Coexistence of Superconductivity and Itinerant Ferromagnetism; Y_9Co_7 as an Example

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***Abstract* - The discovery of superconductivity in Y_9Co_7 that exhibited at the same time the itinerant ferromagnetism was a big surprise because it showed that the two incompatible phenomena might be reconciled at some temperatures under certain thermodynamic conditions. It was 27 years ago. The recently discovered itinerant ferromagnetic superconductors are those strongly correlated electron compounds: UGe_2, UGeRh and likely also $ZrZn_2$. This review is concentrated on the present thinking about and activities on the problem of coexistence of itinerant ferromagnetism and superconductivity with special emphasis on the first weak itinerant spin- fluctuating ferromagnetic superconductor Y_9Co_7.**

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I. INTRODUCTION TO COEXISTENCE OF ITINERANT FERROMAGNETISM AND SUPERCONDUCTIVITY

The first itinerant ferromagnetic superconductor Y_9Co_7, (previously thought to be Y_4Co_3), having the Curie temperature $T_C \cong 4.5$ K and the superconducting transition temperature $T_{SC} \cong 2$ K, was discovered already in 1980 [1], (for more reading and some reviews see [2-7]). A few years ago, there was a considerable amount of work on what is believed to be true coexistence of itinerant ferromagnetism and superconductivity. In UGe_2 [8], URhGe [9] and $ZrZn_2$ [10] superconductivity was observed at very low temperatures inside the ferromagnetic phase, (for further reading see [11,12]).

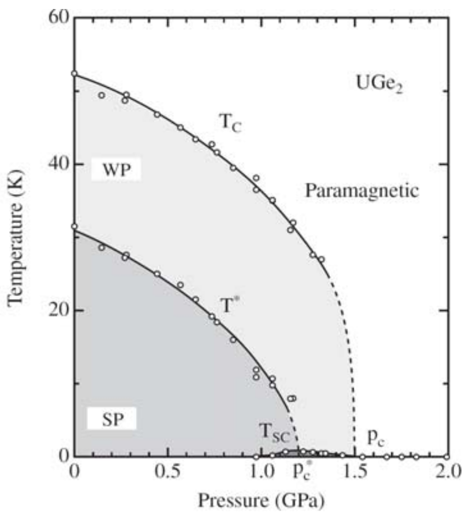

Fig.1. The phase diagram (T_C, T_{SC}-p) of UGe_2 [8,13]. The WP and SP are the weakly and strongly spin polarized phases, respectively.

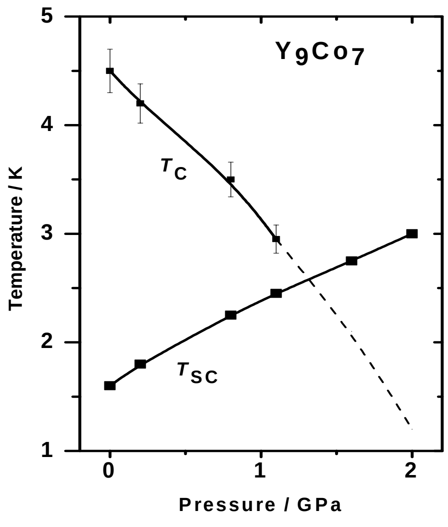

Fig.2. The phase diagram (T_C, T_{SC}-p) of Y_9Co_7. The experimental points are taken from [5,14]. The dashed line is an anticipated extrapolation.

The superconductivity in the UGe_2 was quite surprising. In a ferromagnetic UGe_2 with the Curie temperature T_C = 52 K, pressure-induced superconductivity was discovered within the pressure interval between 1.0 GPa and 1.6 GPa, exhibiting a highest transition temperature $T_{SC} \approx 0.7$ K at $p^*_c \approx 1.2$ GPa (see Fig.1). The superconductivity disappears above $p_c \approx 1.6$ GPa beyond which ferromagnetism is also suppressed. A second phase transition was found below T_C at $T^* \cong 30$ K at ambient pressure. T^* also goes to zero at $p^*_c \cong$ 1.2 GPa. In the pressure region from p^*_c to p_c, in the presence of ferromagnetic state, superconductivity was observed below T_{SC}. The region between the $T_C(p)$ and $T^*_C(p)$ curves is called the weakly spin polarized phase (WP), while the region below $T^*_C(p)$ curve is called the strongly spin polarized phase (SP), because the magnetic moment is enhanced in the strongly spin polarized phase. It is surprising that both the itinerant ferromagnetism and superconductivity are carried by 5f electrons of uranium atoms with a large moment on the order of 1 μ_B/U. This suggests rather a spin-triplet pairing state than the singlet one. The bulk nature of superconductivity was established without any ambiguity by the observation of pronounced specific heat jump at T_{SC} and from the value of Meissner state diamagnetic susceptibility.

