# Neutron Scattering as a Probe for Unconventional Superconducting States

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We show that in the unconventional pairing state of a superconductor, i.e., the state with nodes in its excitation spectrum, the dynamical structure factor $S(\mathbf{q},\omega)$ has anomalous resonance peaks in $\mathbf{q}$ space. These resonance peaks are due to quasiparticle scattering between different nodes on the Fermi surface. The symmetry of the order parameter and the Fermi-surface geometry uniquely determine the resonance pattern. Observation of such peaks by neutron scattering will unambiguously determine the symmetry of the superconducting pairing state. Quantitative calculations are carried out in two dimensions to demonstrate this unusual effect. Applications to high-$T_c$ oxide superconductors are discussed.

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To understand the microscopic mechanism of a superconductor it is crucial to identify the symmetry of the order parameter and the associated normal-fluid excitations. Traditionally one measures the temperature dependence of low-energy excitations to identify the symmetry of a pairing state. If the state is nodeless, i.e., if there exists a minimum gap $\Delta_0$ for the excitations, then the density of normal-fluid excitation decreases exponentially in temperature $\rho_n(T)\sim e^{-2\Delta_0/kT}$. Thus most thermodynamic quantities such as specific heat, magnetic susceptibility, and nuclear relaxation rate, etc., will depend on temperature exponentially. On the other hand, if the pairing state is unconventional, i.e., with nodes in its excitation spectrum, then in general $\rho_n(T)\sim(T/T_c)^n$ at low temperature with the exponent $n$ determined by the symmetry of the order parameter. Thus most properties will follow a power law in temperature from which one can determine the symmetry of the pairing state [1]. Therefore in principle it should be easy to distinguish the nodeless state from the one with nodes.

However, in real materials, particularly in high-$T_c$ superconductors (HTSC), there are strong scatterings which make the lifetime of a quasiparticle finite. The scattering could be due to, for example, the electron-phonon interaction, dynamical spin fluctuations, or impurities [2]. As a result, the density of states below the gap is no longer zero for a nodeless state, nor does it follow a simple power law for the state with nodes. This makes the identification of the intrinsic temperature dependence difficult [3,4]. Many experiments do not have a unique interpretation. They can be equally well fitted by the $s$-wave or the $d$-wave symmetry using different scattering rates and coupling constants [5]. Therefore it is highly desirable to have a direct identification of the pairing state symmetry without relying on the temperature dependence of the quantity measured.

In this Letter we point out that there is a clear way to identify the symmetry of a pairing state by examining the $\mathbf{q}$ dependence of the dynamical structure factor $S(\mathbf{q},\omega)$ at low frequency. The conclusion is that in the state with nodes $S(\mathbf{q},\omega)$ has anomalous resonance peaks at large momentum transfer $\mathbf{q}$. The peak positions are uniquely determined by the Fermi-surface geometry and the symmetry of the pairing state order parameter. Neutron scattering, which has been pivotal in revealing the antiferromagnetic fluctuation around $\mathbf{q}=(\pi,\pi)$ in the normal state [6,7], should be an ideal tool to study such unusual resonances at large $\mathbf{q}$. Recent advances in the field have made it feasible to map out a substantial part of the Brillouin zone at frequencies much smaller than the superconducting gap [7]. It is likely that the anomalous peaks studied in this paper, if they exist, are measurable by neutron scattering. This should provide a unique scheme to identify the superconducting pairing state symmetry.

Considering a BCS singlet pairing state [8], the quasiparticle excitation spectrum is given by

$$E(\mathbf{k})=\{[\epsilon(\mathbf{k})-\mu]^2+|\Delta(\mathbf{k})|^2\}^{1/2}, \qquad (1)$$

where $\epsilon(\mathbf{k})$ is the normal-state conduction band and $\mu$ the chemical potential. For the isotropic $s$ wave, the gap function $\Delta(\mathbf{k})$ is $\mathbf{k}$ independent and nonzero. There exists a minimum energy $2\Delta$ to create a particle-hole pair. So for all quasiparticle scattering processes with $\omega=\epsilon(\mathbf{k}+\mathbf{q})-\epsilon(\mathbf{k})<2\Delta$, the cross section will be exponentially small at low temperature because of energy conservation. Furthermore there is no unique momentum transfer $\mathbf{q}$ which characterizes the superconducting state.

