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Quantum Hall effect anomaly and collective modes in the magnetic-field-induced spin-density-wave phases of quasi-one-dimensional conductors

N. DUPUIS(*) and V. M. YAKOVENKO

*Department of Physics and Center for Superconductivity Research
University of Maryland - College Park, MD 20742-4111, USA*

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PACS. 72.15Nj – Collective modes (e.g., in one-dimensional conductors).

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Abstract. – We study the collective modes in the magnetic-field-induced spin-density-wave (FISDW) phases experimentally observed in organic conductors of the Bechgaard salts family. In phases that exhibit a sign reversal of the quantum Hall effect (Ribault anomaly), the coexistence of two spin-density waves gives rise to additional long-wavelength collective modes besides the Goldstone modes due to spontaneous translation and rotation symmetry breaking. These modes strongly affect the charge and spin response functions. We discuss some experimental consequences for the Bechgaard salts.

Introduction. – The organic conductors of the Bechgaard salts family (TMTSF)₂X (where TMTSF stands for tetramethyltetraselenafulvalene) have remarkable properties in a magnetic field. In three members of this family (X = ClO₄, PF₆, ReO₄), a moderate magnetic field of a few tesla destroys the metallic phase and induces a series of SDW phases separated by first-order phase transitions [1, 2].

According to the so-called quantized nesting model (QNM) [2], the formation of the FISDWs results from the strong anisotropy of these organic materials, which can be viewed as weakly coupled chain systems (the typical ratio of the electron transfer integrals in the three crystal directions is $t_a : t_b : t_c = 3000 : 300 : 10$ K). The SDW opens a gap, but leaves closed pockets of electrons and/or holes in the vicinity of the Fermi surface. In the presence of a magnetic field H , these pockets are quantized into Landau levels (more precisely Landau subbands). In each FISDW phase, the SDW wave vector is quantized, $\mathbf{Q}_N = (2k_F + NG, Q_y)$ with N integer, so that an integer number of Landau subbands are filled. (Here k_F is the Fermi momentum along the chains, $-e$ the electron charge, b the interchain spacing, and $G = eHb/\hbar$.) As a result, the Fermi level lies in a gap between two Landau subbands, the SDW phase is stable, and the Hall conductivity is quantized: $\sigma_{xy} = -2Ne^2/h$ per one layer of the TMTSF molecules [3, 4].

(*) On leave from Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France.

As the magnetic field increases, the value of the integer N changes, which leads to a cascade of FISDW transitions.

A striking feature of the QHE in Bechgaard salts is the coexistence of both positive and negative Hall plateaus. While most plateaus are of the same sign, referred to as positive by convention, a negative Hall effect is also observed at certain pressures (the so-called Ribault anomaly) [5].

We have recently explained the Ribault anomaly within the framework of the QNM by taking umklapp processes into account [6]. Because of umklapp scattering, two linearly polarized SDWs, with wave vectors $\mathbf{Q}_N = (2k_F + NG, \pi/b)$ and $\mathbf{Q}_{-N} = (2k_F - NG, \pi/b)$, coexist in the Ribault phase [7]. The quantum Hall conductivity is quantized: $\sigma_{xy} = -2Ne^2/h$. The integer N is negative (hence a negative Hall plateau) and corresponds to the SDW with the largest amplitude. Experimentally, $N = -2$ and $N = -4$ are the most commonly observed “negative” phases. Our explanation of the Ribault anomaly differs from the one suggested by Zanchi and Montambaux [8] by invoking the pressure dependence of umklapp scattering rather than the electron band structure.

In this letter, we study the long-wavelength collective modes in the FISDW phases that exhibit the Ribault anomaly. The coexistence of two SDWs in these phases [7] gives rise to additional collective modes besides the Goldstone modes resulting from spontaneous rotation and translation symmetry breaking. We point out some analogies with phase modes in two-band or bilayer superconductors [9, 10] and plasmon modes in semiconductor double-well structure [11]. While these modes are generally difficult to observe, collective modes have strong experimental consequences in SDW systems [12]. We discuss how the charge and spin response functions are affected in the Ribault phase.

