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On the possible key experiments to show the *singlet-to-triplet* conversion of the Cooper pairs by the use of a SFCO-method

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Abstract. The problem of electron pairing above Meissner expel arose when high- T_c superconductive (*HTS*) materials were discovered. And scientists are admitting now that there is need to consider 2 processes for *HTS*: electron pairing and long-range phase coherence among Cooper pairs (*pair condensation*) – separately and independently each other. Otherwise, it is admitted, that in *HTS* electrons become paired above Meissner expel (above T_0 , starting from T_c) & start forming *SC*-condensate only at T_0 , while in low- T_c *SC* materials (*LTS*) the pairing and pair condensation start simultaneously ($T_0=T_c$). But, since discovery of *HTS* new data appeared and importance of some earlier data was recognized, indicating analogy among the *HTS* and *LTS* in connection with these 2 processes. Here we show that there are no differences among these materials regarding the said processes: electron pairing and pair condensation are separate & independent even in *LTS*. Difference is in a temperature scale. For *LTS* these processes run in a narrow range ($\sim 30mK$), while in *HTS* the scale is longer (for *YBaCuO*, more than $3K$). This is the reason why separation of T_c from T_0 in *LTS* is so hard so far. The problem is still open also due to lack of methods for “nonperturbative” study of the beginnings of *SC* transition in clean (*tiny*) objects with small signal. Analysis of data permits to conclude that becomes urgent sensitive study of the *Fulde-Ferrell-Larkin-Ovchinnikov* superconductivity in heavy-fermion materials

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that have layered electronic structure. And, the *SFCO* method may stand key tool for sensitive study of the pair formation in *FFLO* state. Such a study may show *singlet-to-triplet* conversion of Cooper pairs. It may also allow separating the *ideal conducting* ($R = 0$) & *ideal diamagnetic* ($B = 0$) states, which are important for true interpretation of a real nature of superconductivity.

1. Introduction

Majority of scientists is admitted now that in high- T_c superconductive (**HTS**) material electrons become paired above Meissner expel (above T_0 , starting from T_c) and start forming the *SC* condensate only at the T_0 , while in low- T_c *SC* material (**LTS**) it is assumed that the pairing process and the Cooper-pair condensation (*long-range phase coherence among pairs*) take place simultaneously (at a same temperature – $T_0=T_c$): due to much larger pair size it is assumed that their wavefunctions are overlapping in a *LTS* material [1]. Shortly after discovery of *HTS* materials [2], however, a “*paramagnetic*” (**PM**) precursor to superconductivity was detected in *LTS* tin (**Sn**) [3]. Weakly expressed this key effect, and related to it “*caloric*” precursor to *SC* transition [4] (seen on *Corak*’s heat capacity vs. temperature curves, before the specific heat’s “*jump*” – first, also detected in *LTS* tin) provide weighty arguments to have the reverse opinion regarding the *LTS* materials, discussed below. There are the reasons why these two fine effects have the same physical origin. To validate such a global correlation between the electromagnetic and thermal properties of the *SC* materials (regardless it is *HTS* or *LTS*) an advanced idea was putted forward by us in [5], admitting existence of two types of Cooper pairs, both in *HTS* and *LTS* materials – *singlet* and *triplet*. According to main conclusions we came to in [5], they show absolutely different non-traditional temperature behaviour upon cooling of the *SC* material. For the *YBaCuO* composition *HTS* material, for example, respective curves are shown in a **figure 1**.

In this connection, it was urgent searches and detection of the said “*paramagnetic*” and “*caloric*” precursors of superconducting state in *HTS* material. We tried to do that, and much more precision recent experiments by the use of a single-layer flat-coil-oscillator (**SFCO**) based highly sensitive our unique technique [6-7] enabled to detect in a *YBaCuO* composition *HTS* material the said “*paramagnetic*” effect [3] (see **figure 2**), at the beginnings of the *SC* phase transition, which precedes Meissner expel. Next, it enabled to detect in this material the said “*caloric*” effect, discussed below. But, before turn to it let’s list *Meissner*-state precursor other phenomena, detected by various groups, which all are important for the true understanding of the real nature of the superconductive phenomenon.

