Stabilization of vortex-liquid state by strong pairing interaction

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We theoretically investigate qualitative features of the field-temperature ($H$-$T$) phase diagram of superconductors with strong attractive interaction lying in the BCS-BEC crossover regime. Starting with a simple attractive Hubbard model, we estimate three kinds of characteristic fields, i.e., the pair-formation field $H^*$, the vortex-liquid-formation field $H_{c2}$, and the vortex-lattice-formation field $H_{melt}$. The region between $H_{c2}$ and $H_{melt}$, as well as that between $H_{c2}$ and $H^*$, is found to be enlarged as the interaction is stronger. That is, a strong attractive interaction can stabilize both the vortex-liquid and preformed-pair regions. We also point out the expected particle-density dependence of the $H$-$T$ phase diagram.

I. INTRODUCTION

So far, the superfluid transition with variable attractive interaction between Fermions has been studied primarily in the field of the ultracold atomic physics. Physical properties have been investigated especially in the BCS-BEC crossover regime, where the interaction between particles is strong enough to create non-condensed preformed pairs [1].

Intriguingly, recent experiments have suggested that a strong attractive interaction can exist in FeSe and related superconductors [2][3], which can pave the way for material realization of the BCS-BEC crossover. In contrast to the electrically neutral ultracold atoms, electrons in a superconductor are charged and thus naturally coupled with the gauge field of an external magnetic field. Therefore, FeSe and related materials can provide an opportunity to experimentally scrutinize unexplored effects of the magnetic gauge coupling on superconductors with strong attractive interaction. In fact, superconducting-fluctuation effects on diamagnetic response observed in FeSe are unusually enhanced compared with those in conventional superconductors [4], which may be understood as caused by the strong attractive interaction [5]. In addition, recent NMR measurements have proposed that a pseudogap caused by the preformed-pair formation can exist, and that the onset temperature of the pseudogap depends on the magnetic-field strength [6]. However, a theoretical understanding of magnetic-field effects on superconductors with strong attractive interaction is still incomplete.

The field v.s. temperature ($H$-$T$) phase diagram of a superconductor with strong fluctuation has been thoroughly investigated in relation to high $T_c$ cuprates [7][8][9] which are believed to belong to superconductors with high particle density. There, it has been clarified by developing the superconducting fluctuation theory [7][10] that the so-called upper critical field $H_{c2}(T)$ in the three-dimensional (3D) type-II superconductor is not a phase transition line but a crossover one separating the vortex-liquid region from the normal phase affected by a weak fluctuation effect, and that, in clean 3D materials, the genuine superconducting ordering occurs as a weak first order transition corresponding to the vortex-lattice melting [11]. The vortex-lattice melting curve $H_{melt}(T)$ can alternatively be determined by examining the elastic energy of the mean-field vortex-lattice state and invoking the Lindemann criterion [9]. In the so-called lowest-Landau-level (LLL) approach to the GL theory, it is believed that $H_{melt}(T)$ should be found as a consequence of the superconducting fluctuation. In fact, the fluctuation effect shows the scaling behavior of the form $T - T_c(H) \sim (TH)^{2/3}$ [10], while the field dependence of the melting temperature also obeys this scaling behavior [12].

In this study, we theoretically investigate qualitative features of the $H$-$T$ phase diagram of superconductors with strong attractive interaction. To obtain a qualitative picture, we start with a simple attractive Hubbard model. Using the T-matrix approximation combined with analysis of the Ginzburg-Landau action, we estimate three types of characteristic magnetic fields: the pair-formation field $H^*$, the vortex-liquid-formation field $H_{c2}$, and the vortex-lattice-formation field $H_{melt}$. The region between $H_{c2}$ and $H_{melt}$, as well as that between $H^*$ and $H_{melt}$, is found to become broader as the attractive interaction gets stronger. Based on this result, we conclude that a strong attractive interaction can stabilize both the vortex-liquid and the preformed-pair regions.

II. MODEL

To consider qualitative magnetic-field effects on electron systems with strong attractive interaction, we begin with a simple attractive Hubbard model on a simple cubic lattice:

$$H = -t \sum_{\langle i,j \rangle, \sigma} \left( c_{i \sigma}^\dagger c_{j \sigma} + c_{j \sigma}^\dagger c_{i \sigma} \right) - U \sum_i c_{i \uparrow}^\dagger c_{i \uparrow} c_{i \downarrow} c_{i \downarrow}. \quad (1)$$

Here, $c_{i \sigma}^\dagger$ is the annihilation (creation) operator of an electron with spin $\sigma$ at site $i$, and $\langle i, j \rangle$ means a nearest-neighbor pair of sites. There are two parameters in our model: the nearest-neighbor hopping amplitude $t(>0)$ and the onsite attractive interaction $U(>0)$. For simplicity, the magnetic-field term is introduced at the stage
of analyzing our Ginzburg-Landau functional (see Sec. IV and Appendix B). Basically, this simplification, equivalent to the electronic semi-classical approximation, corresponds to neglecting the Landau quantization of electron kinetic energy. In the following, the lattice constant is set to unity.

III. ZERO-FIELD PAIR-FORMATION AND PAIR-CONDENSATION TEMPERATURES

As a preliminary step to explore magnetic-field effects, we estimate the zero-field pair-formation and pair-condensation temperatures. Though the results presented in this section is well-known [13], we show them for completeness. As shown in the following, the pair-formation temperature $T^*$ is calculated within the mean-field approximation [14, 15], and the pair-condensation temperature $T_c$ is calculated within the T-matrix approximation [16–19]. The T-matrix approximation can take into account the shift of chemical potential due to superconducting fluctuation, which is important when the attractive interaction is strong, and in addition the particle density is not so high [20].