It should be pointed out that the long-wavelength modes are not the only modes of interest in the FISDW phases. There also exist magneto-rotons at finite wave vectors ($q_x = G, 2G, \dots$) [13]. Within the Zanchi-Montambaux scheme, Lederer has recently shown that these modes exhibit a different behavior in the Ribault phase [14]. The effect of umklapp scattering on the magneto-rotons is not considered in this letter.

Mean-field theory. – In the vicinity of the Fermi energy, the electron dispersion law in the Bechgaard salts is approximated as

$$E(k_x, k_y) = v_F(|k_x| - k_F) + t_\perp(k_y b), \quad (1)$$

where k_x and k_y are the electron momenta along and across the one-dimensional chains of TMTSF, and $\hbar = 1$. In eq. (1), the longitudinal electron dispersion is linearized in k_x in the vicinity of the two one-dimensional Fermi points $\pm k_F$, and $v_F = 2at_a \sin(k_F a)$ is the corresponding Fermi velocity (a is the lattice spacing along the chains). The periodic function $t_\perp(u) = t_\perp(u + 2\pi)$ describes the interchain hopping in a tight-binding approximation:

$$t_\perp(k_y b) = -2t_b \cos(k_y b) - 2t_{2b} \cos(2k_y b) - 2t_{3b} \cos(3k_y b) - 2t_{4b} \cos(4k_y b). \quad (2)$$

We neglect the electron dispersion in the third direction along the z -axis. t_{4b} plays a crucial role in the Ribault phase since (together with the umklapp scattering strength) it determines the ratio of the amplitudes of the two SDWs. For $t_{4b} = 0$, both SDWs have the same amplitude (independently of the value of t_{3b}). A finite t_{4b} lifts this degeneracy. This yields a negative QHE whenever $\text{sgn}(t_{4b}) = \text{sgn}(t_{2b})$ [6]. On the other hand, t_{3b} does not affect directly the Ribault phase [15]. Its only effect is to change the critical value of umklapp scattering above which the Ribault phase becomes stable. As shown by Zanchi and Montambaux [8], a sufficiently strong t_{3b} ($t_{3b} \gtrsim 0.2t_{2b}$) can stabilize a negative phase even in the absence of umklapp scattering. We cannot exclude that both umklapp scattering and dispersion law play a role in stabilizing the

Ribault phase at low pressure. However, whatever the mechanism at work, umklapp scattering (not considered in the Zanchi-Montambaux theory) will always lead to the coexistence of two SDWs in the Ribault phase.

The presence of a magnetic field along the z -direction is taken into account via the Peierls substitution $\mathbf{k} = (k_x, k_y) \rightarrow -i\nabla - e\mathbf{A}$, where \mathbf{A} is the vector potential. The magnetic field also introduces the Zeeman coupling $\sigma\mu_B H$ where $\sigma = +(-)$ for up (down) spins. (μ_B is the Bohr magneton. We take the electron gyromagnetic factor g equal to two.) We consider forward and umklapp scatterings between electrons, with amplitudes g_2 and g_3 , respectively. We do not consider backward scattering, since it does not play an important role in the QNM [6].

In the FISDW phase, the electron spin density has a nonzero expectation value:

$$\begin{aligned} \langle S_x(\mathbf{r}) \rangle &= \sum_{\beta=\pm} m_{\beta N} \cos(\phi_{\beta N}) \cos(\mathbf{r} \cdot \mathbf{Q}_{\beta N} + \theta_{\beta N}), \\ \langle S_y(\mathbf{r}) \rangle &= \sum_{\beta=\pm} m_{\beta N} \sin(\phi_{\beta N}) \cos(\mathbf{r} \cdot \mathbf{Q}_{\beta N} + \theta_{\beta N}), \end{aligned} \quad (3)$$

where $\mathbf{r} = (x, y)$ is the spatial coordinate. Because of the Zeeman coupling with the magnetic field, the SDWs are polarized in the (x, y) -plane. ϕ_N and ϕ_{-N} determine the direction of the spin magnetization, and θ_N and θ_{-N} the position of the SDWs with respect to the crystal lattice. The ratio $|\gamma| = m_{-N}/m_N$ of the amplitudes of the two SDWs in the Ribault phase is not precisely known. It depends on the detailed geometry of the Fermi surface (via t_{4b}) and increases with decreasing pressure. However, the stability of the Ribault phase against the formation of two helicoidal (*i.e.* circularly polarized) SDWs requires $|\gamma| \lesssim 0.5$ [6].