For a state with nodes, there exist planes (lines in 2D) where the gap function $\Delta(\mathbf{k})$ is zero due to the symmetry. At the intersections of these planes and the Fermi surface the quasiparticle excitation energy is zero. Let these points be labeled by $\mathbf{k}_i$; then

$$E(\mathbf{k}_i)=0,\quad \Delta(\mathbf{k}_i)=0,\quad \mathbf{k}_i\in\mathbf{k}_F\,. \qquad (2)$$

Clearly at low temperature the quasiparticles are concentrated around the vicinity of $\mathbf{k}_i$. Therefore the low-frequency scattering processes will be dominated by quasiparticles with these momenta. Obviously there are two distinct types of scattering processes. One involves the scattering of quasiparticles around the same node. The momentum transfer $\mathbf{q}$ of such a scattering is very small (compared with $k_F$) for $\omega < \Delta \ll \epsilon_F$. The second type of scattering carries a quasiparticle from one node to a different node. The momentum transfer of such a scattering is on the order of $2k_F$, which is large. It is this latter scattering process that gives rise to the resonance peaks in $S(\mathbf{q},\omega)$. The positions of these peaks are uniquely determined by the symmetry of the pairing state and the geometry of the Fermi surface. For neutron scattering the small-$\mathbf{q}$ process is difficult to observe because of the underlying Bragg peaks from lattice structure. It is the large-momentum-transfer process which could be measurable.

The neutron-scattering cross section is proportional to the dynamical structure factor $S(\mathbf{q},\omega)$ which is related to the imaginary part of the susceptibility,

$$S(\mathbf{q},\omega) = [1+n(\omega)]\,\mathrm{Im}\chi(\mathbf{q},\omega)\,, \tag{3}$$

where $n(\omega) = 1/[\exp(\omega/T)-1]$ is the Bose distribution function. In the BCS superconducting state, the spin-spin correlation function is given by [9]

$$\begin{aligned}\chi(\mathbf{q},\omega) = &\sum_k \frac{1}{2}\left(1+\frac{\xi(\mathbf{k}+\mathbf{q})\xi(\mathbf{k})+\Delta(\mathbf{k}+\mathbf{q})\Delta(\mathbf{k})}{E(\mathbf{k}+\mathbf{q})E(\mathbf{k})}\right)\frac{f(E(\mathbf{k}+\mathbf{q}))-f(E(\mathbf{k}))}{\omega-[E(\mathbf{k}+\mathbf{q})-E(\mathbf{k})]+i\Gamma}\\ &+\sum_k \frac{1}{4}\left(1-\frac{\xi(\mathbf{k}+\mathbf{q})\xi(\mathbf{k})+\Delta(\mathbf{k}+\mathbf{q})\Delta(\mathbf{k})}{E(\mathbf{k}+\mathbf{q})E(\mathbf{k})}\right)\frac{1-f(E(\mathbf{k}+\mathbf{q}))-f(E(\mathbf{k}))}{\omega-[E(\mathbf{k}+\mathbf{q})+E(\mathbf{k})]+i\Gamma}\\ &+\sum_k \frac{1}{4}\left(1-\frac{\xi(\mathbf{k}+\mathbf{q})\xi(\mathbf{k})+\Delta(\mathbf{k}+\mathbf{q})\Delta(\mathbf{k})}{E(\mathbf{k}+\mathbf{q})E(\mathbf{k})}\right)\frac{f(E(\mathbf{k}+\mathbf{q}))+f(E(k))-1}{\omega+[E(\mathbf{k}+\mathbf{q})+E(\mathbf{k})]+i\Gamma}\,.\end{aligned} \tag{4}$$

Here $\xi(\mathbf{k}) = \epsilon(\mathbf{k}) - \mu$ and $f(E) = 1/[\exp(E/T)+1]$ is the Fermi function. In Eq. (4) the effect of the pair-breaking scattering is incorporated by a finite scattering rate $\Gamma$. In principle, one should do this self-consistently and include the correction of the self-energy into the single-particle Green function [10]. However, our emphasis here is the $\mathbf{q}$ dependence of $\chi(\mathbf{q},\omega)$ at fixed temperature. As we will show later the resonance phenomenon is quite insensitive to the different scattering rates (see Fig. 3).

