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Superconducting state with a finite-momentum pairing mechanism in zero external magnetic field

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In the BCS theory of superconductivity, one assumes that all Cooper pairs have the same center-of-mass momentum. This is indeed enforced by self-consistency if the pairing interaction is momentum independent. Here, we show that for an attractive nearest-neighbor interaction, this is different. In this case, stable solutions with pairs with momenta $\mathbf{q}$ and $-\mathbf{q}$ coexist and, for a sufficiently strong interaction, one of these states becomes the ground state of the superconductor. The possibility for a finite-momentum pairing state emerges only for nodal superconductors and is accompanied by a charge order with wave vector $2\mathbf{q}$. For a weak pairing interaction, the ground state is a d-wave superconductor.

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In the original formulation of the BCS theory of superconductivity,[1] all Cooper pairs are assumed to have the same center-of-mass momentum $\mathbf{q}$. One possible generalization of this theory is to introduce a pair amplitude for each center-of-mass momentum separately. In the BCS theory for conventional superconductors, only one of these order parameters (OPs) is selected and the stable state is the one where all pairs have the same momentum and form the BCS condensate. For films in an external magnetic field, Fulde and Ferrell[2] and, independently, Larkin and Ovchinnikov[3] introduced a superconducting (SC) state with coexisting pair momenta $\mathbf{q}$ and $-\mathbf{q}$, a state that explicitly breaks time inversion symmetry. For unconventional pairing symmetries, the competition between pair momenta is more complex and it has remained unresolved whether a bulk SC ground state with different pair momenta may exist without magnetic field.[4] A SC state with different coexisting pair momenta generally exhibits a spatially inhomogeneous charge density. One example of a superconductor of this type is the recently proposed "pair density-wave" (PDW) state.[5–7] It is characterized in real space by a two-component order parameter $\Delta(\mathbf{r})=\Delta_{\mathbf{q}}e^{i\mathbf{qr}}+\Delta_{-\mathbf{q}}e^{-i\mathbf{qr}}$. This structure bears some resemblance to the Larkin-Ovchinnikov state, but it preserves time inversion symmetry. The PDW is accompanied by a charge-density pattern with wave vector $2\mathbf{q}$. For this reason the PDW state has been proposed to describe the SC state of high-T_c cuprates with coexisting stripe order, especially Nd-doped $La_{2-x}Sr_xCuO_4$ (Ref. 8) and $La_{2-x}Ba_xCuO_4$ for $x=1/8$.[9–12] In particular, the recent experiments on the 1/8-doped material stimulated further theoretical studies to resolve the nature and the origin of the SC state in the charge ordered phase.[13] The PDW might be a candidate state, but so far a microscopic model that yields the PDW as its ground state is lacking.

In this Rapid Communication, rather than attempting a microscopic theory for striped cuprates, we address the general question of whether finite-momentum pairing in zero magnetic field can exist in the ground state of a microscopic pairing Hamiltonian. We formulate an extended version of the BCS theory using Gor'kov's equations and explicitly allow for the coexistence of different finite-momentum pairing amplitudes. We identify conditions for a ground-state solution with finite OPs for the pair momenta $\mathbf{q}$ and $-\mathbf{q}$. This pairing state is realized beyond a critical interaction strength V_c for an attractive nearest-neighbor interaction, and it is characterized by a charge stripe order, a gapless density of states (DOS), and a partially reconstructed Fermi surface. On the other hand, for $V<V_c$, the d-wave superconductor is the stable ground state.

We start from a tight-binding Hamiltonian on a square lattice with N sites and periodic boundary conditions,

$$\mathcal{H}=\sum_{\mathbf{k},s}\varepsilon_{\mathbf{k}}c^\dagger_{\mathbf{k}s}c_{\mathbf{k}s} + \frac{1}{N}\sum_{\mathbf{q}}\sum_{\mathbf{k},\mathbf{k}'}\sum_{s,s'}V(\mathbf{k},\mathbf{k}',\mathbf{q})c^\dagger_{\mathbf{k}s}c^\dagger_{-\mathbf{k}+\mathbf{q}s'}c_{-\mathbf{k}'+\mathbf{q}s'}c_{\mathbf{k}'s}. \quad (1)$$

With nearest- and next-nearest-neighbor hopping amplitudes t and t', respectively, the single-electron dispersion has the form

$$\varepsilon_{\mathbf{k}}=-2t[\cos k_x+\cos k_y]+4t'\cos k_x\cos k_y-\mu, \quad (2)$$

where μ is the chemical potential.

