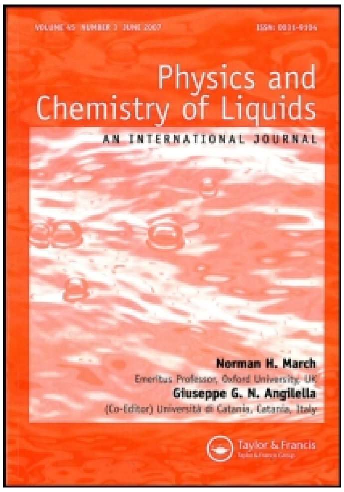

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

Can the Hole Liquid Undergo Wigner Crystallization in High-*T_c* La$_{\text{2-x}}$Sr$_{\text{x}}$CuO$_4$ at Low Density?

N. H. March[ab]; B. V. Paranjape[ab]; F. Siringo[ab]
[a] Theoretical Chemistry Department, University of Oxford, Oxford, UK [b] Department of Physics, University of Alberta, Edmonton, Alberta, Canada

To cite this Article March, N. H. , Paranjape, B. V. and Siringo, F.(1991) 'Can the Hole Liquid Undergo Wigner Crystallization in High-*T_c* La$_{\text{2-x}}$Sr$_{\text{x}}$CuO$_4$ at Low Density?', Physics and Chemistry of Liquids, 24: 1, 131 — 135
To link to this Article: DOI: 10.1080/00319109108030656
URL: http://dx.doi.org/10.1080/00319109108030656

Phys. Chem. Liq., 1991, Vol. 24, pp. 131–135
Reprints available directly from the publisher
Photocopying permitted by license only

LETTER

Can the Hole Liquid Undergo Wigner Crystallization in High-T_c $La_{2-x}Sr_xCuO_4$ at Low Density?

N. H. MARCH*, B. V. PARANJAPE† and F. SIRINGO*

**Theoretical Chemistry Department, University of Oxford, 5 South Parks Road, Oxford OX1 3UB, UK.*

†Department of Physics, University of Alberta, Edmonton, Alberta, Canada

(Received 4 April 1991)

A microscopic interpretation is proposed for the behaviour of the superconducting transition temperature T_c as a function of hole concentration p in $La_{2-x}Sr_xCuO_4$ when oxygen vacancies are suppressed. At high $p \sim 0.32$ a BCS-type formula for T_c is assumed, whereas at low $p \sim 0.06$ it is proposed that the holes undergo Wigner crystallization. Near the Wigner transition, one has a strongly correlated hole liquid, and Cooper pair binding will become increasingly difficult as $p \to 0.06$ from above, because the Fermi distribution is strongly changed from the unit step function, appropriate near $p \sim 0.32$. Some experiments are proposed to test the microscopic model put forward here.

KEY WORDS: Cooper pairs, strongly correlated hole liquid.

The hole concentration p in $La_{2-x}Sr_xCuO_4$ has been used as a variable in mapping out the superconducting phase of this high-T_c material, especially by Torrance *et al*[1]. The phase boundaries of the superconducting state that these and other workers[2,3] observed are reproduced in Figure 1. It is to be noted that no oxygen vacancies were allowed in their experiment, since these vacancies were filled by annealing samples in 100 bars of oxygen pressure.

The purpose of this work is to propose a microscopic interpretation of the boundaries to the superconducting phase shown in Figure 1. We note first with Torrance *et al.*[1] that superconductivity disappears in metallic $La_{2-x}Sr_xCuO_4$ at high hole concentrations, the critical p, denoted by p_c below, being $\simeq 0.32$.

