

Parameters  $\kappa_1$ ,  $\kappa_2$ , and  $\kappa_3$  in magnetic superconductors

H. Matsumoto and H. Umezawa

*Department of Physics, University of Alberta, Edmonton, Alberta, Canada, T6G 2J1*

M. Tachiki

*The Research Institute for Iron, Steel, and Other Metals, Tohoku University, Sendai, Japan*

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The parametrization to describe the magnetic properties of superconductors,  $\kappa_1(t)$ ,  $\kappa_2(t)$ , and  $\kappa_3(t)$ , is extended to the case of magnetic superconductors in such a way that the effects of average polarization are subtracted. In this extension,  $\kappa_1(t)$ ,  $\kappa_2(t)$  approach the same value  $\kappa$  in the limit  $t \rightarrow 1$ . It was shown that when the electromagnetic interplay is the main mechanism in the magnetic superconductors,  $\kappa_1(t)$ ,  $\kappa_2(t)$  [and  $\kappa_3(t)$ ] are related to the nonmagnetic  $\kappa_i(t)$ 's by the simple scaling rule near the critical temperature. In this case, it is also shown that the magnetization curve can be obtained by a suitable scaling of fields and  $\kappa$  from the nonmagnetic ones. Especially, types of the magnetization curve are classified not in terms of  $\kappa$  but in terms of the scaled  $\kappa'$  ( $\equiv \kappa/[1+4\pi\chi(t)]^{1/2}$ , where  $\chi(t)$  is the static spin susceptibility) near the critical-temperature region. Several relations related to the magnetic properties are also presented. The practical usefulness of our formulation lies in the fact that it presents a simple way of obtaining the magnetization curves of magnetic superconductors from those of nonmagnetic ones.

## I. INTRODUCTION

Recently, many rare-earth ternary compounds such as  $RMo_6S_8$  and  $RRh_4B_4$  have been found to show superconductivity as well as magnetism.<sup>1-5</sup> These compounds are well described by the model<sup>6-8</sup> where the localized spins of rare-earth ions interact with the superconducting electrons mostly through the electromagnetic interactions; the spin-dependent interaction (so-called  $s$ - $f$  interaction) is considered to be weak.<sup>1,9-11</sup> Using this model we have made several theoretical predictions,<sup>6,7,12-16</sup> some of which have been favorably supported by experiments.<sup>17-19</sup>

In these materials, both superconductive and magnetic properties are intertwined in the observed phenomena. Therefore it is desirable to have some idea how to separate the properties of the rare-earth ions (the magnetic system) and those of the conduction electrons (the superconducting system).

If two systems are independent, the magnetic system is described by the spin  $J$  of the rare-earth ion, the magnetic moment  $g\mu_B JN$ , the Curie temperature  $T_m$ , the Curie constant  $C [ \equiv (g\mu_B)^2 J(J+1)N/3k_B ]$  and the stiffness constant  $D$  of the exchange interaction, while the superconductive system is described by the magnetic unit  $\phi/\lambda_L^2(0)$  [ $\phi$ : the unit flux  $hc/2e$ ,  $\lambda_L(0)$ ; the London penetration depth at  $T=0$ ], the critical temperature  $T_c$ , the Landau parameter  $\kappa_B = \lambda_L(0)/\xi(0)$  [ $\xi(0)$  is the coherent length at  $T=0$ ], and the BCS coupling constant  $VN(0)$ . The  $\kappa_B$  is related to the Landau parameter  $\kappa$  at  $T=T_c$ : their relation depends on impurity and in the pure limit,  $\kappa = 0.96\kappa_B$ . Because of the interplay

between the superconductivity and magnetism, the above fundamental parameters are intermingled in a complex manner.

In case of the analysis of the magnetic properties of nonmagnetic superconductors, the usefulness of parameters  $\kappa_1(t)$ ,  $\kappa_2(t)$ , and  $\kappa_3(t)$ , defined by<sup>20</sup>

$$H_{c2}(t)/H_c(t) = \sqrt{2}\kappa_1(t) \quad , \quad (1.1)$$

$$\left. \frac{\partial(4\pi M)}{\partial H} \right|_{H=H_{c2}} = \frac{1}{\beta[2\kappa_2^2(t)-1]} \quad , \quad (1.2)$$

$$H_c(t)/[\phi/\lambda_L^2(t)] = \kappa_3(t)/\sqrt{2}(2\pi) \quad , \quad (1.3)$$

(with  $t = T/T_c$ ), has been well established. Here  $\beta$  is a structure constant. They are particularly very useful in determining the Landau parameter  $\kappa_B$  (or  $\kappa$ ), since  $\kappa_1(t)$ ,  $\kappa_2(t)$ , and  $\kappa_3(t)$  approach the same value  $\kappa$  at  $t \rightarrow 1$ .

This paper aims at generalizing the parametrization  $\kappa_1(t)$ ,  $\kappa_2(t)$ , and  $\kappa_3(t)$  in a suitable form for the analysis of the magnetic superconductors. Briefly speaking, we generalize the definition of parameters by subtracting the average polarization effects of the localized spin from magnetic quantities. This parametrization helps us to put the observable quantities (such as magnetization, static susceptibility, etc.) in a simple form. This generalization will be presented in Sec. II in a model independent way. In the consideration in Sec. II, we do not specify the form of the interaction among the superconducting electron and the localized spins. The only assumption used there is that the superconductor is of the type II and that the phase transition at  $H_{c2}$  is of the second

order. Therefore the  $\kappa_i(t)$ 's are formulated on a quite general basis. The  $\kappa_1(t)$  and  $\kappa_2(t)$  approach the same value  $\kappa$  in the limit  $t \rightarrow 1$ .

