FERROMAGNETISM AND SUPERCONDUCTIVITY*

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The experimental and theoretical status on the appearance of superconductivity in strongly correlated electronic system is reviewed with emphasis on ferromagnetism. The discovery of superconductivity in UGe_2, URhGe and $ZrZn_2$ has led to a boost of the thema. Focus is mainly given on UGe_2, this system has been already studied by different technics. Even if the main trends go in the support of triplet pairing among the majority spin band, the new s wave mechanism by the polarization given by localized magnetism is attractive. Experimentally there is no doubt that the driven force is now the improvement of the materials and further discoveries of new cases. On the theoretical side, the activities cover classification of the order parameter by group theory, microscopic and phenomenological approaches and emphasis on the superconducting order parameter and the ferromagnetic domain.

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1. Ferromagnetism and conventional s wave superconductivity

The interplay of ferromagnetism and s wave superconductivity was continuously discussed in the last decades starting with the first paper by Ginzburg [1] in 1957. The subject reaches a peak in the activity when series of magnetic superconductors with rare earth (RE) localized moment ($RERh_4B_4$, $REMo_6S_8$) appear [2]. Basically, antiferromagnetism and superconductivity lives peacefully; on the length of their superconducting coherence length ξ the cooper pair fields an average zero internal magnetic field as the magnetic period d is far smaller than ξ. In these localized magnetic

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systems, the energy gained by the atoms due to the magnetic transition at Néel temperature T_N far exceeds the energy gained by the electrons as they form Cooper pairs at the superconducting temperature T_C even if usually T_C is higher than T_N in these series [3].

In the case of ferromagnetism, the Curie temperature T_{Curie} must be lower than T_C since the exchange field must be weaker than the magnetic field H_P which will break the singlet Cooper pairs ($H_P = 1.8\,T_C$ in Tesla). In the two well-known cases of $ErRh_4B_4$ [4] and $HoMo_6S_8$ [5] when long range magnetic order appears below $T_m < T_C$, the magnetic structure is not ferromagnetic but is with a period $d \leq \xi$ but large by comparison to the interatomic distance. In the intermediate narrow temperature range $T_{Curie} \leq T \leq T_m$, the magnetic arrangement looks ferromagnetic on few atomic distancies but antiferromagnetic on the ξ size. As it costs energy to create this magnetic layer structure, ferromagnetism wins at low temperature and superconductivity simultaneously collapses. Since the transition at T_{Curie} is of first order, regions with superconductivity and modulated magnetic structure and with ferromagnetism may coexist macroscopically. Finally local superconductivity can appear in domain walls [6]. There is no coexistence of ferromagnetism and superconductivity on a microscopic scale.

2. Itinerant magnetism and unconventional superconductivity

The role of ferromagnetic spin fluctuation in triplet Cooper pairing was extensively analyzed for the quantum superfluid He^3 but also for itinerant ferromagnetic metals closed to their quantum critical point *i.e.* the critical density or pressure (P_C) where T_{Curie} collapses. For the ideal case of a spherical Fermi surface [7], superconductivity occurs quite symmetrically on both side of P_C with a collapse of T_c just at P_C and similar maxima in the temperature T_C at $P \sim P_C \pm \delta$ (Fig. 1).

The occurrence of triplet superconductivity seems established for the heavy fermion compound UPt_3 [8], and for the ruthenate case Sr_2RuO_4 [9]; the link with ferromagnetic fluctuations is not demonstrated. For Sr_2RuO_4, there is no doubt that the ground state is paramagnetic.

To search for superconductivity in itinerant ferromagnets close to their QCP, the two channels for material are transition intermetallic systems and $4f$ or $5f$ heavy fermion compounds (mainly Ce, Yb or U systems). For the first ones, the itinerant nature of the magnetism has been well demonstrated and explained by the so-called self consistent spin fluctuation theory [10,11]. For the second ones, the dual localized and itinerant nature of f electron is still under discussions (see this proceedings); experimentally they offer the advantage that often a rather moderate P scan (GPa) tunes the system through P_C with huge variations of the electronic contributions.

