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## Towards room-temperature superconductivity

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By taking advantage of a stability criterion established recently, the critical temperature  $T_c$  is reckoned with help of the microscopic parameters, characterising the normal and superconducting electrons, namely the independent-electron band structure and a *repulsive* two-electron force. The emphasis is laid on the sharp  $T_c$  dependence upon electron concentration and inter-electron coupling, which might offer a practical route toward higher  $T_c$  values and help to understand why high- $T_c$ compounds exhibit such remarkable properties.

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The BCS theory [1], despite its impressive success, does 8 <sup>9</sup> not enable one to predict superconductivity occurring in <sup>10</sup> any metallic compound. Such a drawback ensues from <sup>11</sup> an *attractive* interaction, assumed to couple electrons to-<sup>12</sup> gether, which is not only at loggerheads with the sign of <sup>13</sup> the Coulomb repulsion but in addition leads to questionable conclusions to be discussed below. Therefore this 14 work is intended at investigating the  $T_c$  dependence upon 15 the parameters, characterising the motion of electrons 16 correlated together through a *repulsive* force, within the 17 framework of a two-fluid picture [2] to be recalled below. 18 The conduction electrons comprise bound and inde-19 <sup>20</sup> pendent electrons, in respective temperature dependent concentration  $c_s(T), c_n(T)$ , such that  $c_0 = c_s(T) + c_n(T)$ 21 with  $c_0$  being the total concentration of conduction elec-22 trons. They are organized, respectively, as a many bound 23 <sup>24</sup> electron[3] (MBE) state, characterised by its chemical <sup>25</sup> potential  $\mu(c_s)$ , and a Fermi gas [4] of Fermi energy  $_{26} E_F(T, c_n)$ . The Helmholz free energy of independent <sup>27</sup> electrons per unit volume  $F_n$  and  $E_F$  on the one hand, 28 and the eigenenergy per unit volume  $\mathcal{E}_s(c_s)$  of bound <sup>29</sup> electrons and  $\mu$  on the other hand, are related [4, 5], re-<sup>30</sup> spectively, by  $E_F = \frac{\partial F_n}{\partial c_n}$  and  $\mu = \frac{\partial \mathcal{E}_s}{\partial c_s}$ . Then a stable <sup>31</sup> equilibrium is conditioned[6] by Gibbs and Duhem's law

$$E_F(T, c_n(T)) = \mu(c_s(T)) \quad , \tag{1}$$

<sup>32</sup> which expresses[5] that the total free energy  $F_n + \mathcal{E}_s$  is <sup>33</sup> minimum provided  $\frac{\partial E_F}{\partial c_n} + \frac{\partial \mu}{\partial c_s} > 0$ . Noteworthy is that <sup>34</sup>  $\frac{\partial \mu}{\partial c_s} < 0$  has been shown to be a prerequisite for per-<sup>35</sup> sistent currents[6], thermal equilibrium[3], the Josephson <sup>36</sup> effect[7] and a stable[2] superconducting phase. Likewise, <sup>37</sup> Eq.(1) reads[3, 6] for  $T = T_c$ 

$$E_F(T_c, c_0) = \mu(c_s = 0) = \varepsilon_b/2$$
 , (2)

<sup>38</sup> with  $\varepsilon_b$  being the energy of a bound electron pair[3]. Note <sup>39</sup> that Eqs.(1,2) are consistent with the superconducting <sup>40</sup> transition being of second order[5], whereas it has been <sup>41</sup> shown[3] to be of first order at  $T < T_c \ (\Rightarrow E_F(T, c_0 - c_s) \neq \mu(c_s))$ , if the sample is flown through by a finite <sup>43</sup> current. The binding energy[3] of the superconducting state  $E_b(T < T_c)$  has been worked out as

$$E_b(T) = \int_T^{T_c} \left( C_s(u) - C_n(u) \right) du$$

<sup>44</sup> with  $C_s(T), C_n(T)$  being the electronic specific heat of a <sup>45</sup> superconductor, flown through by a vanishing current[3] <sup>46</sup> and that of a degenerate Fermi gas[4]. A stable phase <sup>47</sup> ( $\Rightarrow E_b > 0$ ) requires  $C_s(T_c) > C_n(T_c)$ , which can be <sup>48</sup> secured[2] only by fulfilling the following condition

$$\frac{\partial E_F}{\partial c_n}(T_c, c_0) = -\frac{\partial \mu}{\partial c_s}(0), \quad \rho'(E_F(T_c, c_0)) > 0 \quad , \quad (3)$$