In Sec. III it will be shown, by use of the boson method, that  $\kappa_i(t)$ 's at  $T$  near  $T_c$  are related to the nonmagnetic  $\kappa_i(t)$  by a simple scaling rule when the electromagnetic interaction is the main agent controlling the interplay of magnetism and superconductivity [that is, if  $s$ - $f$  interaction is negligibly weak or if  $s$ - $f$  interaction effect can be treated by renormalization of parameters such as  $\lambda_L$ ,  $\xi(0)$ , etc.]. In this case, it will be also shown that approximately the same shape of the magnetization curve as that of the nonmagnetic case is obtained by a suitable scaling of fields and  $\kappa$ . In other words, at  $T$  near  $T_c$ , the type of the magnetization curve is classified not by  $\kappa$ , but by a scaled  $\kappa' \equiv \kappa/[1 + 4\pi\chi_n(t)]^{1/2}$  [ $\chi_n(t)$ : static spin susceptibility of the normal state]. Since  $\chi_n(t)$  increases when

$T$  comes close to the magnetic transition temperature, this indicates that the transition from type II/2 to type II/1 or from type II to type I takes place with decreasing temperature. This has already been predicted by the numerical calculation in Ref. 6, and also has been observed by experiments.<sup>18</sup> It will be shown in Sec. III that  $\kappa_i(t) \rightarrow \kappa (i = 1, 2, 3)$  as  $t \rightarrow 1$ . Some of the results of Secs. II and III are obtainable also in the Ginzburg-Landau (GL) theory, which will be presented in the Appendix. Section IV is devoted to the concluding remarks.

## II. FREE ENERGIES AND THE GENERALIZATION OF $\kappa_i$ 's

Let us consider a general expression of free energy for magnetic superconductors. We start with a Hamiltonian

$$H(x) = \psi^\dagger(x) \epsilon \left[ -i \left( \nabla + \frac{ie}{\hbar c} \vec{A}(x) \right) \right] \psi(x) - \lambda \psi_1^\dagger(x) \psi_1^\dagger(x) \psi_1(x) \psi_1(x) + H_I(x) + \frac{1}{8\pi} \vec{B}(x)^2 - \vec{B}(x) \cdot \vec{M}(x) - \frac{1}{2} \vec{M}(x) \gamma_0 (-i \nabla) \vec{M}(x) \quad (2.1)$$

Here  $\vec{A}$  is the vector potential,  $\vec{B} (\equiv \nabla \times \vec{A})$  is the magnetic induction field,  $\vec{M}(x)$  is the magnetic moment given by

$$\vec{M}(x) = g \mu_B \sum_n \vec{S}_n \delta(\vec{x} - \vec{R}_n) \quad (2.2)$$

with  $\vec{S}_n$  being the localized spin and  $\vec{R}_n$  being the lattice point,  $\gamma_0$  is the spin-spin interaction mediated by all the interaction except the dipole interaction, and  $H_I(x)$  is the other interactions among conduction electrons and localized spins. When the  $s$ - $f$  interaction is effective,  $H_I$  should contain the interaction of the form  $I \psi^\dagger \vec{\sigma} \psi \cdot \vec{M}$ .

When the ground-state energy is evaluated, it is convenient to separate (2.1) into two parts; the magnetic energy  $E_m(x)$ ,

$$E_m(x) = \frac{1}{8\pi} \vec{B}^2(x) - \vec{B}(x) \cdot \vec{M}(x) - \frac{1}{2} \vec{M}(x) \gamma_0^s (-i \nabla) \vec{M}(x) \quad (2.3)$$

with  $\gamma_0^s$  being the effective spin-spin interaction modified by  $H_I$  in the superconducting state, and the electronic energy  $E_e(x)$  defined by

$$E_e(x) = \langle 0 | \psi^\dagger \epsilon \left[ -i \left( \nabla + \frac{ie}{\hbar c} \vec{A} \right) \right] \psi - \lambda \psi_1^\dagger \psi_1^\dagger \psi_1 \psi_1 + H_I(x) | 0 \rangle \quad (2.4)$$

which we write

$$E_e(x) = -W_0 - \frac{1}{2c} \vec{j}(\vec{A}_f) \cdot \vec{A}_f + E_{\text{core}}(x) \quad (2.5)$$

Here  $W_0$  is the condensation energy (i.e.,  $\vec{A} = 0$ ) and  $\vec{A}_f = \vec{A} - (\hbar c/e) \nabla f$  with  $f$  being half of the phase of the order parameter. In (2.5), the second term is the bilinear term of  $\vec{A}_f$  and  $E_{\text{core}}(x)$  includes all the higher-power terms. We have

$$\frac{4\pi}{c} \vec{j}(\vec{A}_f) = -\lambda_L^{-2}(t) c (-i \nabla) \vec{A}_f(x) \quad (2.6)$$

where  $c(-i \nabla)$  is a nonlocal kernel relating to the photon self-energy.<sup>21,22</sup>

The entropy of the system in the mean-field approximation is given by

$$TS(x) = TS_e + k_B T N \ln Z \left[ \frac{g \mu_B J}{k_B T} |\vec{H}_m(x)| \right] - \vec{H}_m(x) \cdot \vec{M}(x) \quad (2.7)$$

where  $S_e$  is the entropy of the electrons,

$$Z(y) = \sinh \left[ \frac{2J+1}{2J} y \right] / \sinh \left[ \frac{1}{2J} y \right] \quad (2.8)$$

and

$$\vec{H}_m(x) = \vec{B}(x) + \gamma_0^s (-i \nabla) \vec{M}(x) \quad (2.9)$$

Note that  $\vec{B}(x)$  and  $\vec{M}(x)$  satisfy the following equations:

$$\vec{\nabla} \times \vec{B}(x) = \frac{4\pi}{c} \vec{j}(\vec{A}_f) + 4\pi \vec{\nabla} \times \vec{M}(x) , \quad (2.10)$$

$$|\vec{M}(x)| = g \mu_B J N B_J \left[ \frac{g \mu_B J}{k_B T} |\vec{H}_m(x)| \right] . \quad (2.11)$$

Here  $B_J$  is the Brillouin function.