To calculate $T^*$, we apply to Eq. (1) the mean-field approximation, or equivalently, combine the following two equations with each other: the condition for divergence of the uniform superconducting susceptibility [see Eq. (1) for its definition]

$$\chi^{(SC)}_0 = \infty,$$

and the particle-number conservation for non-interacting particles

$$n = \frac{2}{M} \sum_k \frac{1}{\exp[(\epsilon_k - \mu)/T] + 1}. \quad (3)$$

Here, we define several symbols: particle density (per site) $n$, chemical potential $\mu$, temperature $T$, the total number of lattice sites $M = M_x M_y M_z$, the lattice momentum with the periodic boundary condition $k_\alpha = 2\pi n_\alpha / M_\alpha$ ($-M_\alpha / 2 \leq n_\alpha < M_\alpha / 2$ with $n_\alpha \in \mathbb{Z}$), and the energy dispersion of non-interacting particles $\epsilon_k = -2t(\cos k_x + \cos k_y + \cos k_z)$. The superconducting susceptibility with pair (or center-of-mass) momentum $q$ is defined as

$$\chi^{(SC)}_q = \frac{\chi^{(0)}_q (0)}{1 - U \chi^{(0)}_q (0)}, \quad (4)$$

where

$$\chi^{(0)}_q (i\omega_m) = \frac{T}{M} \sum_{k,n} \epsilon^{(0)}_k (i\epsilon_n + i\omega_m) G^{(0)}_{q-k} (-i\epsilon_n). \quad (5)$$

Here, $\epsilon_n = 2\pi(n + 1/2)T$ ($\omega_m = 2\pi m T$) is the Fermion (Boson) Matsubara frequency, and $G^{(0)}_k (i\epsilon_n) = (i\epsilon_n - \epsilon_k + \mu)^{-1}$ is the non-interacting Green’s function.

As for $T_c$, we apply the T-matrix approximation. This approximation combines the divergence of the susceptibility $\chi^{(SC)}_0 = \infty$, which is the same condition as defining $T^*$, with the particle-number conservation

$$n = \frac{2T}{M} \sum_{k,n} G_k (\pm \epsilon_n) e^{i\pm \epsilon_n 0}, \quad (6)$$

in which superconducting-fluctuation effects are taken into account. Here, $G_k (\pm \epsilon_n)$ is the interacting Green’s function, which is defined as

$$G_k (\pm \epsilon_n)^{-1} = G^{(0)}_k (\pm \epsilon_n)^{-1} - \Sigma_k (\pm \epsilon_n), \quad (7)$$

and $\Sigma_k (\pm \epsilon_n)$ is the self energy defined within the T-matrix approximation as

$$\Sigma_k (\pm \epsilon_n) = -\frac{T}{M} \sum_{q,m} G^{(0)}_{q-k} (\omega_m - \epsilon_n) \times \frac{U^2 \chi^{(0)}_q (i\omega_m)}{1 - U \chi^{(0)}_q (i\omega_m)} e^{i(\omega_m - \epsilon_n)0}. \quad (8)$$

Here, the temperature-independent Hartree shift

$$\Sigma^{(H)} = -\frac{U}{M} \sum_{k,n} G_k (\pm \epsilon_n) e^{i\pm \epsilon_n 0} = -\frac{U n}{2}, \quad (9)$$

is already taken into account by properly choosing the origin of energy; therefore we do not explicitly consider $\Sigma^{(H)}$ [17, 21].

To explain physical meanings of the definitions of $T^*$ and $T_c$, it is convenient to consider the opposite limit to the weak-coupling BCS one in which $T^*$ and $T_c$ take almost the same value. In this strong-coupling limit ($U/t \to \infty$), we can show that $T^* \propto |\mu| \propto U \propto E_b$, where $E_b$ is the two-particle binding energy [21], therefore, $T^*$ can be interpreted as the pair-formation (or pair-breaking) temperature. As for $T_c$, in the same limit, we obtain an asymptotic formula $T_c \propto t^2 / U$, which represents the BEC transition temperature of non-interacting Bosons (or preformed-pairs) with a nearest-neighbor hopping amplitude $t_B \propto t^2 / U$ [13]; accordingly, $T^*$ can be understood as the pair-condensation temperature.

Figure 1 shows an interaction strength v.s. temperature phase diagram obtained from the equations listed above with the particle density fixed to $n = 0.2$. As seen from Fig. 1, the preformed-pair region becomes broader as the interaction gets stronger. In Fig. 1 we also show with a gray dotted line the threshold value $U = U_0 \approx 8.14t$ for the formation of a two-particle bound state [18, 21, 22]. Note that the BCS-BEC crossover occurs close to $U_0$.

As shown in Fig. 2 the chemical potential $\mu$ is remarkably reduced when the attractive interaction $U$ approaches $U_0$. When $U$ is larger than $U_0$, $\mu$ tends to become lower than the band bottom.

In the following, we focus on systems where $U < U_0$ is satisfied so that the decrease in $\mu$ is not so large. More
specifically, we consider two systems with different values of $U$: $U/t = 2.57$ and $U/t = 5.14$ (see the green and yellow dotted lines in Figs. 1 and 2).

**IV. PAIR-FORMATION, VORTEX-LIQUID-FORMATION, AND VORTEX-LATTICE-FORMATION FIELD**

To understand qualitative features of the $H$-$T$ phase diagram, we estimate three kinds of magnetic field values: the pair-formation field $H^*$, the vortex-liquid-formation field $H_{c2}$, and the vortex-lattice-formation field $H_{melt}$. In the following, the direction of magnetic field is fixed in parallel to the $z$ axis, and we assume strongly type-II systems and neglect the difference between the applied magnetic field and the magnetic field in the system ($B = \mu_0 H$). As mentioned in Sec. II, we neglect the Landau quantization of the electron kinetic energy.