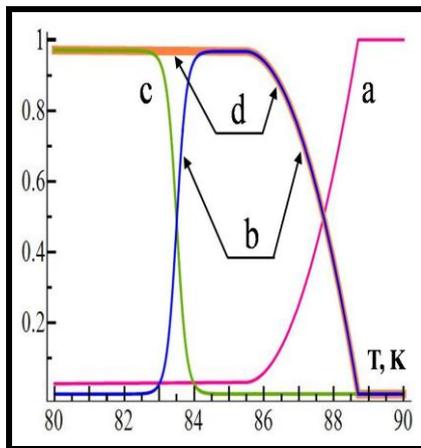


Figure 1. Temperature dependences of normal electrons (a), triplet (b) & singlet (c) Cooper pairs & all pairs (d) [5]

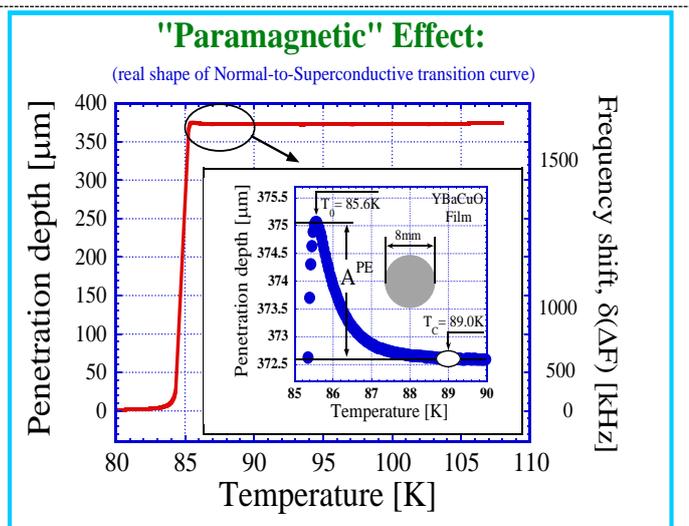


Figure 2. NS transition of the *YBaCuO* film [3]. Inset: enlarged view of the “*ParaMagnetic*” effect (PE) detected at the beginnings of *SC* phase transition, which pre-

cedes Meissner expel. The A^{PE} is the PM effect's height.

Superconductor is a double ideal material, since it becomes *ideal conductor* and gets properties of the *ideal diamagnetic* below some temperature. The latter behaves also as an *ideal conductor* – reverse is not true. But, must SC material obtain such properties at the same temperature? And why transitions of different nature (*1-st is assumed to be connected with electron-pairing, followed by zeroing of the pair momentum, the 2-nd, with pair condensation – due to collection of enough singlet pairs – arguments see in [8]*) should occur at the same temperature? And also, are there other *different-nature* effects happening at the same moment in a Nature? Such questions acquired meaning since the “*paramagnetic*” effect was detected in micron-size tin grains [3] ($T_c \sim 3.72K$), pointing out the real physical onset of the Meissner expel. It precedes the diamagnetic ejection and corrects the shape of the normal-to-superconductive (N/S) phase transition curve. The origin of the above questions relates also with a “*preceding*” effect, opened in a percolating $YBaCuO$ (in ceramics [2] and in films [9] with a granular structure of the material). According to it, *resistive* transition ends before the start of Meissner expel. The effect was seen also in a $BiCaSrCuO$ crystal [10], but doesn't attract a proper attention of the author – perhaps due to lack of assurance in accordance among *temperature*-scales of conducted tests, performed in different set-ups. Questions were deepened when a “*diamagnetic activity*” was revealed in $LaSrCuO$ film [11] – at temperatures much higher transition temperature of material established by the onset point of Meissner expel. A *super-sensitive scanning-SQUID* microscope was used for those tests. Such a flux activity is interpreted by the author as the effect *precursor* to the Meissner state.

