Spiral Phase of a Doped Quantum Antiferromagnet

Boris I. Shraiman
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

Eric D. Siggia[a]
Ecole Normale Superieure, 75231 Paris CEDEX 05, France
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A low density of vacancies in a 2D, spin-$\frac{1}{2}$, Heisenberg antiferromagnet leads (for a range of effective couplings) to a metallic phase with incommensurate antiferromagnetic order, i.e., with the staggered magnetization rotating in a plane with the wave number proportional to the density. This structure originates from the polarization of the antiferromagnetic dipole moments of the vacancies. The excitation spectrum of this spiral state includes an interesting low-lying mode. Implications for neutron scattering and normal-state resistivity are discussed.

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Current theories of high-temperature superconductivity[1] and of neutron scattering[2] on CuO-based compounds have focused attention on the behavior of electrons with hard-core interactions on a two-dimensional square lattice at a density of just less than one per site.[3] The residual interactions are antiferromagnetic (AF) and are described by a generic Hamiltonian which includes vacancy-hopping and spin-exchange terms:

$$H_0=-t\sum_{r,\hat{\mathbf{a}}}c^{\dagger}_{r+\hat{\mathbf{a}},\sigma}c_{r,\sigma}+J\sum_{r,\hat{\mathbf{a}}}\mathbf{s}_r\cdot\mathbf{s}_{r+\hat{\mathbf{a}}}\,,$$

where $\hat{\mathbf{a}}=\hat{\mathbf{x}},\hat{\mathbf{y}}$, the spin-$\frac{1}{2}$ fermion operator $c_{r,\sigma}$ is restricted to single occupancy, and $\mathbf{s}_r=c^{\dagger}_{r,\sigma}\boldsymbol{\tau}_{\sigma\sigma'}c_{r,\sigma'}$ is the local spin.

In the absence of vacancies the spin-$\frac{1}{2}$ Heisenberg model has long-range AF order[4] at $T=0$ and we assume that its long-wavelength properties are described by the nonlinear σ model[5] (NLσ). A good deal is also known about the ground state of the single vacancy.[6-10] Specifically, the single-vacancy ground states form a narrow band, ϵ_k, of width $w\sim O(J)$ in the $t\gg J$ limit [or $w\sim O(t^2/J)$ for $t\ll J$] with energy minima[6-9] lying at $k_v=(\pm\pi/2,\pm\pi/2)$. The band is strongly anisotropic so that the mass, $\mu_\perp\sim w^{-1}$, for k perpendicular to the zone boundary[11] is about a factor of 10 smaller than the parallel mass, $\mu_\parallel$, for $t\approx J$. Near the energy minima, the vacancy states involve a long-range dipolar distortion of the staggered magnetization,[6] $\hat{\boldsymbol{\Omega}}$, and can be assigned[12] an AF dipole moment $\mathbf{p}_a(k)$ which is a vector in both spin and physical spaces with magnitude of order $\min(J^{-1}t,1)$ and k dependence $p_a(k)\sim\sin k_a$.

Presently we consider the effect of a low density of vacancies, $n\ll 1$. We will show that the commensurate Néel state is unstable (at $T=0$ in 2D) for any n, towards a *spiral* state in which the AF dipole moments of the holes order as a result of polarization of the spins of opposite valleys of the Fermi sea (located near k_v) in opposite directions. In the spiral state the staggered magnetization, $\hat{\boldsymbol{\Omega}}$, rotates in a plane with a pitch scaling as n^{-1}. We also find a new low-lying excitation, the "torsion" mode, which is the collective mode associated with the transverse fluctuations of the dipole polarization and hence with the fluctuations of the plane of the spiral in spin space.[13] We expect (in some temperature range) the AF correlation length to scale with n^{-1}. It should be emphasized that the spiral phase appears through a mechanism quite *different* from the usual $2k_F$ spin-density-wave instability of the Fermi surface, and, in contrast, remains metallic with the vacancy Fermi surface ungapped.

