The low-energy excitation spectrum of HTS cuprates is examined in the light of thermodynamic, transport, quasiparticle and spin properties. Changes in the thermodynamic spectrum associated with the normal-state pseudogap disappear abruptly at a critical doping state, $p_{\text{crit}} = 0.19$ holes per Cu. Moreover, ARPES data at 100K show that heavily damped quasiparticles (QP) near $(\pi,0)$ suddenly recover long lifetimes at $p_{\text{crit}}$, reflecting an abrupt loss of scattering from AF spin fluctuations. This picture is confirmed by $\mu$SR zero-field relaxation measurements which indicate the presence of a novel quantum glass transition at $p_{\text{crit}}$. Consistent with this picture resistivity studies on thin films of $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ reveal linear behavior confined to a V-shaped domain focussed on $p_{\text{crit}}$ at $T=0$. The generic phase behavior of the cuprates may be governed by quantum critical fluctuations above $p_{\text{crit}}$ and the pseudogap appears to be caused by short-range AF correlations.

PACS numbers: 74.25.Bt, 74.25.Fy, 74.25.Ha, 74.72.Jt.

1. INTRODUCTION

The highly anomalous properties of the high-$T_c$ superconducting (HTS) cuprates are attributable in part to the presence of a normal-state (NS) pseudogap extending across the underdoped region of the phase diagram. The onset of the pseudogap opens up a gap in the density of states (DOS) which
Tallon, Loram and Panagopoulos

profundely affects all physical properties in both the normal and superconducting (SC) states.\(^1\) Considerable debate has ensued as to the nature of the pseudogap and, in particular, whether it is some form of precursor SC pairing\(^2\) or a NS correlation which competes (or coexists) with SC.\(^3\) We believe the evidence is firmly in support of the latter scenario and present here further supporting evidence. In particular we show that the SC ground state is strongly perturbed by the pseudogap and that the magnetic spectrum undergoes abrupt and major changes at the critical doping state \(p_{\text{crit}}\) at which the pseudogap first appears. (Here \(p\) is the doped hole concentration per planar Cu). Finally, we show from thin-film transport measurements that the famous linear resistivity is confined to a V-shaped region in the \(T-p\) phase diagram that is centered on \(p_{\text{crit}}\). That is, only at \(p_{\text{crit}}\) does the NS resistivity remain linear to \(T = 0\). This behavior is consistent with a quantum critical point (QCP) scenario.\(^4\)

2. THERMODYNAMIC PROPERTIES

Our starting point is to recognize that the thermodynamic and transport behavior discussed here seems to be universal amongst the HTS cuprates. Thus we will alternate between the three compounds \(Y_{0.8}\text{Ca}_{0.2}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}\) (Y-123), \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}\) (Bi-2212) and \(\text{La}_{2-x}\text{Sr}_x\text{CuO}_4\) (La-214) under the assumption that our deductions are universal. Of course there are some features which seem unique to La-214, such as a strong stripe character and a high degree of disorder, but the properties we discuss here are also realized in the other two systems.

Each of these systems exhibits abrupt changes in their thermodynamic properties at \(p_{\text{crit}}\). This is illustrated, in the case of Bi-2212, in Fig. 1 using previously reported data.\(^5\) Panel (a) shows the doping dependence of the jump in the specific heat coefficient, \(\Delta \gamma\), and of the pseudogap energy, \(E_g\), obtained by fitting the temperature dependence of the entropy using a triangular NS DOS \(N(E) = N_0(E/E_g)\) for \(E < E_g\) and \(N(E) = N_0\) for \(E > E_g\). Such a DOS is non-states conserving and has been shown to fit the thermodynamic data very well over a broad range of \(T\) and \(p\).\(^5\)

Panel (b) shows the \(p\)-dependence of the condensation energy \(U_0\). Notably the opening of the pseudogap, as shown by the value of \(E_g\), is abrupt and it has an immediate effect in sharply depressing \(U_0\) and \(\Delta \gamma\) (a measure of the pair density). Accompanying these abrupt changes the superfluid density at \(T = 0\) is also sharply reduced, as shown by ac susceptibility\(^6\) field-dependent specific heat measurements\(^5\) and \(\mu\)SR.\(^7\) This reflects a sudden change in the ground state with the opening of the pseudogap and is not compatible with the precursor pairing models but rather suggests that
Pseudogap and quantum transition phenomenology

Fig. 1. The doping dependence of various parameters for Bi-2212 (a) $\Delta \gamma$ and $E_g$, (b) $U_0$ and (c) the normalized quasiparticle peak at ($\pi,0$) and 100 K.

the pseudogap represents some correlated state that coexists with SC.

