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Static spin susceptibility in magnetically ordered states

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We report that special care is needed when longitudinal magnetic susceptibility is computed in a magnetically ordered phase, especially in metals. We demonstrate this by studying static susceptibility in both a ferromagnetic and an antiferromagnetic state in the random phase approximation to the two-dimensional Hubbard model on a square lattice. In contrast to the case in the disordered phase, a first derivative of the chemical potential (or the density) with respect to a magnetic field does not vanish in a magnetically ordered phase when the field is applied parallel to the magnetic moment. This effect is crucial and should be included when computing magnetic susceptibility in the ordered phase, otherwise an unphysical result would be obtained. In addition, consequently the magnetic susceptibility becomes different when computed at a fixed density and a fixed chemical potential in the ordered phase. In particular, we cannot employ magnetic susceptibility at a fixed chemical potential to describe a system with a fixed density even if the chemical potential is tuned to reproduce the correct density.

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I. INTRODUCTION

Spin susceptibility is a fundamental quantity to study the magnetic property of a system, and it is often computed in the so-called random phase approximation (RPA). While this approximation is usually good enough for a three-dimensional system, it may not be precise enough, especially in a two-dimensional system. However, even in such a case, the susceptibility computed in the RPA is believed to capture at least qualitative properties of the system.

RPA susceptibility is frequently computed in a disordered phase, but it can also be computed in a magnetically ordered phase [1,2]. Moreover, as actually observed in high-temperature cuprates [3], iron-based pnictides and chalcogenides [4], and heavy-fermion materials [5], the ordered phase sometimes coexists with superconductivity. Even in such a complicated situation, the RPA provides feasible computations of magnetic susceptibility [6,7].

Typically, RPA susceptibility is obtained by connecting a simple bubble (or ladder) of noninteracting particle-hole excitations with the electron-electron interaction, that is, its functional form is given typically by

$$\chi \propto (1 - \chi_0 g)^{-1} \chi_0, \quad (1)$$

where g is the interaction strength and χ_0 is the susceptibility in the noninteracting case; χ , χ_0 , and g can be matrices. In a magnetic phase, χ_0 is computed by using the quasiparticle propagator in the ordered phase, and also by considering possible umklapp contributions to the susceptibility when the translational symmetry is broken by a magnetic order. Such a procedure indeed yields the correct result of transverse magnetic susceptibility [1,2,6–11], but special care is needed for longitudinal magnetic susceptibility, which is not well recognized [7,8,10–12].

Spin rotational symmetry is broken in a magnetically ordered phase. As a result, the chemical potential (or the density) is no longer a quadratic function of a magnetic field when the field is applied parallel to the magnetic moment. A

first derivative of the chemical potential (or the density) then becomes finite. Hence this effect should be considered on an equal footing when we compute magnetic susceptibility, because the magnetic susceptibility is a linear-response quantity of a magnetic field.

In this paper, we show how important the contribution of the first derivative of the chemical potential (or the density) is to compute longitudinal susceptibility in a magnetically ordered phase, which we exemplify by employing the two-dimensional Hubbard model for both a ferromagnetic and an antiferromagnetic state. Since the RPA is equivalent to the mean-field approximation or the saddle-point approximation, we can directly compute the magnetic susceptibility in mean-field theory for the Hubbard model. We provide the correct expression of the static susceptibility in the RPA as well as results when the first derivative of the chemical potential (or the density) is neglected. In addition, we point out that the longitudinal magnetic susceptibility is different when computed at a fixed density and a fixed chemical potential in a magnetically ordered phase. Consequently, when the density is fixed, the susceptibility obtained at a fixed chemical potential cannot be applicable even if the chemical potential is tuned to reproduce the correct density.

This paper is organized as follows. In Sec. II we present the model and derive the self-consistency equations for both a ferromagnetic and an antiferromagnetic phase. The corresponding magnetic susceptibility is computed in Secs. III and IV, respectively. We show in Sec. V that the susceptibility for a fixed chemical potential is reproduced in a conventional diagrammatic approach. Concluding remarks are given in Sec. VI.

II. MODEL AND SELF-CONSISTENCY EQUATIONS

To exemplify our issue, we employ the two-dimensional Hubbard model on a square lattice,

$$\mathcal{H} = - \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow} + \mathcal{H}_z, \quad (2)$$

where the transfer integrals t_{ij} are finite between the first- (t) and second- (t') neighbor sites and zero otherwise; U represents the on-site Coulomb repulsion. \mathcal{H}_z is the Zeeman term, for which we consider a static and uniform (staggered) magnetic field when we compute longitudinal magnetic susceptibility in a ferromagnetic (an antiferromagnetic) state. That is, it is described as

$$\mathcal{H}_z = -h \sum_j \frac{1}{2} (n_{j\uparrow} - n_{j\downarrow}) e^{i\mathbf{q}\cdot\mathbf{r}_j} \quad (3)$$

with $\mathbf{q} = \mathbf{0}$ [$\mathbf{q} = \mathbf{Q} \equiv (\pi, \pi)$]. Here h is an effective magnetic field given by $h = g\mu_B H$; g is a g factor, μ_B is the Bohr magneton, and H is an external magnetic field. The magnetic field is infinitesimally small, and we take the limit of $h \rightarrow +0$ when we compute the susceptibility.

Since the RPA is equivalent to the mean-field approximation, we compute the RPA susceptibility in mean-field theory. Defining the magnetization and the density operator as

$$m_j = \frac{1}{2} (n_{j\uparrow} - n_{j\downarrow}), \quad (4)$$

$$n_j = n_{j\uparrow} + n_{j\downarrow}, \quad (5)$$

respectively, the interaction term is written as $n_{j\uparrow}n_{j\downarrow} = \frac{1}{4}n_jn_j - m_jm_j$. The density is assumed to be uniform and is given by $n = \langle n_j \rangle$, whereas the magnetization $\langle m_j \rangle$ is uniform in the ferromagnetic state and staggers with a wave vector $\mathbf{q} = \mathbf{Q}$ in the antiferromagnetic state. In mean-field theory, the interaction term is decoupled as

$$n_{j\uparrow}n_{j\downarrow} \rightarrow \frac{n}{2}n_j - 2\langle m_j \rangle m_j - \frac{1}{4}n^2 + \langle m_j \rangle^2, \quad (6)$$

and self-consistency equations for n and $\langle m_j \rangle$ are obtained by minimizing the free energy.