The FISDW phases are characterized by the complex order parameters $\Delta_{\beta N, \alpha}$ defined by

$$\Delta_{\alpha}(\mathbf{r}) = \langle \psi_{-\alpha, \downarrow}^{\dagger}(\mathbf{r}) \psi_{\alpha, \uparrow}(\mathbf{r}) \rangle = \sum_{\beta=\pm} \Delta_{\beta N, \alpha} e^{i\alpha \mathbf{r} \cdot \mathbf{Q}_{\beta N}}. \quad (4)$$

The operators $\psi_{\alpha, \sigma}^{(\dagger)}(\mathbf{r})$ annihilate (create) electrons with spin σ and momenta close to αk_F ($\alpha = \pm$). The order parameters are entirely determined by the mean value of the spin density (eq. (3)). In particular, $|\Delta_{\beta N, +}| = |\Delta_{\beta N, -}|$ ($\beta = \pm$) for sinusoidal (*i.e.* linearly polarized) SDWs [6].

First we consider the zero-temperature condensation energy ΔE at the mean-field level using the quantum limit approximation (QLA), also known as the single gap approximation. This approximation is valid when $v_F G \gg T$ and consists in retaining only the gaps at the Fermi level, neglecting those opening above and below the Fermi level [2, 6]. Introducing

$$\tilde{\Delta}_{\beta N, \alpha} = I_{\beta N} (g_2 \Delta_{\beta N, \alpha} + g_3 \Delta_{-\beta N, -\alpha}), \quad (5)$$

we write the condensation energy as

$$\Delta E = \sum_{\alpha} \left\{ \sum_{\beta} \frac{\Delta_{\beta N, \alpha}^* \tilde{\Delta}_{\beta N, \alpha}}{I_{\beta N}} + \frac{N(0)}{2} |\tilde{\Delta}_{-N, \alpha}|^2 - \frac{N(0)}{2} \sum_{\beta} |\tilde{\Delta}_{\beta N, \alpha}|^2 \left(\frac{1}{2} + \ln \left| \frac{2E_0}{\tilde{\Delta}_{N, \alpha}} \right| \right) \right\}. \quad (6)$$

Here E_0 is an ultraviolet cutoff of the order of t_a , and $N(0) = 1/\pi v_F b$ the density of states per spin. The coefficients $I_n \equiv I_n(q_y b = \pi)$ are well known in the QNM. They depend on the transverse dispersion law $t_{\perp}(k_y b)$ and measure the degree of perfect nesting of the Fermi surface [2, 6].

Minimizing ΔE with respect to the order parameters $\Delta_{\beta N, \alpha}$, we find that the mean-field ground state corresponds to $\theta_N = -\theta_{-N}$ and $\phi_N = \phi_{-N}$. The latter relation shows that both SDWs have the same polarization axis. The condition $\theta_N = -\theta_{-N}$ means that the two

SDWs can be displaced in opposite directions without changing the energy of the system. This property is related to the pinning that would occur for a commensurate SDW. Indeed, for a single SDW with wave vector $(2k_F, \pi/b)$, the condition $\theta_N + \theta_{-N} = 0$ becomes the usual pinning condition $\theta = 0$, where θ is the phase of the SDW [12]. The degeneracy of the ground state results from rotational invariance around the z -axis in spin space [16] and translational invariance in real space. The latter holds in the FISDW phases, since the SDWs are incommensurate with respect to the crystal lattice [17].

According to the mean-field analysis, and in agreement with general symmetry considerations, we therefore expect two (gapless) Goldstone modes: a spin-wave mode corresponding to a uniform rotation around the z -axis of the common polarization axis, and a sliding mode corresponding to a displacement of the two SDWs in opposite directions.