Among other *Meissner-precursor* effects let's stop on the study conducted by *Tonica Valla* [12]. It shows that a “*pseudo-gap*” in the energy level of HTS materials' electronic spectrum is the result of electrons being bound into Cooper pairs above the transition temperature to SC -state, but unable to *superconduct*, because pairs move incoherently. As to LTS (*which act closer to Absolute Zero*), “*superconductivity in these materials occur as soon as pairs are formed*”, *Valla* says. “*In the case of HTS materials, however, electrons, though paired, ‘do not ‘see’ each other above some temperature,*” *Valla* thinks [12], “*so, they can't establish the phase coherence, with all the pairs behaving as a ‘collective’*”.

Listed here data indicate, however, that such is the case for all types of SC materials. And also, NO indication on any microscopic physical mechanism for establishment of the phase coherence among SC pairs in *Valla's* works, and in papers of other scientists. While, as a hint to possible mechanism for absence (*at higher temperatures*) and presence of the phase coherence among the pairs (*at further cooling*) might be discussed presence of 2 types of pairs in SC materials (regardless it is high- or low- T_c) – *singlet* and *triplet*, with different angular momentum [8]. They have different *temperature-behavior* upon cooling of a material (qualitatively illustrated by **figure 1** [5]). Such an approach may result in the *ideal conductivity* (it starts with a pair formation from the T_c , and acts for both types of pairs [8]). That is because pairs are quasiparticles with a Zero momentum, so, with an infinitely large *de Broglie* wavelength, due to which they can move in a material without scattering, ignoring defects of a crystalline structure. At a later cooling of a material, starting from temperature T_0 , such an advanced approach may also result in the *superconductivity (ideal diamagnetism)* – but, for only *singlet* pairs.

And finally, it is very important to note that recently existence of the superconductive Cooper pairs with parallel spin direction was detected experimentally for the first time, in works [13-14].

2. Results and discussion

So, even based on the listed above data (*among which, many ours*) one may believe, that there are no visible differences between the HTS and LTS materials regarding the said 2 processes. Apparently, the electron pairing and the Cooper-pair condensation (*phase coherence*) are separate and independent even in a LTS material. Difference is in a temperature scale. In a LTS material these processes are running in a very narrow temperature range ($\sim 30mK$ [3-4]), while in HTS the scale is much longer. For the $YBaCuO$, for example, the scale of the “*paramagnetic*” effect estimated by *Gantmakher* (*that indicates the scale of the event selection for the above 2 physical processes*) is about $1K$ [15], while much more precise experiments by our group evidence that it is even broader (more than $3K$ [3] – **figure 2**).

Possibly, this is the reason why separation of the T_c from the T_0 in a *LTS* material was so much hard so far – that is done only qualitatively up to now [3-4]. The problem is still open due to lack of methods for “*non-perturbing*”, sensitive study of the *SC* transition in thin *LTS* objects with small signals – especially at very beginnings of the phase transition, where even a sensitive *SQUID* technique can not “*notice*” such small changes in a normal-state “*skin*”-depth (i.e., start of the “*PM*” effect – **figure 2**).