The pressure dependence of T_C and T_{SC} of Y_9Co_7 is shown in Fig.2. A general character of the dependence is similar to the crossover of $T^*(p)$ with $T_{SC}(p)$ dependences for UGe_2, (*i.e.* of the SP itinerant ferromagnetic phase with the superconducting phase), but the values of T_{SC} are much higher.

Besides Y_9Co_7 the second example of coexistence of itinerant ferromagnetism and superconductivity <u>at ambient pressure</u> came from UGeRh. The superconductivity was discovered in UGeRh [9] with $T_{SC} \approx 0.25$ K, within the weak itinerant ferromagnetic phase below $T_C \approx 9.5$ K with the magnetic moment of 0.42 μ_B per formula unit. It seems to support some common features: the itinerant character at least of a part of the 5 *f* uranium electrons taking place in the both collective phenomena itinerant ferromagnetism and superconductivity. So, the properties of UGeRh are similar to Y_9Co_7 but its T_{SC} value is about ten times larger than T_{SC} of UGeRh mainly due to more than ten times smaller magnetic moment per cobolt atom in Y_9Co_7 (see the next section).

The problem of superconductivity in $ZrZn_2$ is still debatable. The superconductivity was discovered with $T_{SC} \approx 0.3$ K at ambient pressure, and was also confined to the ferromagnetic state for the whole interval up to a critical pressure p_c = 2.2 GPa [10]. The absence of a specific heat jump at T_{SC} and the difficulty to observe zero resistivity below T_{SC} were partially explained soon after by the experiments published in the paper "*Superconductivity induced by spark erosion in $ZrZn_2$*" [15]. The results of resistivity, susceptibility, specific heat and surface analysis measurements on high-quality $ZrZn_2$ crystals showed that cutting by spark erosion leaves a superconducting surface layer. The surface was strongly Zn depleted as shown by EDX analysis. The resistive superconducting transition was destroyed by chemically etching away the surface layer. An explanation of the results was that the superconductivity resulted from an alloy with higher Zr content than $ZrZn_2$. Both Zr and Zn are superconducting with T_{SC} = 0.55 K and 0.88 K, respectively. However, other intrinsic behavior, *e.g.,* that both T_{SC} and T_C may collapse at the same critical pressure p_c = 2.2 GPa, is still thought to support some coexistence state.

In fact, since late seventies $ZrZn_2$ is a well known very weak itinerant ferromagnet (VWIF) with $T_C \approx 28.5$ K and the magnetic moment 0.17 μ_B/Zr atom. Its magnetism is well described by the spin fluctuation theory, (for further reading see, *e.g.*, [12]). Moreover, almost 30 years ago, Enz and Matthias [16] suggested in the article under the title "*p-State Pairing and the Ferromagnetism of $ZrZn_2$*" ..."*that the weak itinerant ferromagnetism of $ZrZn_2$ must be due to electron-phonon interaction. Theoretical arguments are advanced to show that the underlying mechanism is hindered p-state pairing, due to a strongly localized repulsive part of the pair-*

potential that acts as a Hubbard interaction and gives rise to a Stoner instability". In our paper from 1984 [4], we also suggested that such phenomenon might occurs in $ZrZn_2$, $InSc_3$, $TiBe_{1.8}Cu_{0.2}$ and Ni_3Al. Some attempts to find superconductivity for the latter two compounds failed up to now. The recent theoretical results suggest that insertion of boron into cubic $InSc_3$ leads to superconductivity in $InSc_3B$ compound [17]. Thus, it remains to be seen whether higher quality $ZrZn_2$ is superconducting at ambient pressure and/or above.