The one-particle properties mentioned above are incorporated into a semiphenomenological effective Hamiltonian[6]:

$$H_{\text{eff}}=\sum_k\epsilon_k\bar{\Psi}_k\Psi_k-g\sum_{q,a}\mathbf{p}_a(q)\cdot\mathbf{j}_a(q)-g'\sum_{q,k,a}\cos k_a\bar{\Psi}_{k-q/2}\boldsymbol{\tau}\Psi_{k+q/2}\cdot\mathbf{m}(q)+H_{\text{NL}\sigma}\,. \quad (1)$$

The vacancy is represented by a two-component spinor[14] Ψ^a; $\mathbf{m}$ is the local magnetization operator which is conjugate to $\hat{\boldsymbol{\Omega}}$ and enters in the NLσ Hamiltonian $N_{\text{NL}\sigma}=\frac{1}{2}\sum_r[\chi^{-1}\mathbf{m}^2+\rho(\Delta_a\hat{\boldsymbol{\Omega}})^2]$ where $\chi\sim J^{-1}$ is the susceptibility, $\rho\sim J$ is the spin-wave stiffness, and Δ_a is the lattice gradient. The second term in Eq. (1) couples the background magnetization current $\mathbf{j}_a\equiv\hat{\boldsymbol{\Omega}}\times\Delta_a\hat{\boldsymbol{\Omega}}$ with the AF dipole moment of the vacancies

$$\mathbf{p}_a(q)\equiv\sum_k\sin k_a\bar{\Psi}_{k-q/2}\boldsymbol{\tau}\Psi_{k+q/2}\,,$$

and gives rise to the dipolar interactions. The phenomenological coupling constants g and g' are of order $\min(t,J)$. Note that $g=g'=t$ and $\epsilon_k=0$ would correspond[6] to the bare hopping term of H_0; diagrammatically $\epsilon_k\neq 0$ emerges as the coherent part of the self-energy[8,10] while the reduction of g in the $t\gg J$ limit crudely incorporates the downward renormalization[8] of the coherent part of the propagator by incoherent processes. The scaling $g \sim J$ in the $t \gg J$ limit also corresponds to the saturation of the single-vacancy dipole moment[6] at $O(1)$. One observes that since for a low density of vacancies only the states with $k \approx k_v$ are occupied, at small momentum transfer the dipolar coupling is dominant while the coupling to $\mathbf{m}$ is suppressed by an extra power of n.

To understand the physics described by H_{eff} of Eq. (1), let us consider its classical limit

$$H_{\text{cl}} = -g\mathbf{p}_a \cdot (\hat{\mathbf{\Omega}} \times \partial_a \hat{\mathbf{\Omega}}) + \tfrac{1}{2}\rho(\partial_a \hat{\mathbf{\Omega}})^2 .$$

The dipoles clearly order, $\langle \mathbf{p}_a \rangle \neq 0$, leading to a spiral antiferromagnetic phase with $\hat{\mathbf{\Omega}} \times \partial_a \hat{\mathbf{\Omega}} = g\rho^{-1}\langle \mathbf{p}_a \rangle$, where $\hat{\mathbf{\Omega}}$ rotates in the plane perpendicular to the spin direction of $\langle \mathbf{p}_a \rangle$ with a pitch along the spatial direction of $\langle \mathbf{p}_a \rangle$. The magnitude of the inverse pitch, or equivalently the incommensurability wave number Q, is proportional to the total polarization and hence to the density of holes: $Q = |\partial \mathbf{\Omega}| = g\rho^{-1}|\mathbf{p}_a| \sim n$.

The quantum problem is a little more subtle since the Pauli principle prevents $\mathbf{p}_a(k)$ from being identical for all vacancies. We first consider the renormalization of the spin-wave propagator by particle-hole fluctuations as described by the "bubble" diagram. Only the stiffness constant is modified to lowest order in n since the coupling to $\mathbf{m}$ vanishes at the zone face centers. One finds the renormalized static stiffness $\tilde{\rho} = \rho - g^2\chi_d$, where

$$\chi_d \equiv \tfrac{1}{6} \sum_{k,a} \sin^2 k_a \int dt \langle \overline{\Psi}_k \boldsymbol{\tau} \Psi_k(0) \cdot \overline{\Psi}_k \boldsymbol{\tau} \Psi_k(t) \rangle$$

is the static dipole susceptibility. The instability is signaled by $\tilde{\rho} < 0$,

