On the thermal (“caloric”) peculiarity of the SC transition, precursor to specific heat’s “jump”: its relation to the “paramagnetic” effect, precursor to the Meissner ejection — both revealed in HTSC material by the super-high sensitive SFCO method

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Abstract

We have investigated the correlation between the “paramagnetic” peculiarity of the normal-to-superconductive phase transition – detected first in a LTSC tin, in 1989, and then confirmed in HTSC materials with much more higher resolution (by a sensitive SFCO method) – and the other weakly expressed effect, detected also both in LTSC & HTSC materials before their heat capacity’s known “jump”. In a low-$T_c$ superconductive tin this thermal effect is detected a half century ago, by Corak, but passed unnoticed so far. We show in this work that, externally similar these 2 fine effects really and truly have the same physical roots. To prove this assumption theoretically, and to reveal the reasons for their common origin, we use here a concept, which admits existence of two types of the Cooper pairs in SC materials – “singlet” & “triplet”. As we conclude in this paper, they show completely different, non-traditional temperature behavior upon cooling of SC materials. We believe that this study results (along with the “triplet” superconductivity, proven experimentally in Josephson junctions quite recently, with the ferromagnetic barrier) may obtain key importance for the true understanding of the real nature of the superconductive phenomenon (in whole) and to uncover electron “pairing” mechanisms above the Meissner expel (in particular).

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The problem of electron “pairing” above Meissner expel rose after 1986 only, when high-temperature superconductive (HTSC) materials were opened by Bednorz and Muller [1]. And, majority of scientists is admitted now [2], that for truth there is need to consider two physical processes for HTSC materials – electron pairing, and onset of the phase coherence – separately, and independently of one another. So, superconductivity in HTSC material requires both the electron pairing & the Cooper-pair condensation. The later is also known as an onset of the long-range phase coherence among pairs [2]. In other words, it is admitted now [2], that in HTSC material electrons become paired above Meissner ejection (above $T_c$, starting from $T_i$) and start forming the superconductive condensate only at the $T_o$, while in low-$T_c$ (conventional) SC material (LTSC), it is assumed that the pairing process and the onset of the phase coherence take place simultaneously (at the same temperature; $T_o = T_c$): due to relatively large pair size, it is assumed that their wavefunctions are overlapping in a LTSC material.
Shortly after discovery of HTSC materials, however, a “paramagnetic” (PM) precursor to the superconductivity is detected by us in LTSC tin (Sn) [3] (Fig.1). Weakly expressed this effect (in a central press it is reported later [4]) & indirectly related to it another fine effect [5] (Fig.2) – a thermal (“caloric”) peculiarity, seen on heat capacity vs temperature curves, before the specific heat’s known “jump” – provide together serious arguments, however, to hold a contrary opinion regarding low-Tc SC materials. Though such a key effect (also first detected in LTSC tin) is available on Corak’s Figs. since middles of 50-s of the last century, however, to our surprise, it passed unnoticed for a long time. Seems, first we here focus attention to it. So, in this work we try to explain it, & list consequences following from it. Thus, we discuss below both the reasons why these 2 fine effects should have the same physical origin, and also, mark-out their key importance for true understanding of the real nature of the superconductive phenomenon.

![Fig.1](image1.png)

Fig.1. “Paramagnetic” effect detected upon transition to the SC-state of identical tin grains of ~5µm in dia. [3-4], registered by the solenoid-coil based less sensitive technique. Inset: enlarged view of the effect, broken line is device temperature dependence.

![Fig.2](image2.png)

Fig.2. Heat capacity vs temperature curves detected in tin by Corak [5] Inset: enlarged view of a fine effect seen before the specific-heat’s “jump”. Symbol “s” corresponds to the SC-state, while “n” – normal (superconductivity is suppressed by magnetic field).

To explain such a global similarity among thermal and electromagnetic properties of the SC materials (regardless it is HTSC or LTSC) we use here an advanced approach [6-7], admitting existence of 2 types of Cooper pairs – both in HTSC and in LTSC materials – “singlet” and “triplet”. Running ahead of the events let’s note that, according to conclusions we come below, they show fully different non-traditional temperature behavior upon cooling of the material, starting with the birth of the first Cooper pair at temperature Tc. For YBaCuO composition HTSC material, respective curves (plotted by formulas to be derived below) are shown in Fig.8.