3. QUASIPARTICLE WEIGHT AND LIFETIME

The Bi-2212 system has been widely investigated using other techniques, and many key observations on the pseudogap have been assembled for this system. We mention briefly recent scanning tunnelling microscopy (STM) measurements which appear to suggest a very high degree of local inhomogeneity. Pan et al.\(^5\) suggest that such disorder may be a characteristic of all HTS systems. We believe this not to be the case. Inhomogeneity involving separate spatial regions of SC and NS metallic behavior would result in a finite DOS at the Fermi level that is not observed, while spatially separate insulating and metallic regions would result in a reduction of the NS DOS at high temperature, $T > E_g/k_B$. This is not observed either, indeed the specific heat coefficient for Bi-2212 is constant at high $T$, independent of doping.\(^5\) A stronger case, still, can be made with reference to NMR linewidths and transition broadening. Moreover, the abruptness of the various onsets shown in Fig. 1 raise major questions about the very large static spatial inhomogeneity inferred from STM (±30% spread in energy gap, local DOS and doping level). We emphasize that the heat capacity is a bulk mea-
Measurement while STM probes just the outermost CuO$_2$ layer which is likely to be subject to inhomogeneity arising from reconstruction, oxygen disorder and Bi/Sr intersubstitution.

Another technique that has contributed in a major way to our understanding of the cuprates is angle-resolved photoelectron spectroscopy (ARPES). Systematic studies of underdoped samples show that NS quasiparticle (QP) lifetimes are very short near $(\pi,0)$ due to pronounced scattering from antiferromagnetic (AF) spin fluctuations because it is at the zone boundary that the AF wave vector spans the Fermi surface. This is manifested by the almost complete absence of the NS QP peak$^9$ whose weight is redistributed due to these scattering events. On the other hand, near the zone diagonal QP lifetimes are long and the QP peak is fully recovered.$^9$ We have analyzed the leading-edge energy dispersion curves for a large number of reported measurements on Bi-2212$^{9-16}$ at $(\pi,0)$ and at 100K (i.e. in the NS) and we have fitted these to a background curve plus a Gaussian QP peak. The magnitude of the latter has been ratioed with that of the former (as a measure of the loss or redistribution of QP weight) and plotted as a function of doping in panel (c) of Fig. 1. Remarkably the QP weight is very small across the entire underdoped region and is abruptly and fully recovered at $p_{\text{crit}}$ in the lightly overdoped region. (The most highly doped sample is for Bi-2201). While the data used is obtained from a variety of sources with differing experimental conditions, the changes are so marked that they must reflect a real and dramatic alteration in the spin spectrum, presumably the almost complete demise of scattering from AF fluctuations. Elsewhere we have presented evidence from other studies for the collapse of short-range AF correlations at $p_{\text{crit}}$. This includes inelastic neutron scattering in Y-123, induced local moments in Zn-substituted Y-123 samples and two-magnon Raman scattering in Bi-2212.$^{17}$ This accumulated evidence is rather strong and implies that the pseudogap is associated with short-range AF correlations which suddenly disappear at $p_{\text{crit}}$, as does the pseudogap.

4. QUANTUM GLASS TRANSITION

One of the questions which arises from the foregoing is whether $p_{\text{crit}}$ is a QCP. Thermodynamic measurements show that there is no phase transition associated with the characteristic temperature $T^* = E_g/k_B$ which should rather be considered as a cross-over temperature. Various authors have proposed some form of hidden order parameter which is established below $T^*.$$^{18}$ None has thus far been substantiated. However we note that a quantum phase transition at $p_{\text{crit}}$ could be associated with a spin-glass transition with long-range Edwards-Anderson order.$^{19}$
We have recently completed a series of zero-field $\mu$SR studies on the spin fluctuation spectrum in pure and Zn-substituted La-214. These investigations allow the determination (for any given doping state) of the temperature, $T_f$, at which spin fluctuations first enter the $\mu$SR time window ($10^{-10}$ sec) and the temperature, $T_g$, at which they leave the $\mu$SR time window ($10^{-6}$ sec) i.e. at which they become essentially static as far as the $\mu$SR technique is concerned. These two crossover temperatures are plotted in Fig. 2 for La$_{2-x}$Sr$_x$Cu$_{1-y}$Zn$_y$O$_4$ with $y = 0, 1$ and $2\%$. For each Zn concentration the pairs of crossover temperatures, $T_f$ and $T_g$, converge to zero at $p_{\text{crit}}$. Thus at $p_{\text{crit}}$ the rate of slowing down of spin fluctuations diverges, a clear signature of a quantum glass transition. We note, without further discussion, the enhancement in $T_f$ and $T_g$ seen at $1/8^{\text{th}}$ doping which coincides with the local reduction in $T_c$. Evidently, AF spin fluctuations slow markedly in this region, possibly due to incipient stripe formation.

AF spin fluctuations are also slowed by Zn substitution and this, again, moves $T_f$ and $T_g$ to higher values for a given $p$-value. In each case there is no evidence of slow fluctuations (within the $\mu$SR time window) for $p > p_{\text{crit}}$. One interpretation is that the mobile carriers for $p > p_{\text{crit}}$ flip spins so rapidly that the AF spin fluctuation lifetime is suppressed effectively to zero.
emerges, then, the picture that for $p < p_{\text{crit}}$ the AF spin lifetime is long but the QP lifetime at $(\pi,0)$ is strongly attenuated due to scattering from AF spin fluctuations while for $p > p_{\text{crit}}$ the QP lifetime at $(\pi,0)$ recovers while the spin lifetime is strongly attenuated. The crossover is remarkably abrupt.