The pair-formation field $H^*$ is calculated in a similar way to the calculation of $T^*$. To introduce the effect of magnetic field $H$, we only have to replace the condition for divergence of the uniform superconducting susceptibility [Eq. (2)] with that for divergence of a finite-momentum superconducting susceptibility [23]

\[
\chi_{qH}^{(SC)} = \infty,
\]

where $q_H^2 = \sqrt{2\pi \mu_0 H/\phi_0}$ and $\chi_{qH}^{(SC)}$ is given in Eq. (4). Here $\mu_0$ is the vacuum permeability, and $\phi_0 = \pi h/e$ is the flux quantum. $\chi_{qH}^{(SC)}$ approximately describes the susceptibility for states with the lowest-Landau-level index and uniform in the $z$ direction. As for a free-particle number equation to determine the chemical potential, we adopt Eq. (3) since we neglect the Landau quantization of the electron kinetic energy. Therefore, we combine Eq. (10) with Eq. (3) to estimate $H^*$. The curve $(T, H^*(T))$ merges into $(T^*, 0)$ in the low-field limit; thus $H^*$ can be regarded as a natural extension of $T^*$ to the finite-field region.

The vortex-liquid formation field $H_{c2}$ is estimated in a similar way to the calculation of $T_c$. Since we focus on systems with $U < U_0$ (see the green and yellow dotted lines in Fig. 2), where the decrease in $\mu$ is not so large and the $T$ dependence of $\mu$ is not so important, we simply approximate

\[
\mu(T, H) \sim \mu(T_c, 0),
\]

where $\mu(T_c, 0)$ is obtained within the T-matrix approximation (see Sec. III and Fig. 2). This approximation is correct at least in the weak-coupling limit, and we believe that this approximation is a first step to consider magnetic-field effects in the case with strong attractive interaction. After we replace $\mu(T, H)$ with $\mu(T_c, 0)$, we solve Eq. (10) to estimate $H_{c2}$. Similar to the case of $H^*$, the curve $(T, H_{c2}(T))$ merges into $(T_c, 0)$ in the low-field limit; thus $H_{c2}$ can be understood as an extension of $T_c$ to the finite-field region.
Regarding the vortex-lattice-formation field \( H_{\text{melt}} \), we apply an analysis based on the Ginzburg-Landau analysis \[23\] in the lowest-Landau-level approximation \[12,25\], which is valid closer to the \( H_{c2} \) line \[12,26\]. First, as explained in Appendix A, we derive the zero-field Ginzburg-Landau functional \( F_{\text{GL}} \):

\[
F_{\text{GL}} = \sum_q T \left( 1 - U \chi_q^{(0)}(0) \right) |a_q|^2 + \frac{\beta}{2} \sum_i |a_i|^4. \tag{12}
\]

Here, \( a_i = M^{-1/2} \sum_q \exp(iq \cdot r_i) a_q \), and the coefficient \( \beta \) is given as

\[
\beta = \frac{T^3 U^2}{M} \sum_{k,n} \left| G_k^{(0)}(i \varepsilon_n) \right|^4. \tag{13}
\]

As shown in Appendix B, by applying the lowest-Landau-level approximation to Eq. \((12)\) with replacement of the momentum in the \( x-y \) plane by \( q_H \) consistently with Eq. \((10)\) and using the gradient expansion in the \( z \) di-
and liquid region between $H_c$ and $H_{melt}$ are given as follows:

$$F_{GL} \sim \int d^3r \left[ (\alpha_{qH} |\psi(r)|^2 + \gamma |\partial_z \psi(r)|^2) + \frac{\beta}{2} |\psi(r)|^4 \right],$$

(14)

where the order-parameter field $\psi(r)$ involves only the lowest-Landau-level modes in the $x$-$y$ plane. The coefficients are given as follows:

$$\alpha_{qH} = T \left( 1 - U \chi_{qH}^{(0)}(0) \right),$$

(15)

and

$$\gamma = - \frac{T^2 U t}{M} \sum_{k,n} \left[ G_k^{(0)}(i\xi_n) \right]^2 G_k^{(0)}(-i\xi_n)$$

$$\times \left[ \cos k_x + 4t G_k^{(0)}(i\xi_n) \sin^2 k_y \right]^2.$$  

(16)

As shown in Appendix C, based on Eq. (14), the vortex-lattice-formation field $H_{melt}$ is approximately calculated by solving the following equation:

$$\frac{T}{4\pi \sqrt{\beta_s c_{66}}} = \frac{c_{2m}}{\hbar}.$$  

(17)

Here, $h = 2\pi \mu_0 H/\phi_0$ is a dimensionless magnetic field (note that the lattice constant is set to unity), and $c = O(10^{-1})$ is a phenomenological parameter [12]. Also, $c_{66}$ and $\rho_s$ represent the shear modulus of the vortex lattice and the superfluid density defined along the magnetic field, respectively (see Appendix C):

$$c_{66} = \frac{2\gamma \lambda |\alpha_{qH}|^2}{\beta^2},$$

(18)

and

$$\rho_s = \frac{2 |\alpha_{qH}| \gamma}{\beta \lambda \beta_s}$$

(19)

with numerical factors related to the triangular vortex-lattice structure: $\beta \lambda \sim 1.16$ and $\gamma \lambda \sim 0.119$. To obtain $H_{melt}$, we solve Eq. (17) in combination with the approximated chemical potential [Eq. (11)].

