Spontaneous spin-loop-current order mediated by transverse spin fluctuations in cuprate superconductors

Hiroshi Kontani, Youichi Yamakawa, Rina Tazai, and Seiichiro Onari
Department of Physics, Nagoya Furo-cho, Nagoya 464-8602, Japan.
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We predict the theoretical occurrence of nanoscale spontaneous spin-current, called the spin-loop-current (sLC) order, as a promising origin of the pseudogap and electronic nematicity in cuprates. We demonstrate that the spontaneous sLC is accompanied by the exotic odd-parity electron-hole pairs that are mediated by transverse spin fluctuations around the pseudogap temperature $T^*$. The present theory predicts the occurrence of the condensation of odd-parity magnon pairs simultaneously. The sLC order is “hidden” in that neither internal magnetic field nor charge density modulation is induced, whereas the predicted sLC with finite wavenumber naturally gives the Fermi arc structure. In addition, the fluctuations of sLC order work as attractive pairing interaction between adjacent hot spots, which enlarges the $d$-wave superconducting transition temperature $T_c$.

Thus, the sLC state will be the key ingredient in understanding pseudogap, electronic nematicity as well as superconductivity in cuprates and other strongly correlated metals.

Various exotic symmetry-breaking phenomena, such as violations of rotational and parity symmetries, are the central issues in cuprate high-$T_c$ superconductors. However, their microscopic mechanisms still remain as unsolved issues. Figure 1 (a) shows a schematic phase diagram of cuprate superconductors. Below $T_{CDW}$ ~ 200K, a stripe charge-channel density-wave emerges at finite wavevector $q \approx (\pi/2, 0)$ in many compounds [1–4], which produces the Fermi arc structure and causes a reduction in the density-of-states (DOS). However, it cannot be the origin of the pseudogap temperature $T^*$ since $T^* > T_{CDW}$. Short quasiparticle lifetime due to spin or charge fluctuations could reduce the DOS [5–7].

FIG. 1: (a) Schematic phase-diagram of hole-doped cuprate superconductors. The sLC phase is obtained by the present study. (b) Current of spin ($\sigma = \pm 1$) from site 1 to site 2. (c) Schematic single-$q$ sLC orders at $q_{sLC} = (\pi/2, \pi/2)$.

Recently, much experimental evidence for the phase transition at $T^*$ has been accumulated [8–13]. Various fascinating order parameters have been proposed and actively investigated, such as the CDW or bond-order (BO) [14–23], the pair-density-wave [24, 25], and the charge loop-current (cLC) order [26–29]. To understand the mechanism of these exotic orders, higher-order quantum processes have been considered actively [15–19, 21–23] since simple mean-field-level approximations lead to conventional spin-density-wave instabilities.

Let us discuss the symmetry breaking in the correlated hopping between sites $i$ and $j$: $t_{i,j} \rightarrow t_{i,j} + \delta t_{i,j}$, where $\delta t_{i,j}$ is the order parameter. Then, the BO is given by a real and even-parity $\delta t_{i,j} [14, 15, 17–23]$. A spin-fluctuation mechanism [22, 23] predicts the ferro ($q = 0$) $d$-wave BO state at $T^*$ and stripe ($q \approx (\pi/2, 0)$) BO at $T_{CDW}$. The former order explains the experimental nematic transition [10, 13]. However, simple translational symmetry preserving ferro-BO does not explain the pseudogap formation. Also, the cLC order is given by a pure imaginary and odd-parity $\delta t_{i,j} [26–29]$. Both order parameters have been actively investigated.

In contrast, spin current flows if pure imaginary order parameter is odd under space and spin inversions; $\delta t_{i,j} = -\delta t_{j,i} = -\delta t_{j,i}$ as shown in Fig. 1 (b) [29–34]. Here, $\sigma = \pm 1$ represents the spin of the electron. Examples of spin loop-current (sLC) orders at the wavevector $q_{sLC} = (\delta, \delta)$ with $\delta = \pi/2$ is given in Fig. 1 (c). The sLC is a hidden order in the sense that no internal magnetic field appears, and charge density modulation is quite small. On the other hand, the pseudogap and Fermi surface (FS) reconstruction are induced by band-folding if $q_{sLC} \neq 0$.

The sLC is very valid and promising as the origin of the pseudogap; therefore, its emergence has been discussed not only in cuprates [29–31], but also in iridates [32] and heavy-fermion compound [33, 34]. From the microscopic viewpoint, however, the mechanism of the sLC is highly nontrivial, since the realization condition of the sLC order is very severe in the extended $U$-$V$-$J$ Hubbard model within the mean-field theory [30]. In addition, only the case $q_{sLC} = (\pi, \pi)$ was analyzed in previous works.

In this paper, we discover the higher-order many-body effect that induces the exotic electron-hole pairs accompanied by finite spin current at $q_{sLC} \approx (\pi/2, \pi/2)$. The key pairing mechanism is the spin-flipping magnon-
induced self-energy self-consistently [40]. any fine tuning of $U \sim T \chi$ where $|\chi| = 1$ is satisfied for $U > 3.3$ without any fine tuning of $U$ by considering the spin-fluctuation-induced self-energy self-consistently [40].