$$\rho^{-1} g^2 \chi_d > 1 . \qquad (2)$$

For noninteracting particles at zero temperature $\chi_d = 4N_F \langle \sin^2 k_a \rangle$, where N_F denotes the density of states at the Fermi energy and the angular brackets are an average over the Fermi surface. In 2D $N_F = (\mu_{\parallel}\mu_{\perp})^{1/2}/2\pi$ and for t/J both large or small one finds that the left-hand side of Eq. (2) is of order 1 times $(\mu_{\parallel}/\mu_{\perp})^{1/2}$. Thus, provided the anisotropy $\mu_{\parallel}/\mu_{\perp}$ is as large as we expect, the instability occurs at arbitrarily low hole density n. The $n \to 0$ limit is of course singular since the calculated stiffness renormalization only applies at wave numbers[15] $q^2 \ll k_F^2 \sim n$. Even in this limit the absence of a threshold density is an artifact of two dimensionality and $T=0$. In the classical limit $\chi_d \sim n/T$ while in 3D at $T=0$, $\chi_d \sim n^{1/3}$, so that in either case the instability occurs for $n > n_c$.

A state with negative stiffness constant $\tilde{\rho}$ evidently prefers to twist, and we will now construct a mean-field theory for such a phase. Assume[16] $\hat{\mathbf{\Omega}} \times \partial_a \hat{\mathbf{\Omega}} = \hat{\mathbf{z}} Q_a \neq 0$. Then the mean-field version of Eq. (1) reads

$$H_{\text{MFT}} = \sum_k \epsilon_k n_k - gQ_a \sin k_a (n_k^+ - n_k^-) + \tfrac{1}{2}\rho Q_a^2 ,$$

where $n_k^{\pm}$ are the occupation numbers of spin-z states. Minimizing with respect to Q_a leads to a self-consistency condition:

$$Q_a = g\rho^{-1} \sum_k \sin k_a (n_k^+ - n_k^-) , \qquad (3)$$

where

$$n_k^{\pm} = [\exp\beta(\epsilon_k \mp gQ_a \sin k_a - \epsilon_F) + 1]^{-1}$$

and ϵ_F is the chemical potential. Equation (3) acquires a nonzero solution when $g^2/\chi_d \geq \rho$ (note that $\chi_d = g^{-1}\langle \delta \mathbf{p}_a / \delta \mathbf{Q}_a \rangle |_{Q=0}$) and hence we identify the $\tilde{\rho} < 0$ instability [Eq. (2)] with the onset of incommensurate, spiral AF order. For $g^2\chi_d/2 < \rho < g^2\chi_d$ and $n \ll 1$, we find a spatially uniform fully polarized state with $Q_a \sim n$ along the (1,0) or (0,1) directions and a positive stiffness constant. For $\rho < g^2\chi_d/2$ our model suggests phase separation which is unphysical in view of our neglect of the long-range Coulomb potential. A more interesting and highly speculative possibility in this parameter range is an intrinsically disordered phase.[17]

From Eq. (3) it is clear that the dipolar polarization $\langle p_a \rangle \neq 0$ is built up by populating opposite valleys of the Fermi sea with vacancies of opposite pseudospin. The ordering is reminiscent of Stoner ferromagnetism with the important difference that in the present case there is no net spin polarization. In terms of the fields[6,14] $\psi_k^{A,B}$ which create spinless holes on sublattice[18] A or B, any state with $\langle \mathbf{p}_a \rangle \perp \hat{\mathbf{\Omega}}$, as is the case for the spiral phase, has only off-diagonal pseudospin order: i.e., $\langle \psi_k^{\dagger A} \psi_k^B \rangle \sim Q_a \sin k_a$ but $\langle \psi_k^{\dagger A} \psi_k^A \rangle = \langle \psi_k^{\dagger B} \psi_k^B \rangle$. The hole wave function has equal weights on the two sublattices and a fixed phase relation between them. One can also see that for the spiral phase[19]

$$\langle c_\sigma^\dagger(\mathbf{r}') c_{\pm\sigma}(\mathbf{r}) \rangle \sim \exp[i\mathbf{Q} \cdot (\mathbf{r} \mp \mathbf{r}')/2] .$$

