Collective Modes in the Loop Ordered Phase of Cuprate Superconductors

Yan He and C. M. Varma

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Collective Modes in the Loop Ordered Phase of Cuprates

Yan He and C.M. Varma

Department of Physics, University of California, Riverside, CA

Abstract

We show that the two branches of collective modes discovered recently in under-doped Cuprates with a large spectral weight are a necessary consequence of the loop-current state. Such a state has been shown in earlier experiments to be consistent with the symmetry of the order parameter competing with superconductivity in four families of Cuprates. We also predict a third branch of excitations which cannot be discovered by neutron scattering but may be discovered by other techniques. Using parameters to fit the observed modes, we show that quantum fluctuations change the direction of the effective moments in the ground state to lie at an angle to the c-axis as observed in experiments.

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It is generally agreed that the most important theoretical problem posed by the discoveries in the Cuprates is an understanding of the normal states above $T_c(x)$, which present new paradigms in the organization of matter. Specifically these states are the pseudogap state at low dopings and the marginal-fermi-liquid state at intermediate dopings. A novel broken symmetry with a large order parameter, (estimated [3] to be $0.3\mu_B/unitcell$ in Hg(1201) with $T_c = 61K$), has been discovered in four different families of cuprates [1–5] below the pseudogap temperature $T^*(x)$, with $T^*(x) \to 0$ at $x \to x_c$, the quantum critical point. This state can therefore be regarded as a universal feature of the phase diagram of the Cuprates. The symmetry of the state is consistent with the spatial and time-reversal symmetry of a state predicted to arise by formation of ordered loop currents in a microscopic model for the Cuprates [6, 7]. The marginal fermi-liquid properties have been derived from the quantum-critical fluctuations of the observed state[14] and the coupling of the fermions to such fluctuations has been shown [15] to promote d-wave superconductivity with the right scale of T_c.

If there is an unusual order parameter in the pseudogap state, there should also be unusual collective modes there. Recently, inelastic neutron scattering has discovered [8, 9] two branches of weakly dispersive collective modes throughout the Brillouin zone in several underdoped $Hg1201$ for $T \lesssim T^*(x)$. The integrated strength of these modes at low T is estimated to be over 20 times larger than the intensity in the so-called (π, π) resonance [8, 9]. Nearly dispersion free modes have already been indirectly inferred through their dominant coupling to the fermions in ARPES [10] and optical conductivity experiments [11]. The energy and the spectral strength of the newly discovered modes make them the most likely candidates to explain the ARPES and optical conductivity experiments.

Here we present the theory of the newly discovered modes. We show that three branches of excitations must be present in the loop-current ordered state but one of them can not be discovered by neutron scattering. We calculate the frequency dependence of these modes through introducing the quantum-mechancial version of the Ashkin-Teller (AT) model which describes the statistical mechanics of the loop ordered state. Moreover, we resolve the major discrepancy of the order parameter found in polarized neutron Bragg scattering from the predictions of the classical AT model. We show that the quantum-model gives the semi-classical direction of the moments to be lying at an angle to the c-axis and calculate this angle from the parameters needed to fit the observed collective modes. and compare it with

the experiments.

The four possible "classical" domains of the loop current ordered state are shown in Fig. (1). The "classical" order parameter [12] may be characterized by the *anapole* vector [13] $\mathbf{\Omega}$

$$\mathbf{\Omega} = \int_{cell} d^2r(\mathbf{M}(\mathbf{r}) \times \mathbf{r}) \approx \sum_{\mu} \mathbf{M}_{\mu} \times \mathbf{r}_{\mu} \tag{1}$$

where the moment distribution $\mathbf{M}(\mathbf{r})$ is formed due to the currents on the four O-Cu-O triangles per unit-cell as shown. An approximate representation of $\mathbf{\Omega}$ is given by $\mathbf{M}(\mathbf{r}) \approx \mathbf{M}_{\mu}\delta^2(\mathbf{r} - \mathbf{r}_{\mu})$, where $\mathbf{r}_{\mu}$, $\mu = 1, .., 4$ are the locations of the four "sites" in any cell at the centroid of the moment distribution. These sites labelled $S1, .., S4$ in Fig.(1e) have orbital moments $\mathbf{M}_{\mu}$ which are either up or down or zero. The four *classical* domains of Fig.(1e) may be represented by the four values of the angle $\theta = \pi/4, 3\pi/4, 5\pi/4, 7\pi/4$ that $\mathbf{\Omega}$ makes with the $\hat{x}-$axis.