The important feature of coexistence of itinerant ferromagnetism and superconductivity was revealed in the theoretical paper [18]. It was argued that the exchange of longitudinal spin fluctuations can lead to the *p*-wave pairing (triplet pairing), in weak spin-fluctuating itinerant ferromagnets. Then, the *p* state T_{SC} of this phase exhibits a maximum and falls to zero as the magnetic transition is approached from the ferromagnetic side at the so-called quantum critical point (QCP). It is shown in Fig.3. It was considered a coexistence state in an itinerant system in which the same *d* or *f* electrons may be responsible for both superconductivity and the itinerant ferromagnetism.

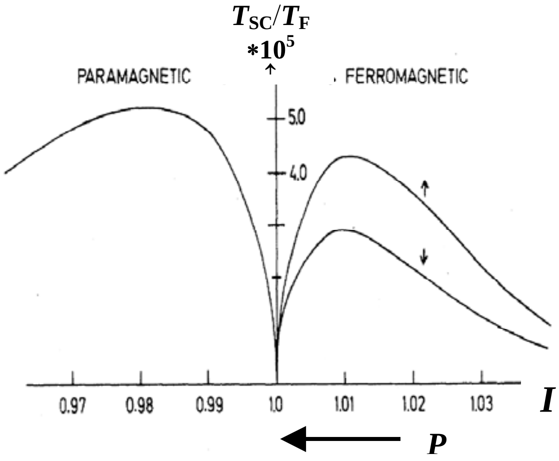

Fig. 3. The *p*- state superconducting transition temperature T_{SC} reduced by the Fermi temperature T_F as a function of the exchange interaction parameter *I* [18], which equivalently can be replaced by an applied pressure *P*.

Taking into account that increasing the external pressure should effectively decrease the exchange interaction parameter *I* one can observe qualitative agreement with the theory of the $T_{SC}(p)$ dependence in UGe_2 and Y_9Co_7 shown in Figs 1 and 2.

In contrast to $ZrZn_2$, there is no doubt about the coexistence state in Y_9Co_7. So it is a pity that almost no contemporary article on ferromagnetic superconductors, or a review of these, cites our papers on the first archetypical example of coexistence of the WIF and superconductivity in Y_9Co_7, even thought the 25th anniversary of this discovery already passed. That is why the goal of the present review is to remind the international audience that investigations of the coexistence of superconductivity and itinerant ferromagnetism started more than 25 years ago with Y_9Co_7.

II. OVERVIEW OF RESULTS FOR Y_9Co_7

The problem of magnetism in the Y-Co system was revived at the beginning of the eighties and much more interest arised since the discovery of superconductivity in Y_4Co_3 with the transition temperature $T_{SC} \cong 2.0$ K [1], later found to be the phase Y_9Co_7 [2]. Neither yttrium nor cobalt are superconducting. In the paper [1] the presence of magnetic ordering below the Curie temperature $T_C \cong 4.5$ and superconductivity below T_{SC} was observed for the first time in a transition metal alloy (Fig. 4). Thus, the intermetallic compound Y_9Co_7 turned out to be the first ever observed example of the coexistence of well experimentally characterized conventional low-temperature superconductivity and very weak itinerant ferromagnetism (VWIF), (for further reading, see [1-7]). A lot of work was done since that time up to about late eighties, and about 90 papers on the subject were published, (for an older review see [2,3,6] and for a recent review, see, *e.g.*, [7]).

The crystal structure of Y_9Co_7 is hexagonal, with the lattice parameters a = 11.53 Å and c = 12.15 Å. The unit cell is enlarged three times in comparison to that of parent Y_4Co_3 [19,20]. The Y_9Co_7 compound can be regarded as a special type of solid solution $Y_{12}Co_8$ ($\square_{2-x}Co_x$), where x = 1.33 and the symbol □ represents the two special sites per unit cell of which 1.33 positions are occupied by the b-type of Co and the other are empty. In high-purity samples T_{SC} was even as high as 3.0 K and the residual resistance ratio R_{300K} / $R_{4.2K}$ about 40 might be reached.