And so, in connection with the problem of electron pairing above Meissner expel, detection of the said “*caloric*” precursor to superconductivity in *HTS* material was so urgent. That is why we now turn to present our data related with the searches and detection of the said fine thermal peculiarity in high- T_c superconductive film. It was not so easy, however, to measure the heat capacity (*moreover, its too much little changes at the beginnings of the SC phase transition*) in so small volume objects, such as *film*-structures. However, we tried to do that and could detect so much small effect in *YBaCuO* composition *HTS* film (**figure 3**). For such fine measurements we have further upgraded the cryogenic laser scanning microscope suggested by our joint group earlier [16-17]. Schematics of an improved imaging technique are shown in **figure 4**. Generally, that is combination of the well-focused laser scanning microscopy [18] and the *SFCO* method-based sensitive measurement technique [6-7]. One of advantages of the *SFCO*-method is its ability to “*notice*” details of the phase transition between the normal and *SC* phases in thin, plate-like objects – with an *angstrom*-scale absolute resolution at measurement of the penetration depth (*by frequency shift of the SFCO-based testing tunnel diode (TD) oscillator* [6-7]), and with about a *nano-watt* resolution at absorption measurements (*by amplitude changes of the same oscillator* [19]). Besides, it can serve as a sensitive temperature probe with better than *0.1mK* resolution [20]. The imaging technique uses a focused laser beam, as a probing signal. It is capable of imaging properties of *HTS film*-structures with $\sim 1\mu\text{m}$ spatial resolution. It operates as follows: the *HTS film*-bridge is illuminated point by point with a focused laser beam ($\Phi_{\text{beam}} \sim 1\mu\text{m}$), which results in a slight local heating. In our case the substrate of the film is transparent, and so, the amount of laser power passed to the flat-coil based thermal sensor depends on peculiarities of the heat capacity (*or heat conductivity*) of the material under test in a lighted point of the film, positioned on the flat face of the detecting coil – leading to some changes in the amplitude (*and frequency*) of the testing *TD*-oscillator. Moving position of the “*X-Y*” stage along both coordinates by stepper motors (with $\sim 0.5\mu\text{m}$ step) and controlling these movements by the **PC** (in LabVIEW environment), we could make micrometer precision positioning of the probing laser beam and scan it over the surface of the film (**figure 4**). This enabled to get *2D*-images of the *N/S* phase transition of thin, plate-like *HTS* materials (e.g., *film*-structures) with $1\text{-}2\mu\text{m}$ spatial resolution. The curve in **figure 3** is the *2D*-raster image of the laser beam power transmitted through the substrate, obtained by summarizing of the *amplitude change* signals all over the film. In other words, in **figure 3** the *2D*-raster image of the heat distribution in the *HTS* film under investigation is shown. Since the created laser scanning microscope is an instrument of the bolometric nature (*the flat pick-up coil detects the residual power of the laser beam that has not been dissipated in the test film*), the signals detected by the flat pick-up coil may be indirectly related to anomalies in the heat capacity (*as in the first approximation, the heat conductivity may not have any visible anomalies in such a narrow range of the temperature in the vicinity of the SC phase transition*).

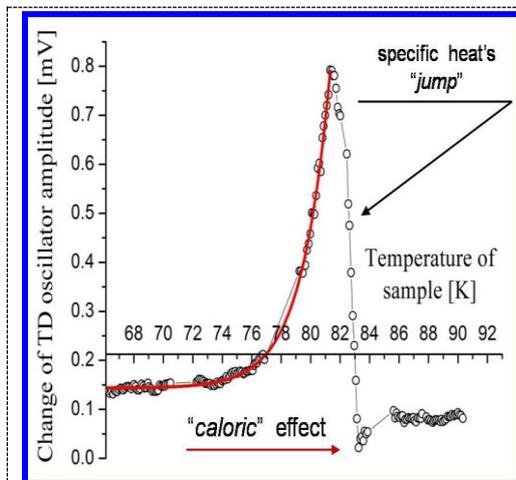


Figure 3. Detection of a “caloric” effect in *YBaCuO* film by an imaging technique created in this work based on the *SFCO* method [6-7]. Circles are measured data, while solid line – exponential fit of data.