In regards to feasibility of separation of the electron pairing and Cooper-pair condensation, it was important searches for the said “caloric” effect in high-Tc superconducting material. But, it is not so easy to measure heat capacity (moreover, its little changes at beginnings of SC transition) in so small volume clean HTSC objects (such as film-structures, single crystals). We did that recently, and could indirectly detect this fine effect also in a YBaCuO composition HTSC material [8] (see Fig.3) – by the step-by-step creation, perfection, and the proper use of a new principle of work imaging technique [9-10], based on the combination of a well-focused laser scanning microscopy [11] and the single-layer flat-coil-oscillator based highly sensitive new measurement method (SFCO-technique), introduced by our group for the high-resolution research in a last decade [12-13].

![Fig.3](image3.png)

Fig.3. Indirect detection of the said “caloric” effect in YBaCuO [8] by a new imaging method [9-10], based on the sensitive SFCO technique [12-13]. Circles are measured data, while the solid line – exponential fit of the exponential data (compare this figure with the Fig.2).

There are other Meissner-state precursor effects too. Before listing them let’s remind, that superconductor is a double ideal material, since it becomes ideal conductor (R=0) and gets properties of the ideal diamagnetic (B=0) material below some temperature. The latter behaves also as an ideal conductor – reverse is not true. But, must the superconductor get such properties at the same moment (temperature)? And, why different-nature transitions (1-st is assumed to be connected with electron pairing, followed by zeroing of the pair momentum,
while the 2-nd, with pair condensation — due to collection of enough “singlet” pairs — proofs see in [6]) should occur at a same temperature? And also, are there any other different-nature effects in Nature, which occur at a same moment? Such basic questions got the meaning since the said “paramagnetic” effect is detected in micron-size tin grains [3–4] (Tc ~ 3.72K – Fig.1) that indicates the real physical onset of the Meissner expel. It precedes diamagnetic ejection, and substantially corrects shape of the normal-to-superconductive (NS) phase transition curve. The origin of above questions relates also with a “preceding” effect, detected in percolating YBa2Cu3O7 (in ceramics [1], and in films [14], with a granular structure of the material). According to it, resistive-transition ends before the start of Meissner expel. It was seen by Morris also in a BiCaSrCuO crystal [15], but doesn’t attract a proper attention of the author – perhaps, due to lack of assurance in accordance among temperature scales of the conducted tests, performed in different setups. Questions were especially deepened when a “diamagnetic activity” was revealed in a LaSrCuO film by Iguchi [16] – at temperatures, much higher the transition temperature of the material, established by the onset point of the Meissner expel. A super-sensitive scanning-SQUID microscope is used for those tests. Such a flux activity is interpreted by the author as the effect, precursor to the Meissner state.

Listing Meissner-state precursor effects, let’s stop also on the study conducted by Valla [17]. It shows that a “pseudo-gap” in the energy level of high-Tc SC materials’ electronic spectrum is the result of electrons being bound into Cooper pairs above the transition temperature to the SC-state, but unable to super-conduct, because pairs move incoherently. As to the LTSC materials (which act much closer to the Zero), it was said that, admittedly, superconductivity in these materials occur as soon as electron-pairs are formed. “In case of high-Tc superconductors, however, electrons, though paired, “do not see each other above some temperature,” Valla says in [17], “and so, they can’t establish the phase coherence, with all the pairs behaving as a ‘collective’”. Listed in this paper numerous data indicate, however, that such is the case for all types of the SC materials.