<sup>49</sup> with  $\rho(\epsilon)$ ,  $\epsilon$  being the independent electron density of <sup>50</sup> states and one-electron energy, respectively, and  $\rho' = \frac{d\rho}{d\epsilon}$ . <sup>51</sup> Since the remaining analysis relies heavily on <sup>52</sup> Eqs.(2,3), explicit expressions are needed for <sup>53</sup>  $E_F(T_c, c_0), \frac{\partial E_F}{\partial c_n}(T_c, c_0), \varepsilon_b, \frac{\partial \mu}{\partial c_s}(0)$ . Because the in-<sup>54</sup> dependent electrons make up a degenerate Fermi gas <sup>55</sup> ( $\Rightarrow T << E_F/k_B$  with  $k_B$  being Boltzmann's constant), <sup>56</sup> applying the Sommerfeld expansion[4] up to  $T^2$  yields

$$E_F(T_c, c_0) = E_F(0, c_0) - \frac{\rho'}{\rho} \frac{(\pi k_B T_c)^2}{6} \\ \frac{\partial E_F}{\partial c_n}(T_c, c_0) = \left(\rho + \rho'' \frac{(\pi k_B T_c)^2}{6}\right)^{-1} , \qquad (4)$$

<sup>57</sup> with  $\rho = \rho(E_F(0,c_0)), \rho' = \frac{d\rho}{dE_F}(E_F(0,c_0)), \rho'' =$ <sup>58</sup>  $\frac{d^2\rho}{dE_F^2}(E_F(0,c_0))$ . As for  $\varepsilon_b, \frac{\partial\mu}{\partial c_s}(0)$ , a truncated Hubbard <sup>59</sup> Hamiltonian  $H_K$ , introduced previously[8–10], will be <sup>60</sup> used. The main features of the calculation[3] are sum-<sup>61</sup> marised below for self-containedness.

The independent electron motion is described by the Hamiltonian  $H_d$ 

$$H_d = \sum_{k,\sigma} \epsilon(k) c^+_{k,\sigma} c_{k,\sigma}$$

 $\epsilon(k), k$  are the one-electron energy  $(\epsilon(k) = \epsilon(-k))$  and a vector of the Brillouin zone, respectively,  $\sigma = \pm$  is the

electron spin and the sum over k is to be carried out over  $_{93}$ the whole Brillouin zone. Then  $c_{k,\sigma}^+$ ,  $c_{k,\sigma}$  are creation and 94 annihilation operators on the Bloch state  $|k, \sigma\rangle$ 

$$|k,\sigma\rangle = c^+_{k,\sigma} |0\rangle \quad , \quad |0\rangle = c_{k,\sigma} |k,\sigma\rangle \quad ,$$

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with  $|0\rangle$  being the no electron state. The Hamiltonian  $H_K$  reads then

$$H_K = H_d + \frac{U}{N} \sum_{k,k'} c^+_{k,+} c^+_{K-k,-} c_{K-k',-} c_{k',+} \quad ,$$

104 62 with N >> 1, U > 0 being the number of atomic sites, 105 63 making up the three-dimensional crystal, and the Hub-106 <sup>64</sup> bard constant, respectively. Note that the Hamiltonian 107 used by Cooper[11] is identical to  $H_{K=0}$ , but with U < 0.  $H_K$  sustains [3] a single bound pair eigenstate, the en-108