V. FIELD–TEMPERATURE PHASE DIAGRAM

Based on numerically calculated $H^*$, $H_{c2}$, and $H_{melt}$, we obtain typical $H$-$T$ phase diagrams (Fig. 3). Since our purpose is to investigate qualitative features of the $H$-$T$ phase diagram, we fix the phenomenological parameter to estimate $H_{melt}$ as $c = 0.5$ throughout our calculation. A slight change in $c$ does not affect the qualitative features. Figures 3(a) and (b) respectively show the weak-interaction ($U/t = 2.57$) and strong-interaction ($U/t = 5.14$) cases with lower density ($n = 0.2$). Comparing Figs. 3(a) and (b), we can see that the vortex-liquid region between $H_{c2}$ and $H_{melt}$, as well as the preformed-pair region between $H_{c2}$ and $H^*$, becomes broader as the interaction becomes stronger; therefore, a strong attractive interaction stabilizes both the vortex-liquid and the preformed-pair regions.

Let us consider physical reasons why both the vortex-liquid and preformed-pair states are stabilized by a strong attractive interaction. First, the stabilization of the preformed-pair state can be understood in the same way as the zero-field case: a strong attractive interaction makes it easy to create non-condensed pairs, or preformed pairs [1]. Second, the stabilization of the vortex-liquid region can be understood based on the superconducting-fluctuation-strength: as the attractive interaction gets stronger toward the BCS-BEC crossover regime, the fluctuation becomes more significant [8, 27], and thus the vortex-liquid region becomes wider.

Figures 3(c) and (d) show the obtained phase diagrams with higher density ($n = 0.5$). Similar to the case with lower density ($n = 0.2$), we can see that both the vortex-liquid and the preformed-pair regions are stabilized when the interaction is strong. Moreover, comparing the higher density case [Figs. 3(a) and (b)] with the lower density case [Figs. 3(c) and (d)], we can see that the vortex-liquid region is broader while the preformed-pair region is narrower when the density is higher. From this result, we conclude that the particle density, in addition to interaction strength, is an important factor in determining the resultant $H$-$T$ phase diagram.

Here, we point out that keeping only the LLL modes among various order parameter’s spatial variations is an approach from the weak fluctuation in the following sense: it is clear that, in the weak-field limit, the LLL mode vanishes so that the fluctuation-induced downward shift of $T_c$ in zero field, $\Delta T_c(0)$, cannot be described within the present approach. To describe $\Delta T_c(0)$, it is necessary to incorporate the higher-Landau-level (HLL) modes in our calculation. In fact, the HLL modes incorporating the vortex-loop fluctuations [8, 28] should lead to not only $\Delta T_c(0)$ and a shift of the $H_{c2}(T)$ line in low fields accompanying it but also a downward shift of $H_{melt}(T)$ and a change of its temperature dependence in low enough fields. Although such effects have been omitted in the present LLL approach, this simplification is not essential to our purpose here of understanding a qualitative picture of the $H$-$T$ phase diagram in superconductors with a strong pairing interaction.

VI. CONCLUSION

To obtain typical $H$-$T$ phase diagrams in electron systems with strong attractive interaction, we estimate the pair-formation field $H^*$, the vortex-liquid-formation field $H_{c2}$, and the vortex-lattice-formation field $H_{melt}$. Based on numerical calculations, we find that a strong attractive interaction can stabilize both the vortex-liquid and the preformed-pair regions. In addition, we point out that the particle density also influences the resultant phase
diagram.

In the preformed-pair and vortex-liquid regions stabilized by strong attractive interaction, thermodynamic and transport properties are expected to be characteristic. In particular, the Hall conductivity in the vortex-liquid region can be enhanced by superconducting-fluctuation effects since the dynamics of the superconducting order parameter can involve a larger propagating part when the interaction is stronger.

In the end of this paper, we discuss the $H$-$T$ phase diagram in FeSe suggested by several experiments. We do not comment on the high-field low-temperature phase ("B-phase") proposed in Ref. since in our calculation we do not take into account the Zeeman coupling of magnetic field, which may be important in the high-field low-temperature region. Let us consider other aspects of FeSe. First, a large pseudogap region above $H_{c2}$ in the $H$-$T$ plane is suggested in Ref. [6]. If we assume that the pseudogap is caused by the preformed pair [16, 19], we can interpret the observed pseudogap region as the preformed-pair region stabilized by a strong attractive interaction as in Fig. 3(b). Second, a crossing of magnetization curves [31] is observed in Ref. [3]. This crossing can be understood as caused by a strong attractive interaction [3] in the vortex-liquid region stabilized also by the strong attractive interaction. Third, the Hall, Seebeck, and Nernst coefficients have shown their maximum or minimum near a temperature $T_c$ defined in the clean limit. This crossing can be understood as caused by a strong attractive interaction as in Fig. 3(b). Though a strong attractive interaction may be related to this behavior, the detailed interaction neglected in the present study is not negligible [36].

As another possible scenario to explain why the vortex-liquid region has estimated much below the actual position of the actual $H_{c2}$ and, upon cooling, begins to vanish close to a vortex-glass transition, which lies near $H_{melt}$ and much below the actual $H_{c2}$.

As another possible scenario to explain why the vortex-liquid region is estimated to be relatively narrow in FeSe, let us consider the two-band structure characteristic of FeSe [32, 33]. If a strong attractive interaction is present in one of these bands while a weak attractive interaction exists in another band, the vortices due to the former band can be pinned by the vortex lattice generated by the latter band. If this is true, the vortex-liquid region can become relatively narrow compared to the case considered in the present work where only a single band with strong attractive interaction exists. This possibility will be examined in details elsewhere.