And also, there is NO indication on any microscopic physical mechanism for establishment of the phase coherence among SC pairs in Valla’s works, as well as in papers of other researchers. While, as a hint to a possible mechanism for absence (at higher temperatures) & presence (later, at cooling) of the phase coherence among pairs might be discussed existence of 2 types of the Cooper pairs in SC materials (regardless it is high-Tc or low-Tc) – “singlet” and “triplet”, with different angular momentum [6]. As is indicating our study below, they have fully different temperature behavior upon cooling of the SC material (qualitatively illustrated by Fig.8 for the YBa2Cu3O7). Such an approach may result in the ideal conductivity (it starts with the pair formation from the Tc, and acts for both types of the pairs). That is because the pairs are quasi-particles with a Zero momentum and so, with an infinitely large de Broglie wavelength, due to which they can move in a material without scattering, “ignoring” defects & impurities of the crystalline structure. At a later cooling of the material only, starting from the temperature Ta, such a reasonable approach may also result in the ideal diamagnetism (superconductivity, Meissner state) – but, for only “singlet” pairs...

And finally, a research team from the Ruhr-University (Bochum), Christian-Albrechts-University (Kiel) & Santa Barbara (headed by Profs. Kurt Westerholt, Hartmut Zabel & Konstantin Efetov) could make an experimental sensation recently; their studies on the “pairing” behavior of electrons have proven the “triplet” superconductivity [18]. In other words, they could detect existence of the pairs with parallel spin direction (for more details see [19]). The said integrated team has studied Josephson junctions with the barriers prepared from the Heusler compound Cu3MnAl. In as-prepared state the Cu3MnAl layers are non-ferromagnetic, and the critical Josephson current density Jc drops exponentially with thickness of Heusler layers dL. On annealing the junctions at 240°C the Heusler layers develop ferromagnetic order and they could observe a dependence Jc(dL) with Jc, strongly enhanced, and weakly thickness dependent in the thickness range 7.0 < dL < 10.6 nm. The team interprets this feature as an indication of a “triplet” component in the SC pairing function, generated by the specific magnetization profile inside thin Cu3MnAl layers.

Analysis of above data (& some other results that we omit due to page limitation) allow to expect, that sooner, there are no serious differences between HTSC & LTSC materials regarding the said two processes. Apparently, electron pairing & the Cooper-pair condensation (phase coherence) are separate & independent even in a LTSC material. Difference is in temperature scales. In LTSC, these processes run in a narrow range (10-20mK – Fig. 1), while in HTSC the scale is much longer. For example, for the YBa2Cu3O composition HTSC material
the scale of the “paramagnetic” effect (that shows the scale of event selection for above 2 physical processes), estimated by Gantmakher, is about 1K [20] – Fig.4, while more sensitive our study, based on the use of SFCO technique, evidence that it is even broader 3K [4] – Fig.5. Possibly, this is the reason why separation of the \( T_c \) from \( T_0 \) was so problematic so far in LTSC materials. The problem is still open also due to lack of methods for the “non-perturbing”, sensitive study of the SC transition in clean (and so, tiny) objects with small signals – especially at very beginnings of the transition, where even a highly sensitive SQUID technique is incapable to “notice” such small changes in a normal-state “skin”-depth (comments on this matter see also in [20]).

2. Correlation between “paramagnetic” & “caloric” peculiarities of the superconductive transition

So, it has been shown by the measurements with a highly sensitive SFCO technique [12-13] that at cooling of SC materials there appears a new effect before the Meissner expel, named “paramagnetic” [4]. In other words, prior to the start of a diamagnetic expel at \( T_0 \), SC matter gets a fine paramagnetic nature, starting with \( T_0 \), and before the huge expel it slightly pulls-in pick-up coil’s MHz-range testing radiofrequency (RF) field, resulting in a fall of a measuring tunnel diode (TD) oscillator frequency \( F_{\text{mean}} \) (Figs. 1 & 5) – that is why effect is named “paramagnetic”. To be more precise note, that \( \Delta F = F_{\text{ref}} - F_{\text{mean}} \) in Fig.5. Besides, frequency, \( F_{\text{ref}} \), of the reference oscillator (used for compensation of a test device temperature dependence [12-13]) is independent of the effect, so, it remains constant during the tests. Note also, the effect is seen both in LTSC (Fig.1) & HTSC (Fig.5) materials – at cooling of the sample, and at its heating, as well. The effect is independent on the sample size or shape. But it gradually disappears in large objects, due to effect’s averaging, due to temperature and/or material inhomogeneity in a sample volume – because the effect is seen in a narrow range (Fig.1). In this connection, let’s state conditions when it was safely detected so far – that is important for true understanding of its nature. So, data related with the PM effect were obtained under conditions when the effect was independent on cooling/heating rates of a sample, and the temperature-stability & material-homogeneity throughout the specimen was high enough [4].