In the ferromagnetic state, $\langle m_j \rangle$ is independent of j , i.e., $\langle m_j \rangle = m$. The self-consistency equations are given by

$$n = \frac{1}{N} \sum_{\mathbf{k}} \left[f\left(\xi_{\mathbf{k}} - Um - \frac{h}{2}\right) + f\left(\xi_{\mathbf{k}} + Um + \frac{h}{2}\right) \right], \quad (7)$$

$$m = \frac{1}{2N} \sum_{\mathbf{k}} \left[f\left(\xi_{\mathbf{k}} - Um - \frac{h}{2}\right) - f\left(\xi_{\mathbf{k}} + Um + \frac{h}{2}\right) \right]. \quad (8)$$

Here

$$\xi_{\mathbf{k}} = -2t(\cos k_x + \cos k_y) - 4t' \cos k_x \cos k_y + \frac{Un}{2} - \mu, \quad (9)$$

and f , μ , and N are the Fermi distribution function, the chemical potential, and the total number of lattice sites, respectively, and the summation of \mathbf{k} is taken over the first Brillouin zone.

In the case of the antiferromagnetic state, the magnetization is described by $\langle m_j \rangle = m_{\mathbf{Q}} e^{i\mathbf{Q}\cdot\mathbf{r}_j}$. Here $m_{\mathbf{Q}}$ is the staggered

magnetization, which is the order parameter of antiferromagnetism. The self-consistency equations are given by

$$n = \frac{2}{N} \sum_{\mathbf{k}}' [f(E_{\mathbf{k}}^+) + f(E_{\mathbf{k}}^-)], \quad (10)$$

$$m_{\mathbf{Q}} = -\frac{1}{N} \sum_{\mathbf{k}}' \frac{Um_{\mathbf{Q}} + \frac{h}{2}}{D_{\mathbf{k}}} [f(E_{\mathbf{k}}^+) - f(E_{\mathbf{k}}^-)], \quad (11)$$

where the summation of \mathbf{k} is taken over the magnetic Brillouin zone, namely $|k_x| + |k_y| \leq \pi$, and

$$E_{\mathbf{k}}^{\pm} = \xi_{\mathbf{k}}^{\pm} \pm D_{\mathbf{k}}, \quad (12)$$

$$\xi_{\mathbf{k}}^{\pm} = \frac{1}{2} (\xi_{\mathbf{k}} \pm \xi_{\mathbf{k}+\mathbf{Q}}), \quad (13)$$

$$D_{\mathbf{k}} = \sqrt{(\xi_{\mathbf{k}}^-)^2 + \left(Um_{\mathbf{Q}} + \frac{h}{2}\right)^2}. \quad (14)$$

A comprehensive mean-field analysis of the Hubbard model [13] clarified the parameter region where ferromagnetic phases with $\mathbf{q} = \mathbf{0}$ and antiferromagnetic phases with $\mathbf{q} = \mathbf{Q}$ are stabilized. Referring to Ref. [13], we fix $U = 3t$ and choose $t' = -0.45t$ and $n = 0.2$ to describe the ferromagnetic state, and $t' = -0.2t$ and $n = 1.1$ for the antiferromagnetic state. Our conclusions, however, do not depend on the choice of parameters as long as the ferromagnetic (or antiferromagnetic) phase is stabilized. In the following, we set $t = 1$ and measure all quantities with the dimensions of energy in units of t .

III. UNIFORM SUSCEPTIBILITY IN THE FERROMAGNETIC STATE

The longitudinal magnetic susceptibility is obtained in the RPA by taking a first derivative with respect to a field in Eqs. (7) and (8), and then by taking the limit of $h \rightarrow +0$. One would assume that a first derivative of μ (or n) with respect to a field should vanish in the limit of $h \rightarrow +0$. This is actually correct at least in the disordered phase. As a result, the longitudinal susceptibility, which is defined by $\frac{\partial m}{\partial h}|_{h \rightarrow +0}$, is obtained as

$$\tilde{\chi}(\mathbf{0}) = \frac{1}{4} \frac{\chi_{\uparrow} + \chi_{\downarrow}}{1 - \frac{U}{2}(\chi_{\uparrow} + \chi_{\downarrow})}, \quad (15)$$

where

$$\chi_{\uparrow(\downarrow)} = -\frac{1}{N} \sum_{\mathbf{k}} f'(\xi_{\mathbf{k}} \mp Um) \quad (16)$$

and f' is the first derivative with respect to energy.

The temperature (T) dependence of $\tilde{\chi}(\mathbf{0})$ is shown in Fig. 1. With decreasing T , $\tilde{\chi}(\mathbf{0})$ grows and diverges at the Curie temperature $T_{\text{FM}} (= 0.187)$. Below T_{FM} , ferromagnetic order m develops. The value of m is determined by the self-consistency equations (7) and (8). As expected, $\tilde{\chi}(\mathbf{0})$ is suppressed below T_{FM} . However, it is enhanced at lower temperature inside the ferromagnetic state. This dependence is obviously unphysical and originates from the wrong assumption that the chemical potential should remain a quadratic function with respect to a field inside the ferromagnetic state. To show this, we plot $\Delta\mu(T, h) \equiv \mu(T, h) - \mu(T, 0)$ in Fig. 2. The chemical potential μ has a quadratic dependence of h in the vicinity

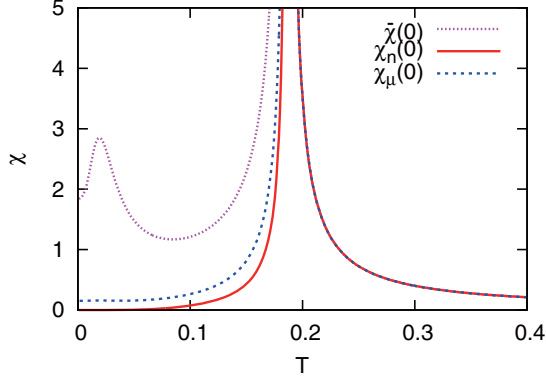


FIG. 1. Longitudinal magnetic susceptibility as a function of temperature at a fixed density; $n = 0.2$, $t' = -0.45$, and $U = 3$. The Curie temperature is $T_{\text{FM}} = 0.187$, below which the ferromagnetic moment develops. $\tilde{\chi}(\mathbf{0})$ is obtained from Eq. (15) and correct only in $T > T_{\text{FM}}$. $\chi_n(\mathbf{0})$ is given by Eq. (17) and correct in the whole temperature region. For $\chi_\mu(\mathbf{0})$ [Eq. (19)], the chemical potential is tuned at each temperature to reproduce the fixed density $n = 0.2$. $\chi_\mu(\mathbf{0})$ provides the correct result only in $T > T_{\text{FM}}$.

of $h = 0$ in the disordered phase because of the spin-rotational symmetry of the system. Its curvature around $h = 0$ becomes larger upon approaching T_{FM} and becomes infinite just at T_{FM} . Below T_{FM} , a linear term emerges with a singularity at $h = 0$. The emergence of the linear term is due to the breaking of the spin rotational symmetry, that is, the system has a different response when an infinitesimally small field is applied parallel and antiparallel to the direction of the ferromagnetic moment. Therefore, the emergent linear term in h is crucially important to describe the response in the ordered phase, and Eq. (15) is valid only in the disordered phase where $m = 0$. While $\tilde{\chi}(\mathbf{0})$ is enhanced below $T \lesssim 0.05$ in Fig. 1 for the present choice of the parameters, it could diverge inside the ferromagnetic phase, especially when U is chosen to be a larger value.

