Neutron Scattering Signature of d-density Wave Order in the Cuprates

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An ordered d-density wave (DDW) state has been proposed as an explanation of the pseudogap phase in underdoped high-temperature superconductors. The staggered currents associated with this order have signatures which are qualitatively different from those of ordered spins. We apply the order parameter theory to an orthorhombic bilayer system and show that the expected magnitude as well as the momentum, energy, and polarization dependence of the consequent neutron scattering is consistent with the findings of a recent experiment.

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a. Introduction. The pseudogap phase of the underdoped high-temperature superconducting cuprates is notable in its departure from the behavior of a conventional metal. Recently, it was proposed \( \text{[2]} \) that this unusual behavior is due to a new broken symmetry, termed d-density wave (DDW) \( \text{[3]} \), which competes with superconductivity. The crucial feature of this order is a staggered pattern of orbital currents that spontaneously appear at the onset of the pseudogap phase. The underdoped superconducting state coexists with this order. This proposal of the existence of an order parameter is fundamentally different from the proposal of staggered current fluctuations \( \text{[4]} \).

We discuss here the experimental signatures of this unusual state vis-à-vis a recent neutron scattering experiment in underdoped YBCO\( _{6.6} \) \( \text{[5]} \). This experiment has identified rods of Bragg scattering (within the energy resolution window of 1 meV) arising from small magnetic moments which increase in strength below the pseudogap temperature, with a further increase below the superconducting transition temperature. The in-plane wavevector is \( Q = (\pi/a, \pi/a) \), where \( a \) is the in-plane lattice spacing, and the intensity is at the level of the background for perpendicular wavevector transfer \( q_z = 0 \), implying an “antiferromagnetic” coupling between the layers within a bilayer. Both of these features are precisely the same as those observed in the undoped antiferromagnet, where the signal is from ordered Cu moments. Such scattering from ordered Cu moments deep in the superconducting state would be surprising to say the least.

However, there are four important ways in which these experimental results differ from the corresponding ones for the undoped antiferromagnet: (1) the magnitude of the observed moments is of order \( 2 \times 10^{-2} \mu_B \) at 10 K, which is 50 times smaller than that observed in the undoped antiferromagnet; (2) the intensity decreases rapidly with scattering wavevector \( q \) in a manner which is inconsistent with ordered Cu spins; (3) aside from the elastic peak, there is no significant intensity up to 20 meV. In other words, there is no evidence for the Goldstone modes (magnons) which must be present if spin-rotational symmetry is broken. (4) Finally, it is found that there is no 3D order down to the lowest temperatures studied despite the fact that the in-plane spatial correlations are resolution limited, with a correlation length greater than 200Å.

We argue that these four are the most robust features of the experiment and that they fit the explanation of DDW order but are inconsistent with spin order. In order to understand the experiment, we first introduce the theoretical framework of the DDW. Next, we address the ratio of the intensities for various Bragg reflections and then examine the ratio of the spin-flip to non-spin flip scattering for polarized neutrons.

b. Currents associated with DDW order. The DDW order parameter \( \text{[3]} \) is a spin singlet particle-hole condensate given by the equal time correlation function \( (\alpha, \beta \text{ are spin indices}): \langle c_{\mathbf{k} \uparrow} \mathbf{Q} c_{\mathbf{k} \downarrow} \mathbf{Q} \rangle = i P_{\mathbf{Q}} \mathbf{Y}(\mathbf{k}) \delta_{\alpha \beta} \), where \( P_{\mathbf{Q}} \mathbf{Y}(\mathbf{k}) \) is a real order parameter, and \( c_{\mathbf{k} \alpha} \) is an electron destruction operator. The internal degree of freedom of the condensate is given by the \( d_{x^2−y^2} \) function \( \mathbf{Y}(\mathbf{k}) = \frac{1}{2} (\cos k_x a + \cos k_y a) \). Charge density is not modulated in the DDW because \( \sum_{\mathbf{k}} \mathbf{Y}(\mathbf{k}) = 0 \) ! It is actually the current density that is modulated, as we discuss below. A conventional charge density wave would occur if the internal orbital degree of freedom of the particle-hole condensate were of s-wave character, e.g. if \( \mathbf{Y}(\mathbf{k}) \) were a constant. The terminology is a very convenient way of classifying the internal orbital degree of freedom of a particle-hole condensate.