Since in Ref. 1 it is emphasized that at low temperatures and around p_c one has a normal metal–superconductor transition, we shall assume in this regime a BCS-like formula[4]

$$k_B T_c = E_M \exp\left(-\frac{2}{g(E_f)V}\right) \tag{1}$$

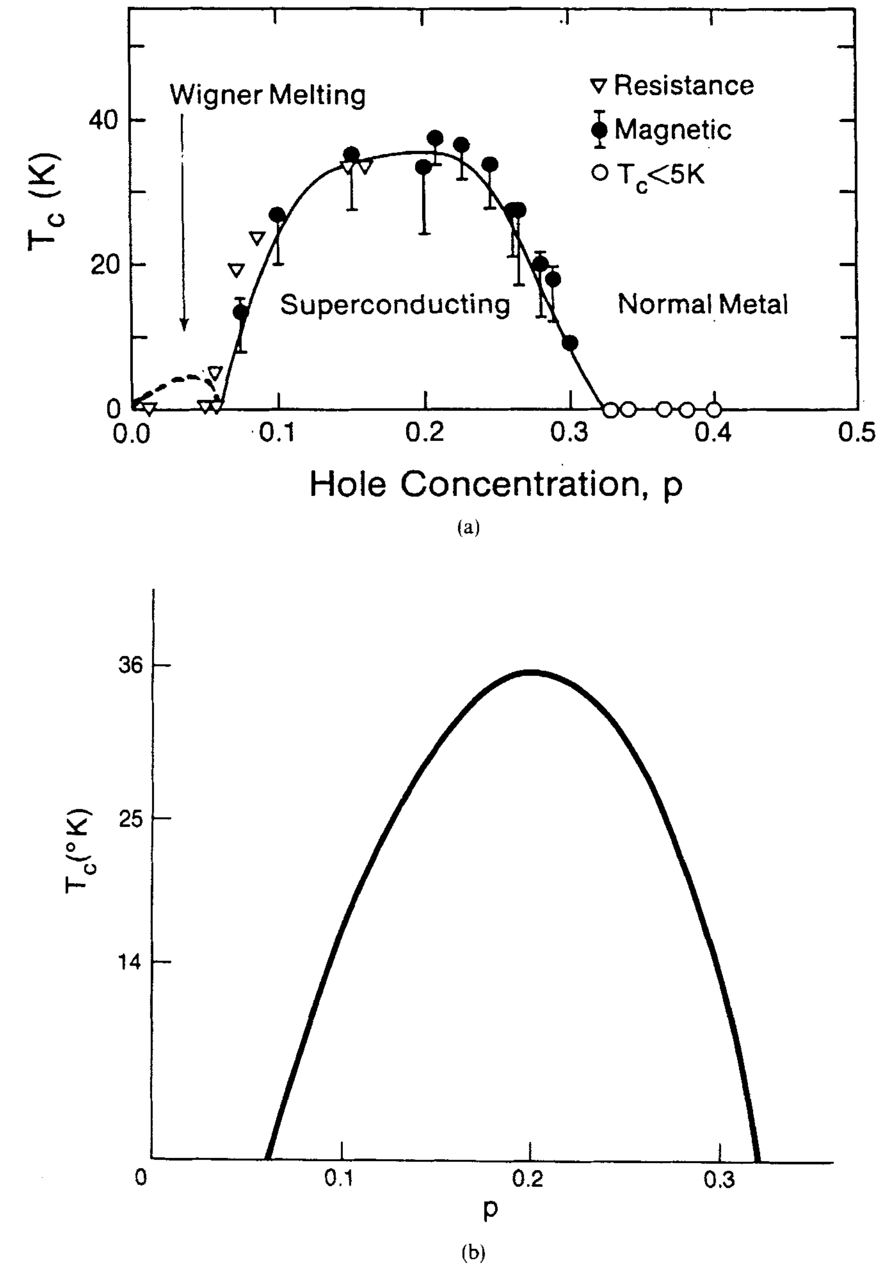

Figure 1 (*a*) The superconducting transition temperature T_c as a function of hole concentration p, after Torrance *et al*[1]. Use has been made of resistance measurements[2] and magnetic measurements[1,3] in establishing this curve. The schematic form of the melting curve of the Wigner hole crystal proposed here is shown by the dashed line. A strongly correlated hole liquid will then exist between these phase boundaries.

