

# Chapter 4

## Alex and the Origin of High-Temperature Superconductivity

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### Introduction

The discovery of high-temperature superconductivity (HTSC) in the cuprates by Müller and Bednorz [1] was a remarkable event in science which radically and irreversibly changed the course of research in condensed matter physics and materials science. Their chemical complexity gave a shock to physicists who were accustomed to simple solids. Oxides have long been thought to be just insulating, but the demonstration of metallic oxides having exciting properties opened up a whole new field of research on the transport properties of oxides.

This discovery was led by Alex's intuition that HTSC must be found in the system with the strong Jahn-Teller (JT) effect resulting in strong electron-phonon ( $e$ - $p$ ) coupling [2]. The traditional search for the HTSC material followed the path by Matthias [3] and sought after compounds of  $d$ -band metals with high Fermi density of states as prescribed by the BCS theory [4]. But the JT effect does not occur in metals because of strong dielectric screening. The genius of Alex was to go after the oxides with low electron density where the JT effect survives, even though the low electron density does not favor HTSC in the conventional thinking. Although superconductivity was already observed in  $\text{LiTi}_2\text{O}_4$  [5] and  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$  [6], it was a total surprise that high values of  $T_C$  were observed in the cuprate compounds.

However, the nature of the parent phase that it is an antiferromagnetic (AFM) Mott insulator [7] led many toward very different paths than those initially conceived by Alex. Actually the history of the HTSC research is teaching us how to do,

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or not to do, research on hugely unknown subjects. So many theories were published even before experimental facts were established. For example, the failure of the electronic calculation based on the density functional theory (DFT) to account for the parent compound as a Mott insulator convinced many that strong electron correlation plays a major role in the HTSC mechanism. To formulate the theory the  $t$ - $J$  Hamiltonian derived from the one-band Hubbard model was used, assuming that doped holes reside on Cu ion [8]. However, it was soon discovered that holes were on oxygen, not on copper [9]. Then the Zhang-Rice model was quickly invented to justify the one-band model [10], even though the three-band model was more realistic [11]. Indeed the validity of the Zhang-Rice model is questionable when carriers are mobile at high levels of doping. Furthermore, the value of  $U$ , the on-site Coulomb repulsion, is not extremely large but only comparable to the band-width,  $W$ . Because the  $t$ - $J$  model is derived from the Hubbard model in the limit of  $U/t \rightarrow \infty$  [12], this fact casts serious doubt on resorting to the  $t$ - $J$  model instead of the full Hubbard model. Thus the one-band  $t$ - $J$  Hamiltonian stands precariously on the two thin layers of fragile assumptions. But the fact that the one-band  $t$ - $J$  model is a minimalist model which can be solved with reasonable approximations kept it in the mainstream of the theory community for a long time [13].

Even though Alex is not a theoretician, he saw through the unrealistic nature of the theories with the  $t$ - $J$  model, based upon the experimental facts and his vast experience. He maintained his firm conviction that only the lattice can provide such high transition temperatures, and the only role of the magnetic order is to prevent superconductivity from occurring. In the last three decades a large number of theories, many of which are based upon wild ideas and speculations, were proposed, and were forgotten. In the meantime abundance of new experimental facts became known, discrediting much of these theories. Through the course of events Alex has been the unyielding and unmoving center of force, and guided us all. After more than 30 years of research the true nature of HTSC is finally emerging as discussed here. They are beginning to indicate that Alex may have been right all along, although the battle has not yet been finally won.

