Chapter 18
Exciting Times in Condensed-Matter Physics

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My first personal encounter with Alex Müller dates back to the second half of the seventies of the last century. On the occasion of a meeting of MECO (Middle European Cooperation Organisation) in Switzerland, I presented a poster on the induced Jahn-Teller effect in $PrCu_2$ [1]. Induced-moment magnetic ordering, i.e., magnetic order in a number of rare-earth (RE) compounds where the ground state of the localized 4f electrons on the RE ions is a singlet state, had occasionally been observed [2]. Not yet reported then was the structural counterpart, i.e., spontaneous lattice distortions due to a cooperative Jahn-Teller effect and $PrCu_2$ was shown to be the first such case [3]. As we now know, the Jahn-Teller effect later became a rather important aspect in the scientific activities of Alex but at that time, he didn't seem to be particularly impressed by my explanations.

Next, a few years later, we met at the IBM research laboratories at Yorktown Heights where Alex spent some sort of sabbatical leave from the Rüschlikon labs near Zurich. He mentioned that he was now interested in superconductivity of granular Aluminium, which used to be prepared by evaporation of the pure metal in a suitable atmosphere of Oxygen [4]. It had been found that the critical temperature T_c of that material was up to a factor three higher than that of very pure Al but by most, this was not considered as a major breakthrough because T_c was still of the order of only 3–4 K. By experiment it was shown that the granular structure of the material led to the appearance of low-frequency phonon modes [5] and therefore, a softening of the lattice and hence, according to MacMillan [6], an enhancement of the electron-phonon interaction and thus to the observed increase of T_c.

Probably unknown to the world outside the Rüschlikon lab, Alex seems to have continued with his search for new superconductors exhibiting higher T_cs by enhancing the electron-phonon interaction of potentially promising materials.

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Among the highest T_cs for superconductivity found at the time were for compounds featuring the cubic A-15 structure, of which V_3Si, V_3Ga and Nb_3Sn were particularly prominent examples. It had been observed that these materials underwent martensitic phase transformations due to a structural instability [7] a few K above the superconducting transition. Because of the particular electronic band structure, this instability was interpreted as being driven by a band Jahn-Teller distortion involving the redistribution of electrons between the d-sub-bands of different symmetries [8]. In an collaboration between the ETH in Zürich and the University of Geneva, invoking dilatometric measurements on V_3Si, it was shown that the cubic to tetragonal distortion was abruptly terminated by the onset of superconductivity and therefore it was concluded that not the structural effect per se but rather the strong electron-phonon coupling was favouring the occurrence of superconductivity in this case [9, 10]. At about the same time, theoretical work aiming at investigating the similarities between localized pairs of electrons termed bipolarons, caused by a deformation-induced attraction and itinerant Cooper pairs in a superconductor showed that both pairings are a consequence of the general Hamiltonian capturing the electron-phonon interaction [11]. The caveat here is, however, that if the coupling strength exceeds a certain value, the result is a bipolaronic insulator. This new point of view naturally suggested the possible formation of Jahn-Teller polarons [12] and, according to later-told stories it was this approach which guided Alex in his efforts to find suitable materials.