Here, we analyze the single-orbital square-lattice Hubbard model $H = \sum_{k,\sigma} \epsilon_k c^\dagger_k c_k + U \sum_i n_{i\uparrow} n_{i\downarrow}$. We denote the hopping integrals $(t_1, t_2, t_3) = (-1, 1/6, -1/5)$, where $t_j$ is the $j$-th nearest hopping integral [40]. Hereafter, we set the unit of energy as $t_1 = 1$, which corresponds to $\sim 4000$ [K] in cuprates, and fix the temperature $T = 0.05$ (≈ 200K). The FS at filling $n = 0.85$ is given in Fig. 2 (a). The spin susceptibility in the random-phase-approximation (RPA) is $\chi^s(q) = \chi^0(q)/(1 - U \chi^0(q))$, where $\chi^0(q)$ is the irreducible susceptibility without $U$ and $q \equiv (q, \omega)$. The spin Stoner factor is defined as $\alpha_S \equiv \max_q \{U \chi^0(q)\} = U \chi^0(Q_s, 0)$. When $\alpha_S = 0.99$ ($U = 3.27$), then $\chi^s(Q_s, 0) \sim 80$ [\mu_B^2/ev], which is still smaller than Im$\chi^s(Q_s, E = 31$meV) $\sim 200$ [\mu_B^2/ev] at $T \sim 200$K in 60K YBCO [41]. Thus, $\alpha_S > 0.99$ in real compounds. Owing to the Mermin-Wagner theorem, the relation $\alpha_S \lesssim 1$ is naturally satisfied for $U > 3.3$ without any fine tuning of $U$ by considering the spin-fluctuation-induced self-energy self-consistently [40].

![FIG. 2: (a) The FS of the present model at $n = 0.85$. $Q_a$ is the major nesting vectors. $Q_d = (\delta, \delta)$ and $Q_a = (\delta, 0)$ (\delta$\approx \delta_{QS}$) are minor nesting vectors. They correspond to the sLC/BO wavelength in the present theory. (b) Obtained eigenvalue $\lambda_q$ for the BO at $n = 0.80 \sim 0.88$. They have peaks at $q = 0$ and $q = Q_a$. (Inset) Relations $\alpha_S - 1 = -0.44p^2$ (full line) and $\alpha_S = 1.01 - 0.2p$ (broken line).

From now on, we investigate possible exotic density-wave (DW) states for both charge- and spin-channels with general wavevector ($q$), which is generally expressed as $D^\rho_q(k) = \langle c^\dagger_{k-\sigma} c_{k+\rho} \rangle - \langle c^\dagger_{k-\sigma} c_{k+\rho} \rangle_0 = f_q(k)\delta_{\sigma,\rho} + g_q(k) \cdot \sigma_{\sigma,\rho}$ [30], where $k_\pm = k \pm q/2$, and $f_q(k)$ ($g_q(k)$) is the charge (spin) channel order parameter, which we call the form factors in this paper. Below, we assume $g_q(k) = g_q(k)e_2$ without losing generality. The DW is interpreted as the electron-hole pairing condensation [30].

Here, $f_q(k)$ is given by the Fourier transformation of the spin-independent hopping modulation $\sum_{r,\tau} t_{ij} c_r^{\dagger}(r-\tau) c_{r+\tau}(r+\tau) q/2$. When $\delta t_{ij} = \pm \delta t_{ij}$, the relation $f_q(k) = \pm f_q(-k)$ holds. Also, $g_q(k)$ is given by the spin-dependent modulation $\delta t_{ij} = -\delta t_{ij}$. The even-parity $f_q(k)$ and the odd-parity $g_q(k)$ respectively correspond to the BO state and the sLC state. Both states preserve the time-reversal symmetry.

To find possible DW in an unbiased way, we generalize the DW equation [23] for both spin/charge channels:

$$\lambda_q f_q(k) = -\frac{T}{N} \sum_p I^q_{\pi}(k, p) G(p_-) G(p_+) f_q(p),$$

$$\eta_q g_q(k) = -\frac{T}{N} \sum_p I^q_{\pi}(k, p) G(p_-) G(p_+) g_q(p),$$

where $\lambda_q (\eta_q)$ is the eigenvalue that represents the charge (spin) channel DW instability, $k \equiv (k_1, \epsilon_m)$, $p \equiv (p, \epsilon_m)$ ($\epsilon_n, \epsilon_m$ are fermion Matsubara frequencies). These DW equations are interpreted as the “spin/charge channel electron-hole pairing equations”.

