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Theory of the One- and Two-dimensional Electron Gas

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THEORY OF THE ONE- AND TWO-DIMENSIONAL ELECTRON GAS

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INTRODUCTION

The study of organic conductors and superconductors has always been intimately associated with low-dimensional physics: polymers or compounds involving planar molecules have anisotropic structure and associated anisotropic physical properties. Frequently, there is a temperature range over which the behavior is essentially one-dimensional, since thermal broadening is sufficient to mask any departure from the planar Fermi surface which is the hallmark of one-dimensionality. Below a crossover temperature T_x there may be a region of two-dimensionality before all aspects of the crystal structure finally make themselves felt. As a consequence, the past ten to fifteen years have seen an extensive development of the theory of the one-dimensional electron gas¹⁻⁴: by now we have a detailed understanding of a wide variety of mathematical models, and the analytical or numerical techniques are available for the study of new models, as they become relevant. It has steadily become clear that, although quasi one-dimensional systems are well-suited to the study of structural phase transitions, it is much easier to find superconductivity, and particularly high-temperature superconductivity, in higher dimensions. The arguments leading to this conclusion involve structure in time, momentum space and real space: in other words retardation, nesting and angular momentum.

In most models of superconductivity, pairing is produced by an attractive interaction which comes about because the charge carriers polarize their surroundings--the (relatively large) organic molecules, the lattice or the spin or charge density of the other carriers. Typically, the induced interaction is weaker than the Coulomb repulsion but it can be effective because the polarization takes time to relax. In three dimensions, this "retardation" leads to superconductivity but in one dimension it serves to enhance the formation of charge-density waves^{3,4}. In particular phonon exchange³ and Little's excitonic mechanism^{4,5} both suffer from this effect. The upshot is that, for retardation to lead to superconductivity, the crossover temperature T_x should be greater than the Debye energy of the phonons or the analogous characteristic energy of other modes responsible for the effective interaction. In this sense, superconductivity always is a two- or three-dimensional phenomenon. (This is quite apart from the fact that there is no long-range order at finite temperatures in one dimension.)

The charge- or spin-density wave states which compete with superconductivity have a characteristic wave vector q which should span a significant portion of the Fermi surface in order to provoke an electronic instability; the Fermi surface should be "nested." This condition is relatively easy to satisfy for the nearly flat Fermi surface of a quasi one-dimensional system but otherwise is quite exceptional. Thus higher dimensionality gives superconductivity a considerable edge in the competition with structural phase transitions.

Finally there is the possibility of new mechanisms of superconductivity or new pairing states when the constrictions of one-dimensional motion are removed. One example is the exchange of spin-density wave fluctuations, which may be relevant for organic superconductors⁶. The effective interaction oscillates in real space, and the motion of two electrons must have a specific impact parameter in order to take advantage of the regions of attraction. This may be accomplished by pairing electrons or holes on different organic stacks or by constructing a pair state with finite angular momentum⁶. Neither possibility is available in one-dimension. All of these considerations may come together when the exchange of soft modes related to a nearby structural phase transition produce a retarded interaction which leads to pairing with a finite impact parameter. This is the case for spin fluctuations in organic superconductors⁶.

Given all of this, it is hardly surprising that the organic superconductors⁷ are much less anisotropic than the previously studied organic conductors, and that the ET compounds with a T_c of 8K are more two-dimensional than the TMSF compounds for which T_c is about 1K. This situation alone makes it clear that, in the future, the two-dimensional electron gas will receive much more attention from theorists. However, the recent discovery of high T_c superconductivity in oxides, with their quasi two-dimensional CuO_2 planes, guarantees that this will be the case.

In line with these remarks, the remainder of these lectures will be concerned with two problems (a) the competition between $2k_F$ and $4k_F$ charge-density waves in a one-dimensional electron gas and (b) a two-dimensional model of high T_c superconductivity in the oxides.

$2k_F$ and $4k_F$ CHARGE-DENSITY WAVES

Over the past ten years, there has been an extensive investigation of materials which show $4k_F$ charge-density waves, sometimes in competition with the more conventional variety which have wave vector $2k_F$. This work is surveyed in the lectures of Dr. Pouget at this workshop⁸. There is general agreement that $2k_F$ charge-density waves are a consequence of a Fermi surface instability, a weak coupling effect, whereas $4k_F$ charge-density waves are essentially the first Bragg peak of a Wigner crystal and require strong coupling^{1,2,3}. However, there is relatively little understanding of the crossover from one to the other as the coupling constants or other parameters of the system are changed. This is not a purely academic question: some systems, such as TTF-TCNQ display both effects— $4k_F$ appears to dominate at high temperature, but $2k_F$ is the first to condense into an ordered state⁸. Clearly, an understanding of the competition between the two phenomena is essential for a full description of the phase diagram of such materials.