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Appendix A: Derivation of Ginzburg-Landau functional

Here we derive the zero-field Ginzburg-Landau functional given by Eq. (12). By using the functional integral representation [14, 27, 37], we can formally rewrite the grand-canonical partition function $Z$ as

$$Z = \int \left[ \prod_{k,\sigma,n} dc_k^\sigma(\varepsilon_n) dc_{k\sigma}(\varepsilon_n) \right] e^{-(S_0 + S_{int})},$$

where

$$S_0 = \frac{1}{T} \sum_{k,\sigma,n} (-G_k^{(0)}(\varepsilon_n))^{-1} c_k^\sigma(\varepsilon_n) c_{k\sigma}(\varepsilon_n),$$

$$S_{int} = -\frac{U}{TM} \sum_{q,m} \phi_q^*(\omega_m) \phi_q(\omega_m),$$

and

$$\phi_q(\omega_m) = \sum_{k,n} c_{-k}^\dagger(\varepsilon_n-\omega_m) c_{k+q} \phi_q(\varepsilon_n + \omega_m).$$

Here, $c_k^\sigma(\varepsilon_n)$ and $c_{k\sigma}^\dagger(\varepsilon_n)$ are the Grassmann numbers, and $G_k^{(0)}(\varepsilon_n) = (i\varepsilon_n - \varepsilon_k + \mu)^{-1}$ is the non-interacting Green's function.

Introducing the order-parameter field $a_q(\omega_m)$ and $a_q^*(\omega_m)$ with the Hubbard-Stratonovich transformation, we can obtain the following expression:

$$e^{-S_{int}} = \int \left[ \prod_{q,m} \frac{da_q(\omega_m) da_q^*(\omega_m)}{\pi} \right] e^{-\sum_{q,m} |a_q(\omega_m)|^2} \times e^{U \sum_{q,m} [a_q(\omega_m) \phi_q(\omega_m) + c.c.]}.$$

Using this expression, we can transform the partition function as

$$\frac{Z}{Z_0} = \langle e^{-S_{int}} \rangle_0 = \int \left[ \prod_{q,m} \frac{da_q(\omega_m) da_q^*(\omega_m)}{\pi} \right] e^{-\sum_{q,m} |a_q(\omega_m)|^2} \times \left( e^{U \sum_{q,m} [a_q(\omega_m) \phi_q(\omega_m) + c.c.]} \right)_0. \quad (A6)$$
Here, $Z_0 = \int \prod_k \langle \varphi_k \rangle e^{-\beta H_0} \langle \varphi_k \rangle e^{-\beta H_0}$ is the non-interacting partition function, and $\langle \cdots \rangle_0$ represents the grand-canonical ensemble average with respect to the non-interacting part $H_0$. Expanding the last term in Eq. (A6) with respect to the order-parameter field $a_q(\omega_m)$ and $a_q^*(\omega_m)$ up to the fourth order and neglecting Bosonic quantum fluctuation, we can finally obtain the following form:

$$Z_0 \sim \int \left[ \prod_q \frac{d\varphi_q d\varphi_q^*}{\pi} \right] e^{-\mathcal{F}_{GL}/T}, \quad \text{(A7)}$$

where we write $a_q = a_q(0)$ for simplicity. Here, $\mathcal{F}_{GL}$ is the Ginzburg-Landau functional, the explicit form of which is given as

$$\mathcal{F}_{GL} = \sum_q T \left[ 1 - U \chi_q^{(0)}(0) \right] |a_q|^2 + \frac{\beta}{2} \sum_i |a_i|^4, \quad \text{(A8)}$$

where $a_i = M^{-1/2} \sum_q \exp(iq \cdot r_i)a_q$ is the real-space order-parameter field,

$$\chi_q^{(0)}(i\omega_m) = \frac{T}{M} \sum_{k,n} G_{k+q,n}(i\varepsilon_n + i\omega_m)G_{-k,n}(-i\varepsilon_n), \quad \text{(A9)}$$

and

$$\beta = \frac{T^3 U^2}{M} \sum_{k,n} |G_{k,n}(i\varepsilon_n)|^4. \quad \text{(A10)}$$

### Appendix B: Lowest-Landau-level approximation of Ginzburg-Landau action

In the following, we explain how we obtain the approximated expression of the Ginzburg-Landau functional [Eq. (14)]. Neglecting the Landau quantization of electrons, the external magnetic field affects the energy eigenstate of the order-parameter field $a_i$. At large length scales, the lattice structure is not important so that we can focus on the long-wavelength parts of $a_i$ and can replace $a_i$ defined on lattice with $\psi(r)$ defined in continuum space (note that the lattice constant is set to unity). Then, to perform our calculation in a finite magnetic field parallel to the $z$ axis, we can rewrite Eq. (12) as

$$\mathcal{F}_{GL} \simeq \int d^3r \left( \psi^* \alpha_Q \psi + \gamma |\partial_z \psi|^2 + \frac{\beta}{2} |\psi|^4 \right), \quad \text{(B1)}$$

where

$$\alpha_Q = T \left[ 1 - U \chi_Q^{(0)}(0) \right] \quad \text{(B2)}$$

with $Q = -i\nabla - 2\pi A/\phi_0$ is the gauge-invariant gradient in the directions perpendicular to the field, and

$$\gamma = -\frac{T^2 U}{M} \sum_{k,n} \left[ G_{k,n}^{(0)}(i\varepsilon_n) \right]^2 G_{-k,n}^{(0)}(-i\varepsilon_n) \times \left[ \cos k_z + 4tG_{k,n}^{(0)}(i\varepsilon_n) \sin^2 k_z \right]^2. \quad \text{(B3)}$$

Here we introduce magnetic-field effects through a minimal coupling of the vector potential $A(r)$ to the order-parameter field $\psi(r)$.