Long-wavelength collective modes. – Collective modes can be studied by expressing the partition function as a functional integral over bosonic fields describing spin fluctuations [18]. The standard mean-field theory is recovered from a saddle-point approximation. Collective modes are obtained studying small (Gaussian) fluctuations around the saddle point [19]. The static mean-field order parameters $\Delta_\alpha(\mathbf{r})$ (eq. (4)) then become space- and time-dependent fluctuating variables $\Delta_\alpha(\mathbf{r}, \tau) = \Delta_\alpha(\mathbf{r}) + \delta\Delta_\alpha(\mathbf{r}, \tau)$. Here τ is an imaginary time. We use the Matsubara formalism and obtain real-time quantities by standard analytic continuation. Taking advantage of the decoupling of phase and amplitude modes in the long-wavelength limit, we do not consider the latter. Order parameter fluctuations $\delta\Delta_\alpha(\mathbf{r}, \tau)$ then correspond to fluctuations of the phase variables $\theta_{\pm N}(\mathbf{r}, \tau)$ and $\phi_{\pm N}(\mathbf{r}, \tau)$. Skipping technical details [20], we only quote the final result for the collective modes, restricting ourselves to longitudinal (*i.e.* parallel to the chains) fluctuations.

We find two Goldstone modes with a linear dispersion law $\omega = v_F q_x$: a sliding mode and a spin-wave mode corresponding to

$$\theta_N(q_x, \omega) = -\theta_{-N}(q_x, \omega), \phi_N(q_x, \omega) = \phi_{-N}(q_x, \omega), \quad (7)$$

respectively. $\theta_{\pm N}(q_x, \omega)$ and $\phi_{\pm N}(q_x, \omega)$ are the Fourier transforms of the phase variables $\theta_{\pm N}(x, t)$ and $\phi_{\pm N}(x, t)$ (after analytic continuation to real time t). Equation (7) agrees with previous conclusions drawn from the mean-field analysis. Notice that the oscillations of the two SDWs are in-phase in the gapless spin-wave mode, and out-of-phase in the gapless sliding mode.

We also find a gapped sliding mode, $\omega^2 = v_F^2 q_x^2 + \omega_1^2$, and a gapped spin-wave mode, $\omega^2 = v_F^2 q_x^2 + \omega_2^2$, with

$$\omega_1^2 = \frac{12}{g_2 N(0)} \frac{r}{1-r^2} \frac{|\tilde{\Delta}_{N,+} \tilde{\Delta}_{-N,+}|}{|I_N I_{-N}|} \frac{3+5\tilde{\gamma}^2}{3\tilde{\gamma}^2}, \omega_2^2 = \frac{12}{g_2 N(0)} \frac{r}{1-r^2} \frac{|\tilde{\Delta}_{N,+} \tilde{\Delta}_{-N,+}|}{|I_N I_{-N}|} \frac{1-\tilde{\gamma}^2}{\tilde{\gamma}^2}, \quad (8)$$

where $r = g_3/g_2$ and $\tilde{\gamma} = \tilde{\Delta}_{-N,+}/\tilde{\Delta}_{N,+}$. $\tilde{\gamma}$ is related to $\gamma = \Delta_{-N,+}/\Delta_{N,+}$ ($|\gamma| = m_{-N}/m_N$) by $\gamma = (\tilde{\gamma} I_N - r I_{-N})/(I_{-N} - r \tilde{\gamma} I_N)$. In the Ribault phase, $|\tilde{\gamma}| \simeq |\gamma|$ [6]. With no loss of generality, we choose $\Delta_{\beta N, \alpha}$ and $\tilde{\Delta}_{\beta N, \alpha}$ to be real. The gapped sliding mode corresponds to a displacement of the two SDWs in the same direction (*i.e.* $\text{sgn}[\theta_N(x, t)] = \text{sgn}[\theta_{-N}(x, t)]$), while the gapped spin-wave mode corresponds to opposite rotations of the SDWs (*i.e.* $\text{sgn}[\phi_N(x, t)] = -\text{sgn}[\phi_{-N}(x, t)]$). In particular, when $|\tilde{\gamma}| = |\gamma| = 1$ (a situation reached when $|I_N| = |I_{-N}|$), gapped modes correspond to $\theta_N = \theta_{-N}$ and $\phi_N = -r\phi_{-N}$. Using the physical parameters of the Bechgaard salts, we find that ω_1 and ω_2 are larger than the mean-field order parameters $|\tilde{\Delta}_{\pm N, \pm}|$, so that the gapped modes appear above the quasi-particle excitation gap (generally within the first Landau subband above the Fermi level). Therefore, we expect these modes to be strongly damped due to the coupling with quasi-particle excitations.

