Neutron experiments as a test of anisotropic pairing in $YBa_2Cu_3O_{7-\delta}$ and $La_{2-x}Sr_xCuO_4$

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(Received 11 June 1992)

We show that the anomalous temperature T dependences at low frequencies ω observed in neutron measurements of the structure factor $S(\mathbf{q},\omega)$ are compatible with a $d_{x^2-y^2}$ pairing state. We further demonstrate that convincing verification of this anisotropic pairing also requires the observation of a $\mathbf{q}$ dependence which, in low-T neutron data, differs significantly from that above T_c. In the absence of such evidence, establishing the existence of the $d_{x^2-y^2}$ state remains problematical.

Recent attention in the high-temperature superconducting oxides has focused on spin dynamical experiments[1–3] below T_c, which have been interpreted[4,5] as support for an anisotropic pairing state. It is the purpose of the present paper to demonstrate that the most unambiguous magnetic evidence in support of anisotropic pairing lies in the behavior of the wave vector $\mathbf{q}$ dependence of the structure factor $S(\mathbf{q},\omega)$, measured in neutron experiments. In this context, we examine recent neutron data on both the LaSrCuO and YBaCuO families and compare with predictions based on a $d_{x^2-y^2}$ state. In order to test unambiguously for the presence of gap nodes, the *magnitude* and *location* of these contributions to $S(\mathbf{q},\omega)$ must be quantitatively characterized. We are able to determine the scale of these nodal effects and thus go beyond Ref. 4 by using (i) new neutron data below T_c and (ii) a calculational scheme[6] which matches on to normal state data in two different cuprates at and above T_c.

It was shown in Ref. 4 that gap nodes yield large resonant peaks in $\mathrm{Im}\chi(\mathbf{q},\omega)=S(\mathbf{q},\omega)(1-e^{-\beta\omega})$. As was pointed out in two important papers,[4,5] this nodal structure will also lead to a low-T upturn in the anisotropy ratio $(1/T_1)_{ab}/(1/T_1)_c$ at the Cu site below T_c. That this nonmonotonic behavior is consistent with recent NMR data[2,3] in $YBa_2Cu_3O_7$ provides important support for the d-wave state. However, because these calculations were based on a band structure which is more suitable for the LaSrCuO family than for YBaCuO, it is essential to reexamine this previous work and to provide predictions for the spin dynamics which build in both the correct band structure and the closely related $\mathbf{q}$ dependence in $S(\mathbf{q},\omega)$.

Recent neutron data[1] on LaSrCuO have shown that the superconducting transition coincides with a maximum in $\mathrm{Im}\chi(q,\omega)$ for sufficiently low $\omega\sim4$ meV and with q fixed at the incommensurate wave vector corresponding to that of the peak in the normal state cross section.[7] These current experiments represent the strongest indications that the superconducting state is affecting the neutron measured spin dynamics in this cuprate family. There is some ambiguity[1] about whether or not $\mathrm{Im}\chi$ at these low ω vanishes at the lowest T; a nonzero value lends some support to an anisotropic pairing state. For $YBa_2Cu_3O_{6.6}$, experiments by Sternlieb *et al.*[1] show that at small ω, the zone center integrated contribution to $\mathrm{Im}\chi(q,\omega)$ first decreases and then increases below T_c with decreasing T. For larger ω, $\mathrm{Im}\chi(q,\omega)$ decreases monotonically with rising temperature. The upturn at low temperatures (which has not, however, been confirmed by the Grenoble group[8]) suggests that the material might be an anisotropic superconductor. Data from Thurston *et al.*[1] and Sternlieb *et al.*[1] on the $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_{6.6}$ systems are shown in the insets to Figs. 1 and 2, respectively.