In order to study this problem⁹, first imagine an extended Hubbard model with transfer integral t , on-site interaction U , and interactions V_1 and V_2 between holes on first and second neighboring sites. As a first step, we shall consider the $U \rightarrow \infty$ limit, which essentially removes the spin degrees of freedom from active consideration, since it is no longer possible to have two electrons on the same site¹. This is an advantage, since it

allows us to focus on the charge-density which is our immediate concern. As is well known¹, the resulting Hamiltonian can be rewritten as a spin chain

$$H = \sum_n \{ -t(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) + V_1 S_n^z S_{n+1}^z + V_2 S_n^z S_{n+2}^z \} \quad (1)$$

where the S_n are Pauli matrix vectors and, physically, a spin is up ($S^z = +1$) if site n is occupied or down ($S^z = -1$) if site n is unoccupied. Equation (1) describes a Heisenberg-Ising model with second neighbor interactions. In the realm of spin chains, the notation (J_{xy}, J_1, J_2) would be more appropriate for (t, V_1, V_2) , but the latter is a better reflection of the Hubbard model convention. The repulsive interactions V_1 and V_2 correspond to antiferromagnetic coupling, since they discourage occupancy of the two sites that they couple—i.e. they discourage parallel spins. They compete since V_1 would like all even sites or all odd sites to have the same spin, whereas V_2 favors opposite spins. In the absence of a magnetic field, the mean magnetization is zero, which corresponds to an average of half an electron per site. The spin analog of a $4k_F$ charge-density wave is antiferromagnetic order (with periodicity two lattice sites) and a $2k_F$ charge-density wave is equivalent to (2,2) order whose extreme form is $(+,+, -,-, +,+, -,-, \dots)$ (with periodicity four lattice sites). Indeed, when $t = 0$, it is easy to show that the ground state of H is antiferromagnetic for $J_1 > 2J_2$ and a (2,2) phase for $J_1 < 2J_2$. The model can be solved exactly along another boundary¹⁰, $V_2 = 0$. There it is the usual Heisenberg-Ising model which has long-range antiferromagnetic order for $V_1 > t$ but power law correlation functions (as at a critical point) otherwise. The problem then is to work out the behavior when both t and V_2 are nonzero. We expect that for larger t , there is a region of values of V_1 and V_2 with critical point behavior. This is indeed the case. Although, so far as we know, there is no exact analytical solution, it is possible to work out the characteristic (fixed point) Hamiltonian which describes the critical region. But to match this to original Hamiltonian and hence to find critical exponents and phase boundaries, it is necessary to do a numerical calculation. The most appropriate method is to diagonalize finite chains of N sites and to use renormalization group arguments to take the $N \rightarrow \infty$ limit. A detailed description of this technique and of the method of exploring the ordered phases is given in reference 9. The important conclusion of the $4k_F - 2k_F$ crossover is that there are two kinds of $4k_F$ order. At small t , $2k_F$ and $4k_F$ density waves have their maximum amplitude on the lattice sites. But there may also be a shift of phase by π so that the peak lies on a bond, midway between two sites. What happens is that a region of $4k_F$ bond order separates the $2k_F$ site-ordered phase from the $4k_F$ site-ordered phase and from the critical region. The $4k_F$ bond order extends into the $2k_F$ phase where they co-exist until the latter finally dominates at $t = 0$.

There is a natural physical interpretation of this picture, which is easy to visualize by drawing the electron lattices as shown in reference 3. A direct transition from $4k_F$ to $2k_F$ site order would require a uniform shift of half of the electrons, all even ones or all odd ones. What actually happens as t decreases is that a bond $4k_F$ charge-density wave is unstable to a (period doubling) dimerization which produces the $2k_F$ site order.

HIGH T_C SUPERCONDUCTIVITY IN OXIDES

The discovery of high T_C superconductivity in doped La_2CuO_4 , $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and related compounds¹¹⁻¹³ has provoked an unprecedented burst of research into the properties of these materials and a search for new materials of even higher transition temperature. They are relatively easy to make (although good samples are not so easy to come by) and laboratories

all over the world are equipped to make some kind of measurement of relevance for superconductivity.