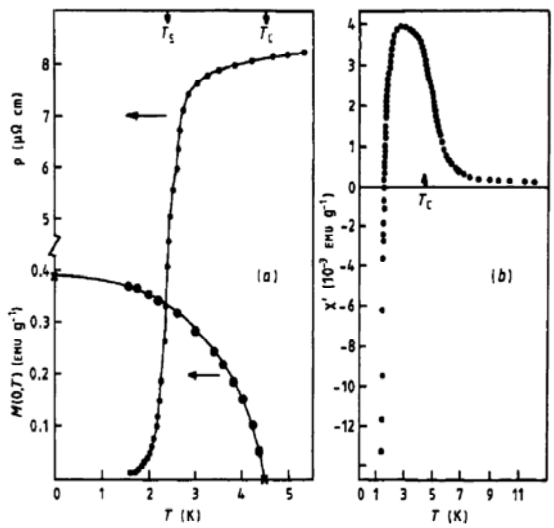

Fig. 4. The temperature dependences of (a) resistivity ρ and the extrapolated to zero magnetic field magnetization *M* (0,T) and (b) the dynamic susceptibility χ' of Y_9Co_7 [1,4,5]..

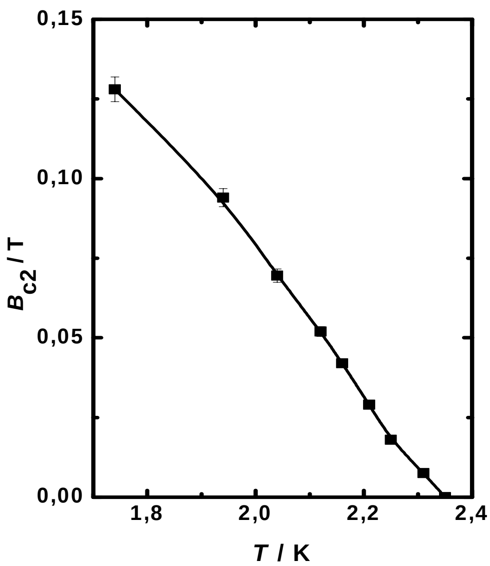

Fig.5. The temperature dependence of the upper critical field of Y_9Co_7 [5]. The extrapolated to zero temperature upper critical field is 0.38 T.

The temperature dependences of the resistivity ρ, magnetization *M* and *ac* susceptibility χ' were measured for a number of different specimens to reveal the most characteristic features of Y_9Co_7 and are shown in Fig. 4. The susceptibility χ' shows a clear diamagnetic response in the superconducting state, although the response is less than for an ideal Meissner state. However, on for many pure samples the complete Meissner state was reached later below 1.0 K with diamagnetic response $\chi' = -1.4 * 10^{-2}$ emu g^{-1}. This value is very close to the theoretical value for the perfect Meissner state $\chi' = -(1/(4\pi d)$, where the density is d = 5.85 gcm^{-3}. Thus, the compound shows the bulk superconductivity at low temperature, a rounded maximum of χ' at T_C = 4.5 K, and also obeys the modified Curie-Weiss law above 2 T_C (see Fig.1 in [4] and Fig.4 in [5]):

$$\chi(T) = \chi_0 + \frac{C}{T-\theta} \qquad (1)$$

with χ_o =(2.25 ± 0.01) $*10^{-6}$ cm^3g^{-1}, the Curie constant C =(2.29 ± 0.01) $*10^{-4}$ $cm^3g^{-1}K$ and the paramagnetic Curie-Weiss temperature θ = (13.6 ± 0.05) K. The effective magnetic moment p_{ef} = (0.14 ± 0.01) μ_B/Co atom was obtained from the expression for the Curie constant [4]. In [4], such properties were compared with those of $ZrZn_2$, $InSc_3$, $TiBe_{1.8}Cu_{0.2}$ and Ni_3A and the conclusion that Y_9Co_7 is a typical very weak itinerant ferromagnet was reached.