The charge (spin) channel kernel function is $I^q_{\pi}(s) = I^q_{\pi}^{(+)} + (-)I^q_{\pi}^{(-)}$: $I^q_{\pi}^{(+)}$ at $q = 0$ is given by the Ward identity $-\delta \Sigma_{\sigma}^q(k) / \delta \Sigma_{\rho}^q(k')$, which is composed of one single-magnon exchange term and two double-magnon exchange ones: The former and the latter are called the Maki-Thompson (MT) term AL terms; see Fig. S1 in the Supplemental Materials (SM) A [42]. The lowest order Hartree term $-U \delta_{\sigma,\rho}$ in $I^q_{\pi}^{(+)}$ gives the RPA contribution, while the AL terms are significant for $\alpha_S \lesssim 1$ [23, 35]. The significance of the AL processes have been revealed by the functional-renormalization-group (fRG) study, in which higher-order vertex corrections are produced in an unbiased way [22, 43]. Note that the MT term is important for the superconducting gap equation and for the transport phenomena [40].

Figure 2 (b) shows the charge-channel eigenvalue $\lambda_q$ derived from the DW eq. (1) [23, 43]. Hereafter, we put $U$ to satisfy the relation $\alpha_S = 1 - 0.44p^2$ with $p = 1 - n$, shown as full line in the inset. The obtained form factor $f_q(k)$ at $q = 0, Q_d$ belongs to $B_{1g}$ symmetry BO, consistently with previous studies [22, 23].

Next, we discuss the spin-fluctuation-driven sLC order, which is the main issue of this manuscript. Figure 3 (a) exhibits the spin-channel eigenvalue $\eta_q$ derived from the DW eq. (2). Peaks of $\eta_q$ are located at the nesting vectors $q = Q_d$ (diagonal) and $q = Q_a$ (axial). The obtained form factor $g_q(k)$ at $q = Q_d$ (diagonal sLC) is shown in Fig. 3 (b). The odd-parity solution $g_q(k) = -g_q(-k)$ means the emergence of the sLC order. The reason for large $\eta_{Q_d}$ is that all hot spots contribute to the
diagonal sLC as shown in Fig. 3 (b). Figure 3 (c) shows the form factor in real space \( \text{Im} q \rho (r) \) with \( r = (x, y) \).

To understand why sLC state is obtained, we simplify Eq. (2) by taking the Matsubara summation analytically by approximating that \( I_q \) and \( g_\mu (k) \) are static:

\[
\eta_q g_\mu (k) = \frac{1}{N} \sum_p I_q^\mu (k, p) F_q (p) g_\mu (p),
\]

where \( F_q (p) = -T \sum_m G(p_+) G(p_-) = \frac{n(\epsilon_{p_+}) - n(\epsilon_{p_-})}{\epsilon_{p_+} - \epsilon_{p_-}} \) is a positive function, and \( n(\epsilon) \) is Fermi distribution function; see the SM A [42]. In general, the peak positions of \( \eta_q \) in Eq. (3) are located at \( q = 0 \) and/or nesting vectors with small wavelength (\( q = Q_\alpha, Q_\beta \) in the present model). The reason is that \( I_q \sim T \sum Q \lambda^\alpha (p_+) \lambda^\beta (p_-) \) by AL terms is large for small \( |q| \), and \( F_q (p) \) is large for wide area of \( p \) when \( q \) is a nesting vector.

To understand why odd-parity form factor is obtained, we show the "electron-hole pairing interaction" \( I_{q=0}^\alpha (k, k') \) on the FS in Fig. 3 (d). Here, \( k \) represents the position of \( k \) shown in Fig. 3 (e). Since \( I_{q=0}^\alpha (k, k') \) gives large attractive interaction for \( k \approx k' \) and large repulsive one for \( k \approx -k' \), the odd-parity form factor \( g_\eta (k) \) is naturally obtained. Figure 3 (e) summarizes the origin of odd-parity sLC: Red (blue) arrows represent the attractive (repulsive) interaction.

The strong \( k, k' \)-dependence of \( I_{q=0}^\alpha (k, k') \) originates from the AL1 and AL2 terms in Fig. 3 (f), or Fig. S1 (a) in the SM [42]. Owing to the spin-conservation law, AL terms in \( I = I_{\uparrow\downarrow} + I_{\downarrow\uparrow} \) originate from the spin-flipping processes due to transverse spin fluctuations in Fig. 3 (f), in proportion to \( \chi_\pm (Q_s) \chi_\pm (Q_s) \). (In \( I^s \), the spin non-flipping AL processes in proportion to \( \chi_\pm (Q_s) \chi_\pm (Q_s) \) are canceled out.) Therefore, \( I = [AL1] - [AL2] \). The AL1 term with the p-p (anti-parallel) pair Green functions causes large attractive interaction for \( k \approx k' \), and the AL2 term with the p-p (parallel) ones does for \( k \approx -k' \), as explained in Ref. [43] in detail. Thus, \( k, k' \)-dependence in Fig. 3 (d) and resultant odd-parity solution is understood naturally. In contrast, \( I = 3([AL1] + [AL2])/2 \), so the even-parity BO is obtained [23, 43].

\[
\lambda_{SC} \Delta(k) = T \sum_p V_{SC} (k, p) |G(p)|^2 \Delta(p),
\]
Thus, the sLC/BO discussed here and the spin nematic and (b) show the Fermi arc structures in the cases of gap due to the diagonal sLC order. Figures 5 (a) where FIG. 5: (a) Fermi arc structure due to the single-

diagonal sLC with period 4

terms of the electron-hole pairing. Another physical arc nor pseudogap, as explained in the SM B [42].