Meanwhile I had followed up another puzzling development in superconductivity, namely the occurrence of superconductivity in a strongly paramagnetic background which was first reported for $CeCu_2Si_2$ [13]. Because this observation was really unexpected and not in line with the then current understanding of superconductivity, it was confronted with strong doubts by many of the community. In a collaboration between my group at ETH Zürich and friends at Los Alamos National Laboratory, we succeeded to confirm this feature of pairing of electrons with extremely large effective masses, i.e., heavy or rather slow itinerant electrons in metals, to also be exhibited by the actinide compound UBe_{13} [14]. The data also indicated some unconventional type of superconductivity, in some way analogous to the previously reported superfluid states of ^3He. The discussion of the data started what later turned into the hunt for power laws in the temperature dependencies of relevant physical quantities, such as the specific heat, below T_c [15]. Such power laws are expected to replace the conventional exponential reduction of electronic excitations in the superconducting state with no gap nodes. At the time, the existence of superconductors exhibiting nodes in the gap, was not very popular but many new experimental data pointed in that direction and induced corresponding work in the theoretical sector. The observation [16] of two subsequent transitions between two superconducting states in $U_{1-x}Th_xBe_{13}$, depending on x, added another strong argument for the unconventional character of this superconductivity, in the sense that in this case it is not the electron-phonon interaction that causes the instability of the ensemble of itinerant electrons. On the occasion of a talk describing these developments in the realm of the Physikalische Gesellschaft Zürich, Alex asked whether this type of superconductivity might also be favourable for high critical temperatures. Since large effective electron masses imply a low value of the Fermi energy E_F, the characteristic energy of the itinerant-electron subsystem, my answer was, of course: rather unlikely. In retrospect it is clear that Alex already had some hints on the onset of superconductivity in the cuprates that he was investigating with Georg Bednorz.

As far as I remember, I had already heard that something was brewing before the public announcement in the form of an article that appeared in Zeitschrift für Physik in October 1986 [17]. As expected, the reported discovery of La-based cuprates being superconductors with a T_c exceeding 30 K was so unexpected that it first met with a lot of skepticism. It didn't take long, however, until the first confirmations appeared in preprints and Conference talks [18–20] and it was clear that the discovery was real and meant serious business. The chapter of high-T_c superconductivity had been opened. This also became clear at a Conference on *Valence Fluctuations and Heavy Fermions* at Bangalore in early January 1987. It was there where Phil Anderson addressed the stunned audience by proposing a novel mechanism as the cause for high-T_c superconductivity in doped La_2CuO_4 [21].

At this point I should like to add some remarks on how a discovery is made. To many the fact, once established, may seem to be a lucky punch which everybody might have landed. But this is almost never the case. In our perspective it is interesting to note, that the same class of cuprate materials had been synthesized and studied by a Russian group of chemists in 1979 [22]. They noted that some of them exhibited metallic conductivity, i.e., $\partial\rho/\partial T > 0$ at room temperature, in particular compounds of the type $La_{2-x}MO_4$ with M = Ba or Sr. They had correctly identified the crystal structure as being of tetragonal K_2NiF_4-type but were mainly interested in the temperature dependence of the resistivity as a function of the varying c/a ratio upon replacing La by Sr or Ba. Everything for a great discovery was in their hands but it didn't happen. It may well be that they did not dispose of cryogenic equipment for cooling the samples to low temperatures. What is more decisive, however, is that they did not have the vision for exploring the possibility of superconductivity in materials of this type as Bednorz and Müller did 6 years later. In other words, in most cases experimental discoveries are either made because you are looking for it or, you recognize it when unusual results are looking at you. Nevertheless, I always felt that it was somewhat unfair that these Russian authors were never cited for their work in the flood of papers on $La_{2-x}M_xCuO_4$ that appeared in the literature after the beginning of 1987.

Meanwhile in December 1986, Gerd Binnig and Heini Rohrer, both also members of the IBM research lab in Rüschlikon, were awarded with the Nobel Prize for inventing and pioneering the scanning tunneling microscopy (STM). During a celebration of their achievement in a leading hotel in Zürich in January 1987, rumours were spread that one would meet again for the same reason at the same location in early 1988. For the few present that were in the game, it was clear what was meant and, as is well known now, it did happen. Two consecutive Nobel prizes exclusively awarded to scientists of a relatively small research laboratory of a private company, a really amazing fact.

The new field of high-T_c superconductivity got an immediate boost in early 1987 via the announcement of the observation of the onset of superconductivity in a not yet identified material containing Y, Ba, Cu and O at 93 K, i.e., distinctly above the boiling point of liquid nitrogen [23]. Again, it didn't take long, barely a few weeks, for numerous confirmations and the identification of the actual compound, $YBa_2Cu_3O_7$, to appear in the literature. At ETH, in spring 1987, Fritz Hulliger and myself embarked on a study of this type of cuprates with a series of compounds where Y was replaced by rare-earth (RE) elements [24]. Contrary to conventional superconductors, the presence of large magnetic moments due to the RE ions had virtually no influence on the value of T_c, especially true for the heavy RE elements beyond Gd where all T_c values exceed 90 K. Neutron diffraction experiments revealed that some of these compounds exhibit magnetic order at temperatures below 1 K without losing superconductivity, i.e., the coexistence of the two long-range order phenomena [25]. These early observations were, of course, no convincing evidence for unconventional superconductivity in these cuprates, but at least among the first hints in this direction.