For the superconducting state with singlet pairing, we use the BCS-type mean-field decoupling scheme and approximate $\langle c^\dagger_{\mathbf{k}\uparrow}c^\dagger_{-\mathbf{k}+\mathbf{q}\downarrow}c_{-\mathbf{k}'+\mathbf{q}\downarrow}c_{\mathbf{k}'\uparrow}\rangle \to \langle c^\dagger_{\mathbf{k}\uparrow}c^\dagger_{-\mathbf{k}+\mathbf{q}\downarrow}\rangle\langle c_{-\mathbf{k}'+\mathbf{q}\downarrow}c_{\mathbf{k}'\uparrow}\rangle$. The system is then represented by the spin-independent imaginary time Green's function $\mathcal{G}(\mathbf{k},\mathbf{k}',\tau) =-\langle T_\tau c_{\mathbf{k}s}(\tau)c^\dagger_{\mathbf{k}'s}(0)\rangle$, and the anomalous propagators $\mathcal{F}(\mathbf{k},\mathbf{k}',\tau)=\langle T_\tau c_{\mathbf{k}s}(\tau)c_{-\mathbf{k}'s'}(0)\rangle$ and $\mathcal{F}^*(\mathbf{k},\mathbf{k}',\tau) =\langle T_\tau c^\dagger_{-\mathbf{k}s}(\tau)c^\dagger_{\mathbf{k}'s'}(0)\rangle$ for $s\neq s'$. The Heisenberg equations of motion for the normal and anomalous Green's functions lead to the following Gor'kov equations:[14]

$$\mathcal{G}(\mathbf{k},\mathbf{k}',\omega_n)=\mathcal{G}_0(\mathbf{k},\omega_n) \times\left[\delta_{\mathbf{k}\mathbf{k}'}-\sum_{\mathbf{q}}\Delta(\mathbf{k},\mathbf{q})\mathcal{F}^*(\mathbf{k}-\mathbf{q},\mathbf{k}',\omega_n)\right]^{-1}, \quad (3)$$

$$\mathcal{F}(\mathbf{k},\mathbf{k}',\omega_n) = \mathcal{G}_0(\mathbf{k},\omega_n)\sum_{\mathbf{q}} \Delta(\mathbf{k},\mathbf{q})\mathcal{G}(-\mathbf{k}',-\mathbf{k}+\mathbf{q},-\omega_n), \tag{4}$$

where $\mathcal{G}_0(\mathbf{k},\omega_n)=[i\omega_n-\varepsilon_{\mathbf{k}}]^{-1}$ is the Green's function in the normal state and $\omega_n=(2n-1)\pi T$ is the fermion Matsubara frequency for temperature T. The order parameter $\Delta(\mathbf{k},\mathbf{q})$ is determined by the self-consistency condition

$$\Delta(\mathbf{k},\mathbf{q}) = -\frac{T}{N}\sum_n \sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}',\mathbf{q})\mathcal{F}(\mathbf{k}',\mathbf{k}'-\mathbf{q},\omega_n). \tag{5}$$

For the interaction, we choose a simple ansatz that allows for unconventional pairing; we assume an attractive interaction between electrons on neighboring sites. The Fourier transform of this attractive interaction can be decomposed into s, p, and d pairing channels. With the restriction to singlet pairing only the s and the d channels remain, which is equivalent to the interaction $V(\mathbf{k},\mathbf{k}',\mathbf{q})=V_s(\mathbf{k},\mathbf{k}',\mathbf{q})+V_d(\mathbf{k},\mathbf{k}',\mathbf{q})$ in momentum space, with factorizable extended s- and d-wave components $V_s(\mathbf{k},\mathbf{k}',\mathbf{q})$ and $V_d(\mathbf{k},\mathbf{k}',\mathbf{q})$, where

$$V_{s,d}(\mathbf{k},\mathbf{k}',\mathbf{q}) = V g_{s,d}(\mathbf{k}-\mathbf{q}/2)g_{s,d}(\mathbf{k}'-\mathbf{q}/2). \tag{6}$$

Here, $V>0$ is the attractive pairing interaction strength and $g_s(\mathbf{k})=\cos k_x+\cos k_y$ and $g_d(\mathbf{k})=\cos k_x-\cos k_y$. Thus,

$$\Delta(\mathbf{k},\mathbf{q}) = \Delta_s(\mathbf{q})g_s(\mathbf{k}-\mathbf{q}/2) + \Delta_d(\mathbf{q})g_d(\mathbf{k}-\mathbf{q}/2). \tag{7}$$

The vector $\mathbf{q}$ labels mean-field solutions that correspond to order parameters in real space with phase winding numbers q_x and q_y in x and y directions, respectively.