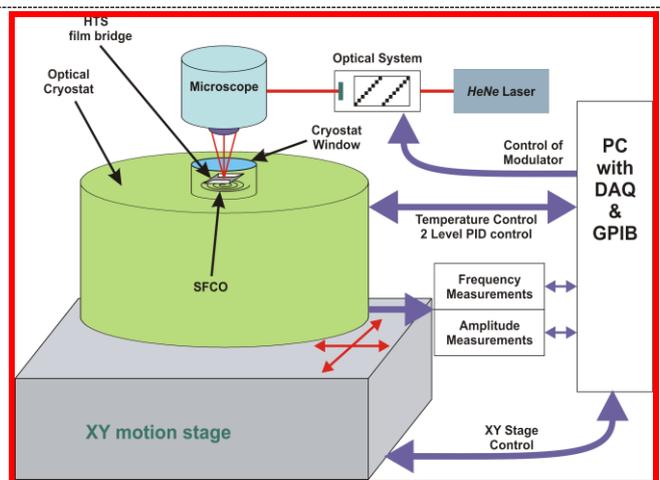


Figure 4. Schematics of the created and used cryogenic laser scanning microscope, based on the focused ($\Phi_{\text{beam}} \sim 1\mu\text{m}$) laser scanning microscopy [18] and the single-layer flat geometry pick-up coil-based sensitive tunnel diode oscillator (SFCO-technique) [6-7].

Superconductivity is a macroscopic quantum effect where electrons of opposite spin & momentum condense into pairs – according to original explanation by *Bardeen, Cooper & Schrieffer (BCS)* [21]. Large fields destroy superconductivity by coupling to the orbital motion of electrons. This critical field (*orbital limit*) separates the uniform superconducting state from the normal metallic state. Magnetic field can, however, couple predominantly to the spins of electrons. Superconductor is then in a *paramagnetic limit*. It was shown in 1964 by *Fulde & Ferrell* [22] & independently, *Larkin & Ovchinnikov* [23], that this superconductive state would be basically different from the *conventional BCS* case. In this new state, magnetic field tries to polarize opposite spins of *SC*-pairs. In response, the *SC* order parameter develops nodes in a real space, leading to the alternating regions of *SC*-layers and spin-polarized magnetic walls. This *FFLO* state manifests itself as a “wedge” in the field-temperature (*B-T*) phase diagram at very low temperatures, below the critical field. In **figure 5** the constructed in 2003 *B-T* phase diagram is shown, obtained from the heat capacity measurements for the plane parallel orientation [24-25]. Blue squares denote the critical field, separating the normal-metallic state from the superconductive one. The “wedge” at low temperatures & high magnetic fields is the *FFLO* phase, with the red squares separating uniform from non-uniform superconductivity. The exact shape of *B-T* diagram and the *FFLO* “wedge” area as well depend on many microscopic parameters, such as the Fermi surface geometry, dimensionality of the host crystal, impurities, and ratio of orbital-to-paramagnetic effects [22-24]. The material in which first was observed the *FFLO* state in 2003 is the *CeCoIn₅*.

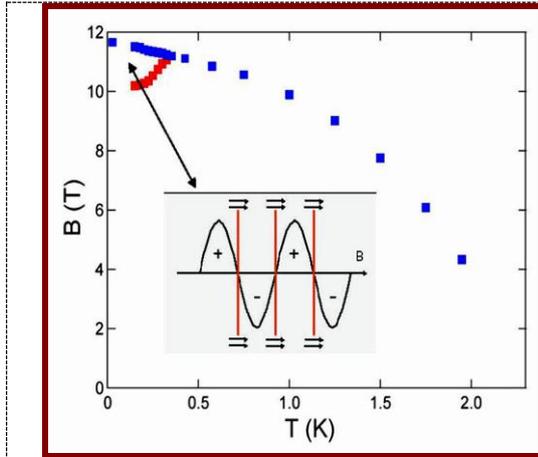


Figure 5. Critical field (blue) & FFLO transition line (red). Inset: schematics of FFLO order parameter with magnetic walls [24-25].