A. Fixed density

We first consider the situation in which the density is fixed. To get the correct RPA susceptibility inside the ordered phase,

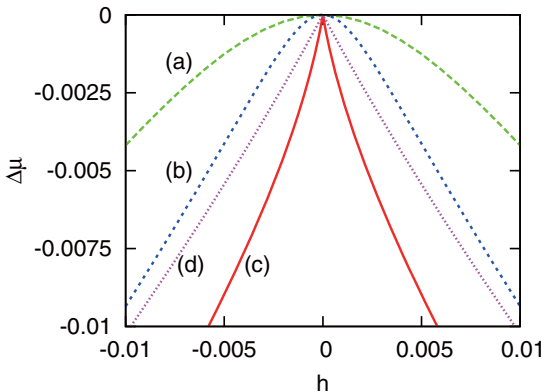


FIG. 2. h dependence of the chemical potential for several choices of temperatures: (a) $T = 1.1T_{\text{FM}}$, (b) $T = 1.04T_{\text{FM}}$, (c) $T = 0.99T_{\text{FM}}$, and (d) $T = 0.9T_{\text{FM}}$.

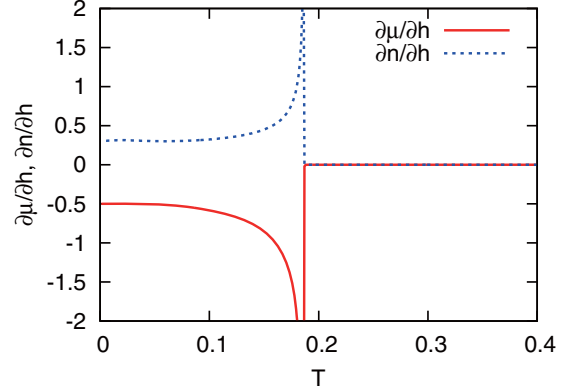


FIG. 3. Temperature dependence of the first derivative of μ and n with respect to a magnetic field h ; $T_{\text{FM}} = 0.187$, $n = 0.2$, $t' = -0.45$, and $U = 3$.

a first derivative of μ should be kept when differentiating Eqs. (7) and (8) with respect to h . Solving coupled equations, we obtain

$$\chi_n(\mathbf{0}) = \left. \frac{\partial m}{\partial h} \right|_{h \rightarrow +0} = \frac{\chi_\uparrow \chi_\downarrow}{\chi_\uparrow + \chi_\downarrow - 2U \chi_\uparrow \chi_\downarrow}, \quad (17)$$

$$\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0} = -\frac{1}{2} \frac{\chi_\uparrow - \chi_\downarrow}{\chi_\uparrow + \chi_\downarrow - 2U \chi_\uparrow \chi_\downarrow}. \quad (18)$$

In the disordered phase, we have $\chi_\uparrow = \chi_\downarrow$. Hence Eq. (17) is reduced to Eq. (15) and $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0} = 0$. However, inside the ferromagnetic phase, it is clear that the functional form of Eq. (17) is very different from Eq. (15), and in addition $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0}$ becomes finite. We plot the temperature dependence of $\chi_n(\mathbf{0})$ in Fig. 1. $\chi_n(\mathbf{0})$ is suppressed monotonically inside the ferromagnetic phase with decreasing temperature. This is because the system becomes less susceptible to an infinitesimally small field parallel to the magnetic moment when the magnetic moment grows with decreasing temperature. The enhancement of Eq. (15) inside the ferromagnetic phase (Fig. 1), therefore, should be an artifact due to the discarding of the contribution from $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0}$. As already implied in Fig. 2, the contribution of $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0}$ is indeed sizable below T_{FM} . The temperature dependence of $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0}$ is plotted in Fig. 3. The quantity $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0}$ is zero down to $T = T_{\text{FM}}$. It diverges at $T = T_{\text{FM}}$ only on the side of low temperature and is suppressed with decreasing T , keeping a value comparable to $\chi_n(\mathbf{0})$ at low temperature (see also Fig. 1).

B. Fixed chemical potential

We now consider the situation in which μ is fixed. In this case, we differentiate Eqs. (7) and (8) with respect to h for a fixed μ . We then obtain

$$\chi_\mu(\mathbf{0}) = \left. \frac{\partial m}{\partial h} \right|_{h \rightarrow +0} = \frac{1}{4} \frac{\chi_\uparrow + \chi_\downarrow + 2U \chi_\uparrow \chi_\downarrow}{1 - U^2 \chi_\uparrow \chi_\downarrow}, \quad (19)$$

$$\left. \frac{\partial n}{\partial h} \right|_{h \rightarrow +0} = \frac{1}{2} \frac{\chi_\uparrow - \chi_\downarrow}{1 - U^2 \chi_\uparrow \chi_\downarrow}. \quad (20)$$

Equation (19) is already known in the literature [1,14].

In the disordered phase, we have $\chi_\uparrow = \chi_\downarrow$, yielding $\frac{\partial n}{\partial h}|_{h \rightarrow +0} = 0$ and $\chi_\mu = \chi_n = \tilde{\chi}$. Consequently, the magnetic susceptibility at a fixed density is the same as that at a fixed chemical potential in the disordered phase.