For theorists, the challenge is to understand the mechanisms--the source of the attractive interaction and the details of pairing with such a large binding energy. BCS theory¹⁴ is well-established as a good description of superconductivity in virtually all metals. Over the years, this has come to mean two things: (a) pairing is produced by electron-phonon coupling (b) the BCS approximation to many-body theory, in which the electrons condense at T_c into pairs with zero total momentum, is a valid description. Although (a) has been questioned on a number of occasions, it has always proven to be relevant for metals. At the present time, there is reason to believe that another mechanism may be appropriate for organic superconductors⁶ and heavy fermion superconductors¹⁵, but this has yet to be established to the satisfaction of all concerned. Another interesting system is liquid He³, which is a fermion superfluid very much like a superconductor except that the He³ atoms are uncharged¹⁶: phonon exchange is irrelevant because there is no lattice, and pairing is a consequence of the combined effect of the Van der Waals interaction between He³ atoms and the exchange of ferromagnetic spin fluctuations. The latter, which are important because He³ is an almost ferromagnetic Fermi liquid, have the distinctive consequence that the pairs are spin triplets rather than singlets as in metals.

The second element of BCS theory, Cooper pairing, is valid for liquid He³ but has been questioned for the high T_c oxides because Δ/E_F (the gap divided by the Fermi energy) is not exceptionally small. This ratio can be as low as 10^{-4} in metals and 10^{-2} in He³. However much larger gaps have been reported for the oxides¹⁷, although Δ/E_F should still be a good expansion parameter in most cases. If BCS theory proved to be inapplicable, the most likely alternative is that the attractive interaction is strong enough to produce real pairs (with negative energy) at a relatively high temperature. The pairs are bosons which undergo a separate superfluid transition at T_c . As we shall see, this could happen at sufficiently low carrier density in the oxides.

The common feature of doped La_2CuO_4 and $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ are quasi one-dimensional CuO_2 planes, shown in Fig. 1. Experiments suggest that the superconductivity is mainly associated with these planes and that the surroundings mainly serve to control the carrier density. The simplest representation is to start with a basic state (or vacuum) consisting of Cu^+ , with all 3d states occupied and O^{2-} which has filled 2p states. The number of holes per unit cell will be denoted by $1 + \delta$, where δ is determined by doping, oxygen defects and the charge states or band structure of the environment. We shall mainly be concerned with doped La_2CuO_4 . Simple valence arguments suggest that the undoped material contains La^{3+} , Cu^{2+} and O^{2-} so that $\delta = 0$. This picture is much too simple to describe what actually happens, but it is a good starting point for estimating the carrier density. The Hamiltonian is assumed to be an extended Hubbard model with the following parameters¹⁸:

ϵ_p, ϵ_d are single site energies for O(2p) and Cu(3d) holes

U_p, U_d are interaction energies of two holes on an O(2p) or Cu(3d) site

V is the interaction between holes on neighboring Cu, O sites

t is the hopping parameter characterizing hybridization of the $\text{Cu}(d_{x^2-y^2})$ and $\text{O}(2p_x)$ or $\text{O}(2p_y)$ orbitals.

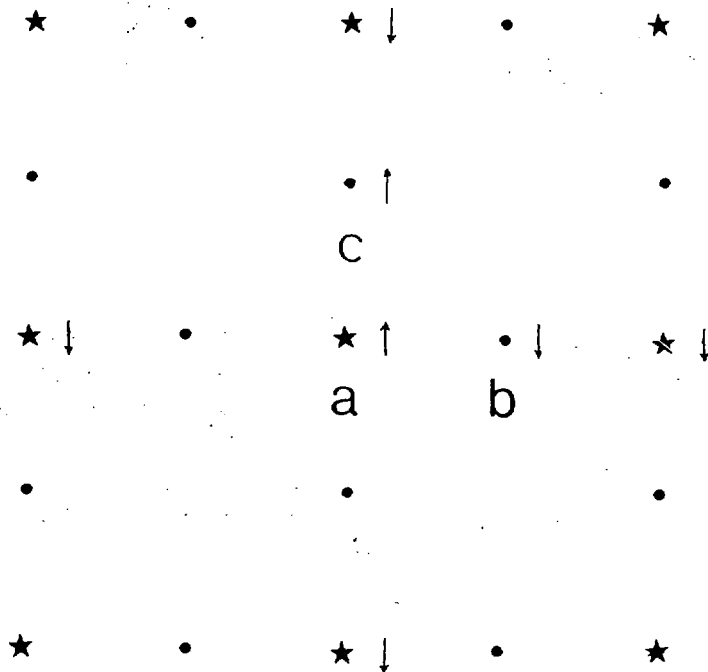


Fig. 1 The structure of the CuO_2 planes. The stars indicate Cu sites and the dots oxygen sites. The particular configuration of spins is discussed in the text.