Along with this effect, combination of the 4-probe method & our sensitive SFCO technique enabled to establish also disappearance of a resistance of SC materials before the start of the Meissner expel [6] – resistive transition ends \( R(\approx 0) \) before the start of Meissner expel. So, physical reasons for the ideal conductivity \((R=0)\) & for the ideal diamagnetism \((B=0)\) may not be the same in Nature. That is why, there are all grounds to believe, that there might be close relationships between the said “paramagnetic” & “caloric” precursors to the superconductive transition, and the ideal conductivity \((R=0)\).

Below, we are going to give theoretical proofs why the “paramagnetic” effect [3-4, 20] (Figs. 1 & 4-5) detected prior to Meissner expel, and the “caloric” effect [5, 8] (Figs. 2 & 3) detected before the specific heat “jump” – both seen not only in LTSC, but also in HTSC – should have the same physical reasons & origin.

2.1 Theory

To explain a nature of the “paramagnetic” effect an idea admitting presence of 2 types of pairs is used by us in [6]. Analyses of the shape of effect & further development of this idea resulted in the creation of a phenomenological theory of the SC phase transition by Sedrakian [21]. In addition, in [6] it is concretized by us that, per-
haps, some of pairs are “singlet” (with $\sigma=0$ spin), while remains are “triplet” ($\sigma=1$). That means, only later ones may contribute to the paramagnetism of the SC matter.

So, we suppose that in SC materials we actually deal with a quasi-particle system with 3 values of a spin – $l = 0$, 1, $1/2$ (let’s give this system a name 1-st model). And, because of magnetic moment is an additive physical quantity, for such a 1-st model SC system we have:

$$M = (n_{s}/j)n \cdot M_0 + (n_{1/2}/j)n \cdot M_{1/2}$$

(1)

where, temperature dependences of the values for $n_{1/2} = n_s(T)$, $n(T)$ and $n(T)$ are given by the formulas (2)–(4), (10) and (11) below, respectively.

If the Cooper-system wouldn’t get a certain spin value at its formation (that is, direction of electron spin does not depend on being of electron in a pair, or not), then the spin values of the pairs of such a 2-nd model system would not depend on temperature, and therefore, Cooper pairs would be the single-type at all temperatures. But, subject to both “triplet” a “singlet” pairs are available in a matter at starts of the SC transition (with different behavior), their amount should depend on temperature. Any other differences between these two kinds of Cooper pairs are not revealed empirically so far.

Now, our goal is to match these 2 model systems by comparing their heat capacities via their magnetic moments. For that it is enough to calculate expected difference of magnetic moments of the said model systems. By the energy difference of these 2 systems in the external magnetic field ($E = -\Delta MH$) we will then calculate difference of heat capacities of these 2 model systems.

According to the above assumption and conclusion given by us in [6], at beginnings of formation of the Cooper-pair system “triplet” pairs should be created first – that is because their stability is higher, so, their chance to be “survived” in a field of thermal fluctuations is higher. This certainly promotes entering of a testing RF-field into the sample – just which is registered by us in Figs. 1 and 5 in the form of the “paramagnetic” effect. Let’s also note that “singlet” pairs (without a spin) in no way may contribute to the paramagnetism of SC matter. While, experimental data & reasoning, presented in [6] & [21] lead to the suggestion, that further formation of the “singlet” Cooper-pair system in a coherent state, seems, is responsible for the establishment of the ideal diamagnetic state in a SC matter (in other words, only the “singlet” pairs may result in Meissner repulsion).