The classical statistical mechanics of the Loop-Current state may be derived from the Ashkin-Teller model, which is given in terms of a pair of Ising spin per unit-cell σ_{i3}, τ_{i3}, whose eigenvalues, ± 1 specify the x and y components of the direction of the vector $\mathbf{\Omega}$ and whose eigenvectors are $|10, 10\rangle$, $|01, 10\rangle$, $|01, 01\rangle$, $|10, 01\rangle$; the first two numbers specify eigenstates of σ_{i3} and the second two of τ_{i3}.

$$H_{AT} = \sum_{(ij)} \frac{J}{4}(\sigma_{i3}\sigma_{j3} + \tau_{i3}\tau_{j3}) + \frac{J_4}{4}\sigma_{i3}\tau_{i3}\sigma_{j3}\tau_{j3}. \tag{2}$$

Quantum fluctuations among the four possible directions of order together with dissipation lead [14] to a scale invariant spectrum which leads to the observed Marginal Fermi-liquid properties in the quantum-critical regime. These quantum fluctuations are specified in the basis of the eigenstates of the operator $\mathbf{\Omega}$. The 4 classical loop current states are eigenvalues

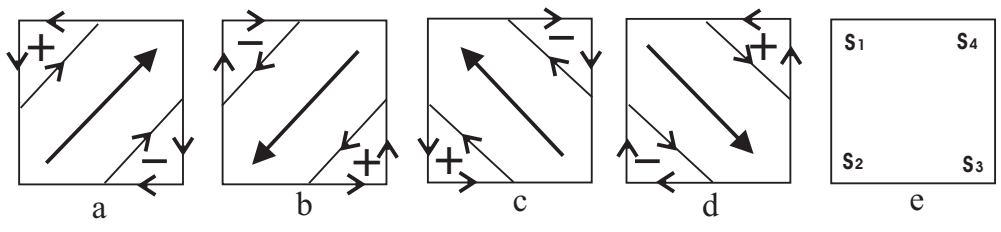

FIG. 1: The four Possible "classical" domains of the loop ordered state with their directions of the order parameter $\mathbf{\Omega}$ are shown. Fig. (e) presents the notation for the location of the moments within the unit-cell.

of operators $\mathbf{\Omega}_i = (\Omega_{i,x}, \Omega_{i,y})$ defined at cells i. According to Eq. (8) of Ref.(15),

$$(\Omega_x + i\Omega_y)|\theta >= \exp^{i\theta} |\theta > \tag{3}$$

The quantum fluctuations are generated from a term in the microscopic hamiltonian:

$$H_Q = \sum_i 2t\Omega_{zi}^2 \tag{4}$$

where Ω_{zi} is the generator of rotations in the space of the four states of $\mathbf{\Omega}_i$, i.e

$$U(\pm\frac{\pi}{2})|\theta >= \exp(\pm i\frac{\pi}{2}\mathbf{\Omega}_{zi})|\theta >= e^{-i\pi/4}|\theta \pm \pi/2 > . \tag{5}$$

The factor $e^{-i\pi/4}$ is necessary in a four state model to ensure that the rotation operator U has a unit determinant. It can then be shown that

$$\Omega_z \;=\; (-i/\sqrt{2})\big[U - U^+ + U^2/\sqrt{2}\big] \tag{6}$$

$$\Omega_z^2 \;=\; 5/4 - (U + U^\dagger)/\sqrt{2}, \tag{7}$$

Note that $(U + U^\dagger)$ rotates the states $|\theta >$ both clockwise and anti-clockwise by $\pi/2$ but with matrix elements which are complex. This is represented in terms of the AT model by an operator $(\sigma_1+\sigma_2+\tau_1+\tau_2)$; the relative coefficients between $\sigma_1(\tau_1)$ and $\sigma_2(\tau_2)$ are dictated by the factor $e^{-i\pi/4}$ in (5).