There are some similarities between collective modes of the SDWs and phase modes occurring in two-band or bilayer superconductors [9, 10]. g_3/g_2 plays the same role as the ratio between the intraband (or intralayer) and interband (or interlayer) coupling constants. To some extent, there are also analogies with plasmon modes occurring in conducting bilayer systems [11]. While the corresponding phase modes in superconducting systems have not yet been observed, plasmon modes in semiconductor double-well structures have been observed recently via inelastic-light-scattering experiments [21].

Spectral functions. – Now we consider the spectral functions of the collective modes of the SDWs: the spin-spin correlation function $\chi_{yy}^{\text{ret}}(\mathbf{q}, \mathbf{q}', \omega)$ and the optical conductivity $\sigma(\mathbf{q}, \omega) = (i/\omega)\Pi^{\text{ret}}(\mathbf{q}, \omega)$, where Π^{ret} is the current-current correlation function. Π^{ret} and χ_{yy}^{ret} are the retarded parts of the imaginary time correlation functions

$$\Pi(\mathbf{r}, \tau; \mathbf{r}', \tau') = \langle j_{\text{DW}}(\mathbf{r}, \tau) j_{\text{DW}}(\mathbf{r}', \tau') \rangle, \quad \chi_{yy}(\mathbf{r}, \tau; \mathbf{r}', \tau') = \langle S_y(\mathbf{r}, \tau) S_y(\mathbf{r}', \tau') \rangle. \quad (9)$$

j_{DW} is the current along the chains carried by SDW fluctuations. For real $\Delta_{\beta N, \alpha}$, the mean-field magnetization (eq. (3)) is along the x -axis so that S_y corresponds to transverse spin fluctuations. To lowest order in phase fluctuations [20]

$$j_{\text{DW}}(\mathbf{r}, \tau) = -\frac{ie}{\pi b} \frac{\partial}{\partial \tau} \frac{\theta_N(\mathbf{r}, \tau) - r\gamma\theta_{-N}(\mathbf{r}, \tau)}{1 + r\gamma},$$

$$S_y(\mathbf{r}, \tau) = 2 \sum_{\beta} \Delta_{\beta N, +} \cos(\mathbf{r} \cdot \mathbf{Q}_{\beta N}) \phi_{\beta N}(\mathbf{r}, \tau). \quad (10)$$

j_{DW} is a function of $\theta_{\pm N}$, while S_y is a function of $\phi_{\pm N}$. Thus, Π is determined by the sliding modes, while χ_{yy} is determined by the spin-wave modes.

In the limit $\mathbf{q} = 0$, the dissipative part of the conductivity is given by [22]

$$\text{Re}[\sigma(\omega)] = \frac{\omega_p^2}{4} \left(\delta(\omega) \frac{3(1 - \tilde{\gamma}^2)}{3 + 5\tilde{\gamma}^2} + \delta(\omega \pm \omega_1) \frac{4\tilde{\gamma}^2}{3 + 5\tilde{\gamma}^2} \right). \quad (11)$$

We have introduced the plasma frequency $\omega_p = \sqrt{8e^2 v_F / b}$. Equation (11) satisfies the conductivity sum rule $\int_{-\infty}^{\infty} d\omega \text{Re}[\sigma(\omega)] = \omega_p^2 / 4$. Quasi-particle excitations above the mean-field gap do not contribute to the optical conductivity, a result well known in SDW systems [12]. Because both modes contribute to the conductivity, the low-energy (Goldstone) mode carries only a fraction of the total spectral weight. We obtain Dirac peaks at $\pm\omega_1$ because we have neglected the coupling of the gapped mode with quasi-particle excitations. Also, in a real system (with impurities), the Goldstone mode would broaden and appear at a finite frequency (below the quasi-particle excitation gap) due to pinning by impurities. In the clean limit, which is appropriate in (TMTSF)₂X salts, the presence of impurities does not restore any significant spectral weight to quasi-particle excitations above the mean-field gap [12]. Therefore, the fraction of spectral weight carried by the two modes is correctly given by eq. (11). By measuring the optical conductivity $\sigma(\omega)$, we can therefore obtain the ratio $|\tilde{\gamma}| \simeq |\gamma|$ of the amplitudes of the two SDWs. We have shown in ref. [6] that $|\tilde{\gamma}|$ can vary between ~ 0 and ~ 0.5 in the Ribault phase. Thus, the Goldstone mode can carry between $\sim 100\%$ and $\sim 50\%$ of the total spectral weight. When this fraction is reduced below $\sim 50\%$ (*i.e.* when $|\tilde{\gamma}| \gtrsim 0.5$), the Ribault phase becomes unstable against the formation of a helicoidal phase [6]. The low-energy spectral weight is, therefore, also a measure of the stability of the sinusoidal Ribault phase against the formation of a helicoidal phase. For a helicoidal structure, we cannot distinguish between a uniform spin rotation and a global translation, so that there is only one type of modes. Thus, in the helicoidal phase, we find a gapless Goldstone mode and a gapped mode.