## Lattice Effects

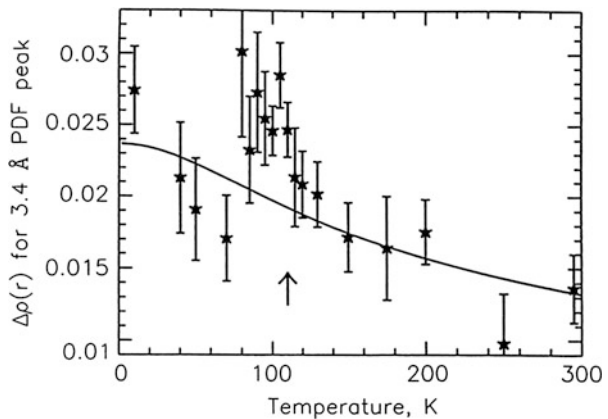
Soon after I became involved in the HTSC research I found that the local structure of HTSC oxides is significantly distorted from the average lattice structure. This discovery was made by using the method of the atomic pair-density function (PDF) determined by pulsed neutron scattering [14–16]. The PDF,

$$\rho_0 g(r) = \frac{1}{4\pi N r^2} \sum_{i,j} \langle \delta(r - |\mathbf{r}_i - \mathbf{r}_j|) \rangle, \quad (4.1)$$

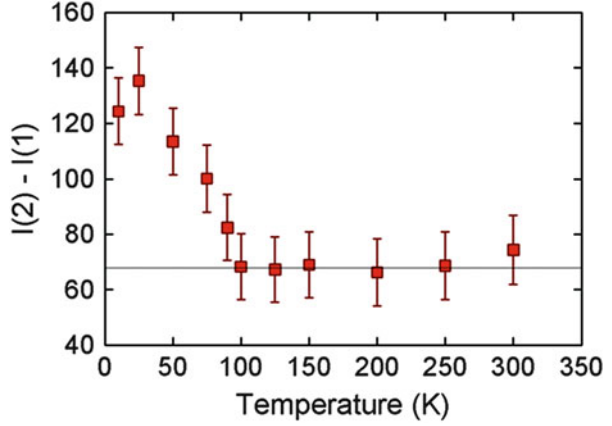
where  $\mathbf{r}_i$  is the position of the  $i$ th atom and  $\rho_0$  is the average atomic number density, describes the true distribution of distances between atoms without the assumption of lattice periodicity. The PDF can be determined from the normalized diffraction intensity, the structure function  $S(Q)$  where  $Q$  is the momentum transfer of scattering, through the Fourier-transformation [17, 18]. This method has been widely used in the structural analysis of liquids and glasses [17], but recently we started to apply it in the study of crystalline materials with defects or with local deviations from the ideal lattice [18]. The key to this new use of the old method was the availability of synchrotron based radiation sources, *i.e.* the synchrotron x-radiation sources and the pulsed neutron sources. For the Fourier-transformation to be accurate  $S(Q)$  has to be determined up to high values of  $Q$ . The availability of high energy probes from these synchrotron based sources made this method much more accurate than it used to be. The discovery of HTSC in the cuprate was timely for the development of the PDF method. We were able to demonstrate the power of this method through the study of the cuprates.

In particular we discovered that the portion of the PDF corresponding to the distance between the apical oxygen and the in-plane oxygen showed an anomalous temperature dependence as shown in Fig. 4.1 [19]. A possible explanation of this observed change was proposed later [20]. This implies intimate involvement of the lattice dynamics in the HTSC phenomenon of the cuprates. Similar observations of the lattice anomaly at or near  $T_C$  were reported also with ion-channeling [21, 22] and EXAFS [23, 24]. Encouraged by the numerous observations of the anomalous lattice behavior, Y. Bar-Yam, J. Mustre de Leon, A. R. Bishop and I organized a conference on the ‘‘Lattice Effects in High- $T_C$  Superconductors’’, in January 1992 at Santa Fe [25]. The meeting was a major success, and helped to establish the lattice effect as one of the major subjects of the HTSC research. This conference prompted various meetings on the lattice effect. The series of meetings on the stripes [26] by

**Fig. 4.1** Variation of the PDF peak height of  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  at 3.4 Å with temperature [19]. The arrow indicates  $T_C$  (=110 K). The line shows the normal behavior



**Fig. 4.2** Temperature dependence of the spectral weight of the inelastic neutron scattering intensity at  $Q = (3.25, 0, 0), I$  (51–55 meV)–I (56–68 meV) for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  [31].  $T_C = 93$  K for this sample



A. Bianconi are the most visible ones, still being continued today. The stripe conference series focuses not only on the lattice effects but also on the spatial separation of spin and charge, the representative of which is the spin-charge stripe structure [27, 28]. Various anomalous lattice effects in the cuprates observed up to 1995 were reviewed in [29].