A more convenient representation for calculations on the problem is $SU(4)$; we introduce the direct products,

$$S^i = \sigma^i \otimes I, \quad T^i = I \otimes \tau^i, \quad K^{ij} = \sigma^i \otimes \tau^j. \tag{8}$$

The quantum AT model in this representation is

$$H_{QAT} = \sum_i t(S_i^1 + T_i^1 + S_i^2 + T_i^2) + t'(K_i^{11} + K_i^{22}) - \sum_{\langle i,j\rangle} \frac{J}{4}(S_i^3 S_j^3 + T_i^3 T_j^3) + \frac{J_4}{4} K_i^{33} K_j^{33}. \tag{9}$$

The second term above expresses the effect of U^2 which rotates $\mathbf{\Omega}$ by π. The magnitude of $4t/J$ (in the present notation) was estimated [15] to be of $O(1)$. It is hard to estimate t', which we leave as a free parameter.

We can parameterize the most general quantum state by the three pairs of angles $\theta_i, \phi_i, i = 1, 2, 3$:

$$\begin{aligned} |\psi\rangle \;&=\; \cos\frac{\theta_1}{2}\Big(\cos\frac{\theta_2}{2}|10,10\rangle + \sin\frac{\theta_2}{2}e^{i\phi_2}|01,10\rangle\Big) \\ &+\; \sin\frac{\theta_1}{2}e^{i\phi_1}\Big(\cos\frac{\theta_3}{2}|10,01\rangle + \sin\frac{\theta_3}{2}e^{i\phi_3}|01,01\rangle\Big) \end{aligned} \tag{10}$$

Special Case $J_4 = 0$: To understand the results for the general case, it is useful first to consider the simple case, $J_4 = 0$, when we simply have two decoupled transverse field Ising models [16]. In this case we have $\phi_i = 0$ for $i = 1, 2, 3$ and $\theta_2 = \theta_3$. Minimizing the Hamiltonian we find $\sin\theta_1 = \sin\theta_2 = \sin\theta = -t/J$. The ground state is

$$|G\rangle \;=\; \Big(\cos\frac{\theta}{2}|10\rangle + \sin\frac{\theta}{2}|01\rangle\Big)_\sigma \bigotimes \Big(\cos\frac{\theta}{2}|10\rangle + \sin\frac{\theta}{2}|01\rangle\Big)_\tau \tag{11}$$

There are three eigenvalues at any k, two of which are degenerate,

$$\omega_{1,2} = 2(J^2 - t^2 f_{\mathbf{k}})^{1/2};\;\; f_{\mathbf{k}} = (\cos(k_x a) + \cos(k_y a))/2, \;\; \omega_3 = 4J. \tag{12}$$

$\omega_{1,2}$ correspond to the propagation of the flipped states in the σ and τ sectors respectively due to the transverse-fields. ω_3 is the energy for flipping the angle of $\mathbf{L}$ by π. The modes in this approximation were derived earlier [6] by a simple procedure and called the "mixing modes" and the "phase mode" in a semi-classical calculation with no four-spin coupling.
General case: Since the 1 and the 2 components of all operators appear symmetrically, $\phi_i = \pi/4$ for all i in the ground state. This leaves 3 real coefficients in the general wavefunction. Also we are interested only in the case $J_1 = J_2$ and the Hamiltonian is invariant under interchange of σ and τ. So the general wave-function has only two parameters. It turns out through minimizing numerically the ground state energy that the wave-function (11) with just one parameter (beside ϕ) is a good approximate (correct to within a few percent) ground state wave-function for the interesting range of parameters.

To obtain the collective modes in the general case, it is convenient to introduce the Holstein-Primakoff transformation generalized to $SU(4)$. We then transform the Hamiltonian to the Boson representation, perform a generalized Bogoliubov transformation and calculate the ground state wavefunction and the eigenvalues and the eigenvectors of the excitations. Details of this procedure will be given in a longer manuscript. We give here only the results.