All samples of Y_9Co_7 accessible up to now were produced by arc melting and then annealed for a long time, first at about 850 K for two weeks, and subsequently at 750 K for about six weeks. This heat treatment is very important to yield a phase-pure material. For high quality specimens, the X-ray and neutron diffraction patterns showed a single and stoichiometric phase Y_9Co_7, with only a very small amount (less than 1 at.%) of oxides Y_2O_3 and Y_8O_5, which are both non-magnetic and non-superconducting compounds [1,2]. Unfortunately, no bulk or thin film single-crystalline material was manufactured up to now, even though a number of attempts were carried out all over the world. Some bulk single crystalline material prepared by Czochralski method quickly self-destroyed into non-superconducting powder. The superconducting properties are sensitive to the heat treatment during sample preparation whereas the magnetic properties are affected to a lesser extent.

The temperature and magnetic field dependences of magnetization were measured and analyzed at low temperatures in [5]. The extrapolated zero-field magnetization values M(0, T) for the magnetic isotherms evaluated from the proper analysis of Arrot's plots is:

$$M\,(0,T) = \lim_{H\to 0} M\,(H, T) = [(T_C - T)\,/\,T^*]^{\,\beta}\ , \qquad (2)$$

with the self-consistently fitted Curie temperature T_C = (4.47 ± 0.03) K, the critical exponent β =0.48 ± 0.01 and the scaling parameter $T^*= M_0^{1/\beta}T_0$= 0.76$*10^{-3}$ emu. The $M(0,T)$ plot is shown in Fig. 4(a). The M $(0,T)$ obeys a quadratic behavior in accordance with the spin fluctuation model of weak itinerant ferromagnetism [4,5]. From the extrapolated zero-field zero-temperature magnetization $M(0,0)$= 0.383 emu g^{-1}, (see Fig. 4a), the saturation magnetic moment was calculated to be μ_S (0) = 0.012 μ_B / Co atom. Taking into account the moment residing only on Co 2b site according to the NMR results, (*c.f.* the next paragraph), we have a saturation moment μ_S (0) ≅ 0.1 μ_B / Co_b atom. This value is the smallest among all weak itinerant ferromagnets known up to now. It was also established that spin fluctuations play a decisive role in the temperature dependences of the resistivity, susceptibility and in other properties [3-7]. In addition, the parameter $\alpha = N(E_F)\mu_B^2\,\chi_o^{-1}$ was calculated to be 1.5 $*10^{-3}$ [4,5], where $N(E_F)$=1.8 states (eV atom spin)$^{-1}$ is the density of states calculated from the specific heat coefficient [21]. Thus the condition for the existence of a ferromagnetic ground state, $\alpha + 1 > 1$, is just barely fulfilled.

From the expression for the slope of the upper critical field near T_{SC}, which is 0.23 T/K (Fig. 5), the set of superconducting zero-temperature parameters was calculated within the frame of the Ginzburg-Landau-Abrikosov-Gorkov (GLAG) theory and making use of the specific heat coefficient γ = (3.4 ± 0.1) mJ mol^{-1} K^{-2}, T_{SC} and the specific heat jump at T_{SC}. Obtained were: the GLAG coherence length of about 315 Å, the GLAG penetration depth of about 1750 Å, the lower critical field of 92 Oe, the upper critical field of 0.38 T, as well as the superconductig energy gap of 0.7 meV and the effective electron-phonon coupling parameter 0.25 [6,22]. On the basis of those parameters, Y_9Co_7 was classified as a conventional low-temperature superconductor.

From these data one can conclude that VWIF and superconductivity coexist in Y_9Co_7 within some temperature interval below T_{SC}, say 1 K $<T <$ 4 K. It is believed that such coexistence of superconductivity and itinerant ferromagnetism is not possible down to zero temperature, but it seems conceivable that the superconductivity could replace the itinerant ferromagnetism at low temperatures. Some evidence came from measurements of pressure dependence of *ac* susceptibility [2, 14]. They showed that pressure suppresses the itinerant ferromagnetism, but enhances the superconducting transition temperature yielding the highest value of $T_{SC} \cong 3.0$ K (see Fig.2).