The first term in Eq. (4) is due to the quasiparticle scatterings. At low temperature it dominates the last two terms which are due to creation and destruction of particle-hole pairs. It is clear from Eq. (4) that there exist resonances in $\chi(\mathbf{q},\omega)$ when

$$\omega = E(\mathbf{k}+\mathbf{q}) - E(\mathbf{k})\,. \tag{5}$$

For small $\omega$, this is possible only when $\mathbf{q}$ is near $\mathbf{q}_0 = \mathbf{k}_i - \mathbf{k}_j$. Since $\mathbf{k}_{i,j}$ are on the Fermi surface $\mathbf{q}_0$ is of the order of $2k_F$, far away from the Brillouin-zone center.

To demonstrate this effect, we did explicit calculations on a two-dimensional square lattice with the nearest-

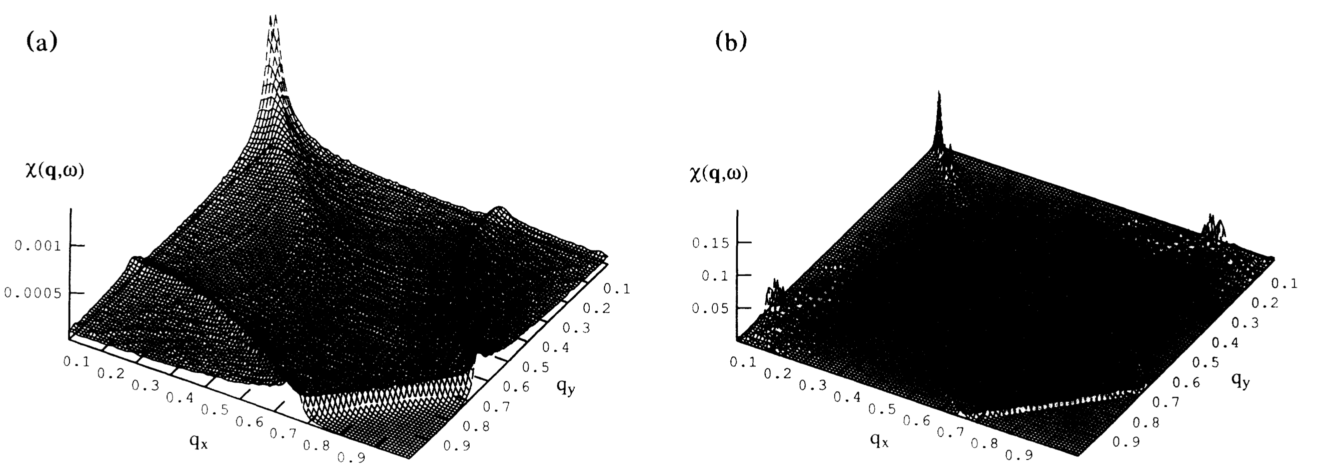


FIG. 1. The imaginary part of the dynamical susceptibility $\mathrm{Im}\chi(\mathbf{q},\omega)$ plotted as a function of $\mathbf{q}$ in the first quadrant of the Brillouin zone. The parameters used are $\mu = -1.0$, $T_c = 0.02t$, $\omega = 0.5T_c$, $T = 0.2T_c$, $\Gamma = 0.05T_c$, and $2\Delta/T_c = 3.5$, where $\Delta$ is $\max[\Delta(\mathbf{k})]$ in the $d$-wave case. (a) The $s$-wave case. $\mathrm{Im}\chi(\mathbf{q},\omega)$ has the same features as in the normal state except that the magnitude is 2 orders of magnitude smaller. The ridge structure is the remnant of the tight-binding Fermi-surface structure which is also present in the normal state. (b) The $d$-wave case with $\cos(k_x) - \cos(k_y)$ symmetry. One clearly sees the anomalous resonance peaks around momentum transfer $(2k_F,2k_F)$, $(2k_F,0)$, and $(0,2k_F)$. These peaks are not present above $T_c$. Note that the amplitudes of these peaks are 2 orders of magnitude larger than the background, which is of the same order as in the $s$-wave case. (See Fig. 3 for a detailed comparison.) Because of the nesting, the peak at $(2k_F,2k_F)$ is stronger than that at $(2k_F,0)$. Also, one observes that the spread of the peak in one direction is much narrower than in the other.