To explore the low-energy excitations of a spiral state with given Q_a semiclassically, we examine the long-wavelength distortions of $\hat{\mathbf{\Omega}}$, $\mathbf{m}$, and $\mathbf{p}_a$ in Eq. (1). Define the linearized staggered magnetization operators ζ_r ($[\zeta^\dagger, \zeta] = 0$) in a rotating frame:

$$O_r^\dagger \hat{\mathbf{\Omega}} = [1 - \zeta^\dagger \zeta/2,\ (\zeta - \zeta^\dagger)/2i,\ (\zeta + \zeta^\dagger)/2] ,$$

where O_r is a uniform O(3) rotation around $\hat{\mathbf{z}}$ corresponding to the spiral state Q_a. Similarly, the long-wavelength transverse fluctuations of the dipole density are parametrized[20] by

$$O_r^\dagger \mathbf{p}_a = |\langle \mathbf{p}_a \rangle| [u_x,\ u_y,\ 1 - \tfrac{1}{2}(u_x^2 + u_y^2)] ,$$

with $u_x \equiv \frac{1}{2}(\pi^\dagger + \pi - \zeta^\dagger - \zeta)$, $u_y \equiv (\pi - \pi^\dagger)/2i$, and $[\pi_r, \pi_{r'}^\dagger] = \delta_{r,r'}$. Introducing the magnetization operator η conjugate to ζ, $[\eta_r, \zeta_{r'}^\dagger] = \delta_{r,r'}$, substituting into Eq. (1), and expanding for small k and Q we find

$$H=\sum_k[\rho\mathbf{Q}^2\pi_k^\dagger\pi_k+\tfrac{1}{2}\rho\mathbf{Q}\cdot\mathbf{k}(\pi_k-\pi_{-k}^\dagger)(\zeta_k^\dagger+\zeta_{-k})+\chi^{-1}\eta_k^\dagger\eta_k+\rho\mathbf{k}^2\zeta_k^\dagger\zeta_k]\,. \tag{4}$$

We observe that the imaginary part, $\zeta^\dagger-\zeta$, decouples from π: The corresponding branch (I) of the spectrum has the usual spin-wave form $\omega_I^2=c^2\mathbf{k}^2$ (where $c^2\equiv\rho/\chi$) with the zero mode being a global rotation of $\hat{\mathbf{\Omega}}$ in the plane of the spiral (the phase mode of the spiral). The out-of-plane distortion of $\hat{\mathbf{\Omega}}$, the $\zeta^\dagger+\zeta$ mode, mixes with the polarization fluctuations π leading to more complex dynamics:

$$\omega_{R,T}^2(k)=\tfrac{1}{2}(c^2k^2+\rho^2Q^4)\{1\pm[1-4\rho^2c^2Q^4k_\perp^2/(c^2k^2+\rho^2Q^4)^2]^{1/2}\}\,, \tag{5}$$

where $k_\perp^2\equiv k^2-(\mathbf{k}\cdot\mathbf{Q})^2/Q^2$. The upper branch ($R$) is spin-wave-like for $k\gg Q$; however, mixing with π introduces a gap $\omega_R=\rho Q^2$ at $k=0$. The transverse fluctuations of the dipole polarization dominate the lower branch, the torsion mode ω_T, which lies entirely on the energy scale $\rho Q^2\sim n^2J$. Notice that for $\mathbf{k}\parallel\mathbf{Q}$, $\omega_T=0$: This is an artifact of the $k,Q\ll 1$ expansion. Higher-order terms[15] in Eq. (4), $O(Q^2k^2)$, would in general include a stiffness D (or diffusivity) term for the polarization and the corrected dispersion relation is obtained by replacing $k_\perp^2$ in Eq. (5) by $\tilde{k}_\perp^2=k_\perp^2+\rho^{-1}D(k^2-Q^2)^2$. The remaining zeros of $\omega_T(k)$ occur at $k_a=\pm Q_a$ and are associated with uniform rotation of the plane of the spiral in spin space.

The transverse fluctuations of the dipole polarization arise as a collective mode involving slowly varying perturbations of the Fermi distributions of the vacancies and the mode structure, Eq. (5), can be also derived using the kinetic theory approach of Landau.[21] It should be emphasized that the dipole (or torsion) mode is limited to small momentum transfers[22] $q<k_F\sim Q^{1/2}$.

