### Landau-Ginzburg theory for phase transitions in magnetic-superconducting sandwiches\*

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The phase diagram of a magnetic-superconducting sandwich below the Curie temperature of the magnetic side is considered. A coupling parameter between the two order parameters is determined from the microscopic properties. For strong coupling the superconducting transition is first order. It is shown that one is always in the weak-coupling limit when the magnetic material has localized spins.

### I. INTRODUCTION

The possibility of coexistence of superconductivity and magnetism has recently attracted a new interest due to experiments on supercondueting systems containing magnetic impurities.<sup>1-3</sup> Entel and Klose' calculated the dependence of the second-order superconducting transition temperature on the concentration of magnetic impurities. Taking into account short-range magnetic correlations in the vicinity of the Curie temperature, they found that the system can go normal between two superconducting regions.

In this article we consider the proximity effect between a superconducting film and a magnetic slab. We use a Landau-Ginzburg theory to examine the competition between the magnetic and the superconducting order parameters in the magnetic slab. It is found that the supercondueting transition may be of the first or of the second order, depending on the strength of the coupling between the two order parameters.

To lowest order, the coupling between the spontaneous magnetization and the gap function is biquadratic. $4$  Landau-Ginzburg models with biquadratic coupling between two order parameters have already been discussed in the literature. Imry an eady been discussed in the interature.  $\lim_{x \to a}$  in  $e^{i}$  al.<sup>5</sup> considered a one-dimensional model including fluctuations and found that there is a possibility of mixed phases as well as of pseudo-firstorder transitions. Levin  $et$   $al$ <sup>6</sup> used a similar model to examine the incompatibility of BCS pairing and Peierls distortion. Although these models describe a homogenous system while we consider sandwiches, the main results are in a sense very similar. However, there is much more experimental flexibility in the proximity-effect situation because the coupling strength can be varied and adjusted by using different types of magnetic layers.

The parameters which determine the strength of

the coupling are assumed to be temperature independent. These parameters can be calculated from a microscopic theory.<sup>7</sup> They do indeed have a weak dependence on the temperature except in the vicinity of the Curie temperature  $T_K$ . This is due to the spin-flip scattering rate  $1/\tau_s$  appearing in them.  $1/\tau_s$  has an anomalous behavior near  $T_K$  because of long-range correlations among the spins of the magnetic impurities. $8$  As a result, the strength of the coupling is changed in the vicinity of  $T<sub>K</sub>$  and this affects the order of the transition.

In Sec. II we find the conditions which determine the order of the superconducting transition. This is done as follows: We construct a free-energy functional that yields Landau-Ginzburg equations for the magnetic-superconducting sandwich. We then look for a second-order superconducting transition. The second-order transition temperature  $T_w$  given by this solution is the one resulting from the usual Werthamer approximation,<sup>9</sup> modified by the presence of the ferromagnetic order. It is found that a second-order supercondueting transition exits only when the coupling between the two order parameters is weak.<sup>5</sup> We then show that when the coupling is strong,<sup>5</sup> there exists a solution with a finite superconducting order parameter at  $T_w$  which has a lower free energy. In this case a first-order transition occurs at a temperature higher than  $T_w$ .

Section III includes a discussion of the coupling strength in real systems, as a function of the ratio  $T_K/T_{c0}$ , where  $T_{c0}$  is the transition temperature of the bulk superconductor.

# II. DETERMINATION OF THE ORDER OF THE **TRANSITION**

We assume that the superconducting slab of the sandwich is of finite width  $d_s$  and occupies the region  $0 \le x \le d_s$  while the magnetic slab is semi-

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infinite and occupies the region  $-\infty < x < 0$ . The assumption that the magnetic slab is infinite is made in order to simplify the calculations; the results hold as long as the magnetic slab is much

thicker than the depth of penetration of Cooper pairs. The interface between the two slabs is at  $x = 0$ . The functional of the free energy of the sandwich is

$$
F = A_n \left\{ \int_{-\infty}^0 dx \left[ \left( \frac{d\Delta_n}{dx} \right)^2 + k_n^2 \Delta_n^2 + \frac{\beta}{2} \Delta_n^4 + \gamma \Delta_n^2 h^2 + ah^2 + \frac{b}{2} h^4 \right] - \lambda_n \Delta_n^2(0) \right\}
$$
  