In the present theory, we discussed the sLC/BO in terms of the electron-hole pairing. Another physical interpretation of the sLC/BO is the “condensation of odd/even parity magnon-pairs”, which is the origin of the nematic order in quantum spin systems [46–48]. In fact, the two-magnon propagator shown in Fig. 4 (c) diverges when the eigenvalue of DW equation reaches unity, as we explain in the SM C [42]. That is, triplet (singlet) magnon-pair condensation occurs at $T = T_{\text{sLC}}(T_{\text{CDW}})$. Thus, the sLC/BO discussed here and the spin nematic order in quantum spin systems are essentially the same phenomenon.

Here, we discuss the band-folding and hybridization gap due to the diagonal sLC order. Figures 5 (a) and (b) show the Fermi arc structures in the cases of (a) single-$\mathbf{q}$ and (b) double-$\mathbf{q}$ orders. We set $g_{\text{max}} \equiv \max_{\mathbf{q}} \{g_{\mathbf{q}_4}(\mathbf{k})\} = 0.1$. Here, the folded band structure under the sLC order with finite $q_{\text{sLC}}$ is “unfolded” into the original Brillouin zone by following Ref. [49] to make a comparison with ARPES results. The Fermi arc due to the single-$\mathbf{q}$ order in Fig. 5 (a) belongs to $B_{2g}$ symmetry. The resultant pseudogap in the DOS is shown in Fig. 5 (c). The unfolded band structure in the single-$\mathbf{Q}_4$ sLC order is displayed in Fig. S4 in the SM B [42].

In the single-$\mathbf{q}$ diagonal sLC order, the uniform magnetic susceptibility shows $B_{2g}$ anisotropy due to the Van-Vleck contribution [50], consistently with the $B_{2g}$ nematicity observed in Hg-based cuprates [11]. In addition, anisotropy in $g$-vector in the sLC state can lead to the anisotropy of magnetic susceptibility [34].

Next, we investigate the spin current in real space, which is driven by a fictitious Peierls phase due to the “spin-dependent self-energy” $\delta t_{ij}^\sigma = bg_{ij}$. As shown in Fig. 3 (c), $\delta t_{ij}^\sigma$ is purely imaginary and odd with respect to $i \leftrightarrow j$. The conservation law $n_i^\sigma = \sum_j J_{ij}^\sigma$ directly leads to the definition the spin current operator from site $j$ to site $i$ as $J_{ij}^\sigma = -i\sum_n \sigma(h_n^\sigma \sum_j c_n^\sigma c_{j,\sigma} - (i \leftrightarrow j))$, where $h_n^\sigma = t_{ij}^\sigma + \delta t_{ij}^\sigma$. Then, the spontaneous spin current from $j$ to $i$ is $J_{ij}^\sigma = (J_{ij}^\sigma + J_{ji}^\sigma)_{\mathbf{k}}$

Here, we calculate the spin current for the commensurate sLC order at $q_{\text{sLC}} = (\pi/2, \pi/2)$, which is achieved by putting $n = 1$. Then, the unit cell are composed four sites A-D. Figure 5 (c) shows the obtained spin current $J_{ij}^\sigma$ from the center site ($j = \text{A-B}$) to different sites in Fig. 1 (c), by setting $g_{\text{max}} = 0.1$. The obtained current is $|J_{ij}^\sigma| \sim 10^{-2}$ in magnitude in unit $|t_1|/h$. The derived spin current pattern between the nearest and second-nearest sites is depicted in Fig. 1 (c). The spin current is exactly conserved at each site.

The Fermi arc structure and the DOS in Fig. 5 is independent of the phase shift $g_{ij} \rightarrow e^{i\theta_{ij}} g_{ij}$. In contrast, the real space current pattern depends on the phase shift. We discuss other possible sLC patterns in the SM D [42]. The charge modulation due to the sLC is just $|\Delta n_i| \sim 5 \times 10^{-4}$ for $g_{\text{max}} = 0.1$ since $|\Delta n_i| \propto (g_{\text{max}})^2$. Thus, experimental detection of translational symmetry breaking by sLC order may be difficult. However, the cLC is induced by applying uniform magnetic field parallel to $g_{ij}$. In the present sLC state, under 10 T magnetic field, the induced cLC gives $\Delta H \sim \pm 0.1$ Oe when the mass-enhancement factor is $m^*/m \sim 10$, which may be measurable by NMR or $\mu$SR study.

Finally, we stress that the fluctuations of the sLC order will contribute to the pairing mechanism. In fact, the pairing interaction mediated by the spin-channel sLC fluctuations between singlet pairs ($\mathbf{k}_+,-\mathbf{k}_+$) and ($\mathbf{k}_-,-\mathbf{k}_-$) is $V^\text{SC}(\mathbf{k}_+,-\mathbf{k}_+) \propto -g_{\mathbf{q}}(\mathbf{k})g_{\mathbf{q}}(-\mathbf{k})\chi_{\text{sLC}}(\mathbf{q})$ for $\mathbf{q} \approx \mathbf{Q}_4$, where $\chi_{\text{sLC}}(\mathbf{q})$ is the sLC susceptibility; see the SM A [42]. Since $g_{\mathbf{q}}(\mathbf{k})$ is real and odd-function, the sLC fluctuations give positive (=attractive) pairing interaction between adjacent hot spots, which will be important for slightly over-doped cuprates with $T_{\text{sLC}} \lesssim T_c$.