In early summer 1987, the Swiss community involved in research on superconductivity reached the conclusion that the already planned *Conference on Superconductivity in d- and f-band metals*, to be held in Switzerland in 1988, should be restructured to include the materials class of copper oxides. The new Conference title *Materials and Mechanisms of Superconductivity, M^2S-HTSC* was quickly adopted and a committee with Alex and myself as co-chairs, supported by colleagues from the Swiss community, set to work. As many still remember, the result was the memorable Interlaken meeting from February 29 to March 4, 1988. The conference attracted more than 1100 participants from 39 countries, an extraordinary large number for this type of meetings, and got the then still unusual attention of all types of media. The audience also included more than 30 delegates from the Soviet Union, again a before unheard number of colleagues from that part of the world together in a single event in the West. Most memorable were the late night sessions where everybody could register to present new data which daily came in through the at the time very new channel of telefax. These sessions often ended between 2 and 3 a.m. and morning sessions started again at 08:30 or 9 a.m. It was a tough week for everybody and, although we had to cope with ample snowfall, we even managed to order some sunshine for the excursion up to Eiger, Mönch and Jungfrau.

During 1987, yet another class of cuprate superconductors made the headlines. Based on indications for superconductivity of $Bi_2Ba_2CuO_6$ around 20 K [26], it was soon realized that by inserting a plane of Ca atoms into the original structure, resulting in the compound $Bi_2Ba_2CaCu_2O_8$, T_c could be raised to above 80 K [27]. The trick with inserting Ca planes and, in addition, enhance the number of the notoriously essential Cu–O planes in cuprate structures, turned out to be a key element for raising T_c. For the Bi-based cuprates, adding even one more Ca and Cu–O plane, respectively, T_c values exceeding 100 K were reached. Early in 1988 and even during the Interlaken meeting, results from studies of yet another class of cuprates starting with $Tl_2Ba_2CuO_6$ and, applying the Ca/Cu–O trick with now five components Tl, Ba, Ca, Cu and O, were reported and a T_c of 125 K was finally reached [28, 29].

It took a few years until a renewed excitement with respect to raising T_c even further emerged. Based on results of a new compound, $HgBa_2CuO_{4+\delta}$, exhibiting an onset for superconductivity at 94 K, by far the highest T_c for a cuprate compound with only one Cu–O plane per structural unit [30]. In that work it was mentioned that the above mentioned Ca-trick doesn't work. Since we had done a few excursions into sample preparation and had some experience in synthesizing cuprate materials [31], we thought we would try it anyway. The courage paid off and we could announce a new record-high critical temperature of 133.5 K [32] for $HgBa_2Ca_2Cu_3O_{8+\delta}$. Later work revealed that under the application of external pressure, T_c could be enhanced substantially, however, the results obviously depended a lot on the chosen samples and their composition [33, 34].