+
$$
A_s \left\{ \int_0^{ds} dx \left[ \left( \frac{d\Delta_s}{dx} \right)^2 - k_s^2 \Delta_s^2 + \frac{\beta_s}{2} \Delta_s^4 \right] + \lambda_s \Delta_s^2(0) \right\}.
$$
 (1)

Here  $\Delta_n$ ,  $\Delta_s$  are the superconducting order parameters in the magnetic ("normal") and the superconducting slabs, respectively, and

$$
\lambda_n = \frac{1}{\Delta_n} \frac{d\Delta_n}{dx} \bigg|_{0^-}, \quad \lambda_s = \frac{1}{\Delta_s} \frac{d\Delta_s}{dx} \bigg|_{0^+}.
$$
 (2)

The terms containing  $\lambda_n$ ,  $\lambda_s$  in Eq. (1) describe the surface energy which has to be included in discussing a finite slab. It is convenient to separate them in this way. These terms, as well as the boundary conditions at  $x = 0$  will be discussed below.  $h$  is the magnetic order parameter. We assume that  $h$  is position dependent only through its coupling to  $\Delta_n$ , i.e., that the magnetic coherence length is short compared to  $k_n^{-1}$ . The coupling between  $\Delta_n$  and h is biquadratic<sup>4</sup> with  $\gamma > 0$ . In the absence of magnetic order,  $k_n^{-1}$  gives a measure of the depth of penetration of Cooper pairs into the normal metal (see Hauser et  $al$ .<sup>9</sup>).  $k_s^{-1}$  gives the effective variation range of  $\Delta_s$  on the S side.<sup>9</sup> The parameters  $\beta$  and  $\beta_s$  are positive and assumed to be independent of the temperature. We also assume units such that

$$
a = T/T_K - 1,\t\t(3)
$$

where  $T_K$  is the Curie temperature of the magnetic side.  $A_n$  and  $A_s$  are constants.

The boundary conditions satisfied by  $\Delta_n$ ,  $\Delta_s$  $are^9$  (i)

$$
\left. \frac{d\Delta_n}{dx} \right|_{x \to -\infty} = 0, \left. \frac{d\Delta_s}{dx} \right|_{x = t_s} = 0;
$$
\n(4)

(ii)  $\Delta/NV$  is continuous at  $x=0$ , where N is the density of states at the Fermi level and  $V$  is the BCS interaction potential; (iii)  $(D/V)$  ( $d\Delta/dx$ ) is continuous at  $x = 0$ , where D is the diffusion coefficient of the electrons. From the last two conditions we obtain

$$
\lambda_n = (D_s N_s / D_n N_n) \lambda_s \,. \tag{5}
$$

The terms  $\lambda_n \Delta_n^2(0)$  and  $\lambda_s \Delta_s^2(0)$  in (1) are needed in order that the variation of  $F$  will give the Landau-Ginzburg equations for  $\Delta_n$ ,  $\Delta_s$ . These "surface energy" terms describe the influence of the superconducting metal on the normal metal and vice versa.

The variation of F with respect to  $\Delta_n$ ,  $\Delta_s$ , and h, together with  $(2)$ ,  $(4)$  gives

$$
\frac{d^2\Delta_s}{dx^2} = -k_s^2 \Delta_s + \beta_s \Delta_s^3, \qquad (6)
$$

$$
\frac{d^2\Delta_n}{dx^2} = k_n^2 \Delta_n + \beta \Delta_n^3 + \gamma \Delta_n h^2,
$$
\n(7)

$$
h(a + \gamma \Delta_n^2 + bh^2) = 0.
$$
 (8)

Equations (6), (7) can be derived from a microscopic theory. Such a derivation gives explicit expressions for the parameters, and especiall their dependence on  $1/\tau_s$ .<sup>7</sup> Equation (8) can be deduced from a calculation similar to the one of Gorkov and Rusinov, $4$  including spin-spin correlations of the magnetic impurities.