We will discuss this issue in future publications.

In summary we proposed a novel and long-sought for-
maticity mechanism for the nanoscale spin-current order, which violates the parity and translational symmetry while time-reversal symmetry is preserved. The band-folding by the sLC orders results in the formations of the Fermi arc structure and pseudogap at $T \sim T_c$. In the sLC state, a staggered moment is expected to appear under the uniform magnetic field. The sLC order will be the key ingredient in understanding pseudogap phase and electronic nematicity not only in cuprates, but also in iridates [32, 39] and heavy-fermion compound [34].
is an important future issue to incorporate the self-energy effect into the present theory.

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A: DERIVATION OF SINGLET AND TRIPLET DW EQUATIONS

Here, we discuss the linearized density-wave (DW) equation driven by spin fluctuations. For this purpose, we introduce the irreducible four-point vertex function \( I_{q}^{\sigma}(k, k') \). It is given by the Ward identity at \( q = 0 \), that is, \( I_{q}^{\sigma}(k, k') \equiv -\delta \Sigma_\sigma(k)/\delta G_\rho(k') \). Here, we use the one-loop self-energy given as

\[
\Sigma_\sigma(k) = T \sum_q U^2 \chi^\sigma_L(q) G_\sigma(k + q) + T \sum_p U^2 \chi^\sigma_T(q) - \chi^{(0)\sigma}(q) G_{-\sigma}(k + q) \tag{S1}
\]

where \( \chi^\sigma_L(q) \) and \( \chi^\sigma_T(q) \) are longitudinal and transverse susceptibilities. They are given as

\[
\chi^\sigma_L(q) = \chi^{(0)\sigma}(q) (1 - U^2 \chi^\sigma_L(q) \chi^\sigma_L(q))^{-1}, \tag{S2}
\]

\[
\chi^\sigma_T(q) = \chi^{(0)\sigma}(q) (1 - U \chi^{(0)\sigma}(q))^{-1} \tag{S3}
\]

where \( \chi^{(0)\sigma}(q) = -T \sum_p G_\sigma(p) G_\sigma(p + q) \) and \( \chi^{(0)\sigma}(q) = -T \sum_p G_\sigma(p) G_{-\sigma}(p + q) \) are longitudinal and transverse irreducible susceptibilities. Then, the irreducible vertex function \( I_{q}^{\sigma}(k, k') \) given by the Ward identity is composed of one MT term and two AL terms in Fig. S1 (a). Note that \( I_{q}^{\sigma} \) contains the lowest order Hartree term \(-U \delta_\sigma \).

First, we derive the charge-channel (singlet) DW equation in the absence of the magnetic field, where the form factor is independent of spin: \( f_q^1(k) = f_q^1(k) = f_q(k) \). The singlet DW equation was introduced in the study of Fe-based superconductors [43] and cuprate superconductors [23]. It is given as

\[
\lambda_q f_q(k) = -\frac{T}{N} \sum_{k'} I_{q}^{1}(k, k') G(k'_{-}) G(k'_{+}) f_q(k'), \tag{S4}
\]

which is shown in Fig. S1 (b), and \( k_{\pm} \equiv k \pm q/2 \). Here, \( I_{q}^{1}(k, k') = I_{q}^{1,\sigma}(k, k') + I_{q}^{1,-\sigma}(k, k') \). It is given as

\[
I_{q}^{1}(k, k') = -\frac{3}{2} V^s(k - k') - \frac{1}{2} V^c(k - k')
+ \frac{T}{N} \sum_p \left[ \frac{3}{2} V^s(p_+) V^s(p_-) + \frac{3}{2} V^c(p_+) V^c(p_-) \right]
\times G(k - p) G(k' - p) \tag{S5}
\]

where \( p = (p, \omega) \), \( V^s(q) = U + U^2 \tilde{\chi}^s(q) \), and \( V^c(q) = -U + U^2 \tilde{\chi}^c(q) \). The first, the second, and the third terms in Eq. (S5) corresponds to the MT, AL1 and AL2 terms in Fig. S1 (a).

In cuprates, Eq. (S4) gives even-parity solution with wavevector \( q = 0 \) and \( q \approx (\pi/2, 0) \). This singlet and even-parity electron-hole condensation is interpreted as the BO.