The complete results can only be obtained through a numerical diagonalization of a matrix. We compare such results below with the experiments. But first we give the perturbative

corrections for $J_4/J \ll 1, t' = 0$ to the eigenvalues given in Eq. (12) for the case $J_4 = 0$:

$$\delta(\omega_1^2)^{(1)} = 4J^2 \left[2 - (3 + f_{\mathbf{k}})\frac{t^2}{J^2} + (1 + 3f_{\mathbf{k}}/2)\frac{t^4}{J^4} - f_{\mathbf{k}}\frac{t^6}{2J^6}\right] \frac{J_4}{J}$$
$$\delta(\omega_2^2)^{(1)} = 4J^2 \left[2 - (1 - f_{\mathbf{k}})\frac{t^2}{J^2} - (1 + 3f_{\mathbf{k}}/2)\frac{t^4}{J^4} + f_{\mathbf{k}}\frac{t^6}{2J^6}\right] \frac{J_4}{J}$$
$$\delta(\omega_3^2)^{(1)} = 8J^2 \left[4\frac{t^2}{J^2} - (4 + f_{\mathbf{k}})\frac{t^4}{J^4}\right] \frac{J_4}{J} \tag{13}$$

Note that the degeneracy of the modes ω_1, ω_2 is lifted by $J_4 \neq 0$. This reflects that operator K_{33} couples the σ and τ modes. For $t' = 0$, the two modes represent the two linear combinations $\sigma \pm \tau$. For $t' \neq 0$, these modes themselves couple and a more complicated linear combination varying with $\mathbf{k}$ results, as we see through our general results obtained numerically. We can also determine the correction perturbatively in t' for $J_4 = 0$ to get

$$\delta(\omega_1^2)^{(1)} = 4J^2 \left[1 - (2 - 3f_{\mathbf{k}})\frac{t^2}{2J^2} + f_{\mathbf{k}}\frac{t^4}{2J^4}\right] \frac{t'}{J}$$
$$\delta(\omega_2^2)^{(1)} = 4J^2 \left[-1 + (2 + 5f_{\mathbf{k}})\frac{t^2}{2J^2} - f_{\mathbf{k}}\frac{t^4}{2J^4}\right] \frac{t'}{J}$$
$$\delta(\omega_3^2)^{(1)} = 32t^2\frac{t'}{J}$$

Now we present the results for the eigenvalues of the collective modes calculated numerically for any given J, J_4, t, t'. For both $J_4, J > 0$, the dispersion of the upper mode is smaller than the lower. For $J > 0, J_4 < 0, t' = 0$, the energy of the upper mode at $k = 0$ progressively decreases as k increases and it must (from the phase diagram of the classical AT model) [18] go to 0 at the zone boundary. With $t' \neq 0$, there is a level repulsion and anti-crossing of the two modes.

Choosing parameters to reproduce the experiments in $Hg1201$ with $T_c = 65K$, we show the results in Fig. (2). In the experiments, one of the collective modes is at 40 ± 5 meV another one at 50 ± 5 meV at $k = 0$. The dispersion across the Brillouin zone in both the (11) and the (10) directions for the higher energy mode is 5 ± 5 meV, while the lower mode is even less dispersive. We also have the very important constraint from the thermodynamics of the AT model [18, 19] that to have a transition with no divergence in the specific heat $-1 < J_4/J < 0, J > 0$. In Fig. (2), the following parameters are used: $t = 5.5\,\text{meV}$, $J = 33.2\,\text{meV}$, $J_4 = -0.3J$, $t' = -1.7t$, $\sin\theta \approx -0.37$. We calculate $\sin\theta \approx -0.37$ for these parameters. In this case the dispersion width of the first mode is about 10% of the energy gap and second one is around 5% of the energy gap. The result is shown in figure 2. These dispersions change with doping and we use a doping where a lot of data is available.

