Sigma$ [28, 33, 40] (see also [34]), to its original value above $T_c$ provides further good evidence that CDW effects are really important. This effect can be seen directly while observing intensities of CDW x-ray reflections. On the other hand, measurements of the ac Josephson current $I_{\mathrm{Joseph}}(V)$ involving one electrode made of a CDW superconductor should show a simultaneous reduction of the magnitude of $I_{\mathrm{Joseph}}$ and a constancy of the position of the Riedel peculiarity at larger $V = D$ (see equation (13)). Concomitant heat capacity measurements could examine the influence of CDWs on superconductivity quantitatively.

Finally, the observed smallness of the $\Delta C$ discontinuity in $MgB_2$ [41–45] may be due to the presence of CDW-triggered energy gaps if there is a Mg deficiency in the relevant specimens [46]. Another explanation [47] of the heat capacity experiments is based on the assumption of intrinsic non-homogeneity of $MgB_2$. The latter point of view is also supported by the transport measurements [46], where a phase separation into Mg-vacancy-rich and Mg-vacancy-poor regions was claimed to occur. One cannot exclude the possibility that *both* large-period CDWs and electronic phase separation act jointly to produce the $\Delta C$ reduction. In this case a proximity-induced multiple-gap superconductivity [48–52] is a consequence of the appearance of mesoscopic domains in $MgB_2$, whatever the details of their origin.

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