neighbor tight-binding band

$$\epsilon(\mathbf{k}) = -2t[\cos(k_x)+\cos(k_y)]\,. \qquad (6)$$

Three different symmetries of the pairing state were investigated: (1) Isotropic $s$ wave,

$$\Delta(k) = \Delta_0\,; \qquad (7a)$$

(2) $d$ wave with $x^2-y^2$ symmetry,

$$\Delta(k) = \Delta_0[\cos(k_x)-\cos(k_y)]\,; \qquad (7b)$$

(3) $d$ wave with $xy$ symmetry,

$$\Delta(k) = \Delta_0 \sin(k_x)\sin(k_y)\,. \qquad (7c)$$

Figure 1(a) shows the $\mathrm{Im}\chi(\mathbf{q},\omega)$ calculated from Eq. (4) for the $s$-wave case. It is clear there are no special features besides the small-$\mathbf{q}$ peak. The ridge structure is the remnant of the Fermi-surface geometry. Compared with the same quantity calculated at $T_c$, the landscape is the same except that the magnitude is suppressed by 2 orders of magnitude uniformly (see Fig. 3). There is little chance that this ridge structure can be observed in the superconducting state [11].

A representative result of the calculations for the case of $d$ wave with $x^2-y^2$ symmetry is shown in Fig. 1(b). The parameters used are the same as those in Fig. 1(a). The excitation spectrum now has four nodes at $\mathbf{k} = (\pm k_F, \pm k_F)$ where $\cos(k_F) = \mu/4t$ (Fig. 2). Besides the small-$\mathbf{q}$ peak, the scattering between different nodes gives several strong peaks at large $\mathbf{q}$. The locations of these peaks are $\mathbf{q}_0 = (2k_F, 2k_F)$, $(2k_F, 0)$, $(0, 2k_F)$ (the corresponding scattering processes are indicated by arrows in Fig. 2) and their equivalent points in the Brillouin zone. Note the peak position along the (1,1) direction is farther away from zone center than those along the (1,0) or (0,1) direction. This qualitative result is independent of the Fermi-surface shape provided that it has the symmetry of the square lattice.

An important result is that the amplitude of the peaks, in the $d$-wave case, is of the same order of magnitude as that in the normal state, or larger (see Fig. 3). This should help the observation of these peaks.

If the symmetry of the $d$ wave is of $xy$ type, the nodes are at $(\pm k_F', 0)$ and $(0, \pm k_F')$, where $\cos(k_F') = \mu/2t - 1$. Then resonant peaks are located near $\mathbf{q}_0 = (\pm k_F', \pm k_F')$, $(2k_F', 0)$, $(0, 2k_F')$, and their equivalent points. In this case the peaks along the (1,0) or (0,1) direction are farther away from the zone center than those along the (1,1) direction. (A simple drawing of excitation spectrum similar to that of Fig. 2 should convince the reader that this is true.) The result is similar to Fig. 1(b) but rotated along the $z$ axis by $\pi/4$. Therefore the locations of peaks can be used to uniquely determine the symmetry of the pairing state.

The exact values of peak positions depend on $\omega$, determined by $\omega = E(\mathbf{k}_F+\mathbf{q}) - E(\mathbf{k}_F)$. For small $\omega$ the deviation from $\mathbf{q}_0$ is small and one can expand around $\mathbf{q}_0$. For example, in the $d_{x^2-y^2}$ case near $\mathbf{q}_0 = (2k_F, 2k_F)$ one obtains

$$\omega = 2t\sin(\mathbf{k}_F)[(\tilde{q}_x+\tilde{q}_y)^2 + (\Delta_0/2t)^2(\tilde{q}_x-\tilde{q}_y)^2]^{1/2}\,, \qquad (8)$$

where $\tilde{\mathbf{q}}$ are measured with respect to $\mathbf{q}_0$. Because $\Delta_0/2t \ll 1$, the spread of the peak along the $\tilde{q}_x = \tilde{q}_y$ direction is much smaller than that along the $\tilde{q}_x = -\tilde{q}_y$ direction. Therefore in doing neutron scattering one should