5. LINEAR RESISTIVITY AND THE QCP

If there is a quantum glass transition (QGT) at $p_{\text{crit}}$ then one expects, within the phenomenology of quantum critical behavior, a resistivity which is linear in $T$ occurring at $p_{\text{crit}}$. To either side of the QGT the region of linearity would be expected to be pushed to higher temperatures thus describing a V-shaped domain of linear-$T$ resistivity in the quantum critical regime which is focussed on $p_{\text{crit}}$. Within this picture the crossover line $T^*(p)$ would form the boundary of the "V" for $p < p_{\text{crit}}$ while another line $T'(p)$, for $p > p_{\text{crit}}$, would form the other boundary of the "V". This is precisely what is observed in the three model cuprates we are discussing. Below $T^*(p)$ the resistivity $\rho(T)$ is found to be sub-linear while below $T'(p)$ the resistivity $\rho(T)$ is super-linear. We have previously analyzed the resistivity of epitaxial thin films of $Y_{0.7}Ca_{0.3}BaCu_2O_{7-\delta}$ to show this very behavior. In Fig. 3 we show this data in a more illustrative way. We plot a map of the $p$- and $T$-dependence of the resistivity with the high-temperature linear-$T$ dependence divided out. The figure shows that the region where $\rho(T)/(\rho_0 + \alpha T)$ is close to unity indeed forms a V-shape about the critical doping state $p_{\text{crit}} = 0.19$ holes per Cu. As can be seen, the tendency of this region to focus to $p_{\text{crit}}$ at $T = 0$ is cut-off by the appearance of superconductivity. Ideally one would like to suppress SC so as to follow this behavior to low temperature. The use of Zn substitution provides a possible route but the data is complicated by the effects of residual scattering and localization. Nonetheless Fig. 3 illustrates a phenomenology strongly reminiscent of QCP behavior. Similar studies could be carried out in high pulsed magnetic fields (perhaps combined with Zn substitution) to expose this behavior to low temperatures.

6. CONCLUSION

In the light of these compelling evidences for coexisting NS correlations and possible QCP behavior we believe it is time to abandon the previously widespread notion of the pseudogap deriving from precursor SC pairing. Clearly such pairing effects occur close to $T_c$, and indeed have been observed, but the dominant pseudogap behavior that is observed over a large domain of the phase diagram (and to high temperature) seems clearly to be associated with short-range AF spin correlations that result in the opening of a gap
Pseudogap and quantum transition phenomenology

Fig. 3. Colour-scale plot, on the $T - p$ phase diagram, of $\rho(T)/(\rho_0 + \alpha T)$ for the resistivity of thin films of $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$. $T_c(p)$ is shown by the solid curve.

in the NS DOS. This takes away spectral weight otherwise available for SC pairing around the Fermi surface with consequent dramatic weakening of the ground-state SC as well as strongly modifying the NS properties. We hope that the present data (and other related data) will help establish much-needed consensus in these matters.

ACKNOWLEDGMENTS

Thanks are due to the Marsden Fund of New Zealand (JLT) and the Royal Society (CP) for financial support for this programme.

REFERENCES

1. J. R. Cooper and J. W. Loram, J. Phys. I (France) 6, 2237 (1996).
2. V. Emery and S. A. Kivelson, Nature (London) 374, 434 (1995).
3. J. W. Loram et al., J. Phys. Chem. Solids 59, 2091 (1998).
4. J. L. Tallon et al., phys. stat. soli (b) 215, 531 (1999).
5. J. W. Loram et al., J. Phys. Chem. Solids 62, 59 (2001).
6. C. Panagopoulos et al., Phys. Rev. B 60, 14617 (1999).
7. C. Bernhard et al., Phys. Rev. Lett. 86, 1614 (2001).
8. S. H. Pan et al., Nature 413, 282 (2001).
9. C. Kim et al., Phys. Rev. Lett. 80, 4245 (1998).
10. D. S. Marshall et al., Phys. Rev. Lett. 76, 4841 (1996).
11. R. B. Laughlin, Phys. Rev. Lett. 79, 1726 (1997).
Tallon, Loram and Panagopoulos

12. M. R. Norman et al., Phys. Rev. Lett. 79, 3506 (1997).
13. Z. -X. Shen and J. R. Schrieffer, Phys. Rev. Lett. 78, 1771 (1997).
14. P. J. White et al., Phys. Rev. B 54, R15669 (1996).
15. T. Sato et al., Phys. Rev. B 64, 054501-1 (2001).
16. J. Mesot et al., Phys. Rev. B 63, 224516 (2001).
17. J. L. Tallon and J. W. Loram, Physica C 349, 53 (2001).
18. C. M. Varma, Phys. Rev. Lett. 83, 3538 (1999); S. Chakravarty et al., Phys. Rev. B 63, 10000 (2001).
19. S.F. Edwards and P.W. Anderson, J. Phys. F: Metal Phys. 5, 965 (1975).
20. C. Panagopoulos et al., Phys. Rev. B 66, 064501 (2002).