Besides the standard magnetic and transport methods, many of differently prepared polycrystalline Y_9Co_7 samples were examined by a number of experimental techniques all over the world. Among these techniques the specific heat [21], the magnetoresistance [5], the pressure dependence of *ac* susceptibility [2,14], the nuclear magnetic resonance (NMR) [21], the X-ray and neutron diffraction structure determination [19,20], the neutron depolarization [2,23], the muon spin relaxation [24], the electron tunneling spectroscopy [25], the Mössbauer spectroscopy [26] and the photoemission spectroscopy [27] were important to prove the coexistence of spin fluctuating VWIF with superconductivity on microscopic scale.

In the last paragraph of this section we will concentrate only on some selected results of specific heat, magnetoresistance, NMR and on our new results on photoemission electron spectroscopy.

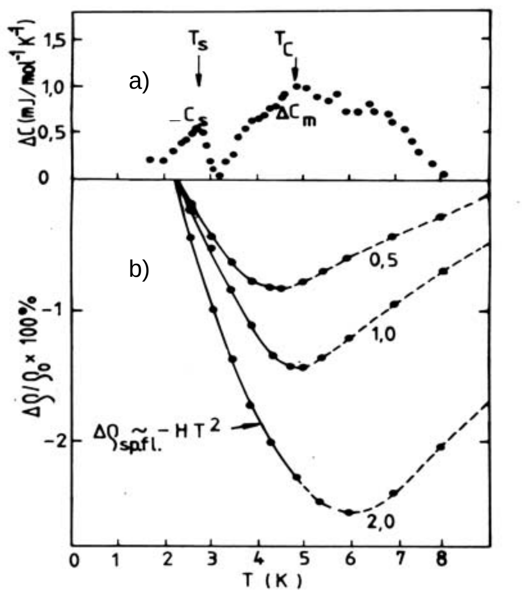

Fig. 6. a) The superconducting ΔC_S and the magnetic ΔC_m contributions to the specific heat versus temperature [21] and b) the temperature dependence of magneto-resistivity in the given magnetic field fitted to the self-consistent spin fluctuation theory [3] (solid line).

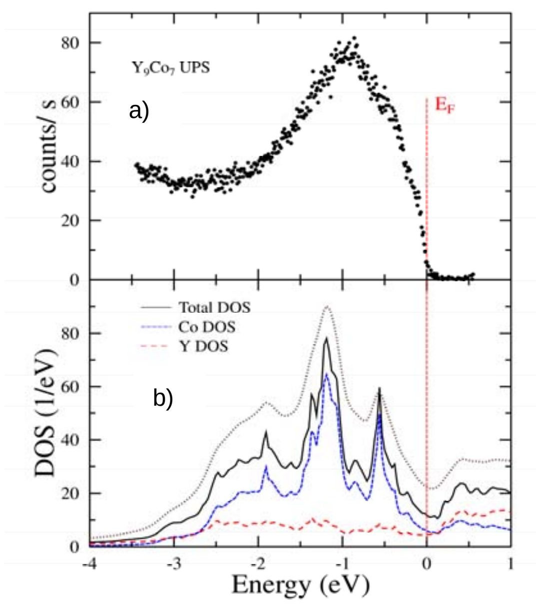

Fig. 7. a) Ultraviolet photoemission spectrum (UPS) at 300 K below Fermi energy E_F [7] and b) the calculated Co- and Y- partial as well as total density of states (DOS) convoluted with the Gaussian of energy resolution width 0.05 eV (dotted line).

The specific heat at low temperatures was measured by the semi-adiabatic heat- pulse method [22]. It is shown in Fig.6 together with the temperature and magnetic field dependence of magnetoresistance [3]. The relatively large temperature widths of the specific heat transitions to the superconducting and to the magnetic state are likely due to the magnetic field caused by ferromagnetic domains of the sample and by inhomogeneous composition of the material. Nevertheless, clear evidence of magnetic transition at $T_C \cong 4.5$ K and the superconducting transition at $T_{SC} \cong 2.0$ K was obtained. The electronic specific heat coefficient $\gamma = (3.4 \pm 0.1)$ mJ mol^{-1}K^{-2}, (close to the value for metallic Co), and the Debye temperature $\theta_D = (215 \pm 5)$ K, (close to that of yttrium), were calculated [21].