So, according to our conclusion in [6] (based on the shape of a measured transition curve – Fig.5) one may say, that due to formation of a Cooper-pair system concentration of “normal” charge carriers (electrons) in a matter decreases (starting already from the 1-st critical temperature, $T_c<89$K), and at approach to the 2-nd critical temperature, $T_c<85.6$K, it already drops to almost the zero (to be more precise, to $n_0(T)$ – see below) – in contrast to the traditional “two-fluid” models of the superconductivity offered in due time first by Gorter & Casimir [22]. While, following by shape of the fine “paramagnetic” effect (Fig.5) one may conclude that the Cooper-pair system (formed due to step-by-step reduced electrons) not only is still unable to eject detecting RF-field of the pick-up coil from sample (which is too weak in our method, & “non-perturbing” [12-13]), but, it still permits the field to continue slowly “intrude” the sample. This process reaches to its peak when the specimen cools down to $T_c<85.6$K. And only below this point temperature dependences of the “normal” charge carriers (electrons – $n_s(T)$) and Cooper pairs, $n(T)$, approximately follow the rules of conventional theories [22-23].

Temperature dependence of “normal” charge carriers, $n_s(T)$, near the fluctuation-temperature region may be presented by the following empirical formulas [6]:

$$n_s(T) \approx n_{res}(T_c) (T/T_c)^{2}, \quad \text{at } T_c<T_0$$

(2)

$$n(T) \approx n_{res}(T_c)(T_c-T)^{2} + n_{res}(T), \quad \text{at } T_0<T<T_c$$

(3)

$$n(T) \approx n, \quad \text{at } T_c<T$$

(4)

where $n$ is the total carrier density.

To get an analytic function for further calculations let’s seam together formulas (2)–(4). In agreement with requirements of continuity and smoothness of the function $n_s(T)$, we suggest for the residual density of “normal” charge carriers $n_{res}(T)$ a linear expression $\beta(T_c-T)$. Note, that $n_{res}(T)$ is negligible in LTSC material. But, as follows from variety of works [24-27], in HTSC materials, being small in general it becomes noticeable in many important practical applications of these materials. So, for $\beta$ one may take, for example, the value $\beta=0.01$ (it may be shown, that $\beta$ can’t be more than 0.06, in general [28]). In that case, the function graph for normalized electron concentration, $\eta(T) = n_0(T)/n$, in a temperature range $T_0<T<T_c$, is shown in the Fig.6.

![Fig.6](image-url)

**Fig.6.** The graph for the normalized electron concentration, $\eta(T) = n_0(T)/n \approx [1-(T-T_c)/(T_c-T_0)]^2 + n_{res}(T)/n$, in a temperature range $T_0<T<T_c$ – for the case, when $\beta=0.01$.

After seaming Eqs. (2)–(4), for $\eta(T)$ we finally get following formulas – for the whole temperature range

$$\eta(T) = \frac{n_{res}(T)}{n} \left(\frac{T}{T_0}\right)^2, \quad \text{at } T<T_0$$

(5)
\[ \eta_1(T) = \left( \frac{T - T_0}{T_1 - T_0} \right)^2 + \frac{n_{1c}(T)}{n}, \]  \text{ at } T_0 < T < T_c, \quad (6) \\
\eta_2(T) = 1, \quad \text{ at } T_c < T, \quad (7)

Hereafter we will deal only with these formulas. The full seamed curve for normalized electron concentration is shown in Fig.7a. The function describing concentration of all paired electrons (Cooper pairs – Fig.7b) is

\[ \eta_s(T) = \frac{n_s(T)}{n} = 1 - \eta_2(T) \]  \quad (8)

As was mentioned, based on high-resolution empirical data on the "paramagnetic" effect [4] (inset curve in Fig.5), a phenomenological theory of SC transition is created in [21]. Starting with an idea of the existence of 2 types of pairs in SC materials, it permits to calculate temperature dependence of a portion of "singlet" pairs, \( \alpha(T) \), with respect to the total number of Cooper pairs.

\[ \alpha(T) = \left\{ 1 - th \left[ 3.5 \cdot (T - 83.5) \right] \right\} / 2. \quad (9) \]

Thus, for concentrations (relative amounts) of "singlet" & "triplet" Cooper pairs in SC material we obtain

\[ n_s(T)/n = \eta_s(T) \cdot \alpha(T)/2, \quad (10) \]
\[ n_t(T)/n = \eta_s(T) \cdot (1 - \alpha(T))/2. \quad (11) \]

Fig.7. Graphs of the functions \( \eta_1(T) = n_s(T)/n \) (a – normal electrons), and \( \eta_2(T) = n_s(T)/n = 1 - \eta_2(T) \) (b – Cooper pairs).