It may be argued that, just as in the case of NMR data, the growth of nodal contributions at low T can give rise to both the nonzero value of $\mathrm{Im}\chi(q,\omega\sim3\text{ meV})$ as $T\to0$ in the LaSrCuO family, as well as the low-T upturn in YBaCuO. The present paper is organized around the following premise. *If* the anomalous T and ω dependences in the neutron data are related to a d-wave pairing, then it follows that the nodal contributions are of sufficient strength so as to lead to *observable* neutron scattering intensities for T sufficiently far below T_c. To establish consistency, a direct observation of the nodal terms, via the $\mathbf{q}$ dependence in $S(\mathbf{q},\omega)$ is required. Our approach is based on choosing a (single) free parameter, the zero tempera-

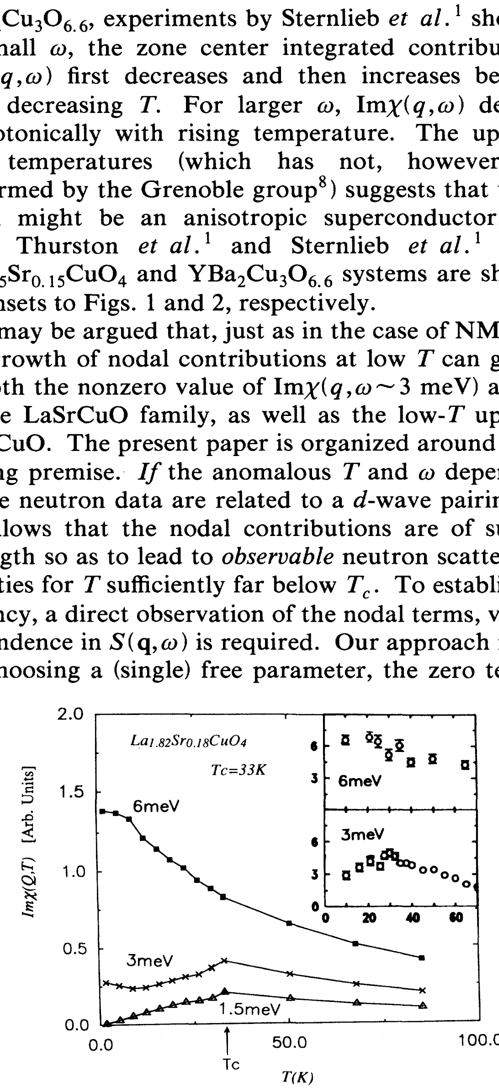

FIG. 1. Calculated temperature dependence of $\mathrm{Im}\chi(Q,\omega)$ in $La_{1.82}Sr_{0.18}CuO_4$ for $Q=(0.67\pi,\pi)$ and various ω. Here $\Delta(T=0)=3$ meV and the exchange amplitude $J_0/J_c=0.6$, where J_c is the critical value for a magnetic instability. Data in inset from Thurston *et al.* (Ref. 1).

ture amplitude of the $d_{x^2-y^2}$ gap $\Delta(\mathbf{k})=\Delta(T)(\cos k_z a - \cos k_y a)$ so as to fit the low-T behavior as a function of ω, and thereby to deduce implications for the $\mathbf{q}$ dependence in the dynamical structure factor. Here for simplicity we assume that the T dependent function $\Delta(T)/\Delta(T=0)$ is given by that of weak coupling BCS theory. We have found that once the (ω,T) dependence in the neutron data are fit, our results for the $\mathbf{q}$ dependence as a function of T are relatively insensitive to whether we use a BCS form or not.

Our superconducting state calculations are based on previous normal state work using a three-band Hubbard model with very strong Coulomb U, as described earlier in Ref. 6. The dynamic susceptibility was found to be of the RPA form:

$$\chi_{rr'}(\mathbf{q},\omega)=\chi^0_{rr'}(\mathbf{q},\omega)+\frac{\chi^0_{rd}(\mathbf{q},\omega)[-J(\mathbf{q})]\chi^0_{dr'}(\mathbf{q},\omega)}{1+J(\mathbf{q})\chi^0_{dd}(\mathbf{q},\omega)} \tag{1}$$

with (weakened) antiferromagnetic[6] superexchange $J(\mathbf{q})=J_0(\cos k_x a+\cos k_y a)$ between copper sites. Here the superconducting Lindhard function is

