

### Electron-hole liquid model for high- $T_c$ superconductors

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We show that the hole concentration dependence of  $T_c$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ - and  $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_y$ -type superconductors can be understood within a model electron-boson hole liquid that possesses a linear sound mode like the one in liquid  $^4\text{He}$ . The electronic specific heat follows a  $T^3$  law near  $T=0$ . The metal-insulator transition found in these compounds is proposed to be connected with the vanishing of the sound velocity at small carrier densities.

There is now strong theoretical<sup>1-6</sup> and experimental<sup>7-10</sup> evidence that the holes in the  $\text{CuO}_2$  plane are responsible for the high- $T_c$  superconductivity. These holes are mobile and can form  $^1S_0$  pairs due to some lattice mechanism still largely unknown. In addition there are mobile, unpaired electrons which reside in some other bands. There seems to be at least moderate evidence that the conduction in the  $c$  direction is due to the electrons and in the  $ab$  plane it is due to the holes.<sup>11</sup> Since the hole pairs are strongly bound<sup>12</sup> and hence small in size, we may treat them in first approximation as bosons<sup>1</sup> with charge  $Q=2e$ . There is some evidence<sup>13</sup> that such pairs exist even above  $T_c$ . For purposes of the present paper we need no model for their binding.

With this as the starting point, we work out next the coexistence of electrons and hole bosons with uniform densities  $n_e$  and  $n_B$ , respectively. The effective masses are correspondingly  $m_e$  and  $m_B$ . We use the plane waves for electrons. From here on the model is roughly the same as the one used previously for electron-hole liquid<sup>14</sup> or metallic hydrogen.<sup>15</sup> There one had two interpenetrating Fermi liquids, whereas here we have a mixture of a Fermi and a

Bose liquid. As a first approximation we replace the lattice with a neutralizing background.

Applying the hypernetted-chain (HNC) theory, one may write an expression for the total energy and derive variationally the Euler-Lagrange equations (EL) (Ref. 15) in  $k$  space in terms of the liquid-structure factors  $S_{ee}(k)$ ,  $S_{BB}(k)$ , and  $S_{eB}(k)$ . The spectrum of elementary excitations is then calculable from these structure factors. Since the details of the EL equations are otherwise largely irrelevant for the present discussion, we only quote the main results needed here from Ref. 15. The most important fact is the small- $k$  behavior of the structure factors

$$\begin{aligned} S_{ee}(k) &\sim \beta\gamma k, \\ S_{BB}(k) &\sim \beta k, \\ S_{eB}(k) &\sim \beta\sqrt{\gamma k}, \end{aligned} \tag{1}$$

where  $\gamma=4n_B/n_e$  and  $\beta$  a constant which can be determined by numerical solution of the EL equations. Applying now the Feynman wave function for excitations, one obtains with Eqs. (1) two branches at small  $k$ :

$$\epsilon_1(k) \sim \frac{\hbar^2 k^2}{2(m_e S_{ee} + m_B S_{BB})} = \frac{\hbar^2 k}{2\beta(\gamma m_e + m_B)}, \quad \epsilon_2(k) \sim \frac{\hbar^2 \beta}{a} \frac{\gamma m_e + m_B}{m_e m_B}, \quad S_{ee} S_{BB} - S_{eB}^2 \sim a k^3. \tag{2}$$

We identify the first one,  $\epsilon_1(k)$ , as a sound mode with velocity

$$u_e = \hbar/2\beta(m_B + \gamma m_e), \tag{3}$$

whereas the second mode,  $\epsilon_2(k)$ , is of plasma type. One should notice that the small- $k$  behavior of these modes is solely dictated by the Coulomb interaction and hence independent of HNC approximation. If we were to switch off the cross Coulomb force which is attractive, we would obtain two separate plasma modes, because one would then have  $S_{eB}(k)=0$  and a  $k^2$  behavior for  $S_{ee}$  and  $S_{BB}$ . The local charge neutrality would require the density parameter to have a value  $\gamma=2$ .