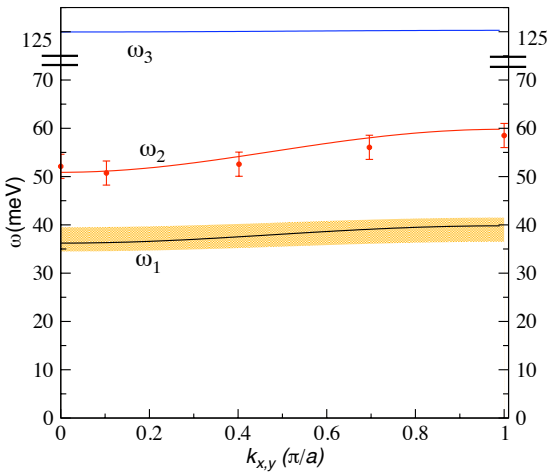

FIG. 2: The collective mode dispersions of Ashkin-Teller model along the $k_x = k_y$ direction. The black curve stands for ω_1 and red curve stands for ω_2. The parameters are taken to fit the experimental results in $Hg1201, T_c = 65K$.. The blue curve gives the calculated third mode.

We can also estimate the transition temperature T_c with these parameters using Baxter's [18] exact result, $\exp(-J_4/2T_c) = \sinh(J/2T_c)$ to be about 200 Kelvin compared to the experimental value of about 250 Kelvin.

The scattering of neutrons from ordered orbital moments leads to a few considerations not usually encountered in scattering due to ordered spins. The moments at the four "sites" in Fig. (1e) are orbital moments and so are represented by the transformation properties of $S = 1$ spins, rather than $S = 1/2$ spins. The neutron spins are represented by the usual spin-1/2 Pauli matrices σ_n, which change the angular momentum of the system by ± 1 in the spin-flip process. So at any "site" the spin-fip due to scattering of a neutron makes the transitions $(1 \to 0), (-1 \to 0)$ and *vice-versa* but there is no transition from the $(1 \to -1)$ or

vice-versa. This should be contrasted with ordered spin-systems, for example for $\mathbf{S} = 1/2$, where the neutrons scatter from states of $S_z = -1/2$ to $S_z = 1/2$ and vice-versa. The high energy mode whose eigenvalue is ω_3 cannot be observed by polarized neutron scattering since, it involves change of angular momentum by 2 but should be observable by other tools.

Semi-Classical Angle of the Effective Moments: The effective direction of the moments is deduced in polarized neutron diffraction experiments by measuring the intensity of the spin-flipped scattered neutrons at a a Bragg vector $\mathbf{Q}$ by sending in the neutron beam at various different polarization directions and using standard selection rules [17]. By this method the *effective* direction of $\mathbf{M}$ has been determined to be 45 ± 20 degrees to the normal to the plane both in a sample of $Y(123)$ and $Hg(1201)$.

To calculate the neutron scattering intensity, we find the expectation value of $\langle \mathbf{M}(\mathbf{Q}) \rangle$, the Fourier transform of the real space magnetic moment operator in the ground state. The representation of $\mathbf{M}(\mathbf{r})$ in terms of the order parameter $\mathbf{\Omega}$ is obtained by inverting Eq. (1) to get

$$\mathbf{M}_i(\mathbf{r}_\mu) = \mathbf{\Omega}_i \times \mathbf{r}_\mu / \mathbf{r}_\mu^2 \tag{14}$$

Here we are treating $\mathbf{r}_\mu$ as classical vectors. Transforming to momentum space

$$\langle \mathbf{M}(\mathbf{Q}) \rangle = \sum_i e^{i\mathbf{Q}.\mathbf{R}_i} \sum_\mu \frac{\langle \mathbf{\Omega}_i \rangle \times \mathbf{r}_\mu}{\mathbf{r}_\mu^2} e^{i\mathbf{Q}.\mathbf{r}_\mu}, \tag{15}$$

where the expectation value is taken in the ground state wave-function, Eq.(11), which we have found to be accurate to a few percent. Using Eq. (6) for the matrix representation of $\mathbf{\Omega}$, we find