Not only the structure, but the lattice dynamics was found to change anomalously across  $T_C$  [30] as shown in Fig. 4.2 [31]. Even though the results by themselves do not represent the “smoking gun” of the phonon mechanism, it is clear that the electron-phonon ( $e$ - $p$ ) coupling plays some prominent role in the HTSC phenomena. It is well known that the conventional  $e$ - $p$  coupling through the charge channel is rather weak in the cuprates, and cannot explain the high  $T_C$  [32]. We argued that the strong electron correlation could result in unconventional  $e$ - $p$  coupling, particularly through the charge transfer induced by the copper-oxygen half-breathing mode [33–35]. Using the vibronic model we showed that  $T_C$  exceeding 100 K can be achieved through the overscreened  $e$ - $p$  coupling [36].

The half-breathing mode modifies the Cu–O  $p$ - $d$  overlap, and the transfer integral,  $t$ . This induces charge hopping between Cu ions, which can be spin-dependent. In ferroelectric oxides, such as  $\text{PbTiO}_3$ , such charge transfer induced by lattice distortion produces electronic polarization which adds to the ionic polarization of the lattice to enhance ferroelectricity [37, 38]. However, in the cuprates the electronic polarization is anti-parallel to the ionic polarization, and they cancel each other [35]. Therefore the macroscopic dielectric  $e$ - $p$  coupling is weak, but the local coupling at the orbital level is very strong. The  $e$ - $p$  coupling through the orbital excitations potentially is a viable mechanism for promoting HTSC.

The strong  $e$ - $p$  coupling for the Cu–O half-breathing mode was then confirmed by the angle-resolved photoemission (ARPES) measurement by Lanzara [39, 40]. The observation was made at the nodal point near  $(\pi, \pi, 0)$ . Even though

the nodal point is not directly relevant to the  $d$ -wave superconductivity, the unexpected strength of the  $e$ - $p$  coupling is enough to wipe out doubts on the relevance of the  $e$ - $p$  coupling. The isotope effects [40] also provides unquestionable evidence of the lattice involvement. The isotope effects are not only large, but also are quite unusual and unconventional. For instance the magnetic screening length shows a larger isotope effects than  $T_C$  [41]. A possible explanation of the unconventional nature of superconductivity was provided by Bianconi [42] who argues that the  $e$ - $p$  coupling is greatly enhanced through the Fano-Feshbach resonance involving the spin-charge stripes which occurs near the Lifshitz transition. The unusual nature of the  $e$ - $p$  coupling in the cuprates is discussed in the article for the celebration of the 80th birthday of Alex [43].

## Iron-Pnictide Superconductors

The discovery of superconductivity in iron pnictides [44, 45] seriously affected the research on the cuprates. Because the highest value of  $T_C$  for the pnictides (56 K) is still lower than that of the cuprates by a factor of two, and the cuprate HTSC has the  $d$ -symmetry [46] whereas the pnictide has the  $s$ -symmetry [47], many are in the opinion that the cuprates and the pnictides are fundamentally different. However, recent developments are beginning to challenge this view as discussed below. If the cuprates and the Fe pnictides are to share the same origin of HTSC, the observations made for Fe-pnictides squarely challenge the common beliefs regarding the nature of HTSC, primarily deduced from the properties of the cuprates:

### **Belief #1: Mottness (Strongly Correlated Electron Physics) is Central to HTSC, so the Parent Phase has to be a Mott Insulator**

Whereas the parent phases of the Fe-pnictides are again antiferromagnetic [48] just as in the cuprates, they are metallic, and not the Mott insulator. The core level spectroscopy also suggest that electrons are not strongly correlated in the iron pnictides [49].