The Goldstone mode carries no current, and all the spectral weight is pushed up above the quasi-particle excitation gap [20].

Due to the presence of SDWs, the spin-spin correlation function χ_{yy}^{ret} is not diagonal in momentum space, but has components $\chi_{yy}^{\text{ret}}(\mathbf{q} + \alpha \mathbf{Q}_{\beta N}, \mathbf{q} + \alpha' \mathbf{Q}_{\beta' N}, \omega)$, where $\alpha, \alpha', \beta, \beta' = \pm$ (see eqs. (9)-(10)). \mathbf{q} corresponds to the momentum of the spin-wave mode and tends to zero for long-wavelength fluctuations. We therefore consider the spectral function $\text{Im Tr}_{\mathbf{q}} \chi_{yy}^{\text{ret}}$, where $\text{Tr}_{\mathbf{q}}$ is a partial trace corresponding to a given spin-wave mode momentum \mathbf{q} . In the limit $q_y = 0, q_x \rightarrow 0$, we obtain [22]

$$\text{Im Tr}_{\mathbf{q}} \chi_{yy}^{\text{ret}} = \frac{2\pi}{g_2^2 N(0)} \frac{|\tilde{\Delta}_{N,+} \tilde{\Delta}_{-N,+}|}{|I_N I_{-N}|} \left\{ \frac{\delta(\omega - v_F q_x)}{v_F q_x} \left[\frac{-4r}{(1-r^2)^2} + \frac{(1+r^2)}{(1-r^2)^2} \frac{\tilde{\gamma}^2 + \zeta^2}{|\tilde{\gamma}\zeta|} \right] + \frac{\delta(\omega - \omega_2)}{\omega_2} \frac{3(1+r^2)}{2(1-r^2)^2} \frac{1 - \tilde{\gamma}^2}{|\tilde{\gamma}\zeta|} \right\}, \quad (12)$$

for $\omega, q_x > 0$. $\zeta = I_{-N}/I_N$. Both spin-wave modes contribute to the spectral function. The spectral weight carried by the Goldstone mode diverges as $1/q_x$ as expected for a quantum antiferromagnet [19]. Equations (11) and (12) predict that all the spectral weight is carried by the in-phase modes, *i.e.* the gapped sliding mode and the gapless spin-wave mode, whenever both SDWs have the same amplitude ($|\tilde{\gamma}| = |\gamma| = |\zeta| = 1$).

Conclusion. – We have shown that the Ribault anomaly in the FISDW phases of the Bechgaard salts [5] is characterized not only by a sign reversal of the QHE, but also by a rich structure of collective sliding and spin-wave modes. The presence of two SDWs in this phase gives rise to in-phase and out-of-phase collective oscillations in the long-wavelength limit. The out-of-phase sliding and in-phase spin-wave modes are gapless (Goldstone modes). The in-phase sliding and out-of-phase spin-wave modes are gapped and are expected to appear above the quasi-particle excitation gap in the Ribault phase. Charge and spin response functions are strongly affected by the presence of these modes. In particular, we have shown that the low-energy (Goldstone) sliding mode carries only a fraction of the total spectral weight in the optical conductivity. By measuring the latter, we can obtain the ratio of the amplitudes of the two SDWs that coexist in the Ribault phase. The low-energy spectral weight is also a measure of the stability of the Ribault phase against the formation of helicoidal SDWs. Helicoidal SDWs have been predicted in ref. [6] and are expected to appear at low pressure (since $|\tilde{\gamma}|$ increases with decreasing pressure). We have pointed out some analogies with collective modes in two-band superconductors or bilayer systems [9-11]. The common feature to all these collective modes is that they appear in a two-component system. To our knowledge, it is the first time that such modes are predicted in SDW systems.

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