<sup>67</sup> ergy  $\varepsilon_b(K)$  of which is obtained by solving

$$\frac{1}{U} = \frac{1}{N} \sum_{k} \frac{1}{\varepsilon_b(K) - \varepsilon(K,k)} = \int_{-t_K}^{t_K} \frac{\rho_K(\varepsilon)}{\varepsilon_b(K) - \varepsilon} d\varepsilon. \quad (5) \quad (5)$$

 $\pm t_K$  are the upper and lower bounds of the two-electron band, i.e. the maximum and minimum of  $\varepsilon(K,k) =$ two-electron density of states, taken equal to

$$\rho_K(\varepsilon) = \frac{2}{\pi t_K} \sqrt{1 - \left(\frac{\varepsilon}{t_K}\right)^2} \quad . \label{eq:rho_K}$$

The dispersion curves  $\varepsilon_b(K)$  are plotted in Fig.1. <sup>120</sup> 68 <sup>69</sup> Though Eq.(5) is identical to the equation yielding the 121Cooper pair energy [11], their respective properties are 70 122 71 quite different : 123

- the data in Fig.1 have been calculated with U > 0, 72 rather than U < 0 favoured by Cooper[11] and 73 BCS[1], because, due to the inequality[3]  $U \frac{\partial \mu}{\partial c_s} <$ 74 0, choosing U < 0 entails  $\frac{\partial \mu}{\partial c_s} > 0$ , which has 75 been shown *not* to be consistent with persistent 76 currents[6], thermal equilibrium[3], the Josephson 77 effect[7] and occurance[2] of superconductivity. As 78 a further consequence of U > 0,  $\varepsilon_b(K)$  shows up 79 in the upper gap of the two-electron band struc-80 ture  $(\Rightarrow \varepsilon_b(K) > t_K)$  rather than in the lower 81 gap ( $\Rightarrow \varepsilon_b(K=0) < -t_K$ ) in case of the Cooper 82 pair[11]. Nevertheless the bound pair is thermo-83 dynamically stable, because, as a consequence of 84  $E_F(T_c, c_0) = \varepsilon_b(K)/2$  in Eq.(2), every one-electron 85 state of energy  $\langle \varepsilon_b(K)/2$  is actually occupied, so 86 that, due to Pauli's principle, a bound electron pair 87 cannot decay into two one-electron states of energy 88  $\epsilon(k) < E_F, \epsilon(K-k) < E_F;$ 89
- a remarkable feature in Fig.1 is that  $\varepsilon_b(K) \to t_K$ 90
- 91
- $U < t_K/2$  (accordingly, the dashed curve is no 132 being dealt with as a disposable parameter. 92

longer defined in Fig.1 for  $\frac{Ka}{\pi}$  < .13), in marked contrast with the opposite conclusion reached by Cooper[11], that there is a Cooper pair, even for  $U \to 0$ . This discrepancy results from the threedimensional Van Hove singularities, showing up at both two-electron band edges  $\rho_K (\varepsilon \to \pm t_K) \propto$  $\sqrt{t_K - |\varepsilon|}$ , unlike the two-electron density of states, used by Cooper[11] which is constant and thence displays no such singularity. Likewise the width of Cooper's two-electron band is equal to a Debye phonon energy  $2t_{K=0} = \omega_D \approx 30 meV \ll E_F \approx$ 3eV. Hence the resulting small concentration of superconducting electrons,  $\frac{c_s(T=0)}{c_0} \approx \frac{\omega_D}{E_F} \approx .01$ , entails that London's length should be at least 10 times larger than observed values [12–15];

• at last Cooper's assumption U < 0 implies  $\varepsilon_b/2 \neq$  $E_F(T_c)$ , which is typical of a first order transition but runs afoul at all measurements, proving conversely the superconducting transition to be of second order  $(\Rightarrow \varepsilon_b/2 = E_F(T_c))$  in accordance with Eq.(2)).