$$\begin{aligned}\chi^0_{rr'}(\mathbf{q},\omega)=\frac{1}{N_{\text{site}}}\sum_{nm\mathbf{k}}\Bigg\{&\frac{1}{2}\left[1+\frac{\varepsilon_m(\mathbf{k+q})\varepsilon_n(\mathbf{k})+\Delta(\mathbf{k+q})\Delta(\mathbf{k})}{E_m(\mathbf{k+q})E_n(\mathbf{k})}\right]\frac{f[E_m(\mathbf{k+q})]-f[E_n(\mathbf{k})]}{\omega-[E_m(\mathbf{k+q})-E_n(\mathbf{k})]}\\&+\frac{1}{4}\left[1-\frac{\varepsilon_m(\mathbf{k+q})}{E_m(\mathbf{k+q})}+\frac{\varepsilon_n(\mathbf{k})}{E_n(\mathbf{k})}-\frac{\varepsilon_m(\mathbf{k+q})\varepsilon_n(\mathbf{k})+\Delta(\mathbf{k+q})\Delta(\mathbf{k})}{E_m(\mathbf{k+q})E_n(\mathbf{k})}\right]\frac{1-f[E_m(\mathbf{k+q})]-f[E_n(\mathbf{k})]}{\omega+[E_m(\mathbf{k+q})+E_n(\mathbf{k})]}\\&+\frac{1}{4}\left[1+\frac{\varepsilon_m(\mathbf{k+q})}{E_m(\mathbf{k+q})}-\frac{\varepsilon_n(\mathbf{k})}{E_n(\mathbf{k})}-\frac{\varepsilon_m(\mathbf{k+q})\varepsilon_n(\mathbf{k})+\Delta(\mathbf{k+q})\Delta(\mathbf{k})}{E_m(\mathbf{k+q})E_n(\mathbf{k})}\right]\\&\times\frac{f[E_m(\mathbf{k+q})]+f[E_n(\mathbf{k})]-1}{\omega-[E_m(\mathbf{k+q})+E_n(\mathbf{k})]}\Bigg\}u^*_{mr}(\mathbf{k+q})u^*_{nr'}(\mathbf{k})u_{mr'}(\mathbf{k+q})u_{nr}(\mathbf{k})\;.\end{aligned} \tag{2}$$

Here $\varepsilon_n(\mathbf{k})=E^0_n(\mathbf{k})-\mu$, $f(E)=1/[\exp(E/T)+1]$ and (m,n), (r,r') are band and site indices, respectively. The superconducting quasiparticle energies are given by

$$E_n(\mathbf{k})=\{[\varepsilon^0_n(\mathbf{k})]^2+[\Delta(\mathbf{k})]^2\}^{1/2}\;,$$

where the normal state bandstructure energy $E^0_n(\mathbf{k})$ is determined from the same Coulomb renormalized bandstructure as was used in Ref. 6, appropriately modeled for *both* $YBa_2Cu_3O_{7-\delta}$ and $La_{2-x}Sr_xCuO_4$ systems.

We have emphasized elsewhere[6] that in the normal state the important differences in the Fermi surface shapes of the two cuprates, which enter the spin dynamics via $E^0_n(\mathbf{k})$, are expected to be strongly reflected in neutron experiments. These differences derive from a 45° rotation of the Fermi surface in YBaCuO relative to LaSrCuO. When combined with $J(\mathbf{q})$ in Eq. (1), they lead to commensurate peaks in the former and incommensurate structure in the latter system, as is observed experimentally.

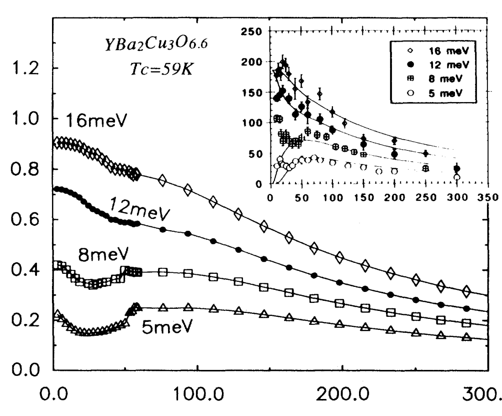

FIG. 2. Calculated temperature dependence of $\mathrm{Im}\chi(Q_0,\omega)$ in $YBa_2Cu_3O_{6.6}$ with $\Delta(T=0)=6$ meV. Here $Q_0=(0.7\pi,0.7\pi)$ is the position of the nodal peak, $J_0/J_c=0.7$ and frequencies ω are as indicated. Data (and solid lines) in inset from Sternlieb *et al.* (Ref. 1).