In the ordered phase, however, we have $\chi_\uparrow \neq \chi_\downarrow$, and $\frac{\partial n}{\partial h}|_{h \rightarrow +0}$ becomes finite as shown in Fig. 3 and diverges at $T = T_{\text{AF}} - 0$. A comparison of Eqs. (17) and (19) should be made in the same condition, namely the same density and the same chemical potential. A physical quantity computed at a fixed chemical potential is frequently used to describe a system with a fixed density by tuning the chemical potential to reproduce the density. Following this standard procedure, we plot the temperature dependence of χ_μ also in Fig. 1. Although the functional forms of Eqs. (17) and (19) are different, both provide similar results in the ordered phase. Nevertheless, in a strict sense, χ_μ does not lead to the correct result χ_n when the density is fixed in the system. Conversely, χ_μ [Eq. (19)] would provide the correct result when the chemical potential is fixed in the system (see the Appendix). In this case, χ_n [Eq. (17)] is in turn not correct even if the density is tuned to reproduce the fixed chemical potential. The reason why the longitudinal magnetic susceptibility χ_μ does not agree with χ_n in the magnetically ordered phase is that μ and n are not symmetric in Eqs. (7) and (8), and thus the field dependences of μ and n (see Fig. 3) are different from each other.

IV. STAGGERED SUSCEPTIBILITY IN THE ANTIFERROMAGNETIC STATE

The longitudinal staggered susceptibility is defined as $\frac{\partial m_Q}{\partial h}|_{h \rightarrow +0}$, where h is a magnitude of a staggered field introduced in Eq. (3) with $\mathbf{q} = \mathbf{Q}$. In the disordered phase, the spin rotational symmetry is preserved and thus μ and n are quadratic functions of h for a small h . In this case, we have $\frac{\partial \mu}{\partial h}|_{h \rightarrow +0} = 0$ and $\frac{\partial n}{\partial h}|_{h \rightarrow +0} = 0$. Thus we do not need to consider a first derivative of μ and n with respect to h in Eq. (11). The staggered susceptibility then becomes

$$\tilde{\chi}(\mathbf{Q}) = \frac{1}{2} \frac{\chi^{(0)}(\mathbf{Q})}{1 - U\chi^{(0)}(\mathbf{Q})}, \quad (21)$$

where

$$\begin{aligned} \chi^{(0)}(\mathbf{Q}) = & -\frac{1}{N} \sum_{\mathbf{k}}' \frac{(\xi_{\mathbf{k}}^-)^2}{D_{\mathbf{k}}^3} [f(E_{\mathbf{k}}^+) - f(E_{\mathbf{k}}^-)] \\ & - \frac{1}{N} \sum_{\mathbf{k}}' \left(\frac{Um_{\mathbf{Q}}}{D_{\mathbf{k}}} \right)^2 [f'(E_{\mathbf{k}}^+) + f'(E_{\mathbf{k}}^-)], \quad (22) \end{aligned}$$

and $m_{\mathbf{Q}} = 0$ here. One might apply the formula Eq. (21) to the antiferromagnetic phase, employing $m_{\mathbf{Q}}$ and n (or μ) determined by solving the self-consistency equations (10) and (11). The resulting $\tilde{\chi}(\mathbf{Q})$ [Eq. (21)] is shown in Fig. 4 as a function of temperature. With decreasing T , $\tilde{\chi}(\mathbf{Q})$ grows and diverges at the Néel temperature $T_{\text{AF}} = 0.380$. Just below T_{AF} , $\tilde{\chi}(\mathbf{Q})$ is suppressed as expected. However, it grows below $T \lesssim 0.2$ inside the antiferromagnetic phase. This apparently unphysical result originates from the wrong assumption that μ and n would still be quadratic in h in the magnetic phase. The enhancement of $\tilde{\chi}(\mathbf{Q})$ could appear as its divergence at

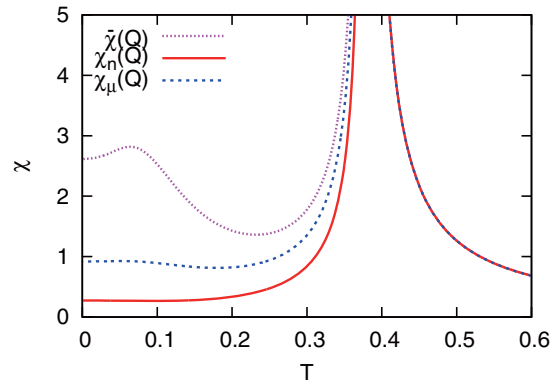


FIG. 4. Longitudinal magnetic susceptibility as a function of temperature at a fixed density; $n = 1.1$, $t' = -0.2$, and $U = 3$. The Néel temperature is $T_{\text{AF}} = 0.380$, below which the antiferromagnetic moment develops. $\tilde{\chi}(\mathbf{Q})$ is obtained from Eq. (21) and correct only in $T > T_{\text{AF}}$. $\chi_n(\mathbf{Q})$ is given by Eq. (23) and correct in the whole temperature region. $\chi_\mu(\mathbf{Q})$ [Eq. (30)] is computed in the condition of a fixed chemical potential; the chemical potential is tuned at each temperature to reproduce the correct density. The result $\chi_\mu(\mathbf{Q})$ is, however, correct only in $T > T_{\text{AF}}$.

a certain temperature below T_{AF} when a larger value of U is taken.

Figure 5 shows $\Delta\mu = \mu(T, h) - \mu(T, 0)$ as a function of h for several choices of T . For $T > T_{\text{AF}}$ we see $\frac{\partial \mu}{\partial h}|_{h \rightarrow +0} = 0$. However, below T_{AF} , $\Delta\mu$ becomes singular at $h = 0$ and acquires a linear dependence of $|h|$ around $h = 0$. This effect is crucially important to obtain the correct RPA expression of the longitudinal magnetic susceptibility inside the magnetic phase. Because the correct expression depends on whether the density is fixed or the chemical potential is fixed, we present it below separately.

A. Fixed density

For a fixed density n , we differentiate both Eqs. (10) and (11) with respect to h and take the limit of $h \rightarrow +0$. Coupled

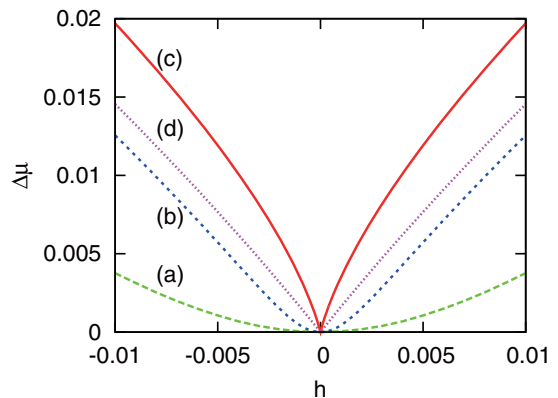


FIG. 5. h dependence of the chemical potential for several choices of temperatures: (a) $T = 1.1T_{\text{AF}}$, (b) $T = 1.03T_{\text{AF}}$, (c) $T = 0.99T_{\text{AF}}$, and (d) $T = 0.9T_{\text{AF}}$; $T_{\text{AF}} = 0.380$, $n = 1.1$, $t' = -0.2$, and $U = 3$.