The bound pair of energy  $\varepsilon_b(K)$  turns, at finite concen-114  $\epsilon(k) + \epsilon(K-k)$  over k, whereas  $\rho_K(\varepsilon)$  is the corresponding <sup>115</sup> tration  $c_s$ , into a MBE state, characterised by  $\mu(c_s)$ . Its <sup>116</sup> properties have been calculated thanks to a variational <sup>117</sup> procedure[3], displaying several merits with respect to <sup>118</sup> that used by BCS[1]:

- it has been shown that  $\mu(0) = \varepsilon_b/2$ ;
  - the energy of the MBE state has been shown to be exact for  $|U| \to \infty$ ;
- an analytical expression has been worked out for  $\frac{\partial \mu}{\partial c_s}(K, c_s = 0)$  as :

$$\frac{\partial \mu}{\partial c_s}(K, c_s = 0) = -\frac{\int_{-t_K}^{t_K} \frac{\rho_K(\varepsilon)}{(\varepsilon_b(K) - \varepsilon)^3} d\varepsilon}{2\left(\int_{-t_K}^{t_K} \frac{\rho_K(\varepsilon)}{(\varepsilon_b(K) - \varepsilon)^2} d\varepsilon\right)^2} \quad . \tag{6}$$

The  $T_c$  dependence on  $c_0$  will be discussed inside a 124 <sup>125</sup> model for which  $\rho(\epsilon)$  is defined as

$$\rho(\epsilon) = \eta \sqrt{\epsilon - \epsilon_b} \Rightarrow c_0 = \frac{2}{3} \eta \left( E_F(0, c_0) - \epsilon_b \right)^{\frac{3}{2}} \quad , \quad (7)$$

<sup>126</sup> with  $\eta = \frac{\sqrt{2}m^{\frac{3}{2}}V}{\pi^{2}\hbar^{3}}$ , whereas  $\epsilon_{b}, m, V = 17\text{\AA}^{3}$  stand for <sup>127</sup> the bottom of the conduction band, electron mass and <sup>128</sup> volume of the unit-cell, respectively.

129 With help of Eq.(4), Eqs(2,3) can be recast into a 130 system of two equations

$$E_F(0, c_0) - \frac{\rho'}{\rho} \frac{(\pi k_B T_c)^2}{6} - \frac{\varepsilon_b(K)}{2} = 0 \left(\rho + \rho'' \frac{(\pi k_B T_c)^2}{6}\right)^{-1} + \frac{\partial \mu}{\partial c_s} (K, c_s = 0) = 0$$
(8)

for  $U \to t_K/2$ , so that there is no bound pair for 131 to be solved for the two unknowns  $c_0(T_c), t_K(T_c)$  with  $T_c$ 



FIG. 1. Dispersion curves of  $t_K$  as a dashed-dotted line and of  $\varepsilon_h(K)$  as solid, dashed and dotted lines, associated with various U values, respectively; those data have been obtained with  $t_K = t \cos{(Ka/2)}$ , where t, a are the one-electron band-156 width and the lattice parameter, respectively.

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159 To that end, starting values are assigned to  $U, t_K$ , 133 which gives access to  $\varepsilon_b(K), \frac{\partial \mu}{\partial c_s}(K, c_s = 0))$  and 160 134 161 thence to  $E_F(0, c_0), \epsilon_b$  and finally to  $c_0$ , owing to 162 Eqs.(2,3,7). Those values of  $c_0, t_K$  are then fed into 136 163 Eqs.(8) to launch a Newton procedure, yielding the so-164 <sup>138</sup> lutions  $c_0(T_c), t_K(T_c)$ . The results are presented in ta-165 ble I. Since we intend to apply this analysis to high- $T_c$ 139 <sup>140</sup> compounds [16], we have focused upon low concentrations  $_{141} c_0 < 0.2$ , which entails, in view of Eqs.(4,7), that  $\left| \frac{\partial \mu}{\partial c_s} \right|$ 142 takes a high value. This requires in turn  $\varepsilon_b(K) \to t_K$ <sup>143</sup> (see Eq.(6)) and thence [3]  $U \to \frac{t_K}{2}$ , in agreement with 144  $\frac{t_K}{U} \approx 2$  in table I.