Symbols “0” & “1” are used to indicate the spin of pair. By the use of formulas (5)–(7), (10) and (11), temperature dependences of normalized “normal” electrons (a), “triplet” (b) & “singlet” (c) pairs, and also all Cooper pairs (d) are plotted in Fig.8. As is seen from the figure, at cooling, starting with the beginnings of the N/S transition \( T=89K \) amount of “triplet” pairs (curve b) is dominant – and up to the \( T=83.5K \). And their amount reaches to its maximum just around the peak of “paramagnetic” effect \( T=85.6K \) – see inset in Fig.5.

Now it is time (we are ready) to discuss contribution to the magnetic moment (magnetization) & the heat capacity of the SC material, conditioned only by spins of “normal” electrons and Cooper pairs, and calculate their temperature dependences. As the real model for the further calculation let’s take above defined 1-st model (material consists of 3 quasi-particles with a following values of the spin – \( I = 1/2, 0, \) & 1), for which amount of each type of particles depends on temperature according to above formulas (2)–(4), (10) and (11) – respectively for the “normal” electrons, “singlet” and “triplet” Cooper pairs. Besides, to have the same temperature everywhere inside the sample, the cooling and heating of the sample was performed too much slowly during the measurements (see the Fig.5, for example, otherwise the “paramagnetic” effect may disappear, due to averaging). That permits to consider the problem under study almost quasistatic, and use advantages of the related calculation methods – to simplify further calculations.

To determine the magnetization of an ideal gas in the external magnetic field, consisting of particles with the given \( I \) value of the spin, let’s suppose, that particles of such a model system are also identical and non-interacting. The statistic sum, \( Z \), of such a system is

\[ Z = \left\{ \frac{sh \left[ I + 1 \right] \mu_s \cdot g \cdot \cdot H}{kT} \right\}^n, \]

and, magnetization of a unite volume of such a model gas system in an external magnetic field, \( H \), is [28]

\[ M_i = n \cdot I \cdot \mu_s \cdot g \cdot B_i \left\{ \frac{I \cdot \mu_s \cdot g \cdot H}{kT} \right\}, \quad (12) \]

where, \( B_i \) is the Brillouin function,

\[ B_i(x) = \frac{2 \cdot I + 1}{2 \cdot I} \cdot \operatorname{cosh} \left( \frac{2 \cdot I + 1}{2 \cdot I} \cdot x \right) - \frac{1}{2 \cdot I} \cdot \operatorname{cosh} \left( \frac{2 \cdot I + 1}{2 \cdot I} \cdot x \right). \]

In this formulas \( g \) is the Lande factor, \( \mu_s \) – the Bohr magneton, \( H \) – the applied external field, \( n \) – total number of electrons, and the \( k \) is the Boltzmann constant.

Thus, we have a system of quasi-particles with 3 different values of the spin – \( I = 1/2, 0, \) & 1. Besides, magnetic moment is an additive physical quantity, so magnetization of such a 1-st model SC system is given by the formula (1) above, where, as is mentioned, temperature dependences of the values \( n_{01}, n_{00}, \) & \( n_1 \) are given by the formulas (2)–(4), (10) & (11), respectively.

So, after necessary calculations [28], for magnetization of the 1-st model system we obtain the next formula

\[ Z = \left\{ \frac{sh \left[ I + 1 \right] \mu_s \cdot g \cdot \cdot H}{kT} \right\}^n, \]
\[ M_{1\rightarrow 2} = n \cdot \mu_{B} \left\{ \eta_{b}(T) \cdot \text{th} \frac{\mu_{B} H}{kT} + \eta_{b}(T) \cdot \frac{1}{2} \right\} \times \frac{g}{c} \left\{ \cosh \frac{3 \mu_{B} gH}{2kT} - \frac{1}{2} \cosh \frac{\mu_{B} gH}{kT} \right\} \]