Recent experiments on AF systems through P_C, show the emergence of a superconducting pocket centered on P_C [12]. They give a strong support for

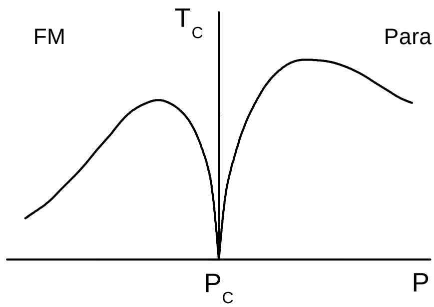

Fig. 1. The pressure variation of the superconducting temperature T_{C} obtained by the spin fluctuation theory of Ref. [7] in arbitrary unit.

spin fluctuation mediated superconductivity. The key point is to achieve the clean limit condition *i.e.* an electronic mean free path $\ell \geq \xi$; in this unconventional superconductors (d singlet type) any type of impurities (even non magnetic) is pair breaking. In parallel with the requirement of high material purity, the thema has highly benefited from the improvement in the pressure technic as well as in progresses on details as the electronic connection. For example in Grenoble, a main breakthrough comes with the development of the microsoldering with tiny gold wires on any metallic material during the sabbatical stay of Dr. Y. Okayama stimulated by Prof. T. Kasuya. The now so-called OK process is currently used by different groups.

3. Experimental breakthroughs

The discovery of superconductivity in the UGe_2 ferromagnet [13], deep inside its ferromagnetic region, was quite surprising; at the pressure $P \sim 12$ kbar where $T_{\mathrm{C}} \sim 0.7\,\mathrm{K}$ reaches its maxima, T_{Curie} is still near 30 K and the sublattice magnetism M_0 near $1\mu_{\mathrm{B}}$. Triplet superconductivity seems likely as the exchange magnetic field (near 100 T estimated in a one electron picture) overpasses the Pauli limit H_{P} by orders of magnitude. The reproductibility of the phenomena was first verified inside the Cambridge–Grenoble collaborations and then rapidly confirmed in Osaka [14], Nagoya [15] and La Jolla [16]. The bulk nature of superconductivity was suggested in flux flow resistivity experiments [17] and established without an ambiguity by the observation of a 30% specific heat jump at T_{C} for $P \sim 12$ kbar [14].

The further discovery of similar phenomena in URhGe [18] even at zero pressure where $T_{\mathrm{C}} = 0.3\,\mathrm{K}$, $T_{\mathrm{Curie}} = 9.5\,\mathrm{K}$ and $M_0 \sim 0.5\,\mu_{\mathrm{B}}$ seems to open the route to decisive experiments to test the order parameter as a large experimental panoply can now be applied at $P = 0$. Common features between UGe_2 and URhGe are their low orthorhombic symmetry the occurrence of uranium zig zag chain, at least the itinerant character of a part of the $5f$ uranium electron and the Ising character of the magnetism.

The observation of superconductivity in $ZrZn_2$ [19] is also restricted to the ferromagnetic side ($P < P_C$). $ZrZn_2$ is known as a weak Heisenberg ferromagnet. At $P = 0$, $T_C = 0.3\,K$, $T_{Curie} \sim 28.5\,K$ and $M_0 = 0.17\mu_B$; its spin dynamics was well analyzed in the spin fluctuation framework [20]. The still puzzling questions are the absence of a superconducting specific anomaly at T_C, the difficulty to observe a zero resistivity below T_C as well as the usual peak in the imaginary part of susceptibility at T_C. The strong support of an intrinsic behavior is that T_C and T_{Curie} may collapse at P_C. Obviously, there is an urgent need of a new generation of $ZrZn_2$ materials. Other attempts to observe superconductivity in other transition intermetallic compounds (for example in Ni_3Al up to 10 GPa) have up to now failed [21].

4. Superconductivity in paramagnetic ε phase of Fe

In its low pressure cubic ($P \leq 13\,GPa$) α phase, Fe is a strong ferromagnet and no superconductivity is expected. However, recently superconductivity was found in its high pressure hexagonal compact ε phase ($P \geq 13$ GPa) [22]. The $\alpha \to \varepsilon$ structural first order transition occurs almost at constant pressure $P_{\alpha\to\varepsilon} \sim 13$ GPa. At least from Moessbauer experiment [23], the ground state of ε Fe is paramagnetic while band structure calculations predict that antiferromagnetism is more stable than ferromagnetism [24].