If $\Delta(\mathbf{k},\mathbf{q})\neq\mathbf{0}$ for a single momentum $\mathbf{q}\neq\mathbf{0}$, then $\mathcal{F}(\mathbf{k},\mathbf{k}',\omega_n)$ and $\mathcal{F}^*(\mathbf{k},\mathbf{k}',\omega_n)$ have off-diagonal terms in momentum space, but $\mathcal{G}(\mathbf{k},\mathbf{k}',\omega_n)$ is still diagonal. If $\Delta(\mathbf{k},\mathbf{q})\neq 0$ for at least two different momenta $\mathbf{q}$, then also $\mathcal{G}(\mathbf{k},\mathbf{k}',\omega_n)$ has off-diagonal terms and the discrete translational invariance is broken. The charge density is obtained from $\rho(\mathbf{r})=1/N\Sigma_{\mathbf{k},\mathbf{k}'}e^{i\mathbf{r}\cdot(\mathbf{k}-\mathbf{k}')}n(\mathbf{k},\mathbf{k}')$, where $n(\mathbf{k},\mathbf{k}')=2T\Sigma_n\mathcal{G}(\mathbf{k},\mathbf{k}',\omega_n)$. Thus, there are charge modulations whenever $\mathcal{G}(\mathbf{k},\mathbf{k}',\omega_n)$ has off-diagonal terms.

Inserting Eq. (4) into Eq. (3) leads to a system of coupled equations for the Green's function $\mathcal{G}(\mathbf{k},\mathbf{k}',\omega_n)$. Assuming that $n(\mathbf{k},\mathbf{k}')\ll n(\mathbf{k})$ for $\mathbf{k}\neq\mathbf{k}'$, $\mathcal{F}(\mathbf{k},\mathbf{k}+\mathbf{q},\omega_n)$ and $\mathcal{F}^*(\mathbf{k},\mathbf{k}+\mathbf{q},\omega_n)$ are approximated by keeping only the term proportional to $\mathcal{G}(\mathbf{k}+\mathbf{q},\mathbf{k}+\mathbf{q},\omega_n)$ in the sum in Eq. (4). This approximation in Eqs. (3) and (4) leads to an analytical solution of the Gor'kov equations to leading order in $n(\mathbf{k},\mathbf{k}')$ for $\mathbf{k}\neq\mathbf{k}'$. The quantitative validity of this approximation will be verified *a posteriori*.

In an ansatz for a self-consistent solution of the Gor'kov equations (3) and (4), we choose Q trial vectors $\mathbf{q}_1,\ldots,\mathbf{q}_Q$ and set $\Delta(\mathbf{k},\mathbf{q})=0$ for all other values of $\mathbf{q}\neq\mathbf{q}_i$. Thereby we test selected combinations of $\mathbf{q}$ vectors for self-consistent solutions. With this ansatz, the energy spectrum of the system consists of $Q+1$ bands $E_\alpha(\mathbf{k})$, where $\alpha=0,\ldots,Q$. The conventional BCS solution is realized for $Q=1$ with just two quasiparticle bands and $\mathbf{q}=\mathbf{0}$. Generally, one obtains a set of $2Q$ coupled self-consistency equations for $\Delta_s(\mathbf{q}_i)$ and $\Delta_d(\mathbf{q}_i)$,

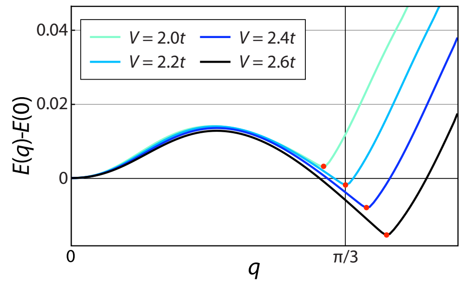

FIG. 1. (Color online) Energy $E=\langle\mathcal{H}\rangle$ as a function of pair momentum $\mathbf{q}=(q,0)$ for different pairing interaction strengths V. Calculations were performed for a 384×384 lattice with fixed electron density $\rho=0.8$ and $t'=0.3t$. For these parameters, the finite-momentum pairing state becomes the ground state for $V>V_c\approx 2.2t$ with $q=\pi/3$.