$$\begin{aligned} \langle \Omega_x \rangle = \langle \Omega_y \rangle &= \frac{1}{\sqrt{2}} r_0^{-1} \cos\theta \\ \langle \Omega_z \rangle &= \frac{1}{\sqrt{2}} r_0^{-1} \sin\theta (\frac{\sin\theta}{2\sqrt{2}} - 1). \end{aligned} \tag{16}$$

r_0 is the magnitude of $\mathbf{r}_\mu$ measured from the center of the cell. Using these we find

$$\begin{aligned} \langle M_x(\mathbf{Q}) \rangle &= -\frac{1}{\sqrt{2}} \sin\theta (\frac{\sin\theta}{2\sqrt{2}} - 1) \Big(e^{i\mathbf{Q}.\mathbf{r}_1} - e^{i\mathbf{Q}.\mathbf{r}_2} - e^{i\mathbf{Q}.\mathbf{r}_3} + e^{i\mathbf{Q}.\mathbf{r}_4} \Big), \\ \langle M_y(\mathbf{Q}) \rangle &= -\frac{1}{\sqrt{2}} \sin\theta (\frac{\sin\theta}{2\sqrt{2}} - 1) \Big(e^{i\mathbf{Q}.\mathbf{r}_1} + e^{i\mathbf{Q}.\mathbf{r}_2} - e^{i\mathbf{Q}.\mathbf{r}_3} - e^{i\mathbf{Q}.\mathbf{r}_4} \Big), \\ \langle M_z(\mathbf{Q}) \rangle &= \sqrt{2} \cos\theta \Big(e^{i\mathbf{Q}.\mathbf{r}_1} - e^{i\mathbf{Q}.\mathbf{r}_3} \Big), \end{aligned} \tag{17}$$

Here we have taken the magnitude of $\mathbf{M}$ to be unity since we wish to compare with experiments only the ratio of $\mathbf{M}$ in different directions.

For the momentum transfer $\mathbf{Q} = (0, a^*, c^*)$, as in the experiment and for the domain chosen (a in Fig.(1))

$$\langle \mathbf{M} \rangle = 2i \sin(a^* r_y) \Big(\frac{1}{\sqrt{2}} \sin\theta (\frac{\sin\theta}{\sqrt{2}} - 2), 0, \sqrt{2} \cos\theta \Big) \tag{18}$$

In the other domains M_x, M_y switch. The tilt angle to the normal is

$$\phi = \arctan \Big[\frac{\sin\theta (\frac{\sin\theta}{\sqrt{2}} - 2)}{2\cos\theta} \Big] \tag{19}$$

For fitting the spectrum of collective modes as shown in Fig.(2), we needed $\sin\theta \approx -0.37$. This yields $\phi \approx 25$ degrees. If we increase t by 20%, the upper mode disperses by about 10 meV, which is also in the experimental range. Then we can get $\phi \approx 33$ degrees compared to the experimental range [1, 9] of 40 ± 20degrees. This means that spin-orbit scattering[20] or additions[21] to the simplest two-dimensional model, which are required to change by general symmetry considerations need not be very large to understand the dependence of the intensity of the neutron scattering on the polarization direction.

It must be noted that the tilt of the moment calculated here is a purely quantum effect. It arises in a four state model due to the fact that (see Eqs. (5) and (6)) that U has necessarily complex matrix elements so that the ground state expectation value $< \Omega_z > \neq 0$, i.e an anapole is generated quantum-mechanically.

A prediction, beside that of the third mode which may be discovered through Raman or inelastic x-ray scattering, is that the frequency of all three branches of the collective modes should $\rightarrow 0$ for $q \rightarrow 0$ as $T \rightarrow T^*(x)$, because the transition is second order. As already noted [3], the observed ARPES spectra in underdoped cuprates

To summarize, it is important for an acceptable theory of the Cuprates that the universal features of the phase diagram and the principal excitations in each part of it be understood consistently and quantitatively from a common set of idea. This paper provides a contribution towards this goal through showing that the dominant new fluctuations in the pseudogap phase are necessary excitations of the loop-ordered phase. An additional result is that the direction of the effective ordered moments are shown consistent with experiments.

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