The linear mode here is similar to one proposed in the acoustic plasmon model of superconductivity.<sup>16</sup> In fact, it may be difficult to distinguish it from the present model experimentally. In the two-dimensional case we obtain two "acoustic" plasmons with small- $k$  behaviors,  $k^{1/2}$  and  $k^{3/2}$ , but their role for superconductivity<sup>17</sup> is not pursued further here.

With these preliminaries we are ready to work out the consequences of this model to the thermodynamics of high- $T_c$  superconductors. The spectrum that we obtained is exactly the one Landau<sup>18</sup> first proposed to explain the properties of liquid  $^4\text{He}$ . In what follows we neglect the plasma branch  $\epsilon_2(k)$  because it presumably lies higher than 1 eV. Clearly then the thermodynamics of our electron-boson system would be identical to the one in  $^4\text{He}$  but now without rotons. If there were no roton excitations in  $^4\text{He}$ , the condition for the  $\lambda$  transition would be that the density of phonons be equal to the density of He liquid at  $T=T'_\lambda$ , i.e.,

$$n_{\text{ph}}(T'_\lambda) = \frac{\hbar^2}{m_{\text{He}}} \frac{2\pi^2}{45} \frac{(k_B T'_\lambda)^4}{(\hbar u)^5} = n_{\text{He}}, \tag{4}$$

where  $u=238$  m/s is the velocity of the first sound. The temperature  $T'_\lambda$  turns out to be 7.3 K. The same equation (4) applies to the boson part of the electron system as well with  $T'_\lambda=T_c$ ,  $m_{\text{He}}=m_B$ , and  $n_{\text{He}}=n_B$ . By taking the ratio of Eq. (4) for the two systems, we obtain a scaling

equation

$$\frac{u_e}{u} = \left( \frac{T_c}{T_c'} \right)^{4/5} \left( \frac{n_{\text{He}}}{n_B(T_c)} \frac{m_{\text{He}}}{m_B} \right)^{1/5}. \quad (5)$$

Inserting the following<sup>19</sup> numbers relevant for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  ${}^4\text{He}$ ,  $T_c = 90$  K,  $n_B \approx 5 \times 10^{21} \text{ cm}^{-3}$ ,  $m_B \approx 20m_e$ , and  $n_{\text{He}} = 0.02185 \text{ \AA}^{-3}$ , one obtains for the electron sound velocity the value  $u_e \approx 5500$  m/s, a number slightly higher than the lattice velocity of sound.<sup>19</sup> Fortunately  $u_e$  is not very sensitive to  $m_B$  and  $n_B$  which are poorly known. So, for example, using  $m_B = 2m_e$  instead gives the value  $u_e = 8600$  m/s. To our knowledge there exists no direct experimental evidence of electron sound except some anomalies found in the attenuation experiments<sup>20,21</sup> near  $T_c$  in Y-Ba-Cu-O superconductors. If one approaches  $T_c$  from above, the attenuation can increase because the new sound mode becomes operative near  $T_c$ . Just like in liquid He, the electron sound mode exists even at  $T > T_c$  until all boson-hole pairs are broken. The behavior of the attenuation coefficient<sup>20,21</sup> is very different from the one in normal superconductors where one has a sudden drop at  $T_c$ .

The coupled Euler-Lagrange equations leading to behaviors given by Eqs. (1) have the property that at densities  $n_B < n_c$  the equations cease to have a solution. This is based on our past experience of the numerical solution of the EL equations. At the critical density  $n_c$  the sound velocity goes through zero. At densities higher than  $n_c$  the sound velocity is the linear function of  $n_B - n_c$ . Applying Eq. (4) again, now for the boson part of the mixture system at two different boson densities  $n_B(1)$  and  $n_B(2)$  and corresponding transition temperatures  $T_{c1}$  and  $T_{c2}$ , we obtain another scaling law

$$\begin{aligned} \frac{T_{c2}}{T_{c1}} &= \left( \frac{n_B(2)}{n_B(1)} \right)^{1/4} \left( \frac{n_B(2) - n_c}{n_B(1) - n_c} \right)^{5/4} \\ &\approx \left( \frac{n_B(2) - n_c}{n_B(1) - n_c} \right)^{3/2}. \end{aligned} \quad (6)$$