(*b*) Schematic form of T_c vs. p. To right of maximum, Eq. (3) plus Eq. (4) extended to include quadratic term in $(p - p_c)$ were employed. To left of maximum, Eqn. (6) was similarly extended with quadratic term.

where E_m is some characteristic energy, the nature of which is to be discussed below, $g(E_f)$ is the density of states at the Fermi-energy E_f while V is a suitable measure of the attractive interaction between holes, leading to Cooper pairs.

In equation (1), which we shall assume can characterize the right-hand part only of the $T_c - p$ plot in Figure 1 when T_c is sufficiently low, i.e. near p_c, let us now utilize a further simple thermodynamic result[5], namely that

$$T_c = (3\gamma/(a - b))^{1/2} \tag{2}$$

where it is assumed that the specific heat per unit volume of a metal has the form $C_s = aT^3$ in the superconducting state while $C_n = bT^3 + \gamma T$ in the normal state. Assuming in the normal state that the electronic specific heat γ is directly proportional to $g(E_f)$, one can use Eq. (2) in Eq. (1) to write

$$\frac{k_B T_c}{E_m} \equiv t_c = \exp\left(-\frac{F(p)}{t_c^2}\right) \tag{3}$$

where one has lumped into the p-dependent function $F(p)$ the dependences of a, b and possibly also E_m on the hole concentration. Equation (3) is the first result of importance for the present argument.

To make a preliminary test of Eq. (3), we have taken E_m to correspond, a little arbitrarily, with the maximum $T_c \sim 36$ K. When this is done, we find the approximate result, over a limited range of p, that

$$F(p) \doteqdot 2.6(p - p_c) \tag{4}$$

or

$$t_c^2 \ln t_c = 2.6(p - p_c) \tag{5}$$

This, we propose, determines, albeit approximately, the shape of the $T_c - p$ boundary in Figure 1 near to the highest hole concentration p_c for which superconductivity exists in this material.

At this point, it is useful to switch attention to the left-hand side of the phase boundary in Figure 1, but again near $T_c = 0$, which will be assumed once more to correspond to a specific hole concentration, denoted below by p_w. Specifically at $T = 0$, one proceeds, by lowering the hole concentration p, to find a 'phase transition' from a state with itinerant holes to a state in which the holes are localized for $p < p_w$.

It seems to us natural to assume that Coulomb repulsion between the holes, which becomes the more important compared to their kinetic (Fermi) energy as p is lowered, will eventually order the holes into a quantal Wigner crystal state[6]. Such strong hole correlations would lead to the filled Fermi sphere picture, so basic to the usual argument for Cooper pairing, which is assumed correct near $p = p_c$, being modified drastically near p_w. In particular, the discontinuity in the momentum distribution

$n(k)$ at $k = k_f$, with k_f the Fermi wave number, which is unity in the usual Cooper pairing theory, will be decreased by hole correlations to a value $q(p)$ say, which reduces to zero as one passes into the Wigner state of localized holes. Of course, whether $q(p)$ falls smoothly to zero at p_w depends on the order of the transition. We expect the hole liquid-solid transition to be first-order but that the discontinuity in q will be small anyway in the strongly correlated hole liquid near $p = p_w$. Then, it is known from heavy Fermion theory[7] that one should compare thermal energies $k_B T$ with qE_f, not E_f, which plainly means that thermal excitation can be expected to play a much greater role near $p = p_w$ than around $p = p_c$, where a BCS-like picture still seems appropriate.