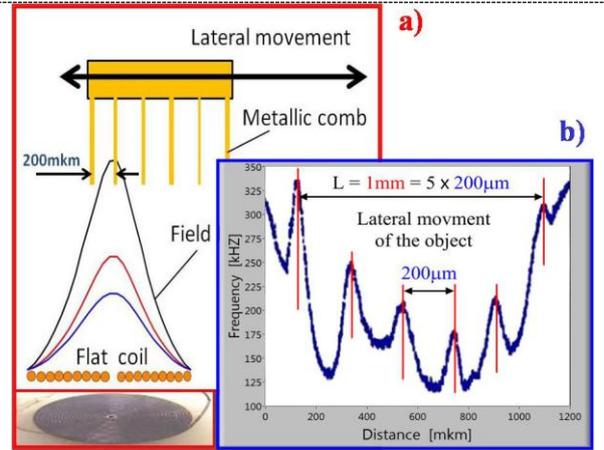


Figure 6. TD-oscillator frequency shift ΔF (b) vs. the lateral position of a 1D grid-shaped metallic object (a) relative to flat coil face.

Taking into account above data, one may come to the conclusion that sensitive study of the FFLO state in heavy-fermion materials becomes urgent. That is important for understanding of microscopic mechanisms of electron pairing. In this regard, we suggest start to newly study it in a $CeCoIn_5$ material by involving methods enabling much higher resolution compared to earlier tests discussed in [24-25]. This material has a critical temperature of 2.3K, and the layered electronic structure. Such anisotropy resists orbital motion of electrons for fields applied parallel to conducting *ab*-planes of this material. Besides, the large spin susceptibility favors paramagnetic limitation. So, this material fulfills delicate balance of properties needed to detect and study the FFLO superconductivity. The SFCO-method [6-7] (with its high capabilities enabling to reveal many fine effects in superconductivity during the last decade [3, 16, 19, 26]), could be serve as a unique instrument for precision study of the Cooper pair formation in FFLO state. That may permit demonstration of the *singlet-to-triplet* conversion of Cooper pairs, and also, separation of the *ideal conducting* ($R = 0$) and *superconducting* (*ideal diamagnetic* – $B = 0$) states. These are among matter of principle questions for true interpretation of the real nature of superconductivity. Such a study may also clarify microscopic mechanisms of the electron pairing (*physical mechanisms for the establishment of the long-range phase coherence among the SC pairs*).

Alongside with the heat capacity measurements, TD-oscillator tests were done in 2003 by Radovan et al., with a solenoid pick-up coil [25]. They provide evidence that the FFLO transition can be qualitatively seen also in penetration depth studies – even with a less sensitive solenoid pick-up coil. However, while studying the SC-phenomenon in $YBaCuO$ composition HTS material during the last 10-15 years we could demonstrate in [3, 6-8, 17, 19, 26], that for the flat geometry (plate-like) objects (*films, crystals*) a flat-coil based TD-oscillator (our SFCO technique) shows higher resolution by at least 3-4 orders of a value, compared to its solenoid-coil based analog used in above pioneering works [24-25]. This permits to believe, that the SFCO technique with about 1mm in diameter pick-up coil may satisfy technical conditions needed for much more sensitive detection and investigation of the FFLO state.

In this connection, our research shows also that the flat-coil oscillator can be activated also with its internal capacitance [27] (*without an external capacitance in its resonant circuit*). That is due to relatively high value of the internal capacitance of single-layer flat coils (compared to their parasitic capacitances with respect to the surrounding radio-technical environment). This opens exotic areas for flat-coil oscillator application. Namely, a “*needle-like*” testing magnetic field of such a flat coil (see **figure 6a**), used as a pick-up in such a stable oscillator, enables a new method (*new approach*) for surface probing, based on replacement of short-range solid-state probes of acting microscopes (*such as needles or cantilevers of the tunneling [28-29] & atomic-force [30] microscopes*) by the long-range action non-solid-state ones. Such a unique probe shows strong dependence of a detected signal on the size of

the spatial-gap between the probe & the surface of the object – crucial for the probe microscopy (**PM**) [31]. This opens a chance for creation “*magnetic-field*” probes with a *RF* power applied to the sample lying in the range of $1nW$ to $5\mu W$. The gap between such “*probe-formative*” coil and the object can be larger than $100\mu m$, compared to $1nm$ gap of the acting probe microscopes [31]. In our tests we reached a lateral resolution $\sim 1\mu m$ – even for the relatively large diameter ($2R_{coil} \sim 14mm$) flat-coil technique.