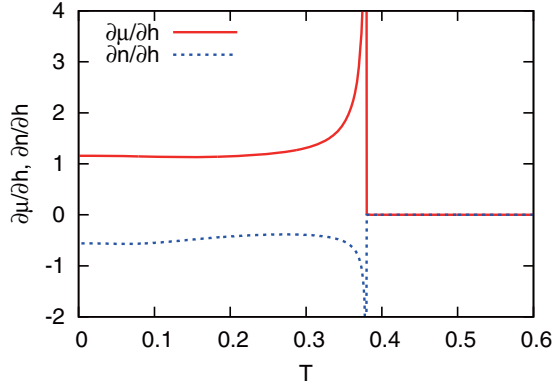


FIG. 6. Temperature dependence of the first derivative of μ and n with respect to a magnetic field h ; $T_{\text{AF}} = 0.380$, $n = 1.1$, $t' = -0.2$, and $U = 3$.

equations of $\frac{\partial m}{\partial h}$ and $\frac{\partial \mu}{\partial h}$ are easily solved, yielding

$$\chi_n(\mathbf{Q}) = \left. \frac{\partial m}{\partial h} \right|_{h \rightarrow +0} = \frac{1}{2} \frac{\chi_n^{(0)}(\mathbf{Q})}{1 - U \chi_n^{(0)}(\mathbf{Q})}, \quad (23)$$

$$\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0} = -\frac{1}{2} \frac{a_{12}/a_{11}}{1 - U \chi_n^{(0)}(\mathbf{Q})}, \quad (24)$$

where

$$\chi_n^{(0)}(\mathbf{Q}) = \frac{a_{11}a_{22} - a_{12}a_{21}}{a_{11}}, \quad (25)$$

and

$$a_{11} = -\frac{2}{N} \sum_{\mathbf{k}}' [f'(E_{\mathbf{k}}^+) + f'(E_{\mathbf{k}}^-)], \quad (26)$$

$$a_{12} = \frac{2}{N} \sum_{\mathbf{k}}' \frac{U m_{\mathbf{Q}}}{D_{\mathbf{k}}} [f'(E_{\mathbf{k}}^+) - f'(E_{\mathbf{k}}^-)], \quad (27)$$

$$a_{21} = \frac{1}{2} a_{12}, \quad (28)$$

$$a_{22} = \chi^{(0)}(\mathbf{Q}). \quad (29)$$

Here h should be set to zero in $E_{\mathbf{k}}^{\pm}$ and $D_{\mathbf{k}}$ [see Eqs. (12) and (14)]. The functional form of Eq. (23) is the same as Eq. (21), but $\chi_n^{(0)}(\mathbf{Q})$ becomes identical to $\chi^{(0)}(\mathbf{Q})$ only for $m_{\mathbf{Q}} = 0$. $\chi_n(\mathbf{Q})$ is plotted in Fig. 4 as a function of temperature. It is the same as Eq. (21) above T_{AF} . Below T_{AF} , $\chi_n(\mathbf{Q})$ is suppressed monotonically with decreasing temperature as it should be. In Fig. 6 we plot $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0}$. It vanishes in the disordered phase, but it becomes sizable in the magnetically ordered phase with divergence at $T = T_{\text{AF}}$ on the side of low temperature.

B. Fixed chemical potential

We next fix the chemical potential and differentiate Eqs. (10) and (11) with respect to h . Taking the limit of $h \rightarrow +0$, we obtain

$$\chi_{\mu}(\mathbf{Q}) = \left. \frac{\partial m}{\partial h} \right|_{h \rightarrow +0} = \frac{1}{2} \frac{\chi_{\mu}^{(0)}(\mathbf{Q})}{1 - U \chi_{\mu}^{(0)}(\mathbf{Q})}, \quad (30)$$

$$\left. \frac{\partial n}{\partial h} \right|_{h \rightarrow +0} = \frac{1}{2(1 + \frac{U}{2} a_{11})} \frac{a_{12}}{1 - U \chi_{\mu}^{(0)}(\mathbf{Q})}, \quad (31)$$

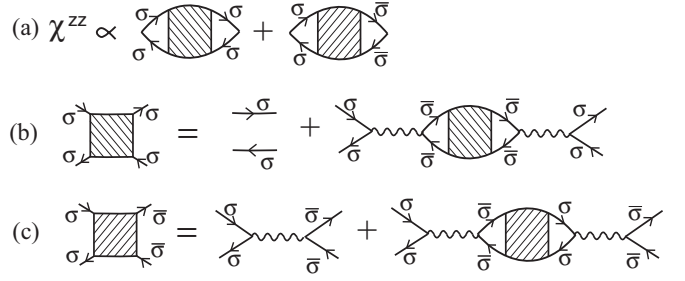


FIG. 7. Diagrams of the longitudinal magnetic susceptibility for the Hubbard interaction. $\bar{\sigma}$ denotes the spin direction opposite to σ .

where

$$\chi_{\mu}^{(0)}(\mathbf{Q}) = a_{22} - \frac{U}{2} \frac{a_{12}a_{21}}{1 + \frac{U}{2} a_{11}}. \quad (32)$$

The functional form of Eq. (30) is the same as Eq. (23) obtained at a fixed density. However, the expression of $\chi_{\mu}^{(0)}$ is very different from $\chi_n^{(0)}$ [Eq. (25)]. They become the same only in the disordered phase, where $m_{\mathbf{Q}} = 0$ and thus $a_{12} = a_{21} = 0$. The temperature dependence of $\chi_{\mu}(\mathbf{Q})$ is shown in Fig. 4. Since the density is fixed in Fig. 4, the chemical potential is tuned to reproduce the correct density at each temperature, as is usually done. Below T_{AF} , $\chi_{\mu}(\mathbf{Q})$ is suppressed, but it does not reproduce the correct result of $\chi_n(\mathbf{Q})$. This wrong result originates from the naive assumption that the susceptibility obtained at a fixed chemical potential could be used for the system with a fixed density after tuning the chemical potential to reproduce the correct density. However, as we have obtained explicitly, the susceptibility at a fixed chemical potential [Eqs. (30) and (32)] is different from that at a fixed density [Eqs. (23) and (25)–(29)] in the magnetically ordered phase. Furthermore, as shown in Fig. 6, the temperature dependence of $\left. \frac{\partial n}{\partial h} \right|_{h \rightarrow +0}$ is very different from $\left. \frac{\partial \mu}{\partial h} \right|_{h \rightarrow +0}$. Therefore, the choice of χ_n and χ_{μ} should be made carefully to describe the system appropriately. Conversely, if we wish to describe the system with a fixed chemical potential, the susceptibility $\chi_{\mu}(\mathbf{Q})$ is the correct one and $\chi_n(\mathbf{Q})$ [Eq. (23)] does not reproduce the correct result even if the density is tuned to reproduce the correct chemical potential at each temperature (see the Appendix).