$$\frac{\Delta_{s,d}(\mathbf{q})}{V} = -\frac{T}{N}\sum_{\mathbf{k}'} g_{s,d}(\mathbf{k}'-\mathbf{q}_i/2)\sum_n \mathcal{F}(\mathbf{k},\mathbf{k}-\mathbf{q}_i,\omega_n). \tag{8}$$

The standard BCS theory provides isotropic solutions of Eq. (8) with $\Delta_s(\mathbf{0})=0$ and $\Delta_d(\mathbf{0})\neq 0$ for an arbitrarily weak interaction strength V. Most remarkably, if V exceeds a certain interaction strength, we identify solutions with $Q=2$ and with the specific set of $\mathbf{q}$ vectors $\{(q,0),(-q,0)\}$. These solutions are anisotropic and have a subdominant extended s-wave contribution $\Delta_s(\mathbf{q})$, which increases with increasing q. Below we will discuss in particular the time inversion symmetric zero-current solutions of Eq. (8), i.e., $\Delta_{s,d}(\mathbf{q})=\Delta_{s,d}(-\mathbf{q})$.

To test the stability of this solution, we solved Eq. (8) iteratively for selected combinations of $\mathbf{q}$ vectors and different initial values of the corresponding OPs. In particular, we investigated the stability of the above solution against decay into the $\mathbf{q}=\mathbf{0}$ state by using the ansatz with the three center-of-mass momenta $\{(q,0),(-q,0),(0,0)\}$. We find that for a pure on-site interaction $V(\mathbf{k},\mathbf{k}',\mathbf{q})\equiv V_0$ (s-wave pairing), finite-momentum pairing is unstable. All OPs with different $\mathbf{q}$'s compete, even the ones with $\mathbf{q}$ and $-\mathbf{q}$. Thus, states with $Q\geq 2$ will always decay into a state with only one finite order parameter for on-site s-wave pairing. However, for the nearest-neighbor interaction (6), additional stable solutions emerge. The finite-momentum pairing solutions are typically stable for a wide range of q values. Here, the OPs for $\pm q$ do not compete, but rather support each other. The range of stability however decreases with decreasing V and eventually disappears.

So far we have verified that stable finite-momentum pairing solutions of the self-consistency equation exist. They refer to local minima of the free energy. To determine the ground state at $T=0$, the global minimum of the energy $E=\langle\mathcal{H}\rangle=\Sigma_{\mathbf{k}}\varepsilon_{\mathbf{k}}n(\mathbf{k})+\Sigma_i[\Delta_s^2(\mathbf{q}_i)+\Delta_d^2(\mathbf{q}_i)]/V$ has to be determined with respect to all q. Figure 1 shows the typical q dependence of E with a minimum at $q=0$ and a further minimum for $q>0$. The minimum at $q=0$ corresponds to the standard d-wave SC state. With increasing V, the energy of the minimum at finite q decreases accompanied by a shift to larger q. This implies the existence of a critical interaction strength V_c, which depends on t' and the electron density

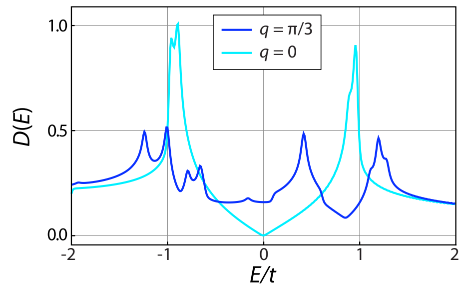

FIG. 2. (Color online) Density of states $D(E)$ of the ground-state solutions for interaction strengths $V=2t$ and $V=2.2t$, corresponding to $q=0$ and $q=\pi/3$, respectively. The other parameters are the same as in Fig. 1. The coherence peaks of the $\mathbf{q=0}$ state are split due to the Van Hove singularity of the two-dimensional tight-binding dispersion.

$\rho=1/N\Sigma_{\mathbf{k}}n(\mathbf{k})$. Above V_c the potential-energy gain from pairing overcomes the concomitant increase in the kinetic energy due to the finite center-of-mass momentum of each pair and, consequently, the finite-momentum pairing state is the ground state. The optimal $\mathbf{q}=q_{\min}(1,0)$ or $\mathbf{q}=q_{\min}(0,1)$ sensitively depends on V, t', and ρ, but it is typically found in between $q\approx\pi/8$ and $q\approx\pi/2$ for a wide parameter range. $q_{\min}$ is near the maximum distance between the nodes on the Fermi surface of the two order parameters $\Delta(\mathbf{k},\mathbf{q})$ and $\Delta(\mathbf{k},-\mathbf{q})$. This suggests that the two order parameters are in competition and their coexistence demands that those regions in momentum space with maximum pairing amplitude of either of the two are optimally separated. We emphasize that this mechanism for the stabilization of the finite-momentum pairing state is not possible for isotropic superconductors.