An argument based on the Clausius–Clapeyron equation then leads us to conclude that since, near $p = p_w$, the entropy of the superconductor at small $T \simeq 0$ will be greater than that of the Wigner crystal, then $(dT_c/dp)_{p=p_w}$ will be finite. This appears not to be inconsistent with the phase diagram given by Torrance *et al*[1], though it would be too strong a statement to say that the experimental data established this result. However, writing

$$T_c = a_w(p - p_w) \tag{6}$$

one can form $t_c^2 \ln t_c$ appearing in Eq. (5), but now near $p = p_w$.

In conclusion, we want to reiterate the following points which we propose must be incorporated in any microscopic model of the superconducting phase boundary found in Ref. 1 and depicted in the upper half of Figure 1:

i) A BCS-like formula for T_c should work at low T_c near $p = p_c$ (see lower part of Figure 1).

ii) The transition at $T = 0$ from itinerant holes to a Wigner crystal of holes as p is lowered through p_w should profoundly influence the left-hand side of Figure 1 near $p = p_w$. Thermodynamics plus experiment are consistent with the proposed form (6), though this is not established by the data available.

iii) Near p_w, the hole liquid is strongly correlated, and the Fermi sphere picture is a poor starting point for discussing Cooper pairing, the discontinuity q in $n(k)$ near $k = k_f$ departing strongly from unity and indeed being zero in the Wigner crystal of holes. This may well explain why Cooper pairs no longer bind for $p \sim p_w$, because the role of the Exclusion Principle is weakened.

Outstanding questions which remain are as follows:

i) What is the nature of the elementary excitations in the superconducting state near $p = p_w$? In particular, is there a linear term in the specific heat in the superconductor in this regime?

ii) Is there an additional phase boundary, starting out at $p = p_w$ and going to the left of Figure 1 representing the melting of the Wigner crystal of holes? If so, what is the behaviour of the holes between these two phase boundaries at low T_c?

iii) In addition to the Coulomb correlations leading to hole localization at $T = 0$

for $p < p_w$, does the local variation in Sr concentration play any role through Anderson localization?

iv) Is the maximum value of T_c related to qE_f where q is the discontinuity in the Fermi momentum distribution at k_f? Measurements of the magnetic susceptibility of the holes in the region between the phase boundaries representing (a) the melting of the Wigner crystal and (b) the transition $T_c - p$ line may be useful in clarifying this point.

Acknowledgements

One of us (NHM) wishes to acknowledge that his contribution to this work was made during a visit to the Physics Department of the University of Alberta, Edmonton during 1989. In particular, Professors H. R. Glyde and F. C. Khanna are thanked for generous hospitality. Another of us (FS) acknowledges a Fellowship award from the Fondazione Angelo Della Riccia, Italy, which made possible his stay in Oxford, U.K. Finally, NHM wishes to acknowledge valuable discussions with Dr. A. Bezinge during a visit to IBM, San José in 1988.

References

1. J. B. Torrance, Y. Tokura, A. I. Nazzal, A. Bezinge, T. C. Huang and S. S. P. Parkin, *Phys. Rev. Lett.*, **61**, 1127 (1988).
2. M. W. Shafer, T. Penney and B. L. Olson, *Phys. Rev.* **B36**, 4047 (1987).
3. R. B. van Dover, R. J. Cava, B. Batlogg and E. A. Reitman, *Phys. Rev.* **B35**, 5337 (1987).
4. See, for example, N. H. March, W. H. Young and S. Sampanthar, *The Many-Body Problem in Quantum Mechanics* (Cambridge: University Press 1967).
5. See, for instance, A. B. Pippard, *Elements of Classical Thermodynamics*, (Cambridge: University Press) 1966.
6. For a review of quantal Wigner crystallization, see C. M. Care and N. H. March, *Advances in Physics* **24**, 101 (1975).
7. See, for example, T. M. Rice, K. Ueda, H. Ott and H. Rudigier, *Phys. Rev.* **B31**, 594 (1985); also R. G. Chapman and N. H. March, *Phys. Rev.* **B38**, 792 (1988).