V. DIAGRAMMATIC APPROACH

It is natural to ask what kind of result is obtained when a diagrammatic approach is employed. The longitudinal magnetic susceptibility is defined by

$$\chi^{zz}(\mathbf{q}, i\omega_m) = \frac{1}{N} \int_0^{1/T} d\tau e^{i\omega_m \tau} \langle T_{\tau} S^z(\mathbf{q}, \tau) S^z(-\mathbf{q}, 0) \rangle, \quad (33)$$

where $\omega_m = 2m\pi T$ is the bosonic Matsubara frequency, with m being an integer, $S^z(\mathbf{q}, \tau) = e^{\tau \mathcal{H}} S^z(\mathbf{q}) e^{-\tau \mathcal{H}}$, and $S^z(\mathbf{q}) = \frac{1}{2} \sum_{\mathbf{k}\sigma} \sigma c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}+\mathbf{q}\sigma}$.

In the disordered phase, χ^{zz} is given by the diagrams shown in Fig. 7 in the RPA. Hence we obtain

$$\chi^{zz} = \frac{1}{4} \left(\frac{\chi_{\uparrow} + \chi_{\downarrow}}{1 - U^2 \chi_{\uparrow} \chi_{\downarrow}} + \frac{2U \chi_{\uparrow} \chi_{\downarrow}}{1 - U^2 \chi_{\uparrow} \chi_{\downarrow}} \right). \quad (34)$$

In the static case, we set $i\omega_m = 0$ and take $\mathbf{q} = \mathbf{0}$ and \mathbf{Q} for the uniform and staggered susceptibility, respectively. We then obtain $\chi_\uparrow = \chi_\downarrow = \chi_0$, which is the same as Eq. (16) for the uniform susceptibility and Eq. (22) for the staggered susceptibility. Hence Eq. (34) is reduced to

$$\chi^{zz} = \frac{1}{2} \frac{\chi_0}{1 - U\chi_0}, \quad (35)$$

and we reproduce the correct results Eqs. (15) and (21) in the disordered phase.

In the ordered phase, we may compute $\chi_{\uparrow(\downarrow)}$ by using the quasiparticle propagator. In the ferromagnetic phase, Eq. (34) then becomes the same as $\chi_\mu(\mathbf{0})$ [see Eq. (19) and Refs. [1] and [14]], but not $\chi_n(\mathbf{0})$ [Eq. (17)].

The situation is delicate in the antiferromagnetic phase. Although the translational symmetry is broken by the magnetic order, the umklapp components of the susceptibility, such as $\langle T_\tau S^z(\mathbf{q}, \tau) S^z(-\mathbf{q} - \mathbf{Q}, 0) \rangle$ and $\langle T_\tau S^z(\mathbf{q} + \mathbf{Q}, \tau) S^z(-\mathbf{q}, 0) \rangle$, do not contribute to the longitudinal susceptibility [8]. Hence one might think that the RPA susceptibility would be obtained simply by replacing the electron Green's function with the Green's function of the two-component field $\psi_{\mathbf{k}\sigma}^\dagger = (c_{\mathbf{k}\sigma}^\dagger, c_{\mathbf{k}+\mathbf{Q}\sigma}^\dagger)$; the summation of \mathbf{k} is then restricted to the magnetic Brillouin zone. This kind of calculation is frequently seen in the literature [7, 8, 10–12]. In this case, however, we obtain $\chi_\uparrow(\mathbf{Q}) = \chi_\downarrow(\mathbf{Q}) = \chi^{(0)}(\mathbf{Q})$ [see Eq. (22)], which is the same as $\tilde{\chi}(\mathbf{Q})$ and does not reproduce the correct result inside the antiferromagnetic phase, as we have seen in Fig. 4. The correct procedure [6, 9, 15] is to take into account the umklapp components such as $\langle T_\tau S^z(\mathbf{q}, \tau) \rho(-\mathbf{q} - \mathbf{Q}, 0) \rangle$ as well as the density fluctuations with $\mathbf{q} + \mathbf{Q}$, namely $\langle T_\tau \rho(\mathbf{q} + \mathbf{Q}, \tau) \rho(-\mathbf{q} - \mathbf{Q}, 0) \rangle$. The density operator may be given by $\rho(\mathbf{q}) = \frac{1}{2} \sum_{\mathbf{k}} \sum_{\sigma} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}+\mathbf{Q}\sigma} = \frac{1}{2} \sum_{\mathbf{k}} \sum_{\sigma} \psi_{\mathbf{k}\sigma}^\dagger \psi_{\mathbf{k}+\mathbf{Q}\sigma}$, where the factor of 1/2 is added to make the formalism simpler. The resulting RPA expression becomes

$$\hat{\chi} = (1 - \hat{\chi}_0 \hat{U})^{-1} \hat{\chi}_0 \quad (36)$$

and

$$\hat{\chi} = \begin{pmatrix} \chi^{zz}(\mathbf{q}, i\omega_m) & \chi^{z\rho}(\mathbf{q}, \mathbf{q} + \mathbf{Q}, i\omega_m) \\ \chi^{\rho z}(\mathbf{q} + \mathbf{Q}, \mathbf{q}, i\omega_m) & \chi^{\rho\rho}(\mathbf{q} + \mathbf{Q}, i\omega_m) \end{pmatrix}, \quad (37)$$

$$\hat{U} = \begin{pmatrix} 2U & 0 \\ 0 & -2U \end{pmatrix}. \quad (38)$$

Here

$$\begin{aligned} \chi^{z\rho}(\mathbf{q}, \mathbf{q} + \mathbf{Q}, i\omega_m) &= \frac{1}{N} \int_0^{1/T} d\tau e^{i\omega_m \tau} \langle T_\tau S^z(\mathbf{q}, \tau) \rho(-\mathbf{q} - \mathbf{Q}, 0) \rangle, \quad (39) \\ \chi^{\rho z}(\mathbf{q} + \mathbf{Q}, \mathbf{q}, i\omega_m) &= \frac{1}{N} \int_0^{1/T} d\tau e^{i\omega_m \tau} \langle T_\tau \rho(\mathbf{q} + \mathbf{Q}, \tau) S^z(-\mathbf{q}, 0) \rangle, \quad (40) \end{aligned}$$