For the finite-q ground-state solutions the charge density $\rho(\mathbf{r})$ has an oscillatory part arising from the off-diagonal terms of the Green's function. For the $\mathbf{q}=(\pm q,0)$ state the charge density forms a sinusoidal stripe pattern with wave number $2q$. Correspondingly, the charge density varies as

$$\rho(\mathbf{r})=\rho+\rho_1\cos(2qx). \quad (9)$$

For all analyzed parameter sets, which led to stable ground-state solutions with finite-momentum pairing, the relative charge modulation with an amplitude ρ_1/ρ was near 2%, which *a posteriori* justifies the assumption of small charge modulations in the above approximation for $\mathcal{F}(\mathbf{k},\mathbf{k}',\omega_n)$. For $q=\pi/3$, the wavelength of the stripe pattern is therefore three lattice constants. The charge modulation in the SC state suggests us to include a self-consistent charge-density-wave (CDW) OP in the mean-field decoupling scheme of the Hamiltonian (1). We have analyzed this extension with coexisting OPs for SC and CDW orders for selected cases. The CDW OP tends to stabilize the state with finite-momentum pairing but it remains small and does not change the solutions qualitatively. Also arbitrary orientations of the Cooper pair's center-of-mass momenta were considered, but in all cases the lowest-energy solutions were obtained for momenta in (10) and (01) directions.

The finite-momentum pairing state has further characteristic properties that are at variance with a BCS-like d-wave superconductor (with $q=0$). The DOS $D(E)$

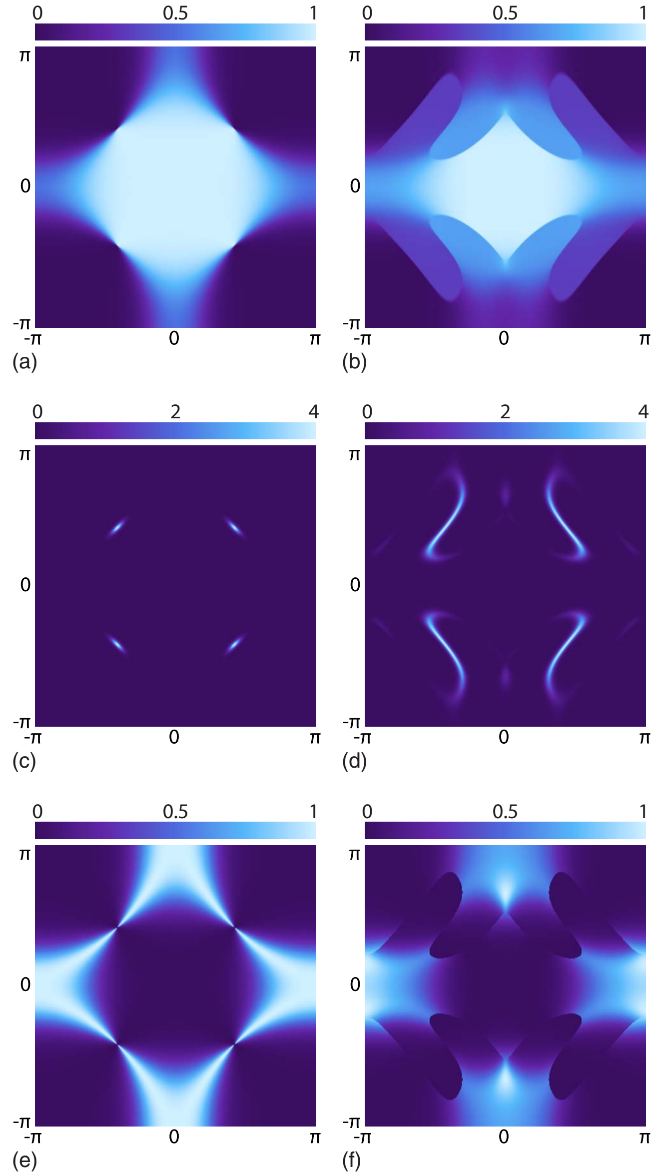

FIG. 3. (Color online) Momentum space properties of the finite-momentum pairing state with $q=\pi/3$ and the same parameters as in Fig. 2 (right panels) and for comparison the d-wave superconductor for $q=0$ (left panels). (a) and (b) Occupation probability function $n(\mathbf{k})$. (c) and (d) Density of states with zero-energy $\mathrm{Im}\,G(\mathbf{k},\mathbf{k},0-i\delta)$ (here, $\delta=0.04t$). (e) and (f) Pair density $P(\mathbf{k})$.