We consider Eqs.  $(6)-(8)$  at temperature below  $T_K$ , i.e.,  $a = -|a|$ . Since the penetration depth of Cooper pairs into the normal metal is finite, it is clear that  $h(x)$  cannot vanish everywhere on the normal side. However, the value of  $h$  at the interface  $x = 0$ ,  $h(0)$  will in general be different from its bulk value  $(h^2 = |a|/b)$  and may even vanish. The only case where  $h^2(0) = |a|/b$  is when  $\Delta_n = \Delta_s = 0$ . When there are, at a certain temperature, and for a certain choice of the parameters  $k_n$ ,  $\lambda_n$ ,  $\beta$ ,  $b$ , and  $\gamma$  several possibilities for the values of  $h(0)$  and  $\Delta_n(0)$ , the system will choose the possibility which gives the lowest  $F$ .

We shall first look for the temperature  $(T_w)$  at which  $\Delta$  can go through a continuous second-order transition. It will be shown that such a transition occurs only when the coupling between  $\Delta_n$  and h is weak,<sup>5</sup>

$$
\gamma^2 < \beta b \tag{9}
$$

In the strong coupling case,

$$
\gamma^2 > \beta b,\tag{10}
$$

we shall show that at  $T_w$  the lowest F is obtained for a solution with  $h(0) = 0$  and a finite  $\Delta_n(0)$ . Thus at a certain temperature higher than  $T_w$ , the sandwich must undergo a first-order transition. In the limiting case

$$
\gamma^2/\beta b = 1 \tag{11}
$$

and at  $T_w$  the solution changes from a secondorder transition solution to a first-order one. Thus when (11) holds,  $T_w$  is a tricitical point.

We begin by classifying the types of solutions that are possible for  $\Delta_n(0)$  and  $h(0)$ . There are two different situations occurring on the normal side at a certain temperature. Either  $h(x)$  vanishes beyond some point  $-|x_0|$  ( $x_0$  may be zero), or  $h(0) \neq 0$ . In the first case

$$
\Delta_n^2(-\left|x_0\right|) = |a| / \gamma. \tag{12}
$$

In this case the first integral of  $(7)$ , using  $(8)$ gives

$$
\left(\frac{d\Delta_n}{dx}\right)^2 - k_n^2 \Delta_n^2 - \frac{\beta}{2} \Delta_n^4 - \frac{\gamma |a|}{b} \Delta_n^2 + \frac{\gamma^2}{2b} \Delta_n^4 = \text{const} = 0, -\infty < x < -|x_0|
$$
\n(13)

$$
\left(\frac{d\Delta_n}{dx}\right)^2 - k_n^2 \Delta_n^2 - \frac{\beta}{2} \Delta_n^4 = \text{const} = \left(\lambda_n^2 - k_n^2\right) \Delta_n^2(0) - \frac{\beta}{2} \Delta_n^4(0),
$$
  

$$
- |x_0| < x < 0.
$$

The constants in the first and second equations are determined by the values of  $\Delta_n$ ,  $d\Delta_n/dx$  at  $x = -\infty$  and  $x = 0$ , respectively. In the second case, where  $h(0) \neq 0$ , the first integral of (7) is

$$
\left(\frac{d\Delta_n}{dx}\right)^2 - k_n^2 \Delta_n^2 - \frac{\beta}{2} \Delta_n^4 - \frac{\gamma |a|}{b} \Delta_n^2 + \frac{\gamma^2}{2b} \Delta_n^4 = 0,
$$
  

$$
-\infty < x < 0.
$$
  
(14)

It is convenient to transform to new variables

$$
h^{2}(0) = (|a|/b)g,
$$
  
\n
$$
\Delta_{n}^{2}(0) = (K/\beta)f, \quad K = \lambda_{n}^{2} - k_{n}^{2}.
$$
\n(15)

There are three types of solutions for <sup>g</sup> and f.

Equations (8) and (14) have two types of solutions:  $(i)$ 

$$
f=0, \quad g=1,\tag{16}
$$

 $(ii)$ 

$$
f = \frac{2(1 - B^2)}{1 - A^2 B^2}, \quad g = \frac{1 - A^2 (2 - B^2)}{1 - A^2 B^2}.
$$
 (17)

Equations (12) and (13) have a solution of one type: (iii)

$$
(f-1)^2 = \frac{A^2 - B^2}{A^2}, \quad g = 0.
$$
 (18)

$$
A^{2} = \frac{\gamma K}{\beta |a|}, \quad B^{2} = \frac{\gamma |a|}{bK}, \quad A^{2} B^{2} = \frac{\gamma^{2}}{\beta b}.
$$
 (19)