When the vortex lattice structure is specified and the vortex density  $n$  is given, one can calculate  $E_m(x)$  and  $E_e(x)$  in principle and then the Gibbs free-energy density under an applied field  $H$  is obtained as a function of  $n$ :

$$G_s(n) = \frac{1}{V} \int_V d^3x \left[ E_m(x) + E_e(x) - TS(x) - \frac{1}{4\pi} \vec{H} \cdot \vec{B}(x) \right] , \quad (2.12)$$

where  $V$  is the volume. Note that

$$\frac{1}{V} \int_V d^3x B(x) = n \phi . \quad (2.13)$$

It is convenient to calculate the average Gibbs free energy of the spin-system interacting average flux  $n\phi$ , since some polarization is induced by  $n\phi$ :

$$F_m(\gamma_0^s, n\phi) = \frac{1}{2} \gamma_0^s m^2(n) - k_B T N \times \ln Z_J \left[ \frac{g \mu_B J}{k_B T} [n\phi + \gamma_0^s m(n)] \right] \quad (2.14)$$

with

$$m(n) = g \mu_B J N B_J \left[ \frac{g \mu_B J}{k_B T} [n\phi + \gamma_0^s m(n)] \right] , \quad (2.15)$$

$$\frac{\partial}{\partial n \phi} F_m(\gamma_0^s, n\phi) = -m(n) . \quad (2.16)$$

Then we can write  $G_s(n)$  as

$$G_s(n) = \frac{n\phi}{8\pi} g(n) + F_m(\gamma_0^s, n\phi) - \frac{H_c^2}{8\pi} - \frac{n\phi}{4\pi} H . \quad (2.17)$$

The  $g(n)$  means the effective magnetic field due to vortices. The  $H_c$  is defined by  $W_0 + TS_e = H_c^2/8\pi$ . The form of  $H_c^2/8\pi$  and  $g(n)$  depend on the models and approximation methods.

The free energy of the normal state is obtained from (2.12) by putting  $\vec{j} = 0$ ,  $H_c^2/8\pi = 0$ ,  $E_{\text{core}} = 0$ , and  $\vec{B} = \vec{H} + 4\pi \vec{m}_n(H)$ , and by  $\gamma_0^s$  replacing  $\gamma_0^f$ . Here  $\gamma_0^f$  is the effective spin-spin interaction in the normal state. The polarization  $m_n(H)$  satisfies

$$m_n(H) = g \mu_B J N B_J \left[ \frac{g \mu_B J}{k_B T} [H + \gamma_0^f m_n(H)] \right] . \quad (2.18)$$

Here

$$\gamma^n = \gamma_0^f + 4\pi \quad (2.19)$$

and is parametrized as

$$\gamma^n = \frac{T_m}{C} . \quad (2.20)$$

The result is

$$G_n(H) = -\frac{H^2}{8\pi} + F_m(\gamma^n, H) , \quad (2.21)$$

where

$$F_m(\gamma^n, H) = \frac{1}{2} \gamma^n m_n(H)^2 - k_B T N \ln Z_J \left[ \frac{g \mu_B J}{k_B T} [H + \gamma^n m_n(H)] \right] . \quad (2.22)$$

The  $n$  dependence of the applied field  $H$  is obtained from  $(\partial/\partial n) G_s(n) = 0$ ,

$$H(n) = \frac{1}{2} \left[ 1 + n \frac{\partial}{\partial n} \right] g(n) - 4\pi m(n) . \quad (2.23)$$

At  $H_{c2}$ , the phase transition is of the second order, therefore the following conditions must be satisfied:

$$G_s(n_c) = G_n(H_{c2}) , \quad (2.24)$$

$$\left. \frac{\partial G_s}{\partial H} \right|_{H_{c2}} = \left. \frac{\partial G_n(H)}{\partial H} \right|_{H_{c2}} . \quad (2.25)$$

Since

$$\frac{\partial G_s}{\partial H} = -\frac{B_s}{4\pi} = -\frac{n\phi}{4\pi} , \quad (2.26)$$

$$\frac{\partial G_n}{\partial H} = -\frac{B_n}{4\pi} = -\frac{1}{4\pi} [H + 4\pi m_n(H)] , \quad (2.27)$$

the condition (2.25) reads as

$$n_c \phi = H_{c2} + 4\pi m_n(H_{c2}) . \quad (2.28)$$

Since  $\gamma_0^s = \gamma_0^f$  at  $H = H_{c2}$ , it follows from (2.15), (2.18), and (2.28) that

$$m(n_c) = m_n(H_{c2}) \quad (2.29)$$

and

$$n_c \phi + \gamma_0^f m(n_c) = H_{c2} + \gamma_0^f m_n(H_{c2}) . \quad (2.30)$$

Using the relations (2.29), (2.30), and  $\gamma_0^s = \gamma_0^f$  at  $H = H_{c2}$  and considering (2.17) and (2.21) with (2.19), we can rewrite the conditions (2.24) as

$$H_c^2 = n_c \phi [g(n_c) - n_c \phi] . \quad (2.31)$$

On the other hand (2.23) for  $H = H_{c2}$  gives

$$\frac{1}{2} \left[ 1 + n_c \frac{\partial}{\partial n_c} \right] [g(n_c) - n_c \phi] = 0 , \quad (2.32)$$

where (2.28) and (2.29) were used. It is convenient to rewrite these two relations as

$$g(n_c) = n_c \phi \left[ 1 + \left( \frac{H_c}{n_c \phi} \right)^2 \right], \quad (2.33)$$

$$n_c \frac{\partial g(n_c)}{\partial n_c} = n_c \phi \left[ 1 - \left( \frac{H_c}{n_c \phi} \right)^2 \right]. \quad (2.34)$$

These are the relations through which  $(H_c/n_c \phi)$  determines the ratio between the critical flux and effective magnetic field and its variational rate at  $H_{c2}$ . These relations hold true in both the magnetic and nonmagnetic cases, because they do not *explicitly* contain the polarization term  $m(H_{c2})$ . This motivates us to generalize  $\kappa_1(t)$  as

$$n_c(t) \phi / H_c = \sqrt{2} \kappa_1(t). \quad (2.35)$$

In other words, the flux  $n_c \phi$  in the magnetic case replaces the  $H_{c2}$  of the nonmagnetic case. Since, as (2.17) shows, the averaged energy of the spin system is already subtracted in the definition of  $g(n)$ , the effect of the spin contributes to  $g(n)$  only through the spin fluctuation, say  $\chi_k(n)$ . Therefore modification of  $\kappa_1(t)$  due to the magnetic effect is expected to be small when  $T \gg T_m$ .