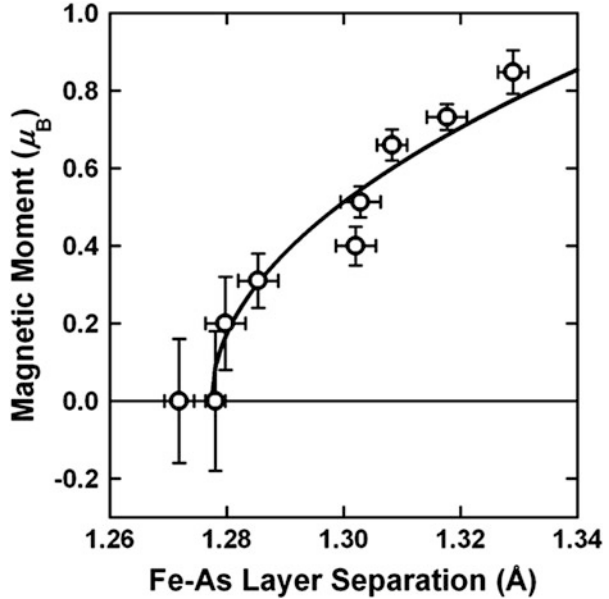
### **Belief #2: Doped Carriers Are Responsible for HTSC**

In the iron pnictides HTSC is observed over a wide ranges of doping concentration, and  $T_C$  is nearly flat within that range [50]. Furthermore HTSC is observed even with isovalent doping, such as P for As [51]. Therefore the role of dopant is not to provide charge carriers, but simply is to suppress the spin ordering. The suppression of spin ordering is not chemical, but physical through the lattice compression, and is described by the Landau theory using the lattice as a variable [52], as shown in Fig. 4.3 [53].

### **Belief #3: HTSC Occurs in a Spin $\frac{1}{2}$ Systems**

If spin fluctuations play an important role in HTSC, spin  $\frac{1}{2}$  systems are strongly preferred because of large spin fluctuations. But Fe pnictides are multi-orbital magnets with a spin larger than  $\frac{1}{2}$ , up to 1 [54].

**Fig. 4.3** Dependence of the magnetic moment on the Fe-As layer separation for CeFe(As,P)O (*open circles*) [51], and the fit by the Landau theory (*solid curve*) [53]

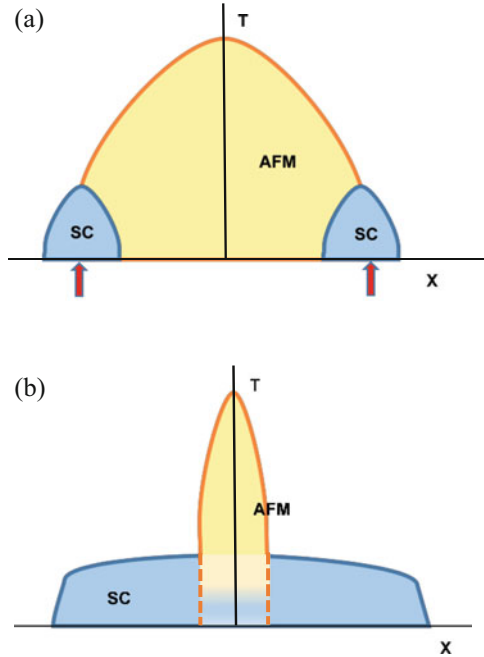


#### **Belief #4: HTSC Occurs in the Vicinity of the Quantum Critical Point**

Near the quantum critical point (QCP) large quantum fluctuations occur, facilitating pairing. Consequently HTSC can occur around the QCP, as illustrated in Fig. 4.4a. Indeed the superconductivity in some heavy fermion systems appears to be strongly linked to quantum fluctuations [55]. However, in Fe pnictides HTSC occurs not in the vicinity of QCP, but over wide ranges of composition, often far from the QCP. In these composition ranges  $T_C$  tends to be flat, rather than having a maximum at the QCP, as illustrated in Fig. 4.4b. In this case it is more likely that HTSC occurs independent of the AFM order, and the role of the AFM order is merely to suppress  $T_C$ .