The situation is more complicated for $0<T<T_c$. Here the neutron scattering form factor consists of three contributions: (1) node-induced resonant peaks, (2) nesting peaks arising from the normal state band structure $E^0_n(\mathbf{k})$, and (3) interaction effects via the antiferromagnetic superexchange $J(\mathbf{q})$. For the LaSrCuO system, the nodal peaks are located at $[(1\pm\delta)\pi,(1\pm\delta)\pi]$ along the zone diagonal direction, where δ corresponds to the normal state incommensurability. By contrast, the nesting peaks occur at $[(1\pm\delta)\pi,\pi]$ and $[\pi,(1\pm\delta)\pi]$, as found by Cheong *et al.*[7] Because most of this Lindhard structure occurs near (π,π), the effects of the exchange interaction $J(q)$ in Eq. (1) are only to increase the peak height, leaving the shape relatively unaffected. It follows from the above discussion that the $\mathbf{q}$ structure in LaSrCuO is expected to change qualitatively at $T\to 0$, when the nodal structure begins to dominate that associated with nesting, and that this change requires that the neutron peaks move to the diagonal direction.

In $YBa_2Cu_3O_{7-\delta}$, the nodal and nesting peaks are located at $(\pm 2k_F,\pm 2k_F)$ and $(\pm 2k_F,0)$ [as well as $(0,\pm 2k_F)$], respectively. The latter (as well as the former) are substantially away from the (π,π) point, as compared to the LaSrCuO system (by roughly a factor of $\sqrt{2}$). This consequence of the 45° Fermi surface rotation implies that the Lindhard function is relatively flat around (π,π). Therefore, in this vicinity, the $\mathbf{q}$ dependence of $S(\mathbf{q},\omega)$ is dominated by that of the exchange interaction $J(q)$. The latter leads in turn to a commensurate peak in the normal state. At sufficiently low T, when the superconducting order is well developed this peak is "gapped out" and the nodal structure becomes the more important. Thus just as in the LaSrCuO family there is a qualitative change in the $\mathbf{q}$ dependent behavior of $S(\mathbf{q},\omega)$ as $T\rightarrow 0$. Here, again, the nodal peaks will show up along the zone diagonal direction, as the peak structure moves from commensurate to incommensurate.

In order to determine the appropriate magnitudes for this *predicted* $\mathbf{q}$-structure evolution, we use existing experiments, shown in the insets of Figs. 1 and 2 to determine the amplitude of the zero temperature gap. The parameter choice $2\Delta(T=0)=6$ meV$\sim 2.1kT_c$ for $La_{1.82}Sr_{0.18}CuO_4$ and $2\Delta(T=0)=2.6kT_c\sim 12$ meV for $YBa_2Cu_3O_{6.6}$ optimizes agreement with both the ω and T dependences of the structure factor.[9] This agreement is viewed as only semiquantitative.

Figure 1 shows the calculated $\mathrm{Im}\chi((1-\delta)\pi,\pi)$ for $La_{1.82}Sr_{0.18}CuO_4$ at the incommensurate nesting peak for different frequencies ω. Experimental data from Thurston *et al.*[1] are shown in the inset. For small ω, i.e., $\omega<\Delta(T=0)$, $\mathrm{Im}\chi((1-\delta)\pi,\pi)$ has a sharp maximum at T_c, similar to that also observed by Mason, Aeppli and Mook.[1] For higher ω, the superconducting gap is negligible so that $\mathrm{Im}\chi((1-\delta)\pi,\pi)$ decreases monotonically as T increases, and there is no sign of the transition at T_c. The small upturn at low temperature for $\omega=3$ meV arises in our calculations from "leakage" due to the gap nodes which are centered at $[(1\pm\delta)\pi,(1\pm\delta)\pi]$. In the neutron experiments, the limiting instrumental resolution may further artificially enhance structure at this nesting peak position via nodal peak contributions which may be inadvertently detected.[10]