From (2.35) and (2.34), we have

$$\frac{\partial}{\partial n_c \phi} g(n_c) = 1 - \frac{1}{2\kappa_1^2(t)}. \quad (2.36)$$

The generalization of  $\kappa_2(t)$  is performed in the following way. The magnetization for a superconductor is given by

$$4\pi M_s = n \phi - H(n). \quad (2.37)$$

Then (2.28) and (2.29) give

$$M_s(H_{c2}) = m_n(H_{c2}) = m(n_c). \quad (2.38)$$

Define

$$\chi_s(H) = \frac{d}{dH} M_s, \quad (2.39)$$

$$\chi_n(H) = \frac{d}{dH} m_n(H), \quad (2.40)$$

and

$$\chi(n) = \chi_B(n) / [1 - \chi_B(n)], \quad (2.41)$$

with

$$\chi_B(n) = \frac{d}{dn \phi} m(n). \quad (2.42)$$

The relation  $\gamma_0^s = \gamma_0^n$  at  $H_{c2}$  and Eq. (2.19) give

$$\chi(n_c) = \chi_n(H_{c2}). \quad (2.43)$$

Equation (2.37) leads to

$$\frac{\partial 4\pi M_s}{\partial H} = \left( \frac{\partial H(n)}{\partial n \phi} \right)^{-1} - 1. \quad (2.44)$$

We thus have

$$\begin{aligned} 4\pi [\chi_s(H_{c2}) - \chi_n(H_{c2})] &= 4\pi [\chi_s(H_{c2}) - \chi(n_c)] \\ &= \left( \frac{\partial H(n)}{\partial n \phi} \right)^{-1}_{n=n_c} \\ &\quad - [1 + 4\pi \chi(n_c)]. \end{aligned} \quad (2.45)$$

From (2.23), we have

$$\frac{\partial H(n)}{\partial n \phi} = \frac{\partial g(n)}{\partial n \phi} + \frac{1}{2} n \phi \left( \frac{\partial}{\partial n \phi} \right)^2 g(n) - \frac{4\pi \chi(n)}{1 + 4\pi \chi(n)}. \quad (2.46)$$

When (2.36) is considered, (2.46) for  $H = H_{c2}$  gives

$$\frac{\partial H(n)}{\partial n \phi} \Big|_{n=n_c} = \frac{1}{1 + 4\pi \chi(n_c)} - \frac{1}{2\kappa_1^2(t)} w(n_c), \quad (2.47)$$

where

$$w(n_c) \equiv 1 - \kappa_1^2(t) n_c \phi \left( \frac{\partial}{\partial n_c \phi} \right)^2 g(n_c). \quad (2.48)$$

Now (2.45) gives

$$4\pi [\chi_s(H_{c2}) - \chi_n(H_{c2})] = [1 + 4\pi \chi_n(H_{c2})] \left[ \frac{w(n_c)}{2\kappa_1^2(t) / [1 + 4\pi \chi_n(H_{c2})] - w(n_c)} \right]. \quad (2.49)$$

Since the deviation of  $w(n_c)$  from one is a small-term proportional to the second derivative of  $g(n_c)$ , we are motivated to rewrite (2.49) as

$$4\pi [\chi_s(H_{c2}) - \chi_n(H_{c2})] = [1 + 4\pi \chi_n(H_{c2})] \frac{1}{\beta(t) (2\kappa_1^2(t) / [1 + 4\pi \chi_n(H_{c2})] - 1)}. \quad (2.50)$$

Then

$$\beta(t) = 1 + \frac{2\kappa_1^2(t)}{2\kappa_1^2(t) - [1 + 4\pi \chi_n(H_{c2})]} \frac{1 - w(n_c)}{w(n_c)}, \quad (2.51)$$

which differs from 1 by a small-term proportional to the second derivative of  $g(n_c)$ . Let us now note that the left-hand side of (2.50) is  $(d/dH)(M_s - M_n)$  where  $M_n$  is the normal-state polarization  $m_n(H)$  and that  $\chi_n$  vanishes to a nonmagnetic case. Comparing (2.50) with (1.2), we see that a natural definition of  $\kappa_2(t)$  is

$$4\pi[\chi_s(H_{c2}) - \chi_n(H_{c2})] = [1 + 4\pi\chi_n(H_{c2})] \frac{1}{\beta(2\kappa_2(t)/[1 + 4\pi\chi_n(H_{c2})] - 1)} \quad (2.52)$$

where the constant  $\beta$  is the value of  $\beta(t)$  at  $t=1$ ;

$$\beta \equiv \beta(1) \quad (2.53)$$

Obviously, this definition leads to the conventional definition of  $\beta$  and  $\kappa_2(t)$  in the nonmagnetic case (i.e.,  $\chi_n = 0$ ).

It follows from (2.50), (2.52), and (2.53) that

$$\kappa_1(1) = \kappa_2(1) \quad (2.54)$$

We define  $\kappa_3(t)$  in the usual way

$$H_c/[\phi/\lambda_L^2(t)] = \frac{\kappa_3(t)}{\sqrt{2}(2\pi)} \quad (2.55)$$

There may exist many kinds of generalizations of  $\kappa$ 's in the magnetic superconductor. In the present generalization, the overall localized spin polarization is subtracted from the definition.

In the nonmagnetic superconductors,  $\kappa_i$ 's approach the same value of  $\kappa$  at  $t=1$ . According to (2.54), in the magnetic case, too,  $\kappa_1(t)$  and  $\kappa_2(t)$  approach the same value of  $\kappa$  at  $t=1$ . The consideration in this section is quite general. In Sec. III, we will study how  $\kappa$  is related to  $\kappa_3(1)$  and how  $\beta$  is modified from that of nonmagnetic case by assuming that the  $s$ - $f$  interaction is very weak so that the electromagnetic interaction is the main agent for the interplay between magnetism and superconductivity.