P. W. Anderson summarized these beliefs, in particular those related to strong electron correlation, into six “dogmas” regarding the nature of HTSC [13]. However, most of them fail to account for the HTSC of Fe pnictides. Particularly the need of the Mott insulator clearly does not apply to Fe pnictides because they are not strongly correlated electron systems. Unless one insists that the mechanisms of HTSC in the cuprates and the Fe pnictides are totally distinct, we are forced to rethink the theories of HTSC. For instance, although it is most likely that the electron correlations play some prominent role in HTSC, Mottness may not be the only way the electron correlation affects the superconductivity. Being a Mott insulator and being a superconductor are two opposite extremes of the transport behavior. The idea of combining the two apparently diametrical ends to create a new state may be a cool idea, but could be a dangerous one.

**Fig. 4.4** Two scenarios for the phase diagram;  $x$  refers to doping, by either hole or electron. (a) QCP (red arrows) promotes SC, (b) AFM suppresses HTSC



## Origin of HTSC

### *Role of the Mottness and AMF Order*

In the context of the argument above some of the results on the electron-doped cuprates are quite revealing. In the initial report on the system,  $(\text{Nd, Pr})_{2-x}\text{Ce}_x\text{CuO}_4$ , the AFM order extends up to  $x = 0.14$ , whereas HTSC is observed for a narrow concentration range of  $0.14 < x < 0.18$  [56]. However, using a special technique to prevent oxygen from entering the apical site Brinkmann et al. were able to suppress the AFM order and extend the range of HTSC down to  $x = 0.04$  [57]. Thus the apparent asymmetry in the phase diagram between the hole-doped and electron-doped HTSC was removed. Furthermore, the recent results on  $\text{Pr}_{1.3-x}\text{La}_{0.7}\text{Ce}_x\text{CuO}_4$  (PLCCO) by ARPES [58] shows that  $T_C$  is almost independent of the doping concentration evaluated from the fermi surface determined by ARPES. Therefore the phase diagram of the cuprates now appears very similar to those of the pnictides as in Fig. 4.4b.

What this means is extremely important. The apparent independence of  $T_C$  from the doping concentration means that the concept of doping, which has been widely accepted without any doubt, may not be relevant to HTSC. The concept of doping presumes that the parent undoped phase is the Mott insulator, and doped charge carriers form a small Fermi surface and become superconducting. A corollary is that when the AFM order is suppressed by doping underlying AFM fluctuations

drive HTSC. However, the new data that  $T_C$  is almost independent of the doping concentration means that we cannot adopt the small Fermi surface picture which assumes strong AFM fluctuations, and we are forced to come back to the large Fermi surface picture where small changes in  $x$  have little effects on the Fermi density of states. In other words local AFM order is not the determining factor of the electronic behavior. We should then choose the scenario of Fig. 4.4b and conclude that the only role of the AFM phase is to suppress HTSC. If this is true, even the undoped cuprates should exhibit HTSC once AFM order is suppressed, for instance by pressure or isovalent substitution. An attempt to test this bold hypothesis is currently under way.

Another common feature of the HTSC in the cuprates and the pnictides is the magnetic resonance peak observed by inelastic neutron scattering [59–62]. The fact that this was observed for the pnictides means that the resonance peak has nothing to do with the Mottness, and could be explained by the itinerant electron picture [63–65].

### ***Possible Mechanism of HTSC***

At this moment we do not have a clear picture on the true origin of HTSC in the cuprates and the Fe pnictides. However, it is possible to speculate on the elements which would constitute the mechanism of HTSC common to both the cuprates and the pnictides.