Neutron scattering from the DDW state is determined by its associated current distribution. This distribution depends on the details of the current paths along which electrons move in a copper-oxide superconductor. As we discuss below, unpolarized neutron scattering is fairly insensitive to these details, but the ratio of non-spin-flip to spin-flip polarized neutron scattering is not.

The charge and spin distribution of an electron at a Cu site is given by the well-known Cu form factor (see below). However, due to mixing of different orbitals, the profile of current flow is more complicated; it will be sensitive to many-body effects when the current is substantial, and it will be affected by the bi-layer coupling and, in YBCO, by the influence of the chains. We make...
a simple and tractable Ansatz for this current distribution, including the effects of orthorhombicity due to the chains. The orthorhombicity, even though it is small in terms of the lattice constants, is well known to give rise to remarkable anisotropy; for example the $a-b$-anisotropy of the superfluid density can be as large as $2-3$, and perhaps even larger [5].

Thus, we define the Fourier transform of the expectation value of the current, $\langle \mathbf{j}(\mathbf{q}) \rangle$, satisfying $\mathbf{q} \cdot \langle \mathbf{j}(\mathbf{q}) \rangle = 0$, by

$$
\langle \mathbf{j}(\mathbf{q}) \rangle \propto \Phi_Q \sum_{\mathbf{G}_i} \delta_{\mathbf{q} \parallel \mathbf{G}_i} f(\mathbf{q})
$$

$$
\times \left[ (\alpha(\mathbf{q}) \frac{x}{q_x} - \beta(\mathbf{q}) \frac{y}{q_y}) - (\alpha(\mathbf{q}) - \beta(\mathbf{q})) \frac{z}{q_z} \right],
$$

where $\mathbf{q}_i = (q_x, q_y)$, $\mathbf{G}_i = (G_x, G_y)$, and $\mathbf{G} = (2\pi H/a, 2\pi K/a, 2\pi L/c)$ is a magnetic reciprocal lattice vector, where the lattice constants are $a = 3.86\, \text{Å}$ and $c = 11.82\, \text{Å}$; we ignore slight orthorhombicity in the lattice constants, but not in the physical quantities in which it is magnified. Note that $f(\mathbf{q}) = \sin(\frac{d \mathbf{q} \cdot \mathbf{a}}{2})$, where $d = 3.25\, \text{Å}$ is the intrablayer separation. The temperature and doping dependence of the currents are determined by the DDW order parameter $\Phi_Q$, which must appear as a thermodynamic phase transition.

If the electrons behaved classically and took straight line trajectories between point-like Cu sites, we would have $\alpha(\mathbf{q}) = \beta(\mathbf{q}) = 1$. In reality, however, these are non-trivial functions of $\mathbf{q}$. Guided by the phenomenology, we choose the simplest model in which these are non-trivial functions of $q_z$ alone. At long scales, the current distribution is essentially classical, $\alpha(0) = \beta(0) = 1$. At short distances, the thickness of the current trajectories becomes apparent. We assume that currents flowing along the $\hat{y}$ axis are more spread out, reflecting the underlying orthorhombicity of YBCO, $\alpha(q_z) \neq \beta(q_z)$ for $q_z$ large. In tetragonal materials, the asymmetry could be due to spontaneous symmetry breaking [6].

The requirement $\alpha(0) = \beta(0) = 1$ automatically leads to zero average current along $\hat{z}$ in the bilayer model because of the bilayer form factor $f(\mathbf{q})$. If we want to interpret $\alpha(q_z)$ and $\beta(q_z)$ to be merely form factors and want to adhere to the original order parameter [7][8][9], we can do so if we stipulate that $\alpha(q_z) - \beta(q_z) \sim q_z^2$, as $q_z \to 0$. This will make the average $z$-component of the current vanish (as it must) for the single layer model for which the same expression for the current holds, except that the factor $f(\mathbf{q})$ is missing.

The orbital currents give rise to magnetic fields of order $10G$, [8][9][10] and a very small magnetic moment, of order $4 \times 10^{-7} \mu_B$, consistent with the experimental estimate of order $2 \times 10^{-2} \mu_B$ [11].

c. Unpolarized elastic neutron scattering. We first recall spin-scattering and then discuss scattering from orbital currents. The well-known scattering cross section of unpolarized neutrons from localized Cu spins in YBCO, when the net spin moment is along the $c$-axis, is

$$
\frac{d\sigma}{d\Omega}(\mathbf{q}) = \sum_{\mathbf{G}_i} \delta_{\mathbf{q} \parallel \mathbf{G}_i} g^2(\mathbf{q}) f^2(\mathbf{q}) \left( 1 - \frac{q_z^2}{q^2} \right),
$$

where $\mathbf{q}$ is the momentum transfer, and the atomic form factor $g(\mathbf{q})$ is the Fourier transform of the normalized density of unpaired electrons in a single ion $\Omega$. We have assumed that the spins are antiferromagnetically coupled within an YBCO bilayer and that different bilayers are uncoupled, so that the scattering cross-section is proportional to that due to a single bi-layer.