The values of the parameters are not well-known but to be specific, it will be assumed¹⁸ that $t = 1.3 - 1.5$ eV, $\epsilon \equiv \epsilon_p - \epsilon_d = 1$ eV, $U_p = 1-3$ eV, $U_d = 5-6$ eV and $V = 1-2$ eV.

Consider first the case $\delta = 0$. When $t = 0$, there is exactly one hole per Cu site (Cu^{2+}) since $\epsilon > 0$. The holes move by hopping via an oxygen atom with an effective hopping parameter $t_d \equiv t^2/(\epsilon+V)$ to give an effective Cu Hubbard model:

$$H_d = t_d \sum_{(i,j)} d_{i\sigma}^+ d_{j\sigma} + U_d \sum_i d_{i\sigma}^+ d_{i+} d_{i-} d_{i-} \quad (2)$$

where $d_{i\sigma}^+$ creates a hole with spins $\sigma = \pm$ in the $d_{x^2-y^2}$ level of cell i , and the sum over (i,j) is restricted to neighboring cells. There have been several studies of this model, the most recent a Monte Carlo calculation by Hirsch¹⁹. The ground state is an antiferromagnetic insulator but at finite temperature the system has long range order only if there is weak hopping between the planes. At $|t_d/U_d|$ increases, the holes delocalize and their effective moment is diminished. Recently it has been established^{20,21} that there is indeed antiferromagnetic order below 250K in $\text{La}_2\text{CuO}_{4-y}$, which lends support to the essential correctness of the approach²².

The band picture regards hopping as the most important process: three sites per cell lead to three bands of which the lowest is half-filled when $\delta = 0$. The system is then susceptible to electronically-driven structural phase transitions, particularly an oxygen breathing mode, which is the most

effective for varying the value of t . This is an essential ingredient in obtaining high T_c by phonon exchange²³. Experimentally, a number of structural phase transitions have been documented but none so far has been clearly attributable to interaction with the conduction electrons, and there is little support so far for this aspect of the phonon exchange mechanism. Indeed, the existence of antiferromagnetic order may be regarded as evidence to the contrary. With repulsive interactions, the charge-density wave related to the structural distortion has its maximum amplitude on the bonds rather than the sites⁴, as described in Sec. 2, and the situation is qualitatively like a spin-Peierls transition⁴, as described in the Coulon's lectures²⁴. But this evidently has been preempted by antiferromagnetism, which might be expected because the exchange integral, which should be a few hundred degrees, is smaller than the Debye energy of the oxygen modes²⁴. An oxygen breathing mode does not modulate t_d so, in the strong-coupling picture which leads to Eq. (2), it is more natural to rephrase the whole argument in terms of Cu phonons²⁵--but the conclusion is the same.

Now suppose that holes are added by doping with Sr or Ba. They will go into $2p_x$ or $2p_y$ states of the oxygen, provided the site energy ϵ_p lies within the gap in the spectrum of H_d . For $t = 0$ and a finite range of values of t , this requires that $U_d > \epsilon + 2V$, since the gap is proportional to U_d for intermediate and strong coupling. Experiments on doped La_2CuO_4 strongly support this picture. In particular, an X-ray absorption edge study²⁶ of $\text{La}_{2-x}(\text{Ba},\text{Sr})_x\text{CuO}_4$ found that the copper remained in the Cu^{2+} state for all x in the range $0 < x < 0.3$ and that changes in the La L_3 edge with doping were consistent with variation of the oxygen charge. Thus, the charge carriers which are responsible for superconductivity are holes in the $O(2p)$ states produced by doping. Their density and Fermi energy are quite low.