Similarly, for the magnetization of the 2-nd model system one may easily obtain the following formula [28]

\[ M_{2\rightarrow 1d} = n \cdot \mu_{B} \cdot \text{th} \frac{\mu_{B} H}{kT}. \]

We remind, that our final goal is to match these two model systems by matching their heat capacities via their magnetic moments. So, expected difference of moments (magnetizations) of these 2 model systems is

\[ \Delta M = n \cdot \mu_{B} \left\{ \eta_{b}(T) \cdot \text{th} \frac{\mu_{B} H}{kT} - \eta_{b}(T) \cdot \frac{1}{2} \right\} \times \frac{g}{c} \left\{ \cosh \frac{3 \mu_{B} gH}{2kT} - \frac{1}{2} \cosh \frac{\mu_{B} gH}{kT} \right\} \]

Fig.9 demonstrates temperature dependence of the function (15). Identity of the curve (b) in Fig.8 with the curve in Fig.9, & their qualitative coincidence with the measured data on “paramagnetic” effect (see inset curve in Fig.5) witness that, apparently, microscopic reasons generating the fine “paramagnetic” effect retate to do main role of the “triplet” pairs over the “singlets” at very beginnings of the formation of the superconductive state. Especially as because the peak of the “paramagnetic” effect is just at the temperature \( T_{0} = 85.6 \text{K} \), at which concentration of the “triplet” pairs reaches to its maximum possible value (compare the Figs. 5 and 8).

The energy difference of these 2 model systems in the external magnetic field \( H \) is given by the following formula, \( E = -\Delta MH \), which allows to determine the difference of the heat capacities of these 2 model superconductive systems by the expression below

\[ \Delta C_{H} = -H \frac{\partial (\Delta M)}{\partial T} \bigg|_{H}. \]

After simple, but long calculations one may get for the function \( \Delta C_{H}(T) \) the following analytic formula

\[ \Delta C_{H} = \frac{1}{2} \mu_{B} \left\{ \frac{-2 \mu_{B} H}{kT} \cdot \eta_{b}(T) \right\} - \frac{g}{c} \left\{ \cosh \frac{3 \mu_{B} gH}{2kT} - 6 \cdot \cosh \frac{2 \mu_{B} gH}{2kT} \right\} \]

\[ \cdot \frac{S}{2kT} \cdot \frac{\partial \eta_{b}(T)}{\partial T} \]

\[ \cdot \frac{g}{c} \left\{ \cosh \frac{3 \mu_{B} gH}{2kT} - \frac{1}{2} \cosh \frac{\mu_{B} gH}{kT} \right\} \]

\[ \cdot \frac{g^2}{2} \left\{ \cosh \frac{3 \mu_{B} gH}{2kT} - 2 \cosh \frac{2 \mu_{B} gH}{2kT} \right\} \cdot [g(T) - 1] \cdot \frac{\partial \eta_{b}(T)}{\partial T} \]

3. Discussion

So, we have compared magnetic moments (magnetizations) of 2 said model superconductive systems conditioned by only their spins. Then, we calculated difference of their heat capacities, \( \Delta C_{H} \), conditioned by different magnetizations. As it turned out, that is enough for description of the fine “calorific” effect, shown in Figs. 2-3, detected both in LTSC & in HTSC materials, before their heat capacity “jump”. Figure 10 shows temperature dependence of the function \( \Delta C_{H}(T) \), plotted by the formula (17). Qualitatively, it explains the fine thermal effect (seen prior to the heat capacity “jump”), detected in LTSC tin by Corak 50 years ago [5] (Fig.2). It is ignored in due time: perhaps, such a key effect is considered as a noise-level signal. Theoretical curve in Fig. 10 agrees also with a similar “calorific” effect detected by us in YBaCuO by a sensitive SFCO method [8] (Fig.3).

Note that a qualitative explanation of the said thermal effect (“paramagnetic” effect as well) stand possible only after we have classified (sorted) the Cooper-pair system by the spin at our calculations: the “singlets” and “triplets”, amount of which sharply changes in so complicated fluctuation temperature region at cooling.
Results obtained here allow to conclude, that a weakly expressed “paramagnetic” effect, detected recently & investigated in details by the created highly sensitive SFCO new test-method, and the no less fine peculiarity before the specific heat’s “jump”, detected long ago by Corak & Satterthwaite, have the same physical reasons – both of the effects result from the different spin values of the Cooper pairs and their different temperature behavior upon cooling of the superconductive material.