To diagonalize the second-order terms of Eq. (B1), we expand the order-parameter field as

$$\psi(r) = \sum_{N,n,q} b_{N,n,q} f_{N,n}(x,y) e^{i \varphi_{q,z}} L_z, \quad \text{(B4)}$$

where $N$ is the Landau-level index, $n_d$ is the degeneracy index for each Landau level with $(\mu_0 H L_d x/\phi_0)$-fold degeneracy, $q_z$ is the $z$-directional momentum, and $f_{N,n}(x,y)$ is the $N$th Landau-level eigenfunction [note that the lattice constant is unity so that $L_1 = M_1 (i = x, y, z)$]. Though, in general, it is not clear whether the second-order terms of Eq. (B1) are diagonalized with the bases appearing in Eq. (B4), at least the lowest-order $Q^2$ terms are exactly diagonalized with these bases. Respecting this fact and substituting Eq. (B4) into Eq. (B1), we obtain the diagonalized second-order terms:

$$\mathcal{F}_{GL} \simeq \sum_{N,n,q} \left( \alpha \sqrt{2N+1} q_z^2 + \gamma q_z^4 \right) |b_{N,n,q}|^2 + \int d^3r \frac{\beta}{2} |\psi|^4, \quad \text{(B5)}$$

where $q_z^2 = l^{-1} = \sqrt{2\pi \mu_0 H/\phi_0}$. Therefore, through the Landau quantization of the order-parameter field, we basically replace squared gauge-invariant gradient $Q^2$ defined in the $x$-$y$ plane with discrete levels $(2N + 1)/l^2$.

As far as we focus our attention on the region relatively near $H_{c2}(T)$, we just take into account the contribution from the lowest Landau-level mode $[8, 12, 26]$; then we can obtain from Eq. (B5) the following expression:

$$\mathcal{F}_{GL} \simeq \sum_{n,q} \left( \alpha q_z^2 + \gamma q_z^4 \right) |b_{0,n,q}|^2 + \int d^3r \frac{\beta}{2} |\psi|^4, \quad \text{(B6)}$$

Conversely using the expansion of the order-parameter field [Eq. (B4)], as well as considering only $N = 0$ mode, we finally obtain

$$\mathcal{F}_{GL} \simeq \int d^3r \left[ (\alpha q_z^2 + \gamma |\partial_z \psi|^2) + \frac{\beta}{2} |\psi|^4 \right], \quad \text{(B7)}$$

where $\psi(r)$ only involves the lowest Landau-level mode ($N = 0$).

### Appendix C: Derivation of vortex-lattice-formation field

In the following, we explain how we estimate the vortex-lattice-formation field $H_{\text{melt}}$ and obtain Eq. (17) starting with Eq. (14). Since the mean-field solution minimizing Eq. (14) is given by the triangular vortex-lattice state $[24]$ within the lowest-Landau-level approximation $[12, 25]$ and then apply the Lindemann criterion to estimate $H_{\text{melt}}$ $[12]$, at which
the first-order melting transition to the vortex-liquid state occurs. Since our formulation is basically based on Refs. [12, 25], we here just present an overview. In the following, we assume the Landau gauge $A(r) = -\mu_0 H y \hat{x}$. In this Appendix, $r$ denotes a coordinate vector $x \hat{x} + y \hat{y}$ in the $x$-$y$ plane.

As a complete orthonormal set of bases diagonalizing the second-order terms of Eq. (14), we consider a set of triangular vortex-lattice states with $z$-directional modulation:

$$\{ \varphi(r|r_0) e^{i q_z z} \}_{r_0, q_z} , \quad (C1)$$

where $\{ \varphi(r|r_0) \}$ represents a two-dimensional triangular vortex lattice with a unit cell shown in Fig. 4 and the position of the vortices is related to $r_0$:

$$r_0 = x_0 \hat{x} + y_0 \hat{y} = \left( \frac{2\pi l^2}{L_y} n_x + \frac{2\pi l^2}{\sqrt{3}L_x} n_y \right) \hat{x} + \frac{2\pi l^2}{L_x} n_y \hat{y} . \quad (C2)$$

Here $l = \sqrt{\phi_0/(2\pi \mu_0 H)}$ is the magnetic length. The degeneracy indices of the lowest Landau level, $n_x$ and $n_y$, satisfy

$$n_x \in \left[ -\frac{L_y}{2k l^2}, \frac{L_y}{2k l^2} \right] , \quad n_y \in \left[ -\frac{\sqrt{3}L_x}{4kl^2}, \frac{\sqrt{3}L_x}{4kl^2} \right] . \quad (C3)$$

with $k = \sqrt{3\pi/l}$. We note that the degeneracy of the lowest Landau level can be calculated as $[L_y/(2k l^2)] \cdot [\sqrt{3}L_y/(2k l^2)] = L_x L_y / (2\pi l^2) = \mu_0 H L_x L_y / \phi_0$. The domain of $r_0$ is equivalent to the unit cell shown in Fig. 4.

As shown in the following, functions $\{ \varphi(r|r_0) \}_{r_0}$ with $r_0$ out of the unit cell are linearly dependent on those with $r_0$ within the unit cell.