![FIG. S1: (a) Irreducible four-point vertex \( I_{q}^{1}(k, k') \) composed of one MT term and two AL terms. (b) Linearized singlet DW equation with the kernel \( I^1 \equiv I^{1,\sigma} + I^{1,-\sigma} \). (c) Linearized triplet DW equation with the kernel \( I^1 \equiv I^{1,\sigma} - I^{1,-\sigma} \). (d) A three-magnon exchange term, which is less important. (e) Full four-point vertex \( \Gamma_q^{(c)}(k, k') \) given by solving the DW equation. The sLC order (BO) emerges when \( \Gamma_q^{(c)}(k, k') \) diverges.](image)

Next, we derive the spin-channel (triplet) DW equation in the absence of the magnetic field, the spin-dependent
form factor is \( g_q(k) \equiv \tilde{g}^i_q(k) = -\tilde{g}^i_q(k) \). It is given as

\[
\eta_q g_q(k) = -\frac{T}{N} \sum_{k'} I^s_q(k,k') G(k') G(k') g_q(k'),
\]

which is shown in Fig. S1 (c). Here, \( I^s_q(k,k') = I^s_{q,-}(k,k') - I^s_{q,+}(k,k') \). It is given as

\[
I^s_q(k,k') = \frac{1}{2} V^s(k-k') - \frac{1}{2} V^c(k-k') + \frac{T}{N} \sum_p \left[ V^s(p+V^s(p-) + \frac{1}{2} V^c(p) V^c(p-) \right.
\]

\[
+ \frac{1}{2} V^c(p) V^c(p-) G(k-p) G(k'-p) + \frac{T}{N} \sum_p \left[ -V^s(p+V^s(p-) + \frac{1}{2} V^c(p) V^c(p- \right.
\]

\[
+ \frac{1}{2} V^c(p) V^c(p-) G(k-p) G(k'-p) + \frac{1}{2} V^c(p) V^c(p-) G(k-p) G(k'-p) + \frac{1}{2} V^c(p) V^c(p-) G(k-p) G(k'-p),
\]

where the first, the second, the third terms in Eq. (S5) corresponds to the MT, AL1 and AL2 terms in Fig. S1 (a). The AL terms with \( V^s(p+) V^s(p-) \) are shown in Fig. 3 (f). In cuprates, Eq. (S6) gives the odd-parity solution at wavevector \( q = (\pi/2, \pi/2) \) and \( (\pi/2, 0) \). This triplet and odd-parity electron-hole pairing is interpreted as the spin-loop-current (sLC).

In both Eqs. (S5) and (S7), the AL terms are proportional to \( \phi^{(2)}_q \) \( T \sum_{p_1, p_2} V^s(p_1) V^s(p_2) \cdot \delta_{p_1+p_2,q} \). The AL terms are significant when the spin fluctuations are large, since both \( V^s(p_1) \) and \( V^s(q-p_1) \) take large value simultaneously when \( p_1 \equiv Q_s \) in the case of \( q \approx 0 \). If we put \( V^s(p) \propto \xi^2/(1+\xi^2(p-Q_s)^2) \) at zero Matsubara frequency, where \( \xi \gg 1 \) is the magnetic correlation length, \( \phi^{(2)}_{q=0} \propto T \xi^2 \) in two-dimensional systems. Therefore, double-magnon exchange (AL) terms induce not only BO, but also the sLC order when \( \xi \gg 1 \). A three-magnon exchange term shown in Fig. S1 (d) is proportional to \( \phi^{(3)}_q \propto T^2 \sum_{p_1, p_2, p_3} V^s(p_1) V^s(p_2) V^s(p_3) \cdot \delta_{p_1+p_2+p_3,q} \). Then, \( \phi^{(3)}_{q=0} \propto T^2 \xi^2 \) in two-dimensional systems for \( q \sim Q_s \), which is smaller than \( \phi^{(2)}_{q=0} \) at low temperatures \( T \ll E_F \). Thus, the AL process would be the most significant, which is also indicated by functional-renormalization-group studies [22].

The electron-hole pairing order is generally expressed in real space as follows [30]:

\[
D^{s}_{i,j} = \{c^i_{\sigma\rho \alpha\beta} c^j_{\sigma\rho \alpha\beta}\}_{\sigma,\rho,\alpha,\beta} = f_{i,j} \delta_{\sigma,\rho} + g_{i,j} \cdot \sigma_{\sigma,\rho},
\]

where \( D^{s}_{i,j} = \{D^{s}_{i,j}\}^* \), and \( f_{i,j} \) \( g_{i,j} \) is spin singlet (triplet) pairing. The BO is given by real even-parity function \( f_{i,j} = f_{j,i} \), and the sLC is given by pure imaginary odd-parity vector \( g_{i,j} = -g_{j,i} \). Both orders preserve the time-reversal symmetry. Note that \( f_q(k) \) and \( g_q(k) \) in Eqs. (S4) and (S6) correspond to \( f_{i,j} \) and \( g_{i,j} \), respectively.

Finally, we discuss the effective interaction driven by the BO/sLC fluctuations. By solving the DW equation (S4), we obtain the full four-point vertex function \( \Gamma^s_q \) \( G(k,k) \) that is composed of \( I^s_q \) \( G(k,k) \) shown in Fig. S1 (e), which increases in proportion to \( (1-\eta_q)^{-1} \). Thus, we obtain the relation \( \Gamma^s_q \approx f_q(k)\{f_q(k')\}^* f_q(k) \approx (1-\eta_q)^{-1} \), which is well satisfied when \( \lambda_q \) is close to unity. Here, \( \tilde{f}^s_q = T^2 \sum_{k, k'} \{f_q(k)\}^* f_q(k')/T \sum_k |f_q(k)|^2 \).