We have used this formula in Table I and Fig. 1 to compare our prediction with experimental data from Refs. 8 and 10. The trend is clearly correct in both cases. We can now interpret the density  $n_c$  to be the point where the metal-insulator transition takes place, because below  $n_c$

TABLE I. Superconducting transition temperature  $T_c$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ . The first column lists the  $x$  values and the third column corresponding experimental  $T_c$  values from Ref. 8. The middle column is the prediction of Eq. (6) assuming that  $n_B$  is proportional to  $x$ , as is explained in Ref. 8 with  $n_c \approx 0$ . Temperatures are in K.

$x$	$T_c$	Expt.
1.00	90	90
0.88	74	78
0.80	64	70
0.60	42	48
0.40	23	24

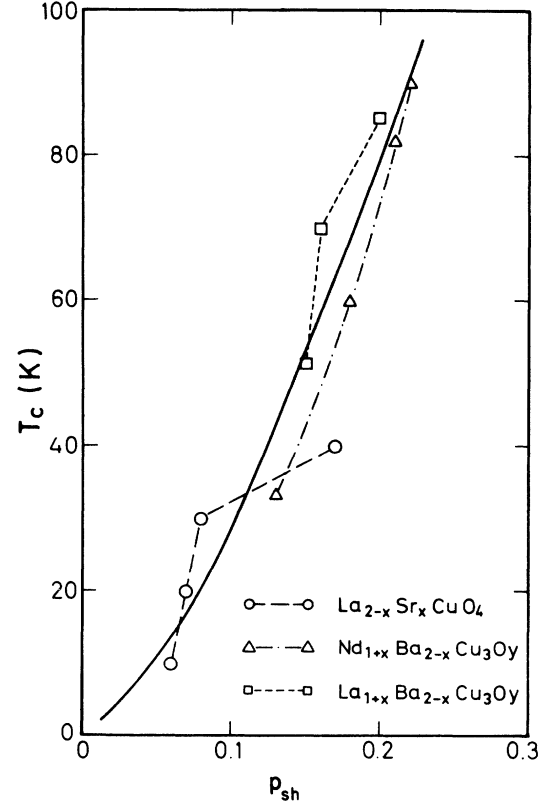


FIG. 1. Transition temperature  $T_c$  as a function of  $p_{\text{sh}}$ , the mobile sheet holes per Cu site. Experimental data for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ,  $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_y$ , and  $\text{La}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_y$  are from Ref. 10. The solid line shows the theoretical result calculated from Eq. (6) assuming  $n_B - n_c$  to be proportional to  $p_{\text{sh}}$ . The uppermost point of Tl compound is used to fix the scale of the solid line.

the electrons and bosons can no longer coexist. In an analogy with electron-hole liquid, the exciton gas is the stable phase below  $n_c$ . Since electrons are mobile mainly in the  $c$  direction and the hole bosons in the  $ab$  plane, it is likely that their excitonic state is nonmobile "heavy fermion" and hence this phase would be an insulator. One may indeed speculate that the large effective mass found in heavy-fermion systems may be of excitonic origin of this type. It would occur in the cases when the energy of electron-hole boson liquid never gets below the excitonic limit.

We can also understand the fact that in many of the compounds  $T_c$  gets lower after some maximum  $T_c$  when the carrier density is further increased. According to Ref. 10 this happens when their density parameter  $p_{\text{sh}} > 0.22$ . Here we have assumed  $n_B - n_c$  to be proportional to  $p_{\text{sh}}$ . This diminishing of  $T_c$  can be understood by the simple fact that for all electron systems the effect of Coulomb correlations get smaller when the density is increased, hence the electrons and bosons decouple. In this case the EL equations would predict  $S_{eB}(k) = 0$  and appearance of two separate plasma modes instead. Therefore at high carrier densities, one can have normal BCS-like superconductivity, if any. In particular, the high- $T_c$  superconductors are characterized by the existence of electron sound.