In the mean time, most worldwide research efforts, both in theory and experiment, had been devoted to characterize the normal and the superconducting state of these cuprate compounds and to identify possible pairing mechanisms for the onset of superconductivity at these, from the traditional point of view, extraordinary high temperatures. Starting already in 1986, the first theory-based claims for a superconducting state with intrinsic gap nodes appeared in the literature [35]. The first reliable experimental indications appeared a few years later, mostly in the form of the above mentioned power laws in the temperature dependencies of relevant parameters below T_c, such as the NMR Knight shift and the penetration depth of external magnetic fields [36]. A more rigorous experimental verification of the suggested d-wave, i.e., to be precise, the $d_{x^2-y^2}$ symmetry of the gap was suggested by Sigrist and Rice [37], based on an idea of Geshkenbein and Larkin for the case of heavy-electron superconductors [38], to probe the relative phase of the gap at different points of the Fermi surface. Results of the first successful experimental attempt were reported by Wollman et al. [39] and not much later, our group at ETH presented data obtained with a somewhat different experimental set-up, which confirmed the claimed gap-node configuration [40]. At about the same time, a very elegant but technologically more demanding experiment by Tsuei and Kirtley [41] led again to the same conclusion. Shortly after these developments, Alex Müller reacted and argued that in order to capture the overall 3-D features of the order parameter, a scenario considering a mix of d- and s-wave symmetry would have to be taken into account [42]. With respect to mechanisms, Alex, mainly based on experimental evidence of isotope effects on T_c of $La_{2-x}Sr_xCuO_4$ [43], never got tired to remind the community that the influence of the lattice, resulting in the formation of Jahn-Teller polarons, should not be neglected.

The discovery of high-T_c superconductivity in copper-oxide materials not only initiated experimental and theoretical research to unravel the secrets of this type of conductors and their superconductivity. A special feature of these cuprate compounds were and are their low-dimensional (low-D) structural subunits in the form of Cu–O planes and chains as, e.g., in $YBa_2Cu_3O_7$. This led to a very intense activity of synthesizing similar materials in the form of transition-metal oxides.

Many of them were not superconductors but they served as a testbed for experimental studies of the behaviour of low-D systems, particularly with respect to their magnetic properties. For example, the mentioned subunits can be regarded as chains, ladders or planes of spin arrays. Of particular interest are compounds that can be regarded as containing individual spin chains or coupled chains to form 2-leg ladders, for instance. Aspects of physical properties of such quasi 1-D cuprate materials were first discussed by Dagotto and Rice [44]. Depending on the choice of the transition-metal element the individual spins adopt a half-integer or integer value. Although in real materials these low-D subunits are components of a three-dimensional crystal structure, in many cases, the anisotropy of the relevant interactions is large enough to hope that some of the typical low-D features that had extensively been studied theoretically since many years, would survive to show up in experimental data of appropriate physical properties of these materials.

For our purposes, outlined below, various Cu-oxide compounds are of particular interest. Examples are Sr_2CuO_3, $SrCuO_2$ and $Sr_{14}Cu_{24}O_{41}$. Structurally they contain linear spin-½ chains, ribbons and 2-leg spin ladders, running along a given crystallographic axis, depending on the material [45]. Therefore they represent physical realizations of specific low-D spin array models whose physical properties can be tested and compared with theoretical predictions. Very early theoretical work [46] had claimed that the spin diffusion constant for spin chains with Heisenberg-type antiferromagnetic (HAFM) coupling diverges for $S = ½$, implying that in this case the transport of energy is not diffusive but ballistic. More recent work invoking other approaches to investigate the flow of energy in such systems concluded that the thermal conductivity κ_s based on itinerant spin excitations, not exposed to extrinsic perturbations, is infinite [47]. The only excitations and thus carriers of energy in a HAFM spin-½ chain are spinons and the corresponding excitation spectrum is gapless. In a real material, for example an insulator, their motion is certainly hampered by lattice defects and possibly lattice excitations (phonons). From our measurements of the thermal conductivity of all the above-mentioned compounds below room temperature, it turned out that at intermediate temperatures, the thermal conduction via spinons along the chain or ladder directions is a significant component and corresponding analyses indicated very long mean free paths for these magnetic excitations, limited by spinon interactions with lattice defects [45].

In [46] it is also claimed that the diffusion constant for spin excitations does not diverge for $S > ½$ and therefore the energy transport is diffusive. We tested this prediction with measurements of $\kappa_s(T)$ on $AgVP_2S_6$, where the V-ions a form a linear array along the **a** axis of the monoclinic crystal structure, and thus can be regarded as spin chain compound with $S = 1$ [48]. In this case, the excitation spectrum is gapped. From our results we concluded that the mean free path of the magnon excitations is much shorter and the corresponding diffusion constant does not diverge but is rather rapidly reduced with decreasing temperature. This direct comparison confirms the characteristically different features of the energy transport of spin arrays with either $S = ½$ or $S = 1$.