A remarkable property of the data in table I is that 167 Our purpose is to determine the  $|\delta c_0| < 10^{-3}, |\delta t_K| < 10^{-5}$  for  $\delta T_c \approx 400 K$ . This can be understood as follows : taking advantage of Eqs.(2,4,7)results into

$$\frac{2E_F(0,c_0)}{\varepsilon_b(K)} - 1 = \frac{\pi^2}{12} \left(\frac{k_B T_c}{\Delta(T_c)}\right)^2 \quad ,$$

146 indeed  $\delta c_0 = c_0(400K) - c_0(1K) \approx 10^{-3}$ , in agreement <sup>147</sup> with the data in table I. Such a result is significant in <sup>148</sup> two respects, regarding high- $T_c$  compounds, for which  $c_0$ can be varied over a wide range : 149

- because of  $\frac{dc_0}{dT_c} \approx 0$ , the one-electron band structure 181 band, which has two consequences : 150 can be regarded safely as  $c_0$  independent, which 151 enhances the usefulness of the above analysis; 152
- the large doping rate up to  $\approx 0.2$  is likely to give rise 184 153 to local fluctuations of  $c_0$ , which, in view of the ut- 185 154 most sensitivity of  $T_c$  with respect to  $c_0$ , will result 186 155

TABLE I. Solutions  $c_0(T_c), t_K(T_c), \Delta(T_c)$  ( $\Delta(T_c)$ ) \_  $E_F(0, c_0(T_c)) - \epsilon_b)$  of Eqs.(8); the unit for  $c_0$  is the number of conduction electrons per atomic site.

$T_c(K)$	$c_0$	$t_K(eV)$	$\Delta(eV)$	U(eV)
1	0.10215	6	1.1976	3.39
400	0.10225	5.9999	1.1984	3.39

$T_c(K)$	$c_0$	$t_K(eV)$	$\Delta(eV)$	U(eV)
1	0.14897	2	1.5402	1.04
400	0.14906	1.9999	1.5407	1.04

$T_c(K)$	$c_0$	$t_K(eV)$	$\Delta(eV)$	U(eV)
1	0.19158	4	1.8214	2.2
400	0.19167	3.9999	1.8219	2.2

into a heterogeneous sample, consisting in domains, displaying  $T_c$  varying from 0 up to a few hundreds of K. Thus the observed  $T_c$  turns out to be the upper bound of a broad distribution of  $T_c$  values, associated with superconducting regions, the set of which makes up a percolation path throughout the sample. However, if the daunting challenge of making samples, wherein local  $c_0$  fluctuations would be kept well below  $10^{-4}$ , could be overcome, this might pave the way to superconductivity at room temperature.

The  $T_c$  dependence upon U will be analysed with

$$\rho(\epsilon) = \frac{4}{\pi t} \sqrt{1 - \left(1 - \frac{\epsilon}{t}\right)^2}$$

unknowns  $c_0, t_K$  are barely sensitive to large variations of  $T_c$ , i.e.  ${}_{168} t_K(E_F, T_c), U(E_F, T_c)$  with  $E_F = E_F(T = 0, c_0)$ 169 and  $c_0 = \int_0^{E_F} \rho(\epsilon) d\epsilon$ . To that end, Eq.(3) will first be <sup>170</sup> solved for  $t_K$  by replacing  $\frac{\partial E_F}{\partial c_n}(T_c, c_0), \frac{\partial \mu}{\partial c_s}(0)$  by their <sup>171</sup> expressions given by Eqs.(4,6), while taking advantage <sup>172</sup> of Eq.(2). Then the obtained  $t_K$  value is fed into Eq.(5)  $_{173}$  to determine U. The results are presented in Fig.2.