The product  $A^2B^2$  is temperature independent and its magnitude compared to 1 determines the strength of the coupling. (The case where  $A^2 = B^2 = 1$ is treated separately later on. )

At the second-order transition the solution is of type (i). Just below second-order transition the solution of type (ii) must hold and at the transition it has to go continuously into the solution of type (i). We now show that this can occur only in the weak coupling case and that the second-order transition temperature is the one obtained by Werthamer approximation.<sup>9</sup> Near the secondorder transition the cubic term in (6) may be neglected. Then by solving (6), using (4), we obtain

$$
\lambda_s = k_s \tan k_s \, d_s \,. \tag{20}
$$

It can easily be verified that this approximate solution causes the free energy of the superconducting side,  $F_s$  [the second term on the righthand side of  $(1)$ , to vanish. Now consider the free energy of the normal side,  $\overline{F}_n^{}[$  the first tern on the right-hand side of  $(1)$ . For the solution of type (i),  $F_n = F_n^{(i)}$  is just the magnetic free energy  $F_{\mu}$ :

$$
F_n^{(i)} = F_M \equiv A_n \int_{-\infty}^0 \left( -\frac{|a|^2}{2b} \right). \tag{21}
$$

The free energy of the normal side corresponding to the solution of type (ii) is  $F_n^{(i)}$ :

(14)  
\n
$$
F_n^{(11)} = F_M + A_n \left[ 2 \int_{-\infty}^0 dx \left( k_n^2 \Delta_n^2 + \frac{\gamma}{b} \right) |a| \Delta_n^2 + \frac{\beta}{2} \Delta_n^4 - \frac{\gamma^2}{2b} \Delta_n^4 \right) - \lambda_n \Delta_n^2(0) \right]
$$
\n
$$
= F_M + A_n \left[ 2 \int_0^{\Delta_n(0)} d\Delta_n \left( k_n^2 \Delta_n^2 + \frac{\gamma}{b} |a| \Delta_n^2 + \frac{\beta}{2} \Delta_n^4 - \frac{\gamma^2}{2b} \Delta_n^4 \right)^{1/2} - \lambda_n \Delta_n^2(0) \right],
$$
\n
$$
= \sum_{n=0}^{\infty} \Delta_n^4 - \frac{\gamma^2}{2b} \Delta_n^4 \Delta_n^2 - \lambda_n \Delta_n^2(0) \right],
$$
\n(22)

where we used (14). Performing the integration and using  $(15)$ ,  $(17)$ , and  $(19)$  we obtain

$$
F_n^{(11)} = F_M - A_n \frac{2K^{3/2}}{\beta} \frac{1}{3(1 - A^2 B^2)}
$$

$$
\times \left[ \frac{\lambda_n}{\sqrt{K}} - \left( B^2 - 1 + \frac{\lambda_n^2}{K} \right)^{1/2} \right]^2
$$

$$
\times \left[ \frac{\lambda_n}{\sqrt{K}} + 2 \left( B^2 - 1 + \frac{\lambda_n^2}{K} \right)^{1/2} \right].
$$
 (23)

Here Thus  $F_n^{(1)}$  is lower than  $F_n^{(1)}$  only in the weak cou-

pling case where  $A^2B^2 \leq 1$ . From (23) we see that the second-order transition temperature  $T_w$  is at  $B^2 = 1$ , i.e., from (19)

$$
\gamma |a|/b = K = \lambda_n^2 - k_n^2. \tag{24}
$$

Equations (5), (20), and (24) are just Werthamer's equations<sup>9</sup> for the superconducting second-order transition. In the absence of magnetic order  $\lambda_n = k_n$  and we obtain the usual solution.<sup>9</sup> (Note that the normal side of the sandwich is semi-infinite.) When there is a magnetic order, the depth of penetration of Cooper pairs into the normal metal is reduced because of the  $\gamma |a| /b$  in (24).

We now consider a system in which the coupling is not weak  $(A^2B^2 \ge 1)$ , at the temperature  $T_w$ , is not weak  $(A^B B^2 \ge 1)$ , at the temperature  $T_w$ ,<br>where  $B^2 = 1$ . We shall first compare  $F_n^{(1)}$ , given by (23), to  $F_n^{(iii)}$ . We then examine the solution of the special case  $A^2 = B^2 = 1$  and finally discuss the behavior of  $\Delta_n$ .