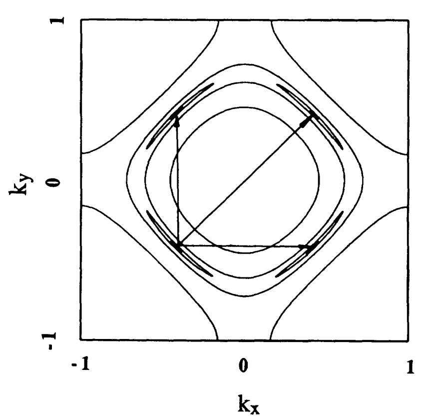


FIG. 2. The contour plot of the excitation spectrum $E(\mathbf{k})$ [Eq. (1)] for the $d$ wave with $\cos(k_x)-\cos(k_y)$ symmetry. Successive contours are in logarithmic scale. The scattering processes which give rise to the resonance peaks shown in Fig. 1(b) are indicated by arrows.

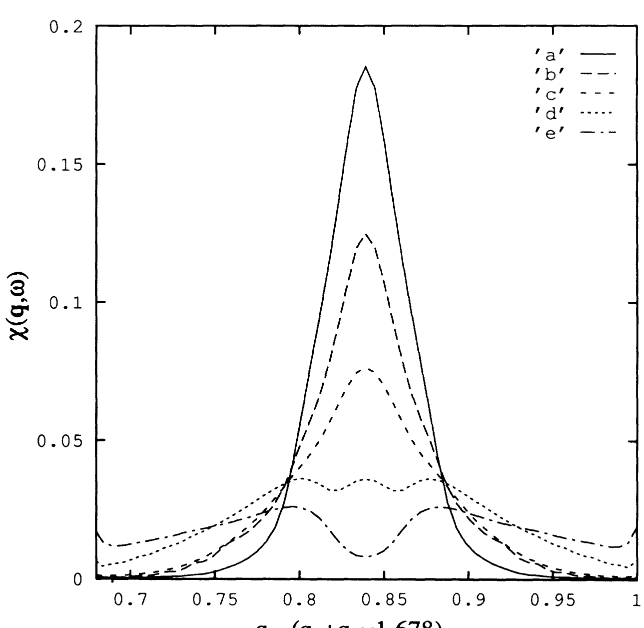


FIG. 3. $\mathrm{Im}\chi(\mathbf{q},\omega)$ plotted along the line $q_x+q_y = 4k_F$ for various $T$ and $\Gamma$ in the $d$-wave case. Other parameters are the same as in Fig. 1. Curve $a$: $t = T/T_c = 0.1$, $\gamma = \Gamma/T_c = 0.1$. $b$: $t = 0.1$, $\gamma = 0.2$. $c$: $t = 0.2$, $\gamma = 0.2$. $d$: $t = 0.5$, $\gamma = 0.2$. $e$: $t = 1.0$, $\gamma = 0.5$.

take this into account.

To examine the robustness of the resonant peak, in Fig. 3 we show a cut of $\mathrm{Im}\chi(\mathbf{q},\omega)$ along the line $q_x+q_y=4k_F$ for several different temperatures and scattering rates. One sees that in the parameter region $T<\omega$, $\Gamma<\omega$ the peak is pronounced. For fixed $\Gamma$ and decreasing $T$, or fixed $T$ and decreasing $\Gamma$, the peak intensity increases. At $T=0.1T_c$, with $\Gamma=0.1T_c$, the peak intensity is almost 10 times larger than that at $T_c$. This is a very unusual temperature dependence as most other quantities in the superconducting state decrease with temperature.

If in the high-$T_c$ oxide superconductors the pairing state has nodes, as recent NMR data seem to suggest [3,5], then we believe these resonance peaks could be measurable and should be pursued. As a result of the high $T_c$ the condition $T<\omega<T_c<\Delta$ can be satisfied. The identification of these peaks will once and for all settle the question concerning the symmetry of the pairing state. This will not only help in interpreting other experiments but will put a strong constraint on the possible mechanism of superconductivity.