The behavior of the static spin-correlation function with temperature and doping should furnish a useful experimental signature of the spiral state. From our discussion of the instability we expect the incommensurability $\mathbf{Q}$ to appear for $T<\epsilon_F\sim n$ (for $n>n_c$) and thereafter to remain constant. The spatial direction of $\mathbf{Q}$ is difficult to predict without a more realistic Hamiltonian; however, we note that in the $\mathbf{Q}\sim(1,1)$ state there would be a net interlayer exchange of $O(Q^2)$ (for the LaCuO based material) which may make it more favorable. The spin correlations are anisotropic because of the softness of the torsion mode in the $\mathbf{k}\parallel\mathbf{Q}$ direction. In the classical limit the fluctuations of $\hat{\mathbf{\Omega}}$, parametrized by the polar angles θ,ϕ (with the uniform rotation about $\hat{\mathbf{z}}$ taken out), have energy

$$E\sim[\rho\mathbf{k}_\perp^2+D(\mathbf{k}_\parallel^2-\mathbf{Q}^2)^2]\,|\theta_k|^2+\rho\mathbf{k}^2|\phi_k|^2\,,$$

where $\mathbf{Q}\cdot\mathbf{k}_\perp=\mathbf{Q}\times\mathbf{k}_\parallel=0$. The spin order along $\mathbf{Q}$ can disappear for $T\sim JD^{1/2}Q\ll\epsilon_F$ as the torsion mode melts, leaving the correlation length $\xi\sim O(n^{-1})$. The presence of domains of different $\mathbf{Q}$ and topological defects in the spiral structure,[23] which may be quenched in, will make the correlations more isotropic. However disordered the torsion mode becomes, it cannot reduce the correlation length beyond $Q^{-1}\sim n^{-1}$ since on shorter scales the spins obey the Heisenberg model which is, we assume, ordered. Note that our arguments in favor of the spiral state do not require long-range order, but only $k_F^{-1}\ll\xi$, which is satisfied for $n\ll 1$.

Holes make a potentially important nonhydrodynamic contribution to spin-wave damping via the imaginary part of the dipole susceptibility $\chi_d(k,\omega)$. For $k\ll k_F$ one finds $\Gamma=\mathrm{Im}(\omega/ck)\sim g^2\mu c/\rho v_F$, provided $c<v_F$, else, $\Gamma\sim g^2\mu v_F/\rho c$ for a window around[24] $k\sim\mu c$. For weakly localized holes where the momentum-conservation constraint is absent, one finds, neglecting numerical factors and characterizing all states by a single length and mass, that $\Gamma\sim g^2\mu n/c$. The latter may be important at $k\ll k_F$ for low densities where $c>v_F$ and Γ is otherwise zero.

The torsion mode lying entirely at frequencies $O(n^2J)$ is easily saturated thermally and is a possible source of the linear-T resistivity characteristic of the normal state of the high-T_c materials. We expect the resistivity to arise from the direct (spin flip) coupling of the holes with the collective torsion mode in a manner analogous to an itinerant ferromagnet.[25] (By contrast, scattering from a spin-diffusion mode would yield a resistivity going as T^2 or higher.) However, a proper calculation has not yet been carried out.

To conclude, we have shown that mobile vacancies in a background with at least short-range AF order have dipolar interactions which induce their collective polarization, leading to a spiral AF phase (even if the vacancies are not strongly localized). The spiral order implies, at a one-particle level, correlations between the wave number and pseudospin which extend throughout the Fermi sea.[26] We expect that along with the ordered spiral phase there might exist (for larger values of effective coupling constant or higher vacancy density) a disordered state with *local* spiral twist. Dipolar interactions may also introduce *pair* correlations and superconductivity in singlet and triplet channels. The competition and coexistence of superconductivity and local spiral order can be explored on the mean-field level using the four-fermion Hamiltonian with dipolar interaction obtained by integrating out the spin waves.[14] Finally, it appears that the hopping-induced dipole moment of the vacancy persists[27] even in a more realistic two-band model[28] of CuO planes, raising hopes that our analysis and predictions may hold for the real materials and may be of experimental relevance.[29]

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Note added.— One interesting extension of the present analysis concerns the double-spiral generalization proposed by Kane *et al.*[30] Indeed, our mean-field theory admits a state with the dipole polarization involving *two* orthogonal spin directions $\hat{\mathbf{e}}^{(x)}\cdot\hat{\mathbf{e}}^{(y)}=0$; $\langle \mathbf{p}_a\rangle=\hat{\mathbf{e}}^{(a)}$. Locally this state is degenerate with the single spiral; however, $\hat{\mathbf{\Omega}}\times\partial_a\hat{\mathbf{\Omega}}=g\rho^{-1}\langle\mathbf{p}_a\rangle$ can no longer be solved *globally* without an amplitude modulation: Hence the mean-field energy is higher. However, the higher symmetry of the double spiral would lead to additional zeros in the torsion-mode spectrum and therefore to the reduction of the zero-point energy.