The important discovery of three non-equivalent magnetic cobalt sites in the unit cell of Y_9Co_7 was a result of NMR spin-echo measurements [21,28]. We and others found there exist itinerant magnetic moments on some cobalt positions and more local magnetic moments on the other cobalt positions, in contrast to the prediction that Y_9Co_7 should be a Pauli paramagnet. Two groups of NMR spin-echo lines were observed: the first exhibited a small but finite hyperfine field and the second had almost zero hyperfine field. Thus, two different types of Co magnetic behavior were suggested as a result of the three non-equivalent Co positions. From the NMR measurements it was established that the itinerant-electron ferromagnetism in the compound is of double character, consisting of a larger magnetic moment on the b-type Co atoms, ($\approx 0.1\ \mu_B$/Co (b) atom), with some more local behavior, and a very small itinerant magnetic moment on the d-type and h-type Co atoms. Both types of magnetic behavior were related to the composition $Y_{12}Co_8$ ($\square_{2-x}Co_x$). Its part $Y_{12}Co_8\square_2$ would be non-magnetic superconductor and the Co_x b-type atoms would carry the larger moments.

The X-ray photoemission spectroscopy (XPS) of the core levels and valence bands of Y_9Co_7 and pure Y and Co were performed with monochromatized Al K_α radiation (1387 eV) at room temperature [27]. The valence band (VB) spectra were detected at room temperature using the ultraviolet photoemission spectrometer (UPS) equipped with the high energy resolution analyzer AR 65, the Auger electron spectrometer (AES) and the low-energy electron diffractometer (LEED), as reported in our recent paper [7]. Therein, the electronic structure calculations were performed using the fully self-consistent Korringa-Kohn-Rostoker Green's function method with the coherent potential approximation (KKR-CPA) to treat chemical disorder [29,30].

Fig. 7 presents the total density of states of Y_9Co_7 compound together with the experimental UPS spectrum of Y_9Co_7. The overall shape of the DOS curve and the UPS spectrum are quite similar, especially for the energy range of -2 eV $< E <$ 0 eV. The DOS consists of three main peaks, which can be attributed basically to Co 3d states and the yttrium Y states contribution is much lower. Nevertheless, Y 4d states become more pronounced for higher energy and the DOS values are comparable to those on Co sites in the vicinity of the Fermi level (E_F). In the whole spectrum one observes a strong hybridization of *d*-states of transition elements, which is partly responsible for a broad DOS minimum appearing near E_F. Interestingly, comparing atomic DOS contributions at E_F, we notice that Co atoms located on the disordered 2b site exhibit important value of DOS at E_F, being about three times larger than the corresponding contributions on Co(6h) and Co(2d) sites. Hence, the only non-vanishing local magnetic moment can be expected on Co(2b) site. This conclusion remains in qualitative agreement with the experimental findings, which showed a very small magnetic moment on the Co 2b site ($\sim 0.1\mu_B$), as detected from NMR measurements [21,28]. Our calculations differ from those recently published [31].

III. SUMMARY AND CONCLUDING REMARKS

The summary of to date results for Y_9Co_7 addresses its itinerant ferromagnetism, its superconductivity and the coexistence of both collective phenomena:

1. The transport, magnetic, specific heat and spectroscopic properties were consistently described and understood as very weak itinerant ferromagnetism with the pronounced effect of spin fluctuations.

2. Y_9Co_7 was classified as the typical conventional BCS superconductor with rather weak electron-phonon interaction, which can be properly described by the Ginzburg-Landau- Abrikosov- Gorkov (GLAG) theory.
3. The very weak itinerant ferromagnetism and superconductivity are strongly linked forming a coexistence state. From the experimental and theoretical data one can conclude that Y_9Co_7 is a very weak itinerant ferromagnetic superconductor and a first archetypical example of the coexistence of both phenomena in a temperature interval 1 K $< T <$ 4 K.

While a number of experiments were performed, more information on the origin of magnetism and superconductivity in Y_9Co_7 is needed, especially from the point of view of electronic band structure at low temperature and for a single crystalline material. Further experimental and theoretical examination of the question whether very weak ferromagnetism might or might not be suppressed by superconductivity in Y_9Co_7 still has to be performed. Examining of the theoretical possibility that weak itinerant ferromagnetism might be particularly amenable to such a take-over bid from superconductivity is a future challenge.

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