The calculation of  $F_n^{(111)}$  is described in the Appendix. The result is

$$
F_n^{(111)} = F_M + A_n 2 \frac{K^{3/2}}{\beta} \left\{ \int_0^{1/A} du \left[ \left( \frac{1}{2} (1 - A^2) u^4 + \frac{\lambda_n^2}{K} u^2 \right)^{1/2} - \frac{\lambda_n}{\sqrt{K}} u \right] + \int_{1/A}^{\sqrt{f}} du \left[ \left( \frac{1 - A^2}{2A^2} + \frac{1}{2} (u^2 - 1)^2 + \frac{\lambda_n^2}{K} u^2 \right)^{1/2} - \frac{\lambda_n}{\sqrt{K}} u \right] \right\}, \quad B^2 = 1, \quad A^2 > 1.
$$
 (25)

We show in the Appendix that the second term on the right-hand side of Eq. (25) is negative. From (23) we have

$$
F_n^{(i)} = F_n^{(i)} = F_M, \quad B^2 = 1, \quad A^2 > 1.
$$
 (26)

Thus in the strong coupling case  $(A^2B^2 > 1)$ , solution (iii), with  $f > 1$  has the lowest  $F_n$  at  $T_w$ . For this solution h vanishes at a certain point  $-|x_0|$  on the normal side. One can show that when  $A^2$  becomes smaller and approaches 1 from above, the point  $x_0$  approaches the interface. At  $A^2 = B^2 = 1$ , we have from (12), (15), (18), and (19) that  $f = 1$ , and  $x_0 = 0$ . The case  $A^2 = B^2 = 1$  is the intermediate case between the strong and weak coupling cases. In this case Eqs. (16), (17) [solutions (i) and (ii)] do not hold since from  $(14)$ ,  $(15)$ , and  $(19)$  we have

$$
[2(1-B^2) - (1-A^2B^2)f]f = 0
$$
 (27)

and thus for  $A^2 = B^2 = 1$ , f can have any value. From (8) we find that in this case

$$
1 - f = g \tag{28}
$$

and therefore  $f$  and  $g$  can have any value between 0 and 1. All the solutions of (28) have the same  $F_n$ :

$$
F_n = F_M, \quad A^2 = B^2 = 1 \tag{29}
$$

from (23) or from the Appendix.

Now consider the behavior of  $\Delta_n(0)$  at  $B^2 = 1$ . From (15) we see that a finite value for  $f$  implies that  $\Delta_n^2(0)$  is finite, as long as K differs from zero. The connection between  $\Delta_n$  and K results from the solution of  $(7)$ . When h vanishes at a certain point on the magnetic side,  $\lambda_n$  (and therefore K) is determined by the first two terms on the right-hand side of (7). If  $\Delta_n$  is sufficiently large so that the

 $\beta \Delta_n^3$  term is important,  $\lambda_n \neq k_n$  and therefore K differs from zero. On the other hand, when  $\Delta_n$ is small and the  $\beta \Delta_n^3$  term is negligible,  $\lambda_n$  approaches  $k_n$  and consequently  $K \rightarrow 0$ . From these remarks we conclude that the behavior of  $\Delta_n$  at  $B^2 = 1$  is as follows: For  $A^2 < 1$ ,  $f = 0$  and K is finite  $\lceil$  Eq. (24) and the transition is of the second order, i.e.,  $\Delta_n(0) = 0$ . As  $A^2$  passes 1 and increases, the transition cannot be of the second order and thus the value of  $\Delta_n(0)$  at  $B^2 = 1$  starts to increase. For  $A \ge 1$ ,  $\Delta_n$  is still very small and therefore although f is finite,  $K \rightarrow 0$ , which is consistent with the fact that  $\Delta_n$  is very small. As  $A^2$  becomes much larger than 1,  $\Delta_n$  becomes larger so that the  $\beta \Delta_n^3$  term in (7) is not negligible and  $K$  differs from zero. A finite value for  $\Delta_n$  implies of course that  $\Delta_n$  is finite. This means that  $F_s$  must be negative (whereas at a secondorder transition it is zero). Note also that since  $h(0) = 0$ ,  $\lambda_n$  and consequently  $\lambda_s$  are smaller than in the case where  $h(0) \neq 0$  [i.e., a solution of type (ii)]. This is because a finite  $h(0)$  reduces the depth of penetration of Cooper pairs. We conclude that in the strong coupling case and at  $B^2 = 1$ ,  $\Delta_n(0)$  is finite and therefore a first-order transition will occur at a certain temperature higher than  $T_w$ . At  $A^2 = B^2 = 1$ the transition is changed from first order to second order and this point is a tricritical point.