[(a)]Permanent address: Department of Physics, Cornell University, Ithaca, NY 14853.

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[11]The AF long-range order doubles the unit cell and reduces the Brillouin zone to the diamond $(\pi,0)$, $(\pi,0)$, $(-\pi,0)$, and $(0,-\pi)$.

[12]It is amusing to note that many aspects of the physics emerge already in perturbation theory for $t/J\ll 1$; e.g., the dipolar structure associated with the hole can be derived from the correlation function $\langle k,\sigma|n(0,t)\partial_a\hat{\mathbf{\Omega}}(r,t)|k,\sigma\rangle$.

[13]This mode may also be interpreted in terms of a rotation about the local $\hat{\mathbf{\Omega}}$ axis which appears as an additional degree of freedom for noncollinear magnetic structures.

[14]The pseudospin of the vacancy originates in the sublattice structure induced by local AF correlations of the spin background. The spinor Ψ^a is derived (Ref. 6) from the more microscopic representation: $c^{a\dagger}_{r,\sigma}=\psi^a_r z^{\dagger a}_{r,\sigma}$, where $\psi^{\dagger a}_r$ creates a fermionic hole and the spinor $z^a_{r,\sigma}$ is a Schwinger spin boson. The explicit sublattice index $\alpha=A,B$ accommodates the staggered order (and labels the twofold degeneracy of the vacancy ground state). The hopping part of H_0 has the form

$$H_{\text{int}}=-t\sum_{\langle rr'\rangle}\psi^{\dagger B}_r\psi^A_{r'}\bar{z}^A_{r'}z^B_r+\text{H.c.}$$

We define $\Psi^a\equiv\mathbf{R}_{aa'}\psi^{a'}$, where $\mathbf{R}$ is an SU(2) rotation relating the spinor z_r, and hence the local direction of the staggered magnetization $\hat{\mathbf{\Omega}}(r)=\bar{z}^A_r\boldsymbol{\tau}z^A_r$, to a fixed basis. The relation between H_{int} and Eq. (1) becomes more evident in the spin-wave approximation after one introduces $z^A_r=(1,a_r)$, $z^B_\tau=(b_r,1)$, passes to the momentum representation, and identifies the staggered magnetization mode as $a^\dagger_q-b_{-q}$ and the net magnetization as $a^\dagger_q+b_{-q}$.

[15]The finite-q corrections to the dipole susceptibility

$$\chi_d(q)\approx\chi_d(0)(1+C^2k_F^{-2}q^2)^{-1}+O(k_F^{-4}q^4)$$

restabilize the system for $q>k_F$.

[16]Equivalently in the CP^1 parametrization (Refs. 6 and 14) one has $\langle\bar{z}_{r+a}z_r\rangle=iQ_a/2$ as the spiral order parameter.

[17]Heuristically if we just include the q dependence of χ_d in $\tilde{\rho}$, the structure factor exhibits a broad peak at $q\sim k_F$ and paramagnetic behavior at longer wavelength where amplitude fluctuations become important.

[18]Shifting k by (π,π) sends $\psi^A\rightarrow\psi^A$, $\psi^B\rightarrow-\psi^B$ and is equivalent to rotating the pseudospin by π around $\hat{\mathbf{z}}$:

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[20]This parametrization is most readily arrived at via the representation in Ref. 14. The details of the analysis will be presented elsewhere.

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[22]One may also consider a collective mode with $q\approx(\pi,0)$ which involves scattering of the vacancies between valleys and consequently a rotation of the *spatial* direction of $\langle\mathbf{p}_a\rangle$. We expect, however, that this mode has an energy scale of order ϵ_F.

[23]One may observe that a dislocation in a spiral structure corresponds to an XY vortex.

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[26]In a hypothetical single-domain sample these correlations imply a new physical effect. Namely, when the magnetic field applied to the sample varies, the hole populations on the opposite sides of the Brillouin zone (Ref. 11) adjust, necessitating a coherent impulse of umklapp momentum which should appear as a displacement of the center of mass along $\mathbf{Q}$.

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