In conclusion, we have shown that in the unconventional superconducting state with nodes the dynamical structure factor should exhibit anomalous resonance peaks in $\mathbf{q}$ space. These peaks are due to the fact that the low-frequency excitations are concentrated around a few nodes on the Fermi surface. Scattering of quasiparticles between different nodes leads to anomalous resonance peaks at large momentum transfers in $\mathrm{Im}\chi(\mathbf{q},\omega)$. The positions of these peaks are uniquely determined by the symmetry of the pairing state and the Fermi-surface geometry. Quantitative calculations on a 2D square lattice show that this feature is relatively robust and independent of detailed parameters of the pairing state. The peak intensity is predicted to increase with decreasing temperature, a rather unusual dependence. We suggest that inelastic neutron scattering should be an ideal tool to search for such an effect in the high-$T_c$ oxide superconductors.

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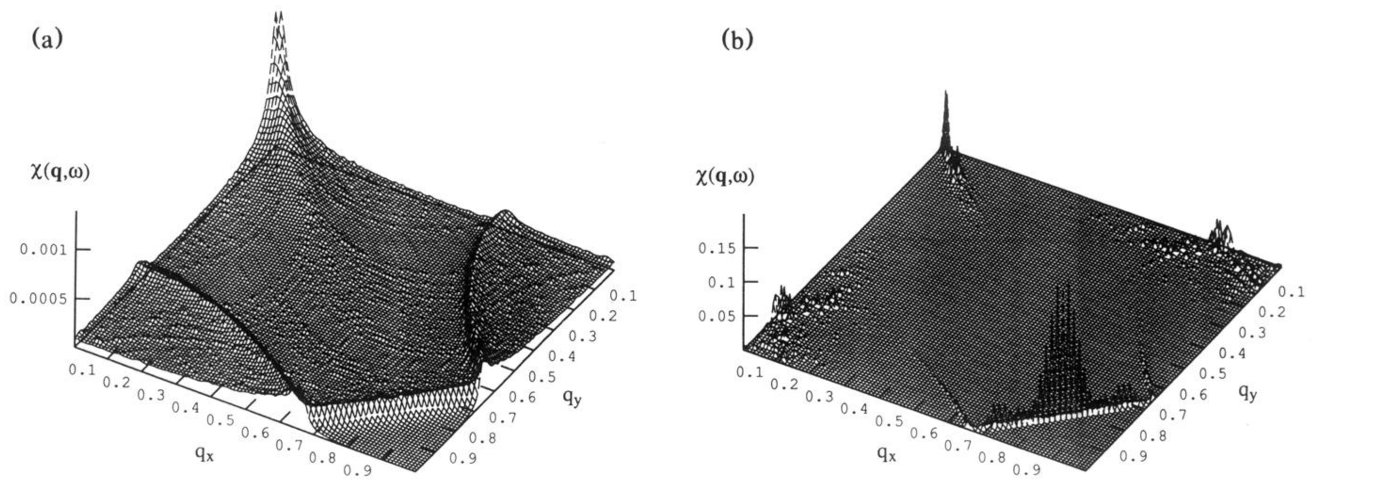


FIG. 1. The imaginary part of the dynamical susceptibility $\mathrm{Im}\chi(\mathbf{q},\omega)$ plotted as a function of $\mathbf{q}$ in the first quadrant of the Brillouin zone. The parameters used are $\mu=-1.0$, $T_c=0.02t$, $\omega=0.5T_c$, $T=0.2T_c$, $\Gamma=0.05T_c$, and $2\Delta/T_c=3.5$, where $\Delta$ is $\max[\Delta(\mathbf{k})]$ in the $d$-wave case. (a) The $s$-wave case. $\mathrm{Im}\chi(\mathbf{q},\omega)$ has the same features as in the normal state except that the magnitude is 2 orders of magnitude smaller. The ridge structure is the remnant of the tight-binding Fermi-surface structure which is also present in the normal state. (b) The $d$-wave case with $\cos(k_x)-\cos(k_y)$ symmetry. One clearly sees the anomalous resonance peaks around momentum transfer $(2k_F,2k_F)$, $(2k_F,0)$, and $(0,2k_F)$. These peaks are not present above $T_c$. Note that the amplitudes of these peaks are 2 orders of magnitude larger than the background, which is of the same order as in the $s$-wave case. (See Fig. 3 for a detailed comparison.) Because of the nesting, the peak at $(2k_F,2k_F)$ is stronger than that at $(2k_F,0)$. Also, one observes that the spread of the peak in one direction is much narrower than in the other.