In Fig. 2 are plotted similar results for $YBa_2Cu_3O_{6.6}$. Here the inset is from Sternlieb *et al.*[1] The temperature dependence of $\mathrm{Im}\chi(\mathbf{q},\omega)$ is indicated for three values of the frequency ω and at the position of the nodal peak where $\mathbf{q}=[(1-\delta/2)\pi,(1-\delta/2)\pi]$, with $\delta=0.6$. This $\mathbf{q}$ value was chosen to indicate the maximum effect of the gap nodes. The upturn at low temperatures for small $\omega=5,8$ meV is a consequence of the growth of these nodal contributions as $T\rightarrow 0$. At higher $\omega=12$ and 16 meV the behavior is more characteristic of the normal state which shows a monotonic increase with decreasing T. The counterpart experiments shown in the inset are obtained by *integrating* $\mathrm{Im}\chi$ over a range of $\mathbf{q}$ values in the vicinity of (π,π). The absence of any experimentally observed upturn at higher frequencies $\omega\sim 12$–16 meV provides a constraint[9] on the allowed values of the zero temperature gap. For our choice of the parameter $2k_f$ we find no evidence of the nodal contribution precisely at the (π,π) point.[11] Hence if experiments are picking up the d-wave nodes at this point, this would appear to be a consequence of the integration over a range of $\mathbf{q}$ values, due to the finite resolution of the experiments. It follows from the above discussion that without more detailed $\mathbf{q}$ scans for $T\sim 5$ K, and $\omega\leq 5$ meV, it is not possible to determine whether these low ω,T anomalies are in any way related to the presence of an anisotropic superconducting order parameter.

The following two figures represent predictions based on the implications of Figs. 1 and 2. The sharp, jagged structure is an artifact of the numerical plotting routine and should be viewed as smoothed out within the plotted envelope function. Figure 3 shows the predicted temperature evolution of the $\mathbf{q}$-dependent structure in $\mathrm{Im}\chi(\mathbf{q},\omega=2.5\text{ meV})$ for $La_{1.82}Sr_{0.18}CuO_4$ from just below T_c to the low-T regime where the nodal contributions are dominant. This figure should be viewed as following necessarily from the premise that the measured low T,ω anomalies of Fig. 1 are associated with d-wave pairing. For this cuprate, the topography of the structure factor essentially reflects that of the Lindhard function. At the highest temperature ($T=25$ K), the nesting peaks are dominant and the maxima are away from the zone diagonal direction. At intermediate temperatures ($T=15$ K), the nodal contributions give added weight to $\mathbf{q}$ along the diagonal and by the lowest T ($=3$ K) the nodal peaks dominate and the nesting structure has been "gapped out."

A similar plot of $\mathrm{Im}\chi(\mathbf{q},\omega=2.5\text{ meV})$ is shown in Fig. 4 for the $YBa_2Cu_3O_{6.6}$ system. Just below T_c (at $T=55$ K), the shape of the structure factor mirrors that of the normal state, but with reduced intensity due to the opening of the superconducting gap. As T decreases to intermediate temperatures ($T=30$ K) the growth of the four nodal peaks is clearly evident; these coexist with the commensurate maximum. At the lowest $T=4$ K, the (π,π) peak has been gapped out and only the nodal structure remains.

In summary, the main contribution of the present paper was to present systematic predictions for the temperature evolution of the $\mathbf{q}$ structure in neutron experiments from the normal state to $T\sim 0$ for two families of the copper oxides. In this way, we may hope to obtain a clear indication of the superconducting gap symmetry.