#### **Spin-Channel $e$ - $p$ Coupling**

The charge-channel  $e$ - $p$  coupling is weak, particularly in the cuprates because of the cancellation of the ionic effect and the electronic charge-transfer effect [35]. However, the spin-channel coupling can be strong due to spin-dependent phonon-induced charge transfer [35]. A recent observation of the “forbidden” spin-dependent phonon, the acoustic phonon near the Bragg peak forbidden by symmetry, for the chalcogenides [66, 67] and the cuprates [68] could be a signature of the unconventional spin-channel  $e$ - $p$  coupling.

#### **Real-Space Polaronic Effect**

When the  $e$ - $p$  coupling is strong the combined  $e$ - $p$  excitations naturally become polaronic [69, 70]. In particular because the optical phonons responsible for the  $e$ - $p$  coupling [31, 35, 39] are relatively dispersionless, they can easily be localized and participate in forming polarons. Whereas strongly localized heavy polarons reduce  $T_C$ , in the strongly hybridized multi-band model [71] mobility can be restored through hybridization.



## Role of the Orbitals

Fe pnictide superconductors are multi-orbital systems with several  $d$ -orbitals coupled by the Hund coupling [50]. Therefore orbital ordering and excitation can play a role. A recent theory by Kontani [72, 73] proposes that if one goes beyond the conventional adiabatic Migdal-Eliashberg theory and considers the vertex correction, the renormalized Coulomb interaction becomes dependent on spin and orbit. Consequently the orbital excitations can strongly couple to spin, and could contribute to HTSC of Fe pnictides. Because orbitals are intimately coupled to the lattice, this spin-orbital coupling induces spin-channel  $e$ - $p$  coupling. In the cuprate, on the other hand, only the  $p$ - $d$   $\sigma$ -band is involved in the ground state. However, the  $d(z^2)$  orbital is close to the fermi level, and is likely to share the doped holes [74]. For this reason possible involvement of the apical oxygen has long been suspected [20, 24, 29, 75]. The  $d(z^2)$  orbital occupation is sensitive to the position of the apical oxygen, which has long been known to affect HTSC [76, 77].

## Alex and HTSC

The involvement of the  $d(z^2)$  orbital and the apical oxygen is precisely the Jahn-Teller effect in an extended sense, which Alex has long been advocating. After 30 years of intense research and many arduous detours, for instance to the Mott state, spin fluctuations and the  $t$ - $J$  model, we are back to where it all started. Incidentally, Sir Nevil Mott himself did not believe that the Mottness alone will lead to the answer, and believed in the polaron picture [78]. Alex, as well as Nevil Mott, knew that the truth comes out of the minorities, and the majority is always in the wrong.

This field is remarkable not only because of the novelty of the phenomenon and intensity of the competition, but also because of the advances in the experimental techniques it forced on the field. The HTSC phenomenon is so different from anything known so far, and demands much higher precision of measurement to study it. For instance the energy resolution of various methods of spectroscopy, including ARPES and STS, has improved tremendously in order to detect subtle features of the electronic states involved in the HTSC phenomenon. It provided a major drive for the improvement of the precision of the atomic pair-density function (PDF) technique we have been developing. It also prompted many theories to be put forth, although some of them are far-fetched. Furthermore, the discovery of HTSC drove many to study complex oxides and find various exciting properties. It established the research on the correlated electron systems as one of the main foci in condensed matter physics. The overall intellectual impact of the discovery of HTSC is immense, and we have to thank Alex for bringing about such a tremendous gift to us.

**Acknowledgment** At this happy occasion of congratulating Alex for his 90th birthday, I warmly thank him for leading the field and encouraging us even at the time the majority rushed to opposite extreme directions. Your conviction always inspired us and helped us to come up with more evidence in the right direction. I also thank my collaborators for their passion and endurance which led to valuable contributions, and the colleagues of the same mind in the field, who are too numerous to list here, for making the research so rewarding and enjoyable. This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Science and Engineering Division.

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