Finally, we present some relations of practical usefulness. From (2.28) it is easy to derive a relation for the temperature derivative:

$$\frac{d[n_c(t)\phi]}{dt} = [1 + 4\pi\chi_n(H_{c2})] \frac{dH_{c2}(t)}{dt} \quad (2.56)$$

where  $\chi_n(H_{c2})$  is the static differential susceptibility for the applied field  $H_{c2}$ .

From (2.26) and (2.27), we have

$$\frac{d}{dH}(G_s - G_n) = m_n(H) - M_s(H) \quad (2.57)$$

When  $T > T_m$  ( $T_m$ : the Curie temperature for normal ferromagnetic state), no spontaneous magnetization appears. Then by taking into account (2.24), we have

$$G_n(H=0) - G_s|_{H=0} = \int_0^{H_{c2}} dH [m_n(H) - M_s(H)] \quad (2.58)$$

Since  $G_n(H=0) = 0$  and  $G_s|_{H=0} = -H_c^2/8\pi$  (the en-

ergy of the Meissner state), we have

$$\frac{H_c^2}{8\pi} = \int_0^{H_{c2}} dH [m_n(H) - M_s(H)] \quad (2.59)$$

Especially if  $m_n(H)$  is linearly approximated,  $m_n(H) = \chi(0)H$ , one has

$$\frac{H_c^2}{8\pi} = \frac{1}{2}\chi(0)H_{c2}^2 - \int_0^{H_{c2}} dH M_s(H) \quad (2.60)$$

### III. SCALING RULE OF $\kappa$ 's

To study the modification of  $\kappa_i(t)$ 's in the magnetic superconductors, we take a specific model. Namely, we assume that the superconducting system and the localized spin system are coupled to each other only through the electromagnetic interplay, and that other interactions are treated by the renormalization of the fundamental parameters of the above system [such as  $VN(0)$ ,  $\lambda_L$ ,  $\xi_0$ ,  $T_c$ ,  $C$ ,  $D$ ,  $T_m$ , etc.]. Therefore the  $H_I$  term is neglected in (2.1) and  $H_c^2/8\pi$  and the nonlocal kernel  $c(\vec{k})$  in (2.6) are the same as the ones in the nonmagnetic case.

In order to obtain the effective magnetic field  $g(n)$  in (2.17), we solve the Maxwell equation (2.10) and the molecular-field equation (2.11):

$$\vec{\nabla} \times \vec{B}(x) = \frac{4\pi}{c} \vec{j}(\vec{A}_f) + 4\pi \vec{\nabla} \times \vec{M}(x) \quad (3.1)$$

$$|\vec{M}(x)| = g\mu_B J N \times B_J \left[ \frac{g\mu_B J}{k_B T} |\vec{B}(x) + \gamma_0(-i\nabla)\vec{M}(x)| \right] \quad (3.2)$$

where  $\gamma_0^s$  are assumed to be equal to  $\gamma_0^f$  and is simply denoted by  $\gamma_0$  and  $4\pi \vec{j}/c$  is given by (2.6). When the vortices form a lattice, we have

$$\frac{\hbar c}{e} \vec{\nabla} \times \vec{\nabla} f(\vec{x}) = \phi \sum_i \delta^{(2)}(\vec{x} - \vec{\zeta}_i) \vec{e}_3 \quad (3.3)$$

with  $\phi = hc/2e$ . Here  $\vec{e}_3$  is the unit vector along the third axis and  $\vec{\zeta}_i$  is the position of the vortex center.

Hereafter we assume that  $\vec{B}$  and  $\vec{M}$  are parallel to the third axis and we omit the vector notation. The spatial averages of  $B(x)$  and  $M(x)$  are  $n\phi$  and  $m(n)$ , and their deviation is denoted by  $\tilde{b}(x)$ ,  $\tilde{m}(x)$ , respectively. Here  $n$  is the vortex density. Then we

can linearize Eq. (3.2) as

$$m(n) = g \mu_B J N B_j \left[ \frac{g \mu_B J}{k_B T} [n\phi + \gamma_0(0)m(n)] \right], \quad (3.4)$$

$$\tilde{m}(x) = \frac{C}{T} \alpha_J(n) [\tilde{b}(x) + \gamma_0(-i\vec{\nabla})\tilde{m}(x)], \quad (3.5)$$

where

$$C = \frac{(g\mu_B)^2 J(J+1)}{3k_B} N$$

and

$$\alpha_J(n) = \frac{3J}{J+1} B_j \left[ \frac{g \mu_B J}{k_B T} [n\phi + \gamma_0(0)m(n)] \right]. \quad (3.6)$$

Then we can solve Eq. (3.1) as follows:

$$\tilde{b}(x) = \sum_i b(x - \zeta_i) - n\phi \quad (3.7)$$

with

$$b(x) = \frac{\phi}{(2\pi)^2} \times \int d^2 k e^{i\vec{k} \cdot \vec{x}} \frac{[1 + 4\pi\chi_k(n)]\lambda_L^{-2}(t)c(k)}{k^2 + [1 + 4\pi\chi_k(n)]\lambda_L^{-2}(t)c(k)} \quad (3.8)$$

and

$$\chi_k(n) = C \alpha_J(n) / [T - C \gamma(k) \alpha_J(n)]. \quad (3.9)$$

Here

$$\gamma(k) = \gamma_0(k) + 4\pi, \quad (3.10a)$$

and it is parametrized as

$$\gamma(k) = \frac{T_m}{C} - \frac{D}{C} k^2. \quad (3.10b)$$

The fluctuation of the internal magnetic field  $\tilde{h}(x)$  defined by  $\tilde{h}(x) = \tilde{b}(x) - 4\pi\tilde{m}(x)$  [the average value is  $n\phi - 4\pi m(n)$ ] is given by

$$\tilde{h}(x) = \sum_i h(x - \zeta_i) - \frac{n\phi}{1 + 4\pi\chi_0(n)}, \quad (3.11)$$

with

$$h(x) = \frac{\phi}{(2\pi)^2} \times \int d^2 k e^{i\vec{k} \cdot \vec{x}} \frac{\lambda_L^{-2}(t)c(k)}{k^2 + [1 + 4\pi\chi_k(n)]\lambda_L^{-2}(t)c(k)}. \quad (3.12)$$