Similarly, there is a formula which applies when the spin moment lies in the $x-y$ plane [11]. If the spin direction in the plane is averaged over magnetic domains or averaged due to spin glass order, this is

$$
\frac{d\sigma}{d\Omega}(\mathbf{q}) = \sum_{\mathbf{G}_i} \delta_{\mathbf{q} \parallel \mathbf{G}_i} g^2(\mathbf{q}) f^2(\mathbf{q}) \left( 1 + \frac{q_z^2}{q^2} \right).
$$

It is worth emphasizing that the quantity in large parentheses in [3] increases with $q_z$, as does its counterpart in the case in which the spins point in some direction in the $a-b$ plane which is not averaged over.

Next, we consider Bragg scattering from orbital currents [12]. The neutron magnetic moment generates a vector potential, $A$, given by $A = \mu \times \frac{(r_r - r_s)}{|r_r - r_s|}$, where $\mu = -1.91(\frac{\mu_B}{m_n})s_n$; $s_n$ is the neutron spin, and $m_n$ is the mass of a neutron; the electron and the neutron coordinates are $r_r$ and $r_s$, respectively. The coupling of the electrons to this gauge field is given in momentum space by the Hamiltonian $H_{\text{int}} = \int \frac{d^3q}{(2\pi)^3} \mathbf{j}(\mathbf{q}) \cdot \mathbf{A}(\mathbf{q})$. The Bragg intensity of unpolarized neutrons is then

$$
\frac{d\sigma}{d\Omega}(\mathbf{q}) \propto \delta_{\mathbf{q} \parallel \mathbf{G}_i} \frac{g^2(\mathbf{q}) f^2(\mathbf{q}) \delta_{\mathbf{q} \parallel \mathbf{G}_i}}{q^2}
$$

$$
\times \left[ \frac{\lambda^2(q_z)}{q_z^2} + \frac{1}{q_y^2} + \left( \frac{\lambda(q_z) - 1}{q_z} \right)^2 \right],
$$

where $\lambda(q_z) = \alpha(q_z)/\beta(q_z)$. We emphasize that there are no approximations in this formula, such as the SU(n) mean field approximation [13]; it simply follows from the assumption of DDW order.

We estimate the factor $\beta^2(q_z)$ by $g^2(\mathbf{q})$ for Cu spins found by Shamoto et al. [14] as an upper bound, because $\beta^2(q_z)$ must fall off faster than $g^2(\mathbf{q})$ as the charge distribution for orbital current loops is more spread out than the atomic orbitals. The case for orbital currents made below will be even stronger with the true form factor.

The most salient feature distinguishing (2) and (3) by the Hamiltonian $H$ is

$$
\frac{d\sigma}{d\Omega}(\mathbf{q}) \propto \sum_{\mathbf{G}_i} \delta_{\mathbf{q} \parallel \mathbf{G}_i} g^2(\mathbf{q}) f^2(\mathbf{q}) \left( 1 - \frac{q_z^2}{q^2} \right),
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$$

$$
\times \left[ \frac{\lambda^2(q_z)}{q_z^2} + \frac{1}{q_y^2} + \left( \frac{\lambda(q_z) - 1}{q_z} \right)^2 \right],
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where $\lambda(q_z) = \alpha(q_z)/\beta(q_z)$. We emphasize that there are no approximations in this formula, such as the SU(n) mean field approximation $\Omega$; it simply follows from the assumption of DDW order.

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The most salient feature distinguishing (2) and (3) by the Hamiltonian $H$ is
\( \alpha(q_z) \) and \( \beta(q_z) \) are of order unity. The latter form follows from the inherent spatial extent of a current loop and leads to a rapid decay of the scattering cross-section with \( q \).