Recently the first two contact resistance measurements [22] on ε Fe was completed by a four lead absolute resistivity (ρ) experiments [25]; it allows to verify the achievement of a zero resistivity below $T_C = 2\,K$ for $P = 22\,GPa$ and to derive the inelastic contribution $A_n\ T^n$ of ρ. Close to P_C, the exponent n is characteristic of the magnetic fluctuation; for 3 dimensional case $n = 5/3$ and $3/2$ respectively, for ferromagnetic and antiferromagnetic QCP [10]. As suspected, the $A_n T^n$ term is large far stronger than the weak temperature observed in the α ferromagnetic phase; the more surprising result is the observation of $n = 5/3$ exponent as predicted for ferromagnetic spin fluctuation. This result as well as the relative weak pressure dependence of T_C (the superconducting pocket extends from 13 to 30 GPa) are explained in band calculations where a ferromagnetic coupling prevails [26]. At least, ε Fe is another case where superconductivity exists in a strongly magnetic fluctuating medium.

These different results have led to a large theoretical activity. Before a rapid overview, we will discuss in more details the data on UGe_2 which is up to now the main studied example.

5. UGe_2: two magnetic states?

The striking point [17] is that under pressure an extra feature appears at $T = T_x$ in the temperature variation of ρ(T) outside the kink observed at T_{Curie} and the drop of ρ to zero at T_C (Fig. 2 and 3). The marked drop

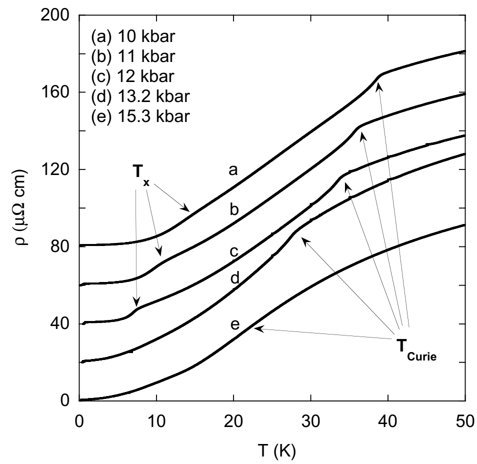

Fig. 2. Temperature variation of the resistivity of UGe_2 at different pressure with the emergence of the two anomalies at $T_{\rm Curie}$ and T_x below $P_x = 12.2\,$kbar.

observed at T_x on cooling is also associated with a jump of the sublattice magnetization [15,17,27] and a drop of the residual C/T term of the specific heat (*i.e.* decrease of the average effective masses) [28]. Up to now as indicated in Fig. 4, the magnetic states $FM1$ and $FM2$ appears ferromagnetic. No evidence of extra charge density wave or spin density wave contributions has yet been detected in neutron or X ray scattering experiments realized in Grenoble [29] or Tokai [30]. $FM1$ and $FM2$ are often labeled weak and strong polarized phase. Magnetization and neutron scattering experiments [31] indicates that when T_x collapses at P_x, the transition may be first order as the change ΔM_0 is drastic at $P_x \sim 12\,$kbar ($\Delta M_0 = 0.4\,\mu_{\rm B}$) while at $P_{\rm C} = 16\,$kbar the ΔM_0 drop is $0.78\mu_{\rm B}$. The transition at $P_{\rm C}$ is of first order [17].

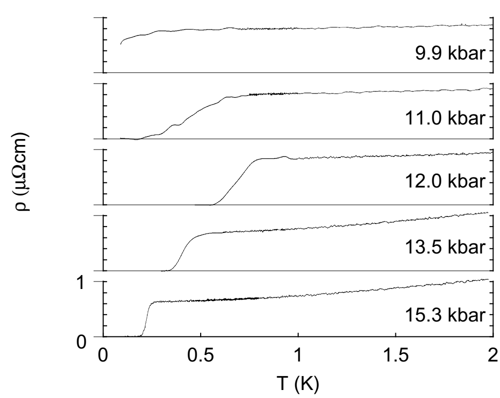

Fig. 3. Observation of the superconducting resistivity anomalies for UGe_2 at different pressures.