Then we can evaluate the Gibbs free energy (2.12). After using the same linear approximation as in Eqs. (3.4)–(3.12) and following the procedure presented in Ref. 6, we get

$$G_s(n) = \frac{n\phi}{8\pi} g(n) + F_m(\gamma_0, n\phi) - \frac{H_c^2}{8\pi} - \frac{n\phi}{4\pi} H, \quad (3.13)$$

where

$$g(n) = n\phi + \tilde{h}(0) + E_{\text{core}}(n), \quad (3.14)$$

$$E_{\text{core}}(n) = \frac{1}{V} \int_V d^3 x E_{\text{core}}(x). \quad (3.15)$$

In (3.14),  $\tilde{h}$ ,  $\tilde{b}$ , and  $\tilde{m}$  are taken up to second order of them. The core energy is evaluated<sup>22</sup> as

$$E_{\text{core}}(n) = \epsilon_1 - \epsilon_2 \tilde{b}^{\text{int}}(n), \quad (3.16)$$

with

$$\epsilon_1 = \frac{\phi}{\lambda_L^2(t)} \frac{1}{4\pi} \quad (3.17)$$

and

$$\tilde{b}^{\text{int}}(n) = \sum_{i \neq 0} b(\zeta_i). \quad (3.18)$$

The  $\epsilon_2$  is determined from (2.31) and (2.32).

Usually the vortices form a lattice. Then  $\tilde{h}(0)$  and  $\tilde{b}^{\text{int}}(n)$ , which contain the summation over vortex lattice points, are rewritten in terms of the sum over the reciprocal lattice  $\vec{K}$  as

$$\tilde{h}(0) = n\phi \sum_{\vec{K} \neq 0} \frac{\lambda_L^{-2}(t)c(K)}{K^2 + [1 + 4\pi\chi_K(n)]\lambda_L^{-2}(t)c(K)}, \quad (3.19)$$

$$\tilde{b}^{\text{int}}(n) = n\phi \sum_{\vec{K}} \frac{[1 + 4\pi\chi_K(n)]\lambda_L^{-2}(t)c(K)}{K^2 + [1 + 4\pi\chi_K(n)]\lambda_L^{-2}(t)c(K)} - b(0). \quad (3.20)$$

Now we inspect the structure of  $g(n)$ . The London penetration depth  $\lambda_L(t)$  is chosen as the units of length, and  $n$  and  $K$  are written as  $\bar{n}\lambda_L^{-2}(t)$  and  $\bar{K}\lambda_L^{-1}(t)$  with dimensionless quantities  $\bar{n}$  and  $\bar{K}$ . Then we have

$$g(n) - n\phi = [\phi/\lambda_L^2(t)] F(\bar{n}, \kappa_B, \epsilon_2; t), \quad (3.21)$$

with

$$F(\bar{n}, \kappa_B, \epsilon_2; t) = \bar{n} \sum_{\vec{K} \neq 0} \frac{c_{\vec{K}}}{\bar{K}^2 + [1 + 4\pi\chi_{\vec{K}}(n)]c_{\vec{K}}} + \frac{1}{4\pi} - \epsilon_2 \left[ n \sum_{\vec{K}} \frac{[1 + 4\pi\chi_{\vec{K}}(n)]c_{\vec{K}}}{\bar{K}^2 + [1 + 4\pi\chi_{\vec{K}}(n)]c_{\vec{K}}} - \bar{b}(0) \right], \quad (3.22)$$

where

$$c_{\bar{K}} = \exp[-\nu(\bar{K}/\kappa(t))^\eta] , \quad (3.23)$$

$$\kappa(t) = \gamma(t)\kappa_B , \quad (3.24)$$

with  $\nu$ ,  $\eta$  and  $\gamma$  being certain functions of  $t$ ,<sup>23</sup>

$$\chi_{\bar{K}} = \frac{c/4\pi}{t/[t_m] - \alpha_J(n) + d(t)\alpha_J(n)\bar{K}^2} , \quad (3.25)$$

with

$$c = 4\pi C/T_m , \quad (3.26)$$

$$d = D/T_m \lambda_L^2(0) , \quad (3.27)$$

$$d(t) = d(\lambda_L(0)/\lambda_L(t))^2 , \quad (3.28)$$

and

$$\bar{b}(0) = \frac{1}{(2\pi)^2} \int d^2\bar{K} \frac{(1+4\pi\chi_{\bar{K}})c_{\bar{K}}}{\bar{K}^2 + (1+4\pi\chi_{\bar{K}})c_{\bar{K}}} . \quad (3.29)$$

The function in (3.22) with  $\chi_{\bar{K}} = 0$  will be denoted by  $F_0(\bar{n}, \kappa_B, \epsilon_2; t)$ . Note that, in the nonmagnetic case,  $F$  in (3.21) is replaced by  $F_0$ .

When  $t \sim 1$ ,  $(H_{c2}/g\mu_B JN) \ll 1$ . Therefore the zero-field approximation for  $\chi_{\bar{K}}[\alpha_J(n) = 1]$  may be a good approximation. Furthermore, since  $\lambda_L(t) \rightarrow \infty$  as  $t \rightarrow 1$ ,  $d(t) \rightarrow 0$  for  $t \rightarrow 1$ , implying that the  $\bar{K}^2$  dependence of  $\chi_{\bar{K}}$  is neglected when  $t_m < 1$  (i.e.,  $\chi_{\bar{K}} \rightarrow \chi_0$ ). Experimentally,  $\chi_0(H_{c2})$  is observable. Therefore we use  $\chi_0(H_{c2})$  as  $\chi_0$ . Scaling  $\bar{n}$ ,  $\bar{K}$ ,  $\kappa_B$ , and  $\epsilon_2$  as

$$\begin{aligned} \bar{n} &= (1+4\pi\chi_0)\bar{n}' , \\ \bar{K} &= (1+4\pi\chi_0)^{1/2}\bar{K}' , \\ \kappa_B &= (1+4\pi\chi_0)^{1/2}\kappa'_B , \\ \epsilon_2 &= (1+4\pi\chi_0)^{-1}\epsilon'_2 , \end{aligned} \quad (3.30)$$

we have

$$F(\bar{n}, \kappa_B, \epsilon_2; t) = F_0(\bar{n}', \kappa'_B, \epsilon'_2; t) .$$