4. Conclusion, final remarks, and suggestions

Analyzing all discussed above, one may conclude that at the moment becomes important unambiguous demonstration of the “singlet”-to-“triplet” conversion of Cooper pairs (and back), and separation of the ideal conducting ($R=0$) and ideal diamagnetic (superconductive – $B=0$) states from each other. That is too much important not only for the true interpretation of the real nature of the superconductive phenomenon (in whole), but also, for the correct understanding of the microscopic mechanisms of the electron “pairing” (the physical mechanisms for the establishment of the long-range phase coherence among the superconductive pairs).

In this connection, one of the most effective experimental ways is investigation of the Fulde-Ferrell-Larkin-Ovchinnikov superconductivity (the FFLO state [29–30] – as sensitive, as it is possible (and, many-sided). One of materials, in which theorists suggest to study this unique phenomenon, is the heavy-fermion superconductor CeCoIn$_5$. First, that was experimentally investigated by Radovan et al. [31], but, with not enough resolution, compared to what we suggest to do below. This material satisfies a delicate balance of properties needed to detect and study the FFLO superconductivity at temperatures below the 350mK. A highly sensitive SFCO test-method [12-13] (with its unprecedented capabilities) one needs to use as a unique scientific research instrument for precision study of the peculiarities of the superconductivity, and the pair formation in FFLO state. Such a research (to be carried out approximately by the scenario implemented in a pioneering experiment [31]) may enable direct demonstration of the “singlet”-to-“triplet” spin-flip of the Cooper pairs, and also, the separation of the ideal conducting ($R=0$) and ideal diamagnetic ($B=0$) states.

At this, distortion of the testing RF-field configuration near the flat coil face (taken by the frequency rise of the testing SFCO technique) one may use for the checking whether the ideal diamagnetic state (with $B=0$) is established or not. While, amount of the absorption of the testing TD-oscillator’s power by the sample (taken by the amplitude fall of the measuring SFCO technique) may be used to check whether the ideal conducting state (with $R=0$) is established in a material, or not. Along with this, establishment of an ideal conducting state may be checked independently by the 4-probe test technique.

Besides, our research shows that a flat-coil based oscillator can be activated also with its internal capacitance [32] (without an external capacitance in its resonant circuit). That is the result of a relatively high value of the internal capacitance of single-layer flat coils, compared to their parasitic capacitance with respect to the surrounding radio-technical environment. This key circumstance opens exotic areas for the flat-coil oscillator application. Namely, a “needle-like” measuring MHz-range magnetic field of such a flat coil [33] (used as a pick-up in such a stable TD-oscillator), enables a novel method (new approach) for the surface probing [34]. Such an unusual probe shows strong dependence of a detected signal both on the lateral position of such a probe with respect to the surface of the object, and on the size of a spatial-gap between the probe and the surface of the object [34] – crucial for the probe microscopy. This opens an opportunity for the designing of the non-solid-state “magnetic-field” probes with the RF power applied to the sample, lying in the power range of 1nW to 5µW. The gap between such a “probe-formative” flat coil and the object can be larger than 1nm [33-34], compared with ~1nm gap of acting tunneling and atomic force probe-microscopes [35-38].

That is why, we believe that such a SFCO-probe may enable to distinguish (both by the amplitude and by frequency of the detecting TD-oscillator) details of the relief of the FFLO “node” structure, in a real space (consisting of the alternating regions of the superconducting layers and the spin-polarized magnetic walls). However, for that aim, we suggest to create and use a SFCO method-based advanced “magnetic-field” probe, with a lithographically made single-layer flat coil of about 1mm in diameter [39] – as an effective “needle-type” probing instrument with better than 100nm predicted lateral resolution. Such a new probe will have considerably large work-distances (approx., 0.1-1 mm) between the probe & the surface of the object, which enables a “laser” control of the local probing area of the object, and, if needed, application of the test and control perturbations.

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