$=\Sigma_{\mathbf{k}}\mathrm{Im}\,G(\mathbf{k},\mathbf{k},E-i0^+)$, where Im G is the imaginary part of the analytical continuation of $\mathcal{G}$ to the real frequency axis, is shown in Fig. 2. For $q=\pi/3$, the DOS bears little resemblance to a d-wave-like gap as the coherence peaks are split and the DOS is finite at the Fermi energy. A similar splitting is observed for current carrying d-wave states,[15,16] which originates from the Doppler shift of the finite-momentum eigenstates.

Figure 3 displays the characteristic momentum space properties of the finite-momentum pairing state and, for comparison, of the $q=0$ d-wave superconductor. In the finite-momentum pairing state, the momentum distribution function $n(\mathbf{k})=n(\mathbf{k},\mathbf{k})$ develops structures with sharp boundaries. These boundaries consist of lines in momentum space with

$E_\alpha(\mathbf{k})=0$, for $\alpha=0$, 1, or 2, which is indicative of a Fermi-surface reconstruction. The zero-energy states generate Fermi-arc-like structures as shown in Fig. 3(d). For $t'=0$, $n(\mathbf{k})$ is similar to the result obtained in Ref. 6. The pair density $P(\mathbf{k})=\Sigma_i P(\mathbf{k},\mathbf{k}-\mathbf{q}_i)$, where $P^2(\mathbf{k},\mathbf{k}')=2\langle c^\dagger_{\mathbf{k}\uparrow}c^\dagger_{-\mathbf{k}'\downarrow}\rangle\langle c_{-\mathbf{k}'\downarrow}c_{\mathbf{k}\uparrow}\rangle$, clearly shows that the finger-shaped $\mathbf{k}$-space structures of $n(\mathbf{k})$ contain unpaired electrons only. The overall number of pairs is smaller in the finite-momentum pairing state than in the $q=0$ state. This seems to contradict the fact that it has the lower energy. The latter, however, consists of both the kinetic energy, which rises in the SC state and acts against the formation of pairs, and the gain of condensation energy. The optimal balance between these two contributions depends on details of the single particle kinetic energy $\varepsilon_\mathbf{k}$ and the interaction potential $V(\mathbf{k},\mathbf{k}',\mathbf{q})$ and does not generally favor a larger number of paired electrons.

In this Rapid Communication we have shown that the extended BCS theory with attractive nearest-neighbor interaction provides self-consistent solutions with the simultaneous formation of electron pairs with center-of-mass momenta $\mathbf{q}$ and $-\mathbf{q}$. It is a microscopic solution that constitutes a stable macroscopic state of the PDW type, which was proposed to describe the striped SC phase in hole-doped 214 cuprates. This finite-momentum pairing state is the ground state beyond a critical interaction strength V_c. V_c depends sensitively on the band filling ρ and ranges from $V_c\approx 1.4t$ for $\rho=0.6$ to $V_c\approx 3.5t$ for $\rho=1$. This is consistent with the result in Ref. 7 that only the uniform phase with fixed $\mathbf{q}$ can be the ground state of the BCS Hamiltonian in the weak-coupling limit.

We emphasize that the solutions of Gor'kov's equations described above are identically reproduced by using an extended Bogoliubov transformation and are in qualitatively accurate agreement with numerical solutions of the Bogoliubov-de Gennes equations in real space. Our results demonstrate as a proof of principle that stable ground-state solutions of the pairing Hamiltonian (1) exist with coexisting finite-momentum pairing amplitudes for center-of-mass momenta $\mathbf{q}=(q,0)$ and $-\mathbf{q}$; these solutions are absent for an attractive contact interaction and their possibility arises only for nodal superconductors. Due to the concomitant striped charge-density modulation with wave vector $2\mathbf{q}$, a connection to the striped superconductor $La_{15/8}Ba_{1/8}CuO_4$ appears tempting. However, without the inclusion of additional correlation effects as the source for a possible spin order pattern, we consider it premature to draw conclusions about the favorable wavelength of the stripes.

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