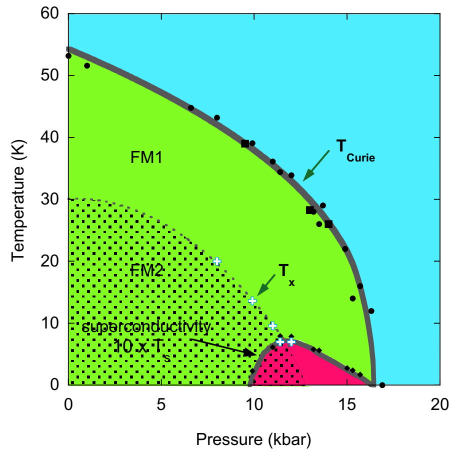

Fig. 4. (T, P) phase diagrame of UGe_2 from Ref. [17] and [31].

Applying a magnetic field along the easy a axis restores a strongly polarized ground state $FM2$ for $H = H_x$ for $P > P_x$ and even the successive phases $FM1$ and $FM2$ at H_M and H_x above P_C (Fig. 5) [27, 31]. Fermi surface (FS) determinations were realized in Osaka [27], [32], and Tsukuba [33,34] with the field oriented along the different principal axis. The

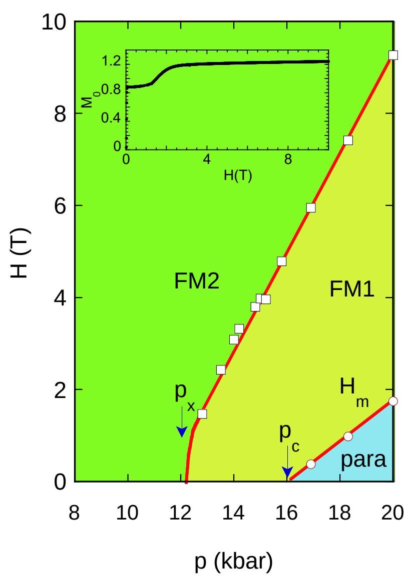

Fig. 5. (H, P) phase diagrame of UGe_2 the insert show the jump of M_0 in μ_B at the transition $FM1 \rightarrow FM2$ at P_x [31].

$H \parallel b$ experiments with very weak polarization probe the different phases. In agreement with band structures calculations, [35], strong differences exist between large majority and small minority spin band at least below P_x while a drastic FS change occurs on entering in the paramagnetic regime. Applying a magnetic field along a leads to scan the different phase $FM2$ and $FM1$. The situation is clear for the $FM2$ state and thus the smooth pressure evolution of its FS established. For $FM1$ there is still not agreement on FS signals. Let us also stress that band structure calculations [35], [36], point out the large $5f$ contributions at the Fermi level; the occurrence of sharp structured density of states will favor nesting and field instability as observed at H_x and H_M. The field transition at H_x from $FM1$ to $FM2$ has clear consequences on the unusual shape of the upper critical field H_{c2} as observed Fig. (6) at $P = 13.5\,\text{kbar}$ [37].

The drop of the resistivity at T_x as well as the coincidence of the maxima in T_C when T_x collapses are reminiscent of the paramagnet α Uranium, where T_x is identified as the charge density wave temperature T_{CDW}. Furthermore, the common point between UGe_2 and α Uranium is their zig zag Uranium chain [38]. This analogies plus the unusual temperature variation of the neutron intensity of ferromagnetic Bragg reflection (at T_x) and a bump in C/T near T_x push to propose a model where CDW may occur below T_x [39]. This model is able to explain the field instability at H_x and the unusual shape of $H_{c2}(T)$ of Fig. (6) [29,37,40]. Up to now, as indicated no superstructure has been detected. In this conference, a zero temperature Stoner model [41] is proposed on a system which has a twin peak structure in the electronic density of states *i.e.* the ingredient for the two meta-magnetic fields H_M and H_x.