The $O(2p)$ holes move by hopping via the Cu sites, and it is straightforward to obtain the effective Hamiltonian to second order in t , treating the $\text{Cu}(3d)$ holes exactly. Assuming translational invariance on the Cu, it is found that¹⁸:

$$H_p = \sum_{k\sigma} t^2 e_{\underline{k}} G_{\underline{k}}(\epsilon + 2V - \mu) b_{\underline{k}\sigma}^+ b_{\underline{k}\sigma} \quad (3)$$

where $G_{\underline{k}}(\omega)$ is the one-particle Green's function and μ the chemical potential of the $\text{Cu}(3d)$ holes,

$$e_{\underline{k}} = 2(2 - \cos k_x a - \cos k_y a) \quad (4)$$

a is the lattice spacing and $b_{\underline{k}\sigma}$ annihilates a hole in an oxygen state of momentum \underline{k} and spin σ . The energy spectrum is partly determined by the shape of $G_{\underline{k}}$. For weak to intermediate coupling, $G_{\underline{k}}$ varies rapidly near the ideal Fermi surface S_F on a scale determined by the mean free path or localization length. In that case, $e_{\underline{k}} G_{\underline{k}}$ has a minimum just inside S_F . For large U_d , $G_{\underline{k}}$ varies slowly with \underline{k} and the minimum in the spectrum occurs at the zone corners, where $e_{\underline{k}}$ is a maximum. Experiment supports the intermediate coupling picture¹⁸, although this may change as more evidence comes in.

Within this picture, there is a natural way of obtaining a strong attraction between $O(2p)$ holes. In perturbation theory in t , there is an attractive exchange interaction between opposite spins, coming from a process in which one spin hops onto the other's site and one or other hops back¹. The interaction is $O(t^{2n})$ where n is the number of bonds between the two sites. The essential point is that the $O(2p) - \text{Cu}(3d)$ exchange

J_{CO} (which is $O(t^2)$) is much stronger than the Cu(3d) - Cu(3d) exchange J_{CC} (which is $O(t^4)$). The first consequence is that antiferromagnetism is rapidly destroyed by doping. The $O(2p)$ holes act as a set of annealed, random, strongly coupled magnetic impurities, and remove the long-range order when $J_{CO}\delta \geq J_{CC}$. Consider now the situation shown in Fig. 1. The $O(2p)$ holes at b and c may be exchanged by first interchanging the holes at a and b and then interchanging those at a and c (which now have opposite spin). The net result is that the spins at b and c have been reversed by exchange of a spin-1 excitation on the Cu, which is a local, strong-coupling version of spin-density wave exchange^{6,15}. It is important to note that this process did not require two holes to be on the same $O(2p)$ site, so there is no restriction in the motion of $O(2p)$ holes which may increase their energy.

It is important to notice that each oxygen hole is coupled to six neighbors by this exchange process which helps to obtain¹⁸ attractive interaction of the order of 1 eV. Then the form of pair wave function depends upon the value of U_p and the location of the Fermi surface¹⁸. Large values of U_p favor d-state pairing¹⁵ for which the gap Δ_k is given by

$$\Delta_k = \Delta_0(\cos k_x a - \cos k_y a) \quad (5)$$

But if U_p is smaller than about 1 eV, a uniform gap, independent of k , would give a lower energy. This is particularly true when U_d is large and the Fermi surface is at the zone corners. (Note that the extended s-state, for which $\Delta'_k \sim (\cos k_x a + \cos k_y a)$ is not very different from a uniform gap, since Δ'_k is constant on the Fermi surface). Another possibility is to construct a wave function pairing holes on different oxygen sites within a cell, to give a uniform gap. In any event, it is not difficult to choose the parameters of the model to obtain transition temperatures of the required order of magnitude for the high T_C oxides.

The nature of the many-body state depends on the density of carriers. In two dimensions even for a spectrum $\epsilon_k \sim k^2$, the density of states is constant for small k , and an arbitrarily weak interaction will produce a bound state of two holes²⁷. As the density increases, the ground state should first be a Bose condensate of these "real" pairs²⁷ and ultimately will cross over to the BCS state¹⁴ which is a condensate of Cooper pairs. The presence of real pairs should be clearly visible in the normal state properties--for example, the susceptibility should be activated. Up to the present, the most extensive investigations have been concerned with samples displaying the highest T_C : $La_{2-x}Sr_xCuO_4$ with $x \sim .15$ and $YBa_2Cu_3O_{7-y}$ with y close to zero. In these cases, the evidence favors the BCS picture. However, it would be interesting to study materials with lower carrier density (smaller x or larger y) to seek evidence of crossover to "real" pairing.

In summary, the model of high T_C superconductors, presented here, has a low density low Fermi energy system of charge carriers, which are holes with most of their weight on the oxygen sites. The high transition temperature is a consequence of a strong coupling produced by exchange of spin fluctuations, largely on Cu atoms. The strength of this coupling is usually small^{6,15}. But the copper oxide superconductors are exceptional because of their electronic structure ($\epsilon_p \sim \epsilon_d$) and lattice structure (oxygen on bridge sites between the Cu atoms).

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