$$\begin{aligned} \chi^{\rho\rho}(\mathbf{q} + \mathbf{Q}, i\omega_m) &= \frac{1}{N} \int_0^{1/T} d\tau e^{i\omega_m \tau} \langle T_\tau \rho(\mathbf{q} + \mathbf{Q}, \tau) \rho(-\mathbf{q} - \mathbf{Q}, 0) \rangle, \quad (41) \end{aligned}$$

and $\hat{\chi}_0$ denotes a bare susceptibility matrix where each element is given by a simple bubble diagram. Setting $i\omega_m = 0$ and $\mathbf{q} = \mathbf{Q}$, we obtain $\chi^{zz}(\mathbf{Q}, 0)$, which reproduces Eq. (30). That is, the effect of $\frac{\partial n}{\partial h}$ is taken into account diagrammatically by considering the contribution from the density-density interaction such as $\chi^{z\rho}$, $\chi^{\rho z}$, and $\chi^{\rho\rho}$.

The diagrammatic method is formulated in the grand-canonical ensemble. Hence it is natural that we can successfully reproduce both results $\chi_\mu(\mathbf{0})$ [Eq. (19)] in the ferromagnetic phase and $\chi_\mu(\mathbf{Q})$ [Eq. (30)] in the antiferromagnetic phase. A remaining problem is how to reproduce $\chi_n(\mathbf{0})$ [Eq. (17)] and $\chi_n(\mathbf{Q})$ [Eq. (23)] obtained at a fixed density in terms of the diagrammatic method. As we have shown explicitly in Secs. III and IV, the longitudinal magnetic susceptibility at a fixed density is different from that at a fixed chemical potential in the magnetically ordered phase. Given that the density is usually fixed in the actual material, it is an important problem to find a general recipe to compute the magnetic susceptibility in the ordered phase at a fixed density.

VI. CONCLUDING REMARKS

We have studied longitudinal magnetic susceptibility by employing the two-dimensional Hubbard model. In the magnetically ordered phase, the spin rotational symmetry is broken, and thus μ and n acquire a linear term in a magnetic field when the field is applied parallel to the direction of the magnetic moment. Because of this effect, a careful analysis is required: the longitudinal magnetic susceptibility becomes different when computed at a fixed density and a fixed chemical potential. We have provided the correct expressions (17) and (23) at a fixed density and Eqs. (19) and (30) at a fixed chemical potential in both ferromagnetic and antiferromagnetic states. It should be noted that the susceptibility obtained at a fixed chemical potential (density) cannot be applied to the system with a fixed density (chemical potential) even though the chemical potential (density) is tuned to reproduce the correct density (chemical potential).

While we have exemplified our issue by employing the two-dimensional Hubbard model in the RPA, we believe that our conclusions do not depend on the choice of models, dimensions, lattices, and approximations even beyond the RPA. This consideration is based on thermodynamics. As in the case of the relation between specific heat at constant volume and that at constant pressure, we can derive the following relation from the thermodynamic principle:

$$\chi_n = \chi_\mu + \left. \frac{\partial n}{\partial h} \right|_\mu \left. \frac{\partial \mu}{\partial h} \right|_n. \quad (42)$$

In addition, one can easily show that the second term in Eq. (42) becomes negative-semidefinite and thus $\chi_n \leq \chi_\mu$. This is because $\left. \frac{\partial n}{\partial h} \right|_\mu \left. \frac{\partial \mu}{\partial h} \right|_n = -\left(\left. \frac{\partial n}{\partial h} \right|_\mu \right)^2 \left. \frac{\partial \mu}{\partial n} \right|_h$, and the stability of the thermodynamic potentials indicates that $\left. \frac{\partial \mu}{\partial n} \right|_h$ should be positive-semidefinite. Our obtained results in Figs. 1, 3, 4, and 6 indeed satisfy Eq. (42) numerically, and we can also check analytically that Eqs. (17)–(20) and Eqs. (23), (24), (30), and (31) fulfill Eq. (42) in the ferromagnetic and the antiferromagnetic case, respectively. Moreover, Figs. 3 and 6 indeed show that $\left. \frac{\partial n}{\partial h} \right|_\mu$ and $\left. \frac{\partial \mu}{\partial h} \right|_n$ have opposite signs. The

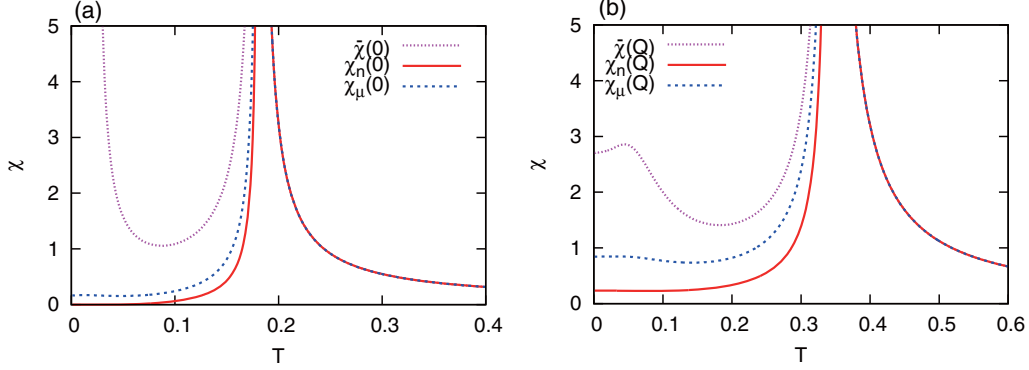


FIG. 8. Longitudinal magnetic susceptibility as a function of temperature for a fixed chemical potential in the ferromagnetic (a) and antiferromagnetic (b) phase. The chemical potential is chosen as $\mu = -1.83$ in (a) which reproduces $n = 0.2$ at $T = 0.15$, and as $\mu = 1.69$ in (b) where $n = 1.1$ at $T = 0.2$. The other parameters are $U = 3$, $t' = -0.45$ in (a) and $t' = -0.2$ in (b). For χ_n , the density is tuned at each temperature to reproduce the fixed chemical potential. The result of χ_μ is correct in the whole temperature region.

thermodynamic relation (42) is, however, not well recognized in the literature. In fact, the contributions from $\frac{\partial n}{\partial h}$ and $\frac{\partial \mu}{\partial h}$ are frequently missed, and an inappropriate formula such as Eqs. (15) and (21) is employed to compute the longitudinal magnetic susceptibility in the magnetically ordered phase [7,8,10–12].