Thus (3.21) gives

$$g(n) - n\phi = \frac{\phi}{\lambda_L^2(t)} F_0(\bar{n}', \kappa'_B, \epsilon'_2; t) . \quad (3.31)$$

Our task now is to express the basic relations for magnetic quantities in terms of  $F_0$ . From (2.23) and

$$m(n) = \frac{\chi_0}{1+4\pi\chi_0} n\phi , \quad (3.32)$$

we have

$$\begin{aligned} \frac{H(n)}{\phi/\lambda_L^2(t)} &\approx \frac{1}{2} \left[ 1 + \bar{n}' \frac{\partial}{\partial \bar{n}'} \right] F_0(\bar{n}', \kappa'_B, \epsilon'_2; t) + \bar{n}' \\ &= \bar{H}(\bar{n}') . \end{aligned} \quad (3.33)$$

A scaled magnetization  $4\pi M(n)$  defined by

$$4\pi M(n) = \frac{4\pi[M_s(n) - M_n(H(n))]}{1+4\pi\chi_n(H_{c2})} \quad (3.34)$$

is approximated by<sup>24</sup>

$$4\pi M(n)/[\phi/\lambda_L^2(t)] \approx \bar{n}' - \bar{H}(\bar{n}') = 4\pi\bar{M}(\bar{n}') . \quad (3.35)$$

Since  $H_c^2(t)/8\pi$  has the same functional form as that of the nonmagnetic case with renormalized parameters,  $H_c(t)/[\phi/\lambda_L^2(t)]$  is proportional to  $\kappa_B$ .

Then we can write

$$\frac{H_c^2(t)}{[\phi/\lambda_L^2(t)]^2} = \left[ \frac{\kappa_3(\kappa_B)}{\sqrt{2}2\pi} \right]^2 = \left[ \frac{\kappa_3(\kappa'_B)}{\sqrt{2}2\pi} \right]^2 (1+4\pi\chi_0) . \quad (3.36)$$

From (2.31), (2.32), and (3.31), we can see that the equation which determines  $\epsilon'_2$  is

$$\left[ \frac{\kappa_3(\kappa'_B)}{\sqrt{2}2\pi} \right]^2 = \bar{n}'_c F_0(\bar{n}'_c, \kappa'_B, \epsilon'_2; t) , \quad (3.37a)$$

$$\frac{1}{2} \left[ 1 + \bar{n}'_c \frac{\partial}{\partial \bar{n}'_c} \right] F_0(\bar{n}'_c, \kappa'_B, \epsilon'_2; t) = 0 . \quad (3.37b)$$

Equations (3.33), (3.35), and (3.37) are exactly the same as those in the nonmagnetic case (with primed variables). This indicates that the magnetization curve ( $4\pi\bar{M}$  vs  $\bar{H}$ ) is classified in the same way as the nonmagnetic case when  $\kappa_B$  is replaced by the scaled  $\kappa'_B$ ,

$$\kappa'_B = \frac{\kappa_B}{(1+4\pi\chi_0)^{1/2}} , \quad (3.38)$$

and that the  $\kappa$  values for this magnetization curve are obtained by

$$\bar{H}_{c2}/\bar{H}_c = \sqrt{2}\kappa_1^0(\kappa'_B, t) , \quad (3.39)$$

$$\frac{\partial(4\pi\bar{M})}{\partial\bar{H}} \Big|_{\bar{H}=\bar{H}_{c2}} = \frac{1}{\beta^0[2[\kappa_2^0(\kappa'_B, t)]^2 - 1]} , \quad (3.40)$$

and

$$\bar{H}_c/[\phi/\lambda_L^2(t)] = \kappa_3^0(\kappa'_B, t)/\sqrt{2}(2\pi) . \quad (3.41)$$

Here  $\bar{H}_c$  is defined by

$$\begin{aligned} \frac{\bar{H}_c^2}{8\pi} &= \int_0^{\bar{H}_{c2}} d\bar{H} [-\bar{M}(\bar{H})] \\ &= (1+4\pi\chi_0)^{-1} \int_0^{H_{c2}} dH [m_n(H) - M_s(H)] \\ &= (1+4\pi\chi_0)^{-1} \frac{H_c^2}{8\pi} . \end{aligned} \quad (3.42)$$

In (3.39)–(3.41), subscripts 0 indicate the functions for nonmagnetic case. Rewriting quantities with bars

in terms of the original quantities, we have

$$\frac{n_c(t)}{H_c(t)} = \sqrt{2}(1+4\pi\chi_0)^{1/2}\kappa_1^0 \left[ \frac{\kappa_B}{(1+4\pi\chi_0)^{1/2}}, t \right], \quad (3.43a)$$

$$\frac{\partial}{\partial H} \left[ \frac{4\pi M_s(H) - 4\pi m_n(H)}{1+4\pi\chi_0} \right] = \frac{1}{\beta^0 \left\{ 2 \left[ \kappa_2^0 \left[ \frac{\kappa_B}{(1+4\pi\chi_0)^{1/2}}, t \right] \right]^2 - 1 \right\}}, \quad (3.43b)$$

and

$$\frac{H_c}{\phi/\lambda_L^2(t)} = (1+4\pi\chi_0)^{1/2}\kappa_3^0 \left[ \frac{\kappa_B}{(1+4\pi\chi_0)^{1/2}}, t \right] / \sqrt{2}(2\pi). \quad (3.43c)$$