Let us consider the ratios of the Bragg scattering intensities at various magnetic reciprocal lattice vectors; we will quote the intensities at the representative values, \( \lambda(q_z) = 1 \) and 3, thereby crudely replacing \( \lambda(q_z) \) by a constant for wavevectors larger than or of order \( 2\pi/c \). From [1], we see that the ratio of the intensity at \((0,0,0.5,1)\) to that at \((1.5,1.5,1)\) is 111 at \( \lambda = 1 \), and 23 at \( \lambda = 3 \). Meanwhile in the spin case, the same ratio is 1.9 if the spins lie in the \( a-b \) plane and 1.4 if they are along the \( c \)-axis. The strong suppression of the intensity at \((1.5,1.5,1)\) in the orbital case is consistent with experiments [5,11] where no peak is observed at \((1.5,1.5,1)\) or \((1.5,1.5,2)\), while peaks are observed at \((0.5,0.5,1)\) and \((0.5,0.5,2)\). The absence of an observable peak at \((1.5,1.5,1)\) is inconsistent with spin order.

Let us now contrast the ratios of the intensities at different values of \( q_z \) for spins pointing within the plane, Eq. [8], and orbital currents, Eq. [4], with the form factor correction described above. The ratio of the intensity at the Bragg reflection \((0.5,0.5,1)\) to that at \((0.5,0.5,2)\) is 0.96 for orbital moments at \( \lambda = 1 \), increasing to 1.5 at \( \lambda = 3 \). For spins pointing in the plane it is 0.5. Experimentally [8] it is found to be 1.16±0.16, suggesting orbital currents as the source of scattering. One can easily see from the known Cu form factor [10] that the correction due to the atomic form factor for nearby Bragg reflections, such as \((0.5,0.5,1)\) and \((0.5,0.5,2)\), is negligible.

Moreover, for spin-moments lying parallel to the plane, the ratio of the intensity at \((0.5,0.5,5)\) to \((0.5,0.5,1)\) should be 1.6. This is clearly not the case in the experiment [11] because there is no intensity above the background at \((0.5,0.5,5)\). For orbital currents, the same ratio is 0.2 (which is only an upper bound) at \( \lambda = 1 \) and 0.1 at \( \lambda = 3 \); as a result, there may not be a measurable intensity at \((0.5,0.5,5)\) as discussed in Ref. [8], thus confirming once again the source of the scattering to be orbital currents.

\[ \frac{d\sigma}{dT}_{\alpha-\beta} \propto \frac{1}{q^4} |\langle \alpha | j | \beta \rangle \cdot q \times \langle j(q) \rangle|^2, \tag{5} \]

where \( |\alpha\rangle \) and \( |\beta\rangle \) are the initial and final spin states of the neutrons. It is easy to see that if the neutron polarization is parallel to the scattering vector, the scattering is entirely spin-flip irrespective of \( \langle j(q) \rangle \).

In contrast, if it is perpendicular to the scattering vector, then the scattering can be either spin flip or non-spin flip depending on \( \langle j(q) \rangle \) and \( q \). If \( q = (H,H,L) \) and the neutrons are polarized perpendicular to \( q \) in the \([1,T,0]\) direction, the ratio of the non-spin-flip to spin flip scattering intensities is:

\[ \frac{\text{NSF}}{\text{SF}} = 2 \left( \frac{\lambda(q_z)}{\lambda(q_z) + 1} \right)^2 \left( \frac{Hc}{La} \right)^2 \left[ 1 + \frac{1}{2} \left( \frac{La}{Hc} \right)^2 \right] \tag{6} \]

While this ratio vanishes at \( \lambda = 1 \), it increases rapidly with \( \lambda \) because \( c \gg a \). At \((0.5,0.5,1)\) and \( \lambda = 3 \) it is of order unity, more precisely 1.4, which is similar to the ratios found in the experiments of Refs. [1,2].

**e. Interlayer Coupling.** Earlier, we assumed that the order parameters from the two layers in a bilayer are opposite in sign. Let us see why this should be so. Interactions which couple the density at a site in one layer to the density at a site in another layer will not couple the DDW order parameters in the two layers because they will average over directions in the \( a-b \) plane, giving zero. However, interlayer tunneling will couple the DDW order parameters at second order.