The specific form of the eigenfunctions $\{ \varphi(r|r_0) \}$ is given as

$$\varphi(r|r_0) = e^{-i\phi_0 r_x l^2} \varphi(r + r_0|0) , \quad (C4)$$

and

$$\varphi(r|0) = \frac{3^{1/8}}{\sqrt{L_x L_y}} \sum_{n = -\infty}^{\infty} e^{ikn x - i\pi n^2/2 - (y - k l^2 n)^2 / (2l^2)} . \quad (C5)$$

Defining primitive lattice vectors $a = (2\pi/k) \hat{x}$ and $b = (\pi/k) \hat{x} + (\sqrt{3}\pi/k) \hat{y}$ as shown in Fig. 4, we obtain from Eq. (C5) the following (quasi)periodicity of $\varphi(r|0)$:

$$\begin{aligned}
\varphi(r + a|0) &= \varphi(r|0) \\
\varphi(r + b|0) &= i e^{i kx} \varphi(r|0) ,
\end{aligned} \quad (C6)$$

As for a general lattice vector $R = ma + nb$, we can show from Eq. (C6) the following quasiperiodicity:

$$\varphi(r + R|0) = e^{i(\pi m_x^2/2 + \pi m_k x)} \varphi(r|0) . \quad (C7)$$

Combining Eqs. (C4) and (C7), we can obtain

$$\varphi(r + R|0) = e^{i(\pi m_x^2/2 + \pi m_k x - r_0 \cdot \hat{z}) \cdot R / l^2} \varphi(r|0) , \quad (C8)$$

which shows that $\varphi(r|r_0)$ and $\varphi(r|r_0 + R)$ are not independent; therefore, we only have to consider a set $\{ \varphi(r|r_0) \}_{r_0}$ where $r_0$ is in a unit cell of the vortex lattice. Moreover, Eqs. (C5) and (C4) lead to the following orthonormal relation:

$$\int_S d^2r \varphi^*(r|r_0) \varphi(r|r_0') = \delta_{r_0, r_0'} , \quad (C9)$$

where $S$ means the entire $x$-$y$ plane.

From Eqs. (C4) and (C7), we can show another relation:

$$\varphi(r + R|r_0) = e^{i(\pi m_x^2/2 + \pi m_k x - r_0 \cdot \hat{z}) \cdot R / l^2} \varphi(r|0) . \quad (C10)$$

Defining a momentum vector corresponding to $r_0$ as

$$k_0 = -\frac{r_0 \times \hat{z}}{l^2} \quad (\Leftrightarrow r_0 = l^2 k_0 \times \hat{z}) , \quad (C11)$$

we can rewrite Eq. (C10) as

$$\varphi(r + R|0) = e^{ik_0 \cdot R \cdot \hat{z}} \varphi(r|0) . \quad (C12)$$

Combination of Eqs. (C7) with (C10) leads to

$$\varphi^*(r + R|0) \varphi(r + R|0) = e^{ik_0 \cdot R} \varphi^*(r|0) \varphi(r|0) \quad (C13)$$

which means that $\varphi^*(r|0) \varphi(r|0)$ is a Bloch function with a lattice momentum vector $k_0$; therefore, we can expand this function as [21]

$$\varphi^*(r|0) \varphi(r|0) = \frac{1}{L_x L_y} \sum_{K} e^{i(k_0 + K \cdot \hat{r})} F_K(k_0) , \quad (C14)$$

FIG. 4. Schematic figure of a unit cell of the triangular vortex lattice (blue area). Primitive lattice vectors $(a$ and $b$) as well as the size of the unit cell are shown. Note that one quantum flux penetrates one unit cell $[(\sqrt{3}\pi/k) \cdot (2\pi/k) = 2\pi l^2 = \phi_0/(\mu_0 H)]$. 

where $\mathbf{K}$ is a reciprocal lattice vector, which can be written with a certain lattice vector $\mathbf{R} = m_a \mathbf{a} + m_b \mathbf{b}$, as

$$
\mathbf{K} = -\frac{\mathbf{R} \times \mathbf{z}}{l^2}.
$$

(C15)

Applying the Fourier transformation to Eq. (C14), we obtain

$$
F_K(k_0) = \exp \left\{ i^2 \left[ -\frac{\mathbf{K} + k_0^2}{4} - \frac{1}{2} \left( \frac{K_x^2}{\sqrt{3}} + K_y + k_0 z - (\mathbf{K} \times k_0)_z \right) \right] \right\}
$$

(C17)

non-Gaussian fluctuation $F_{\text{GL}}^{\text{nonGauss}}$ and concentrate on the Gaussian fluctuation $F_{\text{GL}}^{\text{Gauss}}$. The fluctuation amplitude $a^{(m)}_{k_0 q_z}$ is defined as

$$
a^{(m)}_{k_0 q_z} = \frac{1}{\sqrt{2}} \left( a_{k_0 q_z} \pm a_{-k_0, -q_z} \right),
$$

(C23) and the fluctuation energy of each mode $E_{k_0}^{(m)}$ is obtained as

$$
E_{k_0}^{(m)} = \frac{\alpha q_x}{\beta_A} \left[ 2 \sum_{K} |F_K(k_0)|^2 - \sum_{K} |F_K(0)|^2 \right] \pm \sum_{K} F_K(k_0)^2,
$$

(C24)

where $F_K(k_0)$ is given in Eq. (C17). We can show that $F_K(0) \in \mathbb{R}$, so that $E_{k_0}^{(m)} = 0$, which shows that the fluctuation mode represented as $a^{(m)}_{k_0 q_z}$ is massless (corresponding to the incompressible shear mode of the vortex lattice [12, 24]). Since the massless mode is expected to be dominant in considering the melting transition [12], we take into account the contribution of the massless mode $a^{(m)}_{k_0 q_z}$ and neglect that of the massive mode $a^{(m)}_{k_0 q_z}$. Moreover, to consider the long-wavelength and low-energy contribution of the massless mode, we expand the fluctuation energy $E_{k_0}^{(m)}$ with respect to $k_0$

$$
E_{k_0}^{(m)} = \frac{\gamma_A |\alpha q_x|}{\beta_A} l^4 k_0^4 + \mathcal{O}(k_0^6).
$$