#### III. DISCUSSION

We have considered a superconducting-magnetic sandwich below the Curie temperature using a Landau-Ginzburg theory, and have shown that the competition between ferromagnetism and superconductivity in the normal side of the sandwich may lead to a first-order superconducting transition. The order of the transition is determined by the strength of the coupling between the two order parameters. In the weak coupling case where  $\gamma^2/\beta b < 1$ , a second-order transition is predicted and the transition temperature  $T_w$  is the dicted and the transition temperature  $I_W$  is the one given by the Werthamer approximation.<sup>9</sup> In the strong coupling case,  $\gamma^2/\beta b > 1$ , and at  $T_w$ , the magnetic order parameter vanishes at the interface and  $\Delta_n(0)$  is finite. Thus at a certain temperature higher than  $T_w$  a first-order transition will occur. When  $\gamma^2/\beta b = 1$  there is a tricritical point at  $B^2 = 1$ .

At  $T=T_w$  ( $B^2=1$ ) the coupling parameter may be written from (19) as

$$
\frac{\gamma^2}{\beta b} = \frac{K^2/\beta}{|a|^2/b}, \quad B^2 = 1.
$$
 (30)

This shows that the coupling is qualitatively the ratio between the densities of the superconducting free energy and magnetic free energy of the normal side. The numerator cannot be larger than the density of the free energy of the supereonducting side. Therefore

$$
\gamma^2/\beta b \le \Delta F_s / \Delta F_M,\tag{31}
$$

where  $\Delta F_s$ ,  $\Delta F_u$  are the condensation energies of the superconducting and the magnetic order parameters, respectively.

The inequality (31) implies that it is virtually impossible to attain strong coupling conditions. For a localized spin ferromagnet one has

$$
\frac{\Delta F_s}{\Delta F_M} \sim \frac{T_c^2/T_F}{T_K}
$$

and one would require  $T_c/T_K$  large enough to compensate for the factor  $T_c/T_F$  on the right-hand side, to attain strong coupling.

The situation would be quite different for an itinerant electron ferromagnet. One then has

$$
\Delta{F}_{\ \ M}^{} \sim\ {T\ ^2_K}/{T}
$$

so that the Fermi temperature cancels:

$$
\frac{\Delta F_s}{\Delta F_M} \sim \frac{T_c^2}{T_K^2}
$$

 $\Delta F_M$  1  $_K$ <br>which can be large.<sup>10</sup> It would be interesting to investigate this possibility more closely and in particular to check if the approximations we made can also be justified in this case.

In the theory presented here the magnetic ordering is treated in the mean- field approximation (MFA). Its explicit dependence on position as well as its fluctuations in the vicinity of the Curie temperature are neglected. The theory can be improved by calculating  $\gamma$  and  $\beta$  from a microscopic theory.<sup>7</sup> In this way the behavior of the coupling  $\gamma^2/\beta b$  near the Curie temperature can be obtained.  $\gamma$  and  $\beta$  depend on  $1/\tau_{\gamma}$ . the spin-flip scattering rate<sup>8</sup> which has a maximum at the Curie temperature  $T_K$ . The increase in  $1/\tau_s$  in the vicinity of  $T_K$  tends to lower the value of  $\gamma^2/\beta b$ ,<sup>7</sup> and thus to make the coupling weaker. This can change the quantitative details of the transition but can have no drastic effects when the coupling is weak in the first place.