Such a *SFCO*-probe could also “*notice*” & distinguish details of relief of a normal-metallic object, with $\sim 10\mu m$ spatial-resolution ((see **figure 6**). To demonstrate this we performed an experiment with one-dimensional (**1D**) metallic grid made of 6 copper wires (see **figure 6a**): each wire was $20-30\mu m$ in dia., and was positioned with an average interval of $\sim 200\mu m$ between the wires. Wires distort coil *RF*-field configuration when they move (*or when coil moves relative to the grid*), leading to changes of the oscillator frequency or/and amplitude. The effect is maximal when each wire reaches to the coil center. **Figure 6b** illustrates detected dependence of the oscillator frequency shift, $\delta(\Delta F)$, vs. the lateral position of the metallic-comb relative to the coil face (*relative to “magnetic-field” probe*). Average distance between detected 6 vertical neighboring peaks on the curve in **figure 6b** is $\sim 200\mu m$ – just in agreement with the experimental setup in **figure 6a**. That is why we think, that our *SFCO*-probe may also distinguish and image (*both by amplitude and frequency of the TD-oscillator*) details of the relief of the nodal structure in *FFLO*-state, in a real space (*consisting of alternating regions of the SC-layers and the spin-polarized magnetic walls*). For that aim, we believe the best way is creation and use of a said *SFCO* method-based “*magnetic-field*” probe, with a lithographically made coil of about $1mm$ in dia. [32] – as an effective “*needle-type*” probing instrument with better than $100nm$ predicted lateral resolution. Such a probe may have much larger work-distances (more than $100\mu m$) between the probe and the surface of the testing object, which enables a “*visual*” control of the local probing area of the object, and, if needed, application of test perturbations (for example, exposition to laser radiation).

3. Conclusions

Resuming the said above, let’s underline importance and feasibility of key experiments for sensitive study of the *Fulde-Ferrell-Larkin-Ovchinnikov* superconductivity (*FFLO*-state) [22-23] by the use of a *SFCO*-technique [6-7] – that stands urgent to demonstrate the *singlet-to-triplet* conversion of Cooper pairs. The material in which we suggest to study this phenomenon at temperatures below $350mK$ is a heavy-fermion superconductor *CeCoIn₅*. That was investigated earlier [24-25], but with much less resolution compared to what we offer to do. The unique *SFCO*-method (*with its unprecedented capabilities we demonstrated while studying the SC-phenomenon in YBaCuO during the last 10-15 years* [3, 6-8, 17, 19, 26]) we suggest to use as the main research instrument for precision study of the pair formation in *FFLO* state. Such a study (to be carried out approx. by the scenarios implemented in original works [24-25]) may enable demonstration of the *singlet-to-triplet* conversion of Cooper pairs, and also separation of the *ideal conducting* ($R = 0$) and *superconducting* (*ideal diamagnetic* – $B = 0$) states, which are important not only for true interpretation of the nature of superconductivity (*in a whole*), but also for correct understanding of the microscopic mechanisms of the electron pairing (*physical mechanisms for the establishment of the long-range phase coherence among the superconductive pairs*).

At this, distortion of a testing *RF*-field configuration near the flat coil face (*taken by the frequency increase of the SFCO technique*) may be used for checking whether the *ideal diamagnetic* state (with $B = 0$) is established or not. While, amount of the absorption of a testing *TD*-oscillator power by the sample (*taken by the amplitude increase of the testing SFCO technique*) one may use to check whether the *ideal conducting* state (with $R = 0$) is established or not. Alongside with that, establishment of the *ideal conducting* state one may check also by the traditional *AC 4-probe* measurement technique.

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