Consider the interlayer tunneling Hamiltonian Eq. [10] between layers \((1)\) and \((2)\): \( H_c = -\sum_{\alpha} \sum_{k} (\langle j^{(1)} \rangle \cdot \langle j^{(2)} \rangle + \text{h.c.}) \), where \( t_{\perp}(k) = (t_{\perp}/4)(\cos k_x a - \cos k_y a)^2 \). Let \( |s \rangle \) be the ground state for \( t_{\perp} = 0 \) with equal DDW order parameters in each layer, while \( |a \rangle \) is the ground state for \( t_{\perp} = 0 \) with DDW order parameters equal and opposite. The energy difference between these two states, per site, is given to lowest non-vanishing order in \( t_{\perp} \) by

\[ \Delta E = -\frac{4\pi}{a^2} \Delta \Phi, \]

where \( \Delta E = \sqrt{\Delta_{\text{DDW}}^2 + \Delta_{\text{DSC}}^2} \), and \( \Delta_{\text{DSC}} \) is the maximum of the superconducting gap, i.e. the antisymmetric state has lower energy. The above result also holds for body-centered tetragonal materials, with the modified tunneling matrix element [4]. Moreover, we see why the coupling between bilayers is so small: the tunneling matrix elements between bilayers are much smaller than those within a bilayer because \( c = 11.82\AA \gg d = 3.25\AA \).

Because the coupling between the unit cells is exceptionally small, we expect to see 2D Bragg rods without 3D order for temperatures which are not exceedingly low; recall that the transition to the DDW state is an Ising transition unlike the corresponding spin problem. Indeed, as remarked earlier, Mook et al. [10] find such Bragg rods.

**f. Inelastic scattering.** We note that because DDW order is Ising-like, there are no Goldstone modes, but a gap in the frequency spectrum. This is consistent with the experimental finding of a spin gap above the elastic peak [10], extending up to an energy of 20 meV. For spin-ordering, a Bragg peak will necessarily lead to a Goldstone mode (the Ising anisotropy in these materials is negligibly small; see, for example, Ref. [10]), which is continuously connected to it. It is not possible to have one without the other.

The magnetic resonance peak [10] in inelastic scattering at 34 meV at the same momentum, \( Q \), also testifies against the spin-ordering scenario. If the spins are ordered, this resonance should be at zero energy – i.e. it should not be distinct from the Bragg peak with the same
quantum numbers! Furthermore, in the presence of broken spin-rotational symmetry, excitations are not organized into SU(2) multiplets (but, rather, U(1) multiplets if rotations about one axis are preserved), so the resonance could not possibly be a triplet mode as suggested in Ref. \[4\].

**g. Global Phase Diagram.** According to the interpretation proposed here, DDW order has been observed in neutron scattering in YBCO$_{6.6}$ [5]. On the other hand, neutron scattering in YBCO$_{6.35}$ produces a signal which is 10 times more intense, is due to moments which lie in the $a-b$ plane and is believed to be due to spin glass ordering \[5\]. This not only supports the hypothesis that the effect observed in YBCO$_{6.6}$ is due to something other than spin ordering – namely DDW ordering – but also suggests that DDW order disappears at low doping in favor of spin glass order and, possibly, other competing orders as well \[5\].

In neutron scattering experiments on YBCO$_{6.5}$, Sidis et al. \[2\] find similar magnetic Bragg peaks with a ratio of 0.67 between the intensity at (0.5, 0.5, 1) and that at (0.5, 0.5, 2). This is intermediate between the observed ratio in the putative DDW state of YBCO$_{6.6}$ and the observed ratio in the spin glass of YBCO$_{6.35}$. The measurements of Ref. \[2\] seem to be indicative of the spin glass phase. \textit{It is clear therefore that the doping dependence may be crucial, changing the results as we move from YBCO$_{6.5}$ to YBCO$_{6.6}$.}

With the above ideas as inspiration, we propose the global phase diagram of Fig. \[1\].

**h. Summary.** To summarize, the ratio of spin-flip to non-spin-flip scattering depends on details of the current distribution, but the rapid decrease of the scattering intensity with $q$ is a robust feature which follows from the inherent spatial extent of the current loops (which is much larger than a Cu or even an O orbital). We believe that DDW order is the only way of reconciling the constraints following from the wavevector and polarization dependence of the neutron scattering intensity. These phenomenological considerations, although quite tight, clearly await a microscopic understanding.

\textit{Note added:} Recent striking zero-field muon spin relaxation measurements in YBCO$_{6.67}$ and YBCO$_{6.95}$ have uncovered the onset of static magnetism consistent with our DDW picture, and the phase diagram proposed above \[6\], in particular, a phase transition within the superconducting dome.

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