 $\varepsilon_b(K)$  12  $(\Delta(I_c))$ 174 It can be noticed that there is no solution for  $c_0 > 1$ 175 Which, due to  $\frac{dt_K}{dT_c} \approx 0, \Delta(T_c) \approx 1 eV, T_c = 400K$ , yields 176 decrease and increase, respectively, with increasing  $c_0$ , so that Eq.(3) can no longer be fulfilled eventually. But the 177  $_{178}$  most significant feature is that  $\delta U$  is almost insensitive <sup>179</sup> to large  $T_c$  variation, except for  $E_F \to 0$ , i.e. for  $E_F$  close 180 to the Van Hove singularity, located at the bottom of the

> •  $c_0$  cannot be varied in most superconducting materials, apart from high- $T_c$  compounds, so that U is unlikely to be equal to  $U(c_0)$ , indicated in Fig.2. Conversely, since high- $T_c$  compounds allow for wide  $c_0$  variation,  $c_0$  can be tuned so that  $U = U(c_0)$ ;



FIG. 2. Plots of  $E_F(T_c, c_0), t_K(T_c, c_0), U(T_c, c_0)$  calculated for  $T_c = 1K$  and t = 3eV; the unit for  $c_0$  is the number of conduction electrons per atomic site;  $\delta f$  with  $f = E_F, t_K, U$  is defined as  $\delta f = \left| 1 - \frac{f(300K,c_0)}{f(1K,c_0)} \right|$ ; the scale is linear for  $E_F, t_K, U$  220 but logarithmic for  $\delta E_F, \delta t_K, \delta U$ .

• the only possibility for a non high- $T_c$  material to 187 225 turn superconducting is then offered at the bot-188 226 tom of the band, because  $\delta U$  becomes large due to 227 189  $\frac{\rho'}{\rho}(E_F \to 0) \propto \frac{1}{E_F}$  in Eq.(4). Such a conclusion, <sup>228</sup> that superconductivity was likely to occur in the <sup>229</sup> 190 191 230 vicinity of a Van Hove singularity in low- $T_c$  ma-192 231 terials, had already been reached[3] independently, 193 232 based on magnetostriction data. 194 233

It will be shown now that  $\rho(\epsilon)$ ,  $\rho_K(\varepsilon)$  cannot stem from <sup>235</sup> the same one-electron band. The proof is by contradiction. As a matter of fact  $\rho(\epsilon)$  should read in that case

$$\rho(\epsilon) = \frac{4}{\pi t} \sqrt{1 - \left(\frac{\epsilon}{t}\right)^2}$$

<sup>195</sup> Hence U > 0 entails, in view of Fig.1 and Eq.(2), that <sup>243</sup> <sup>196</sup> there is  $\frac{\varepsilon_b}{2} = E_F > 0$ , which implies  $\rho'(E_F) < 0$  in con-<sup>244</sup> tradiction with Eq.(3). Accordingly, since the two differ-197 <sup>198</sup> ent one-electron bands, defining respectively  $\rho(\epsilon), \rho_K(\varepsilon),$ <sup>199</sup> display a sizeable overlap, they should in addition belong 200 to different symmetry classes of the crystal point group, 201 so that superconductivity cannot be observed if there are 250 [16] N.P. Armitage, P. Fournier and R.L.  $_{202}$  only s-like electrons at  $E_F$  or if the point group reduces  $_{251}$ <sup>203</sup> to identity. Noteworthy is that those conclusions had <sup>252</sup> [17] <sup>204</sup> already been reached empirically by Matthias [17].

The critical temperature  $T_c$  has been calculated for 205 conduction electrons, coupled via a repulsive force, 206 within a model based on conditions, expressed in 207 Eqs.(2,3). Superconductivity occurring in conventional 208 materials has been shown to require  $E_F(T_c)$  being lo-209 210 cated near a Van Hove singularity, whereas a practical route towards still higher  $T_c$  values has been delineated 211 in high- $T_c$  compounds, provided the upper bound of lo-212 cal  $c_0$  fluctuations can be kept very low. The thermody-213  $_{214}$  namical criterions in Eqs.(2,3) unveil the close interplay between independent and bound electrons in giving rise 215 216 to superconductivity. At last, it should be noted that  $_{217}$  Eqs.(2,3) could be applied as well to any second order <sup>218</sup> transition, involving only conduction electrons, such as <sup>219</sup> ferromagnetism or antiferromagnetism.

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