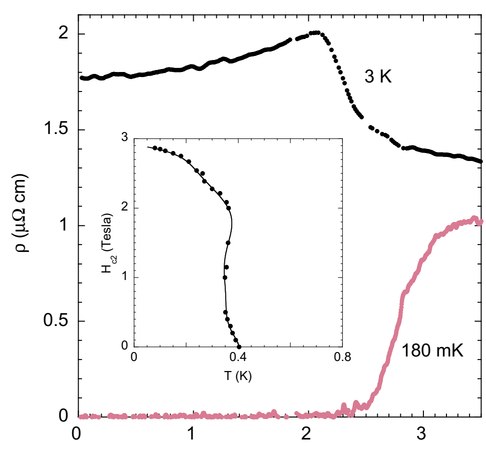

Fig. 6. For $P \sim 13\,\text{kbar}$, the resistivity variation observed on entering in FM2 at $H_x \sim 2\,T$ for $T = 3.0\,\text{K}$ and the observation of $H_{c2}(180\,\text{mK})$. In insert, the upper critical field H_{c2} at the same pressure [17–37].

Up to now, superconductivity and ferromagnetism are assumed to coexist on a microscopic scale. From ac susceptibility measurements, it has been claimed that the superconductivity may coexist in a competitive way with ferromagnetism [15]; as P increases, a volume fraction of the superconductivity may grow while ferromagnetism appear spatially inhomogeneous. To increase the complexity, the Nagoya group [39] suggests now that the size of magnetic domain is smaller than ξ; this proposal comes through the observation of quantized magnetization jump at low temperature and parallelism with phenomena observed for quantum magnetic cluster. The statements of inhomogeneity are reminiscent of previous problems found as underlined before in $HoMo_6S_8$ and $ErRh_4B_4$. They must be also associated with the statement that in UGe_2 the clean limit condition for unconventional superconducting may not be achieved as dirty poly-crystals ($\rho_0 \sim 3\mu\Omega\,cm$) ($\ell = 100$ Å $< \xi 4 = 150$ Å) are superconducting [16]. These restrictions must be of course taken seriously; but there is no decisive proofs on the invalidity of the clean limit, on a segregation between superconducting and ferromagnetic regions (the optima in the jump of the superconducting specific heat jump is found near P_x and not at all on approaching P_C) and on resonant tunneling between quantum spin states at low temperature as observed for macroscopic quantum tunneling.For example, our estimate of ℓ from residual resistivity ρ_0, specific heat C and H_{c2} leads to an order of magnitude greater for ℓ than the derived in 16 for the same ρ_0.

In this new superconducting materials, the positive aspect is the reproducibility of the main data (P, T, H, phase diagrame); microscopic knowledges are far to be established.

6. Theoretical overview

On general symmetry arguments, the different superconducting states in ferromagnetic phases for crystal with cubic [42] and orthorhombic structure [43] have been classified. For the $ZrZn_2$ cubic case [44], it was predicted that the gap nodes change when the magnetization is rotated by the magnetic field. Tests can be easily made in ultrasound attenuation and thermal conductivity experiments. For a orthorhombic point group, only one dimensional representations are possible; this can lead to a magnetic superconducting phase with spontaneous magnetization when superconductivity occurs inside the ferromagnetic region ($T_{\mathrm{Curie}} < T_C$) for the case of a strong spin–orbit coupling [43, 44]. In general no symmetry nodes exists [43], at least with a pairing amplitude with the zero projection of the Cooper pair. If this component is canceled, zeros can occur [45].

A new glance is the interplay between ferromagnetic domain and the superconducting order as the superconducting order parameter is linked to the magnetization [44–46]. It has been emphasized that the superconducting nano-structure will consist of complex conjugate states related to the opposite directions of M in adjacent domain [43].

Another supplementary consideration [47] at least in resistivity experiments, is the domain wall superconductivity *i.e.* how magnetic domains influence the superconducting characteristic of unconventional ferromagnetic superconductors. In each domain a finite average magnetic induction $4\pi\ M_0$ exists (near 2000 Oe for UGe_2). Assuming a thin domain wall ($\ll \xi$) and modeling the domain interface by a step like function $\pm\ M_0$ on each side of the wall, the orbital effect is canceled. On cooling, the superconductivity will first appear locally at domain wall not inside the magnetic domain. Furthermore depending on the relative orientation of M by respect to H, different critical temperatures will occur between two opposite domains. That must give to observable effects near T_C such as an unusual broadening at $H = 0$ which will disappear in H.