The pairing interaction due to the sLC fluctuations is given by the full four-point vertex. It is approximately expressed as \( V^sQ_{s,k,-} = -T g_q(k,-k) \propto \left| g_q(k) \right|^2 \left| f_q(k) \right|^2 \approx (1-\eta_q)^{-1} \). Since \( g \) is odd-function, the sLC fluctuations cause attractive interaction: \( V^sQ_{s,k,-} \approx -\left| g_q(k) \right|^2 \left| f_q(k) \right|^2 \approx (1-\eta_q)^{-1} \).

**B: ADDITIONAL NUMERICAL RESULTS OF DW EQUATIONS**

In Fig. 4 (a) in the main text, the \( p \)-dependences of the sLC and BO eigenvalues are calculated based on the DW equations, using \( \alpha_S \) shown in Fig. 2 (b). In order to verify the robustness of obtained results, here we calculate the \( p \)-dependences of the eigenvalues in case of another \( p \)-dependence of Stoner factor; \( \alpha_S = 1.01-0.2p \). In this case, magnetic order appears for \( p \leq 0.05 \). The obtained results in Fig. S2 are very similar to Fig. 4 (a) in the main text.

![FIG. S2: Obtained eigenvalues of sLC and BO as function of hole-doping \( p = 1-n \), under the condition \( \alpha_S = 1.01-0.2p \) that is shown as broken line in Fig. 2 (d) in the main text.](image-url)
In Figs. 4 (a) and (b) in the main text, the sLC eigenvalue $\eta = q_0$ is comparable to the BO eigenvalue $\lambda_0 = 0$ for a wide doping range. This result means that the sLC order at $q_{sLC} = Q_d$ and the ferro-BO occur at almost the same temperature $\sim T^*$. Here, we discuss the possibility of coexistence of sLC order and ferro-BO.

Since the ferro-BO does not induce the band-folding and pseudogap, the sLC order will emerge even if the ferro-BO transiting temperature is higher. To verify this expectation, we calculated the triplet DW equation (S6) under the ferro-BO with $f_{q=0}^{\text{max}} = 0$, 0.01, 0.03. Thus, the ferro-BO does not prohibit the emergence of the sLC order. The eigenvalue $\eta_q$ slightly increases with $f_{q=0}^{\text{max}}$, since the spin Stoner factor $\alpha_S$ is enlarged by the ferro-BO [22, 23].

Since the ferro-BO form factor $f_{q=0}^{\text{max}} = 0$ is comparable to the BO eigenvalue $\lambda_0 = 0$), we calculated the triplet DW equation (S6) under the ferro-BO with $f_{q=0}^{\text{max}} = 0$, 0.01, 0.03. Figure S3 shows the eigenvalue of sLC as function of $q$ for $n = 0.85$ and $U = 3.27$ ($\alpha_S = 0.99$) under the ferro-BO form factor obtained by the spin-singlet DW equation (S4). It is verified that the ferro-BO does not prohibit the emergence of the sLC order. The eigenvalue $\eta_q$ slightly increases with $f_{q=0}^{\text{max}}$, since the spin Stoner factor $\alpha_S$ is enlarged by the ferro-BO [22, 23].

**FIG. S3:** Obtained $\eta_q$ for $n = 0.85$ ($\alpha_S = 0.99$) under the ferro-BO with $f_{q=0}^{\text{max}} = 0$, 0.01, 0.03. Thus, the ferro-BO does not prohibit the emergence of the sLC order.

**FIG. S4:** The unfolded band structure in the single-$Q_d$ sLC order corresponds to Fig. 5 (a) in the main text.

Figure S4 shows the “unfolded” band structure in the single-$Q_d$ sLC order at $g^{\text{max}} = 0.1$, which corresponds to Fig. 5 (a) in the main text. The pseudogap closes on the X-Y line owing to the odd-parity form factor. This Dirac point will be smeared out for $T \sim T^* \gg T_{CDW}$ because of very large inelastic scattering at the hot spot [5–7, 40]. In addition, the Dirac point should be masked by the $d$-wave BO below $T_{CDW}$.

**C: BO/sLC ORDER AS MAGNON-PAIR CONDENSATION**

We explain that the sLC order is exactly the same as the magnon-pair condensation. The following spin quadrupole order occurs owing to the magnon-pair condensation [46]:

$$K^{\alpha\beta}_{i,j} = \langle s_i^\alpha s_j^\beta \rangle - \langle s_i^\alpha \rangle \langle s_j^\beta \rangle,$$

where $\alpha, \beta = x, y, z$, and the relation $K^{\alpha\beta}_{i,j} = K^{\beta\alpha}_{j,i}$ holds. We will explain that the even-parity function $a_{i,j} \equiv K^{\alpha\alpha}_{i,j} / 3$ (with $a_{i,j} = a_{j,i}$) corresponds the BO state, and the odd-parity function $b_{i,j}^{\alpha\alpha} \equiv i\epsilon_{\alpha\beta\gamma} K^{\beta\gamma}_{i,j} / 2$ (with $b_{i,j}^{\alpha\alpha} = -b_{j,i}^{\alpha\alpha}$) corresponds the sLC order.