As we have discussed in Secs. III (Fig. 1) and IV (Fig. 4), the enhancement of $\tilde{\chi}(0)$ [Eq. (15)] and $\tilde{\chi}(Q)$ [Eq. (21)] at low temperature inside the magnetic state is not a signal of some instability, but just an artifact due to the employment of the wrong susceptibility. Mathematically, this enhancement comes from a slight enhancement of $\chi_\uparrow + \chi_\downarrow$ [Eq. (16)] and $\chi^{(0)}(Q)$ [Eq. (22)] due to the development of magnetic order. This subtle change is removed by including the effect of $\frac{\partial \mu}{\partial h}|_{h \rightarrow 0}$ in Eqs. (17) and (23) or $\frac{\partial n}{\partial h}|_{h \rightarrow 0}$ in Eqs. (19) and (30) in the mean-field theory of the Hubbard model. However, it should be noted that in a more general situation, an enhancement of the susceptibility inside the magnetic phase could occur even if the effect of $\frac{\partial \mu}{\partial h}|_{h \rightarrow 0}$ (or $\frac{\partial n}{\partial h}|_{h \rightarrow 0}$) is correctly taken into account. For example, with decreasing temperature inside the magnetic phase, there could occur a tendency of a reentrant transition to a normal phase or a continuous transition to a different ordered phase.

Our obtained results are relevant to metallic systems whenever μ and n acquire a linear dependence on a magnetic field. The presence of the linear term in the field is easily recognized by symmetry. The ferromagnetic (antiferromagnetic) state is not symmetric with respect to the change of the field direction, namely $h \leftrightarrow -h$ when h is applied parallel to the direction of the uniform (staggered) magnetism. Therefore, we expect $\frac{\partial \mu}{\partial h} \neq 0$ and $\frac{\partial n}{\partial h} \neq 0$. On the other hand, for the uniform longitudinal magnetic susceptibility inside the antiferromagnetic state, we have $\frac{\partial \mu}{\partial h} = 0$ and $\frac{\partial n}{\partial h} = 0$, because the system is symmetric with respect to the change of the direction of a uniform field inside the antiferromagnetic phase. Another example is the case of the transverse field h_\perp : the system is symmetric with respect to the change of the field direction in both the ferromagnetic and antiferromagnetic state, leading to $\frac{\partial \mu}{\partial h_\perp} = 0$ and $\frac{\partial n}{\partial h_\perp} = 0$. Hence the transverse magnetic susceptibility is computed without considering possible contributions from $\frac{\partial \mu}{\partial h_\perp}$ and $\frac{\partial n}{\partial h_\perp}$ as seen in the literature [1,2,6–11].

For an insulating state, special care may not be needed, because $\frac{\partial n}{\partial h}$ should vanish in Eq. (42) due to the presence of a charge gap, and we obtain $\chi_n = \chi_\mu$. In fact, in an antiferromagnetic insulating state, we would have $E_k^+ > 0$ and $E_k^- < 0$ independent of \mathbf{k} . We can then easily obtain $n = 1$ [Eq. (10)] and $\frac{\partial n}{\partial h} = 0$ [Eq. (31)] at $T = 0$.

As a direct test of the present theory, we propose a susceptibility measurement in two different conditions, i.e., for a fixed density and a fixed chemical potential. Whereas the former condition is easily controlled in experiments, the latter condition may require the state-of-the-art technique in which a magnetic metal touches a charge reservoir, for example, exploiting a field-effect transistor. As seen in Figs. 1, 4, and 8, we predict a sizable difference between χ_n and χ_μ in a magnetically ordered phase.

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APPENDIX A: SYSTEM WITH A FIXED CHEMICAL POTENTIAL

The temperature dependence of the magnetic susceptibility is obtained for a fixed density in Figs. 1 and 4. Hence χ_n provides the correct result. While the density does not change as a function of temperature in actual materials, one can still consider a situation in which a system comes into contact with a charge reservoir. For example, a system is described as having several bands crossing the Fermi energy, and there is essentially only one active band with a large density of states. In that case, we may focus on such a band and invoke a condition of a fixed chemical potential. The temperature dependence of the magnetic susceptibility for a fixed chemical potential is shown in Figs. 8(a) and 8(b) in the ferromagnetic and antiferromagnetic case, respectively. These results are very similar to the results for a fixed density shown in Figs. 1 and 4. However, the correct result here is χ_μ , not χ_n .

- [1] T. Izuyama, D. J. Kim, and R. Kubo, *J. Phys. Soc. Jpn.* **18**, 1025 (1963).
- [2] K. Ueda, *J. Phys. Soc. Jpn.* **44**, 1533 (1978).
- [3] H. Mukuda, S. Shimizu, A. Iyo, and Y. Kitaoka, *J. Phys. Soc. Jpn.* **81**, 011008 (2012).
- [4] G. R. Stewart, *Rev. Mod. Phys.* **83**, 1589 (2011).
- [5] C. Pfleiderer, *Rev. Mod. Phys.* **81**, 1551 (2009).
- [6] H.-J. Lee and T. Takimoto, *J. Phys. Soc. Jpn.* **81**, 104704 (2012).
- [7] W. Rowe, J. Knolle, I. Eremin, and P. J. Hirschfeld, *Phys. Rev. B* **86**, 134513 (2012).
- [8] J. R. Schrieffer, X. G. Wen, and S. C. Zhang, *Phys. Rev. B* **39**, 11663 (1989).
- [9] A. V. Chubukov and D. M. Frenkel, *Phys. Rev. B* **46**, 11884 (1992).
- [10] A. P. Kampf, *Phys. Rep.* **249**, 219 (1994).
- [11] J. Knolle, I. Eremin, A. V. Chubukov, and R. Moessner, *Phys. Rev. B* **81**, 140506(R) (2010).
- [12] J. B. Sokoloff, *Phys. Rev.* **185**, 770 (1969).
- [13] P. A. Igoshev, M. A. Timirgazin, A. A. Katanin, A. K. Arzhnikov, and V. Y. Irkhin, *Phys. Rev. B* **81**, 094407 (2010).
- [14] T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer, Berlin, 1985).
- [15] In the numerator of Eq. (52) in Ref. [9], $+U[\chi_{\mathbf{Q}}^{z\rho}(\mathbf{q},\omega)]^2$ should be $-U[\chi_{\mathbf{Q}}^{z\rho}(\mathbf{q},\omega)]^2$.