One up to date problem is the reason for the stabilization of superconductivity in the ferromagnetic region. It was suggested [48] an exchange type interaction between the magnetic moments of Cooper pair with the magnetization density. That conducts to a collapse of T_C at P_C according to the relation $T_C(P) = T_C(0)(1 - P/P_C)^{1/2}$ (Fig. 7) assuming a linear decrease of T_{Curie}. This results explains rather well the case of $ZrZn_2$. The balance between the enhancement of T_C stimulated by the exchange field (as by the magnetic field for the $A1$ superfluid phase of ^{3}He) and the suppression due to the orbital electron motion has been discussed recently in Ref. [43]. The criteria for such a superconducting stabilization in the ferromagnetic domain may not be fulfilled.

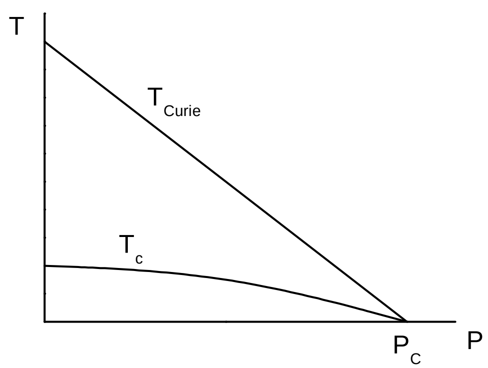

Fig. 7. The proposed (T, P) phase diagrame [48] due to exchange coupling between the magnetism of the Cooper pair and the ferromagnetic magnetization. He reproduces well the phase diagrame of $ZrZn_2$ found in Ref. [19].

An alternative idea for the weak Heisenberg ferromagnetic is to link the disappearance of the transverse magnetic fluctuation for coherent magnons below T_{Curie} with an enhancement of T_C on the ferromagnetic side. The coupling of magnon to longitudinal magnetic susceptibility enhances strongly T_C respectively to the paramagnetic state [49]. The coexistence of spin triplet superconductivity with an itinerant ferromagnetism induced by the Hund'rule exchange is presented in this proceedings [50].

Of course, another possibility is to consider the possible occurrence of s wave superconductivity by bypassing the argument on the strength of the exchange field seen by the conduction electrons. Such a possibility is considered mainly for UGe_2 as the ferromagnetism may come from the localized $5f$ part. It was shown that the coupling of two electrons via a localized spin can be attractive [51] and demonstrate that this s wave attraction holds for the whole FS [52]. The supplementary condition for the occurrence of superconductivity a large density of states at Fermi level *i.e.* a heavy fermion case; the coupling will be also strong [51]. The applicability to UGe_2 is an open question.

The stability of a s wave superconducting ferromagnetic ground state has been also discussed in Refs. [53, 54]. However, the validity of the approaches are under debates [55]. There is also the possibility with complex Fermi surfaces to escape from the strength of the exchange if particular electronic orbits can get strong electron phonon coupling and weak exchange splitting. Such a remark has been made for $ZrZn_2$ [56].

Finally in UGe_2 and also in some AF systems as $CeRh_2Si_2$ reported in this meeting [57], QCP points are unlikely since the transition at P_C appears first order. It may confirm that low energy excitation may not play a positive role on the Cooper pairing. A first approximation, the magnetic coherence length may exceed a critical value for the onset of superconductivity.

7. Future experimental challenge

The discovery of new materials and possible at zero pressure is crucial even in the study of ferromagnetic quantum critical point. Transition intermetallic compounds are more simple for the discussion on the itinerary but they appear quite difficult to study by transport measurements since even deep inside the ferromagnetic region no simple T^2 Fermi liquid law is observed ($ZrZn_2$, Ni_3Al); other ferromagnetic heavy fermion systems has the advantage of a complete pressure scan from localized to itinerant behavior.

Even in the referenced materials (UGe_2, URhGe, $ZrZn_2$) there is a need to systematic measurements with different mean free path (UGe_2) to produce a new set of single crystals (URhGe, $ZrZn_2$) where a large diversity of tests on the order parameter can be realized (superconductivity at $P = 0$).

The field of ferromagnetism and superconductivity in strongly correlated electronic systems is recent. Many points are still to be confirmed. Specific domain structures or spontaneous vortex states are worthwhile to study. Microscopic measurements as NMR and/or spatial low energy spectroscopy will confirm or infirm some of our statements. The realization of detailed magnetization measurements is a simple and important goal. After the new experimental facts which attract again theoretical activities, a new generation of sound experiments is needed.

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