**FIG. S5:** Diagrammatic expression of $\Gamma^{(c)}_{q}(p,p')$, which represents the scattering process of the magnon pair through the interaction $J^{(c)}_{q}(p,p')$. Mathematically, $\Gamma^{(c)}_{q}(p,p')$ diverges when magnon pairs with momentum $q$ condense. Thus, sLC/BO is interpreted as the condensation of odd/even parity magnon pairs.

Here, we explain that $\Gamma^{(c)}_{q}(k,k')$ due to the AL processes represents the scattering between two-magnons. To simplify the discussion, we drop the MT term, and consider only AL terms with two $\chi$’s. Then, we define $\Gamma^{(c)}_{q}(p,p')$ by the following relation; $\Gamma^{(c)}_{q}(k,k') = T^2 \sum_{p,p'} [G(k-p)G(k'+p)] \Gamma^{(c)}_{q}(p,p')G(k'-p)$. Figure S5 shows the diagrammatic expression of $\Gamma^{(c)}_{q}(p,p')$, which represents the scattering process of magnon pair amplitude $b^2 (a)$ through the interaction $J^{(c)}_{q}(p,p')$, which is moderate function of $T$. With decreasing temperatures, $\Gamma^{(c)}_{q}(p,p')$ diverges when singlet (triplet) magnon pairs with momentum $q$ condensate, and the critical temperature corresponds to $\lambda_0 = 1 (\eta_q = 1)$.

Here, we introduce $f_{q}(k) \equiv T \sum_{p} H_{q}(k,p)f_{q}(p)$ and $g_{q}(k) \equiv T \sum_{p} H_{q}(k,p)g_{q}(p)$, where $H_{q}(k,p) =
\(G(p_+)G(p_-)G(p - k)\), and \(f_q(p)\) and \(g_q(p)\) are form factors of the DW equations. Then, the DW equations are rewritten as

\[
\lambda_q \bar{f}_q(k) = T \sum_p J_q^c(k, p) \chi^s(p_+) \chi^s(p_-) \bar{f}_q(p) \tag{S10}
\]

\[
\eta_q \bar{g}_q(k) = T \sum_p J_q^s(k, p) \chi^s(p_+) \chi^s(p_-) \bar{g}_q(k) \tag{S11}
\]

where the kernel function \(J_{q}^{c,s}(k, p)\) is given in Fig. S5.

These equations mean that \(\bar{f}_q(k)\) (\(\bar{g}_q(k)\)) corresponds to the singlet (triplet) magnon pair condensation. Therefore, their Fourier transformations correspond to \(a_{i,j}\) and \(b_{i,j}^z\), respectively.

To summarize, in the present double spin-flip mechanism, magnon-pair condensation \(a, b^z \neq 0\) occurs at \(T = T_{sLC}\). Therefore, the sLC/BO given by the present mechanism is exactly the same as “condensation of odd/even parity magnon pairs”.

**D: POSSIBLE SLC PATTERNS IN REAL SPACE**

\[\text{FIG. S6: Examples of the sLC pattern in real space for } q_{sLC} = (\pi/2, \pi/2), (\pi/2, -\pi/2). \text{ (a) Single-} q \text{ sLC pattern for } \psi = \pi/2. \text{ (b)-(e) Four examples of double-} q \text{ sLC patterns.} \]

Previously, we showed one example of spin current pattern at \(q_{sLC} = (\pi/2, \pi/2)\) in Fig. 1 (c). However, the spin current pattern derived from the form factor \(g_q(k)\) in Fig. 3 (b) is not uniquely determined. In fact, the form factor in real space is given as \(\text{Im}\{g_{i,j} e^{i\psi}\} \sim \text{Im}\{e^{iq \cdot (r_i + r_j)/2} e^{i\psi}\}\), where \(\psi\) is an arbitrary phase. Here, we discuss other possible spin current patterns by choosing \(\psi\).

First, we discuss the real space pattern for \(q_{sLC} = (\pi/2, \pi/2), (\pi/2, -\pi/2)\). We assume that Fig. 1 (c) corresponds to \(\psi = 0\). Then, the single-\(q\) spin current pattern for \(\psi = \pi/2\) is given in Fig. S6 (a). The double-\(q\) spin current order is given by the combination of the sLC order at \(q_{sLC} = (\pi/2, \pi/2)\) and that at \(q_{sLC} = (\pi/2, -\pi/2)\) with arbitrary phase factors. Figure S6 (b)-(c) are given by the combination of Fig. 1 (c) with its \(\pi/2\)-rotation, and Figs. S6 (d)-(e) are given by the combination of Figs. S6 (a) with its \(\pi/2\)-rotation. We stress that the magnitude of spin current \(|J_{i,j}^s|\) in Fig. S6 (b)-(c) has \(C_4\) symmetry, whereas that in Fig. S6 (d)-(e) breaks the \(C_4\) symmetry.