We should also point out that roles of electrons and holes can be interchanged. Therefore the recent findings of electrons forming pairs<sup>22</sup> can also be accommodated within the present model. The formula predicting the  $T_c$  when the velocity  $u_e$ , density  $n_B$ , and effective mass  $m_B$  are known reads, according to Eq. (4), as follows:

$$k_B T_c = \left[ \frac{45}{2\pi^2} \frac{m_B n_B}{\hbar^2} \right]^{1/4} (\hbar u_e)^{5/4}. \quad (7)$$

Next we want to point out that our model predicts the boson part of the electronic specific heat to be the cubic function of  $T$  like the one for the lattice part at low temperatures. This is again a direct consequence of the fact that our model is liquid He without the rotons. Except at the very lowest temperatures, many measurements<sup>19,23</sup> seem to give results in agreement with a  $T^3$  law. Assuming that the electron contribution gives a linear term, we can write the specific heat in the form

$$c = \gamma(0)T + (\beta^e + \beta^L)T^3, \quad (8)$$

where the ratio of the bosonic and lattice specific heats is given by

$$\frac{\beta^e}{\beta^L} = \frac{1}{3} \left( \frac{u_D}{u_e} \right)^3, \quad (9)$$

where  $u_D$  is the Debye velocity. The experimental results concerning the linear term<sup>24</sup> seem to be sample dependent, so we postpone any further discussion of the  $\gamma(0)$  term. We have analyzed the experimental data at temperatures from 3 to 10 K using the Debye model for the lattice specific heat. The result depends very much upon the Debye temperature  $\Theta_D$  and the set of experimental numbers used. The values of  $u_e$  for Y-Ba-Cu-O scatter from 2000 m/s for  $\Theta_D = 500$  K, Ref. 23, to 4000 m/s for  $\Theta_D = 400$  K, Refs. 19 and 25. The values are now smaller than the ones obtained from liquid He but the order of magnitude is the same. The results for Tl and Bi com-

pounds would be similar. The existence of this extra bosonic  $T^3$  term helps to increase  $\Theta_D$  to better fit<sup>19</sup> the lattice specific heat for  $T > T_c$ . Unfortunately we cannot make a more definite statement at this point until more accurate specific-heat data become available. To separate out the lattice contribution would require data for a single specimen from  $T \geq T_c$  down to  $T \approx 0$ . Near  $T_c$  a substantial fraction of pairs are broken. This will modify the  $T^3$  behavior and add a BCS correction to the electronic specific heat. This requires a more thorough treatment of the mixture system including the deviations from linearity in the excitation spectrum.

Finally we should point out that the electron component does not change the supercurrent which is then determined by the superfluid density of the boson component alone. This has been verified experimentally for  $^4\text{He}$ - $^3\text{He}$  mixtures<sup>26</sup> and hence by analogy should also work here.

As a conclusion we may say that the present model can serve at least as a qualitative framework for understanding some of the properties of the new superconductors. The thermodynamics of the bosonic part of high- $T_c$  compounds is identical with liquid  $^4\text{He}$  without rotons. The most important corrections would be due to the anisotropy which would give directional dependence of the electron sound. Furthermore, near  $T_c$  one should add the effect of BCS quasiparticles as well. According to the present model, the difference between a high- $T_c$  and a normal superconductor is the large gap in the former case and the high carrier density in the latter case. Also, in the normal superconductors no electron sound would exist.

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<sup>1</sup>A. S. Alexandrov, J. Ranninger, and S. Robaszkiewicz, Phys. Rev. B **33**, 4526 (1986).

<sup>2</sup>P. W. Anderson, B. Baskaran, Z. Zou, and T. Hsu, Phys. Rev. Lett. **58**, 2790 (1987).

<sup>3</sup>S. K. Kivelson, D. S. Rokhsar, and J. P. Sethna, Phys. Rev. B **35**, 8865 (1987).

<sup>4</sup>J. M. Wheatley, J. C. Hsu, and P. W. Anderson, Phys. Rev. B **37**, 5897 (1988).