# APPENDIX: FREE ENERGY OF THE MAGNETIC SIDE OF A SOLUTION OF TYPE (iii)

Solution (iii) describes a situation where the magnetic order vanishes at the interface  $x = 0$ . In this case

$$
(f-1)^2 = (A^2 - B^2)/A^2.
$$
 (A1)

Therefore this solution exists only for  $A^2 > B^2$ . Since  $h(0) = 0$ , this implies that h vanishes at a certain point  $-|x_0|$  in the magnetic slab  $(|x_0|)$  may be zero). The free energy  $F_n^{(111)}$  of the magnetic  $\textsf{side is [from (12), (13)]}$ 

$$
F_n^{(111)} = A_n \left\{ -\lambda_n \Delta^2(0) + \int_{-\infty}^{-|x_0|} dx \left[ 2 \left( k_n^2 \Delta_n^2 + \frac{\beta}{2} \Delta_n^4 \right) + \gamma \Delta_n^2 h^2 - |a| h^2 + \frac{b}{2} h^4 + \frac{\gamma |a|}{b} \Delta_n^2 - \frac{\gamma^2}{2b} \Delta_n^4 \right] \right\} + \int_{-|x_0|}^{0} dx \left[ 2 \left( k_n^2 \Delta_n^2 + \frac{\beta}{2} \Delta_n^4 \right) + \frac{|a|^2}{2b} \right] \right\}.
$$
 (A2)

At the point  $-|x_0|$  we have

$$
\Delta_n^2(-\left| x_0\right| )=\left| a\right| / \gamma.
$$

Using Eqs. (8), (12), (13), (15), and (19), Eq. (A2) becomes

$$
F_n^{(111)} = A_n \left( \int_{-\infty}^0 dx \left( -\frac{|a|^2}{2b} \right) + 2 \frac{K^{3/2}}{\beta} \left\{ \int_0^{1/A} du \left[ \left( -u^2 + \frac{1}{2} u^4 + B^2 u^2 - \frac{1}{2} A^2 B^2 u^4 + \frac{\lambda_n^2}{K} u^2 \right)^{1/2} - \frac{\lambda_n}{\sqrt{K}} u \right] \right\}
$$
  
+ 
$$
\int_{1/A}^{\sqrt{f}} du \left[ \left( \frac{B^2}{2A^2} - u^2 + \frac{1}{2} u^4 + \frac{\lambda_n^2}{K} u^2 \right)^{1/2} - \frac{\lambda_n}{\sqrt{K}} u \right] \right). \tag{A3}
$$

The first term on the right-hand side is just the magnetic free energy  $F_{M}$ . We now show that when  $B^2 = 1$ , (i.e., at  $T = T_w$ ) and in the strong coupling case  $A^2B^2>1$ , i.e.,  $A^2>1$ , the integrals give a negative contribution. For  $B^2 = 1$  the integrals are

$$
\int_0^{1/A} du \left[ \left( \frac{1}{2} u^4 (1 - A^2) + \frac{\lambda_n^2}{K} u^2 \right)^{1/2} - \frac{\lambda_n}{\sqrt{K}} u \right] + \int_{1/A}^{\sqrt{f}} du \left[ \left( \frac{1 - A^2}{2A^2} + \frac{1}{2} (u^2 - 1)^2 + \frac{\lambda_n^2}{K} u^2 \right)^{1/2} - \frac{\lambda_n}{\sqrt{K}} u \right].
$$
\n(A4)

The two integrands are never positive in the region of integration. The first integrand vanishes at  $u = 0$  and is negative for  $0 \le u \le 1/A$  since  $A^2 > 1$ . At  $u = 1/A$  the two integrands are equal. From

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Eq. (A1) it seems as though  $f$  has two values

$$
f = 1 \pm \left(\frac{A^2 - B^2}{A^2}\right)^{1/2}.
$$
 (A5)

However, for  $B^2 = 1$  and  $A^2 > 1$  only the solution with the + sign is valid. This is because in order that the + sign is valid. This is because in order that  $h(0) = 0$ ,  $A^2 f \ge 1$  [from Eqs. (8), (15), and (19)]. For  $A<sup>2</sup>$  1, this condition is not satisfied by the other solution of (A5). As a result the second integrand in (A4) is also negative and vanishes at  $u = \sqrt{f}$ . Thus the two integrals in (A3) are negative for  $B^2=1$  in the strong coupling case.

When  $A^2 = B^2 = 1$ ,  $x_0 = 0$  and  $f = 1$  [from (12), (18), and (19)]. In this case the second integral in (A4) disappears while the first integrand vanishes. Thus for  $A^2 = B^2 = 1$ ,  $F_n^{(iii)} = F_{M}$ .

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