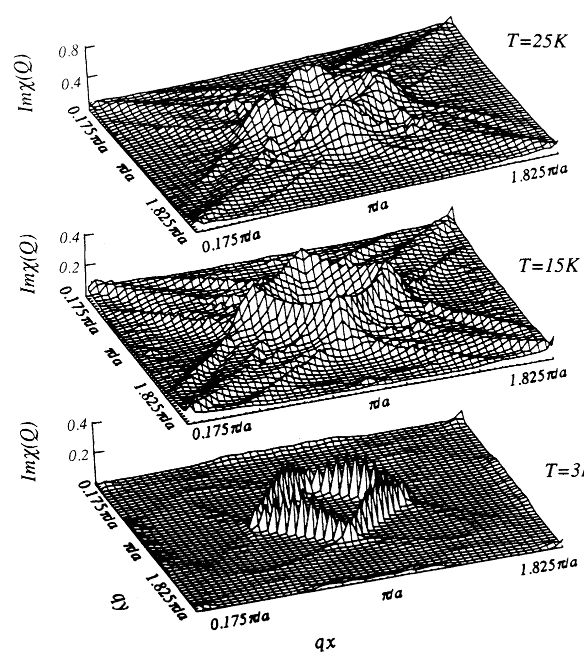

FIG. 3. Predicted wave-vector dependence of $\mathrm{Im}\chi$ at $\omega=2.5$ meV for $La_{1.82}Sr_{0.18}CuO_4$ at various temperatures below T_c. Parameters are chosen as in Fig. 1.

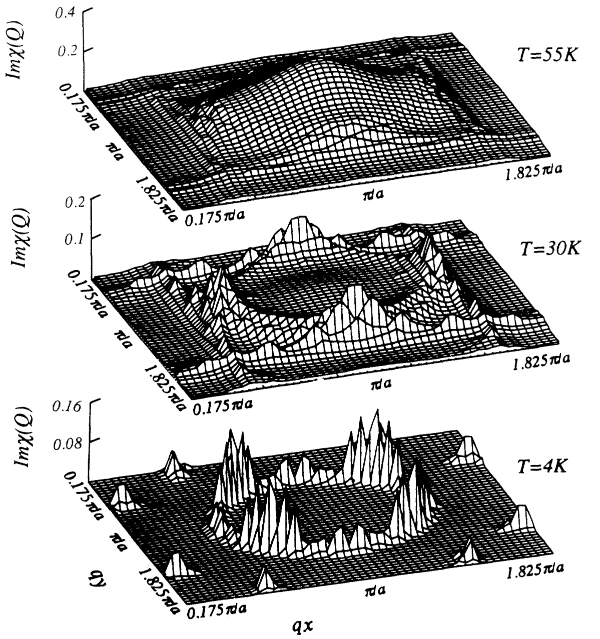

FIG. 4. Predicted wave-vector dependence of Imχ at $\omega=2.5$ meV for $YBa_2Cu_3O_{6.6}$ at various temperatures below T_c. Parameters are chosen as in Fig. 2.

While related perditions are in the literature,[4,5] ours is the first to present these predictions in a quantative context using models for which the normal state **q** structure is in reasonable semiquantitative agreement with experiment for the two different cuprates. In addition, our calculations for this **q** dependence are based on parametrizations which yield agreement with low T,ω anomalies observed in recent neutron data.[1] Thus whether the low-temperature upturns in Imχ vs T observed[1] in the YBaCuO family, are associated with the $d_{x^2-y^2}$ state can now be directly tested. Note that these upturns in the neutron data are closely related to those of the NMR anisotropy which are argued[4,5] to be among the strongest evidence in favor of the d-wave pairing state. It is also clear that these previous calculations of the NMR relaxation should be revisited using the present models which can address the differences in the normal state spin dynamics of the two cuprates. Our analysis of the neutron data suggests that the zero temperature gap $2\Delta(T=0)/kT_c$ lies somewhere[9] between 1.5 and 3.0 for both cuprates. By contrast the best fits to the NMR data[5] suggest that this ratio lies somewhere between 6.0 and 8.0.