After the year 2000, the number of experimental and theoretical studies probing, explaining and predicting the magnetic properties of spin-chain and -ladder compounds as those mentioned above has grown enormously. Theoretical studies mainly concentrated on aspects of quantum criticality and many-body effects. Experiments served to test predictions of model calculations or to explore new and unexpected features of these systems [49]. Particular attention was devoted to spin-1/2 antiferromagnets with a singlet ground state and boson-like S = 1 triplet excitations termed triplons, where the application of external magnetic fields is expected to lead to quantum phase transitions (QPT). For example, by closing the gap between the singlet and the triplet states, the triplons can undergo a Bose-Einstein-type condensation (BEC) at low temperatures and the BEC is reflected by a magnetic order of the spin component orthogonal to the applied-field direction. Other aspects worth studying are the influences of frustration and disorder on the ground state of the system.

Our group at ETH embarked on experimental investigations of Cu-containing compounds (S = ½) whose structures feature either chain- ($BaCu_2Si_2O_7$) or ladder-type ($BiCu_2PO_6$) subunits in the sense outlined above. The experiments employed methods of nuclear magnetic resonance (NMR) probing either ^{29}Si or ^{31}P nuclei, depending on the compound under investigation. With the introduction of maximum disorder without changing the overall structure by randomly replacing Si by Ge to obtain the random Heisenberg chain (RHC) system $BaCu_2SiGeO_7$, we succeeded to identify the theoretically predicted random-singlet (RS) state [50]. NMR is well suited to explore the low-energy dynamics of an RHC system, not accessible by standard neutron-scattering techniques. Experimental data revealing the details of the NMR spin-lattice relaxation allowed to extract the distribution of relaxation functions in a random Heisenberg-type spin chain directly. Taking into account additional information from magnetization measurements and quantum Monte-Carlo (QMC) calculations finally justified the experimentally-based conclusion that the RS state is indeed the ground state of such systems [51].

As regards $BiCu_2PO_6$, we first explored the possibility to directly observe impurity-induced magnetic features in $BiCu_{2-x}Zn_xPO_6$. Again probing static and dynamic features of the NMR response (line positions and -shapes, and relaxation data) obtained from measurements on single crystals, as well as invoking QMC calculations, the set goal was reached [52]. With essentially the same experimental approach we also attempted to map the [H,T] phase diagram of the pristine compound. In a first approach, the range of external magnetic fields up to 32 T could be mapped, including the field-induced quantum phase transition FI-QPT at approximately 21 T at zero temperature, at about the same time confirmed by other authors employing different experimental techniques [53]. The description of the structure of the magnetic order above this critical field demanded a lot of analysis and calculations employing the density matrix renormalisation group (DMRG) method. Finally we claimed that our data suggest that the magnetization process involves the formation of a soliton lattice [54]. Current efforts aim at unraveling the development of the magnetization pattern upon increasing the magnetic field up to above 40 T.

Dear Alex, I hope that this brief essay doesn't bore you. It is intended to illustrate that your discovery, together with Georg, of superconductivity in copper-oxides gave a big boost to the field of superconductivity in general and in details, but not exclusively. I believe that it also had a very beneficial impact on materials-driven solid-state physics in general. People were encouraged to try to synthesize new materials, often with 5 or 6 chemical elements as constituents and in unexplored compositions. For instance, the discovery of superconductivity in so-called Iron-pnictide compounds [55], currently at temperatures up to 55 K, initiated a similar gold-rush excitement as did your findings in 1986. For other materials it turned out that a number of them have the potential to serve as model materials for studying different aspects of condensed-matter physics. Thus the availability of new materials is certainly one of the reasons why this sector of physics that is briefly outlined above, flourished so much in the last 30 years.

Finally I wish you all the best concerning health, spirit and fun in your life beyond 90.

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