<sup>5</sup>M. J. Rice and Y. R. Wang, Phys. Rev. B **37**, 5893 (1988).

<sup>6</sup>B. K. Chakraverty, D. Feinberg, Z. Hang, and M. Avignon, Solid State Commun. **64**, 1147 (1987).

<sup>7</sup>Y. Tokura, J. B. Torrance, T. C. Huang, and A. I. Nazzari, Phys. Rev. B **38**, 7156 (1988).

<sup>8</sup>J. M. Tranquada, S. M. Heald, A. R. Moodenbaugh, and Youwen Xu, Phys. Rev. B **38**, 8893 (1988).

<sup>9</sup>D. Y. Xing, M. Liu, and C. S. Ting, Phys. Rev. B **38**, 11992 (1988).

<sup>10</sup>M. W. Shafer, T. Penney, B. L. Olson, R. L. Greene, and R. H. Koch, Phys. Rev. B **39**, 2914 (1989).

<sup>11</sup>W. E. Pickett, Rev. Mod. Phys. **61**, 480 (1989).

<sup>12</sup>H. Ye, W. Lu, Z. Yu, X. Shen, B. Miao, Y. Cai, and Y. Qian, Phys. Rev. B **36**, 8802 (1987).

<sup>13</sup>W. W. Warren, Jr., R. E. Walstedt, G. F. Brennert, R. J. Cava, R. Tycko, R. F. Bell, and G. Dabbagh, Phys. Rev. Lett. **62**, 1193 (1989).

<sup>14</sup>T. Chakraborty and P. Pietilainen, Phys. Rev. Lett. **49**, 1034 (1982).

<sup>15</sup>T. Chakraborty, A. Kallio, L. J. Lantto, and P. Pietilainen, Phys. Rev. B **27**, 3061 (1983).

<sup>16</sup>H. Frohlich, J. Phys. C **1**, 544 (1968).

<sup>17</sup>J. Ruvalds, Phys. Rev. B **35**, 8869 (1987).

<sup>18</sup>David R. Tilley and John Tilley, *Superfluidity and Superconductivity* (Van Nostrand Reinhold, New York, 1974), p. 37, Fig. 1.29.

<sup>19</sup>H. E. Fischer, S. K. Watson, and D. G. Cahill, Comments Condens. Matter Phys. **14**, 65 (1988).

<sup>20</sup>T. Deng, L. Zhang, H. Gu, Z. Xiao, and L. Chen, Phys. Lett. **5**, 461 (1988); S. Ewert, S. Guo, P. Lemmens, F. Stellmach,

- J. Wynants, G. Arlt, D. Bonnenberg, H. Kliem, A. Comberg, and H. Passing, *Solid State Commun.* **64**, 1153 (1987).
- <sup>21</sup>S. Hoen, L. C. Bourne, C. M. Kim, and A. Zettl, *Phys. Rev. B* **38**, 11949 (1988); X. D. Shi, R. C. Yu, Z. Z. Wang, N. P. Ong, and P. M. Chaikin, *ibid.* **39**, 827 (1989).
- <sup>22</sup>H. Takagi, S. Uchida, and Y. Tokura, *Phys. Rev. Lett.* **62**, 1197 (1989).
- <sup>23</sup>C. Ayache, B. Barbara, E. Bonjour, P. Burlet, R. Calemczuk, M. Couach, M. Jurgens, J. Y. Henry, and J. Rossat-Mignod, *Physica* **148B**, 305 (1987).
- <sup>24</sup>R. A. Fisher, S. Kim, S. E. Lacy, N. E. Phillips, D. E. Morris, A. G. Markelz, J. Wei, and D. S. Ginley, *Phys. Rev. B* **38**, 11942 (1988).
- <sup>25</sup>E. M. Forgan, C. Gibbs, C. Greaves, C. E. Gough, F. Wellhofer, S. Sutton, and J. S. Abell (unpublished).
- <sup>26</sup>J. G. Daunt, R. E. Probst, H. L. Johnston, L. T. Aldrich, and A. O. Nier, *Phys. Rev.* **72**, 502 (1947).