Although lifetime effects have been omitted from our considerations, recent data on the lifetime in the superconducting state[12] imply that these effects are negligible at the low temperatures where the gap node contributions are dominant. Hence these effects should not alter in any significant way the central predictions of this paper. Of greater concern, however, are bilayer interactions which are known to be important[13] in the YBaCuO family. These contributions, which are more difficult to treat since they also involve assumptions about the nature of interlayer pairing, will be discussed in future work.

We end by noting that the preferred system for observing gap node effects is in slightly deoxygenated YBaCuO. Here the gap size appears to be larger than in the optimally doped LaSrCuO system. Therefore, the neutron frequencies do not need to be extremely small. At the same time, away from O_7, there is sufficient magnetic scattering in the normal state. In $YBa_2Cu_3O_{6.6}$, a scan along the zone diagonal direction at $T\sim4$ K and $\omega\sim3$ meV should reveal a double peaked structure, if this system is indeed a d-wave superconductor.

Note added in proof. Different conclusions have been reached by Bulut and Scalapino [Phys. Rev. B **47**, 3419 (1993)]. We believe these differences are a consequence of strong nesting effects associated with one- vs three-band models and of close proximity to a magnetic instability. It should also be noted that Sternlieb *et al.* [Phys. Rev. B **47**, 5320 (1993)] have now failed to observe any incommensurate neutron structure in $YBa_2Cu_3O_{6.6}$. This should be viewed as a potential conflict wiht the standard d-wave interpretation of NMR anisotropy data.

We thank J. P. Lu, R. Birgeneau, T. Thurston, J. M. Tranquada, and L. Coffey for stimulating conversations. We are grateful to T. Thurston for providing us with raw data prior to publication. This work was supported by NSF-STC Grant No. STC-8809854.

[1] T. E. Mason, G. Aeppli, and H. A. Mook, Phys. Rev. Lett. **68**, 1414 (1992); B. J. Sternlieb *et al.*, Phys. Rev. B **47**, 5320 (1993); T. R. Thurston *et al.*, Phys. Rev. B **46**, 9128 (1992).

[2] J. Martindale *et al.*, Phys. Rev. Lett. **68**, 702 (1992).

[3] P. C. Hammel *et al.*, Phys. Rev. Lett. **63**, 1992 (1989).

[4] J. P. Lu, Phys. Rev. Lett. **68**, 125 (1992); J. P. Lu, Mod. Phys. Lett. B **6**, 547 (1992).

[5] N. Bulut and D. J. Scalapino, Phys. Rev. Lett. **68**, 706 (1991); Phys. Rev. B **45**, 2371 (1992).

[6] Y. Zha, Q. Si, and K. Levin, Physica C **201**, 289 (1992); Qimiao Si *et al.*, this issue, Phys. Rev. B **47**, 9055 (1993).

[7] S.-W. Cheong *et al.*, Phys. Rev. Lett. **67**, 1791 (1991).

[8] J. Rossat-Mignod *et al.*, Physica C **185-189**, 86 (1991).

[9] The neutron data for the LaSrCuO and YBaCuO systems could be fit for gap parameters in the range $1.5<2\Delta(T=0)/kT_c<2.5$ and $2<2\Delta(T=0)/kT_c<2.7$, respectively.

[10] The resolution function of Thurston *et al.* is elliptical in shape with the long axis perpendicular to the scanning direction. Thus scanning along $q_x+q_y=(2+\delta)\pi$, at $[(1+\delta)\pi,\pi]$, the instrument may pick up part of the nodal contribution at $[(1+\delta/2)\pi,(1-\delta/2)\pi]$.

[11] It should be noted however, that if the parameter $2k_f$ is considerably smaller than our estimates, there may be some nodal contribution at the commensurate point. A theory which includes a Fermi surface with two interacting bilayers will, in principle, lead to smaller $2k_f$ for one of the layers.

[12] D. B. Romero *et al.*, Phys. Rev. Lett. **68**, 1590 (1992).

[13] J. M. Tranquada *et al.*, Phys. Rev. B **40**, 4503 (1989).