(C25)

Here $\gamma_A$ is a numerical factor related to the triangular-lattice structure:

$$
\gamma_A = \sum_{K} e^{-i^2 K^2/2} \left\{ 1 + \frac{1}{12} \left[ \frac{3}{8} l^4 K^4 - 3 l^2 K^2 + 3 \right] - \frac{1}{8} \right\} 
\approx 0.119.
$$

(C26)

To derive Eq. (C25), we use the following properties with an arbitrary function $f(K) = f(|K|)$ due to a six-fold rotational symmetry of the reciprocal lattice space:

$$
\sum_{K} (K \cdot k_0)^2 f(K) = \sum_{K} \frac{1}{2} K^2 k_0^2
$$

$$
\sum_{K} (K \cdot k_0)^4 f(K) = \sum_{K} \frac{3}{8} K^4 k_0^4.
$$

(C27)
In the following, therefore, we focus on the following functional:

\[ \mathcal{F}_{GL}^{Gauss(-)} = \sum_{k_0, q, z > 0} \left( \frac{\gamma A |\alpha q u| l^4 k_0^4 + \gamma q_z^2}{\beta A} \right) |a_{k_0 q z}|^2. \]  

(C28)

It has been proved [24] that this form of the dispersion relation of the massless mode of the vortex lattice in type-II limit remains valid when the higher Landau-level modes \((N \geq 1)\) are included.

Since the relative fluctuation \(2^{-1/2} a_{k_0 q z} / |\pi|\) can be regarded as an angular change of the vortex lattice \(\theta_{k_0 q z}\), we can rewrite \(\mathcal{F}_{GL}^{Gauss(-)}\) [Eq. (C28)] as

\[ \mathcal{F}_{GL}^{Gauss(-)} = L_x L_y L_z \sum_{k_0, q, z > 0} \left( c_{66} l^4 k_0^4 + \rho_s q_z^2 \right) |\theta_{k_0 q z}|^2 \]

\[ = \frac{1}{2} \int d^2 r \int_0^{L_z} dz \left[ c_{66} l^4 (\nabla \cdot \theta)^2 + \rho_s (\partial^2 \theta)^2 \right]. \]

(C29)

Here, \(\theta(r, z) = \sum_{k_0, q, z} e^{i(k_0 r + q_z z)} \theta_{k_0 q z}\) is a real-space phase field related to the vortex-lattice displacement field \(u(r, z)\) [12] as

\[
\begin{align*}
u_x &= l^2 \partial_y \theta, \\
u_y &= -l^2 \partial_x \theta.
\end{align*}
\]

(C30)

This relation indicates that the vortex-lattice deformation corresponding to the massless mode \(\alpha q u\) represents an incompressible shear mode: \(\nabla \cdot u(r, z) = 0\) [12]. Also, \(c_{66}\) and \(\rho_s\) represent the shear modulus of the vortex lattice and the superfluid density defined as the response quantity in the \(z\) direction, respectively:

\[ c_{66} = \frac{2\gamma A |\alpha q u|^2}{\beta A^2 \beta}, \]

(C31)

and

\[ \rho_s = \frac{2|\alpha q u|^2}{\beta A^2 \beta}. \]

(C32)

The mean square displacement of the vortex lattice \(d^2 = \langle |u(r)|^2 \rangle\) is calculated as

\[ d^2 = \langle |u|^2 \rangle = l^4 \langle (\nabla \cdot \theta)^2 \rangle = 2l^4 \sum_{k_0, q, z > 0} k_0^2 |\theta_{k_0 q z}|^2. \]

(C33)

Here, \(\langle \cdots \rangle\) means the ensemble average with respect to the low-energy Ginzburg-Landau functional \(\mathcal{F}_{GL}\) [Eq. (C29)], and thus we can obtain the following formula:

\[ d^2 = \frac{l^4}{L_x L_y L_z} \sum_{k_0, q, z > 0} T k_0^2 \left( c_{66} l^4 k_0^4 + \rho_s q_z^2 \right). \]

(C34)

Since the summation about \(q_z\) is convergent, we take \(L_z^{-1} \sum_{q_z} (\cdots) \rightarrow (2\pi)^{-1} \int_{-\infty}^{\infty} dq_z (\cdots)\). On the other hand, since the summation about \(k_0\) is not convergent when \(k_0 \rightarrow \infty\), we simply replace the summation with an integration over an area corresponding to the first Brillouin zone: \(L_x L_y^{-1} \sum_{k_0} (\cdots) \rightarrow (2\pi)^{-1} \int_{0}^{\sqrt{2}/l} dk_0 k_0 (\cdots)\). These replacements lead to the following simple expression:

\[ d^2 = \frac{T}{4\pi \sqrt{\rho_s c_{66}}} \]

(C35)

Using the Lindemann criterion [12], we can expect that the vortex lattice can melt into the vortex liquid when a condition \(d = c \times 1\) is satisfied [note that the magnetic length \(l\) corresponds to the unit-cell size (see Fig. 4)], where \(c = O(0.1)\) is a phenomenological parameter. Introducing a dimensionless magnetic field \(h = 2\pi \mu_0 H / k_0 l^2\) (note that the lattice constant is set to unity), we obtain the equation [Eq. (17)] describing the melting-transition field, or the vortex-lattice-formation field, \(H_{melt}\):

\[ \frac{T}{4\pi \sqrt{\rho_s c_{66}}} = \frac{c^2}{h}. \]

(C36)

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