Comparing these relations with (2.35), (2.52), and (2.55), we have the simple scaling law for  $\kappa_i'$ 's in the region where the linear approximation  $m_n(H) = \chi_0 H$  is valid,

$$\kappa_i(t) = (1+4\pi\chi_0)^{1/2}\kappa_i^0 \left[ \frac{\kappa_B}{(1+4\pi\chi_0)^{1/2}}, t \right], \quad (3.44)$$

and also we have the result

$$\beta = \beta^0. \quad (3.45)$$

Especially  $\kappa_i(t)$  at  $t = 1$  approaches same value  $\kappa$ :

$$\begin{aligned} \kappa_i(1) &= (1+4\pi\chi_0)^{1/2}\kappa_i^0 \left[ \frac{\kappa_B}{(1+4\pi\chi_0)^{1/2}}, t \right] \Big|_{t=1} \\ &= \kappa_3^0(\kappa_B, 1) \equiv \kappa, \end{aligned} \quad (3.46)$$

where we use the fact that  $\kappa_i^0$  approaches the same value as  $\kappa$  in the limit  $t \rightarrow 1$ ,<sup>25</sup> and that  $\kappa_3^0$  is proportional to  $\kappa_B$ .

The above results indicate simple magnetic properties of magnetic superconductors around  $t \sim 1$ .

(1) The result (3.43a) shows that the temperature behavior of  $n_c(t)\phi$  is little affected by the magnetic moments when  $H_c(t)$  (and therefore also the gap energy) is not much modified by the magnetic effect. In other words, when the  $s$ - $f$  interaction is weak, the temperature behavior of  $n_c\phi$  is very similar to that of  $H_{c2}$  of the nonmagnetic case with the same  $\kappa$ . This can be experimentally checked.

In Fig. 1, we present an example of numerical calculation of  $n_c(t)\phi$ ,  $H_{c2}(t)$ , and  $H_{c1}(t)$  curves. Parameters are  $VN(0) = 0.2635$ ,  $\kappa_B = 2.0$ ,  $J = 7.5$ ,  $t_m = 0.16$ ,  $c = 1.6$ ,  $d = 0.01$ , and  $u = 0.13$ , where  $u = (g\mu_B JN)/[\phi/\lambda_L^2(0)]$ . The  $c$  function in (3.23) is taken from Ref. 23. The dot-dashed line is for nonmagnetic case and the solid line is for magnetic case. The dotted line is obtained from the formula for the nonmagnetic case by scaling  $\kappa_B$  as  $\kappa_B' = \kappa_B/(1+4\pi\chi_0)^{1/2}$ . the difference of  $n_c\phi$  between nonmagnetic and magnetic case is small, though  $H_{c2}$  is very suppressed in the

magnetic case. Scaling rule (i.e., coincidence of solid and dotted curves) seems quite good up to  $t \sim 0.5$  for this choice of parameters.

(2) The magnetization curve can be compared with that of nonmagnetic one by a suitable scaling. Namely, in the temperature region where  $m_n(H) \sim \chi_0 H$  ( $0 < H < H_{c2}$ ) is valid, one plots  $H$  vs  $4\pi M \equiv 4\pi[M_s - M_n(H)]/(1+4\pi\chi_0)$  as is shown in Fig. 2. Then this  $H$  vs  $4\pi M$  curve is the same as the nonmagnetic

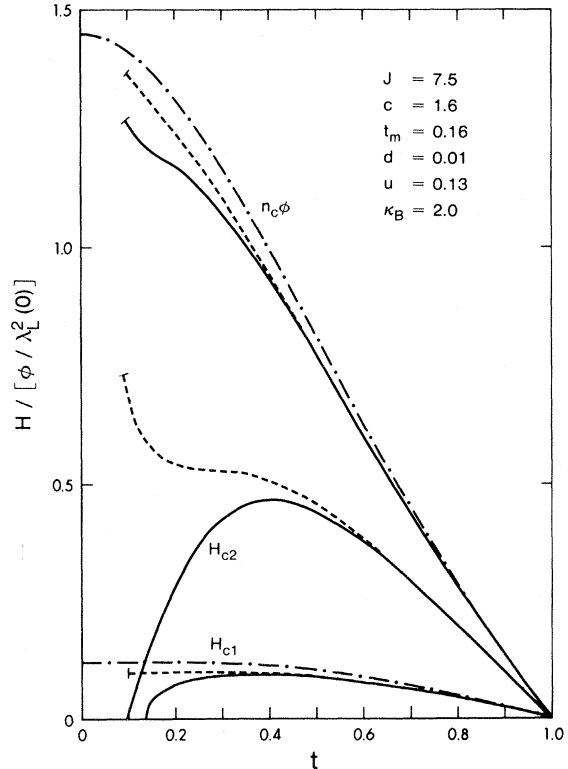


FIG. 1. Scaling rules in the critical fields. Solid line is for the magnetic case, dash-dotted line is for the nonmagnetic case. Dotted line is obtained by the scaling of  $\kappa_B$ .



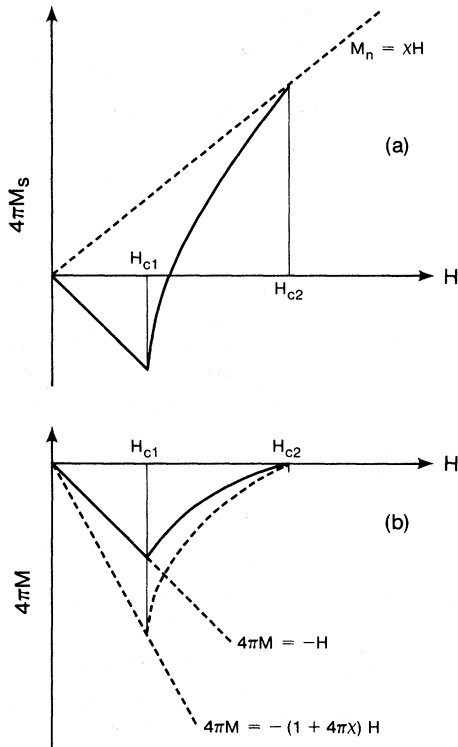


FIG. 2. (a) Schematic magnetization curve for magnetic superconductor. Solid line indicates  $4\pi M_s$  and dashed line indicates  $4\pi m_n$  ( $m_n \sim \chi H$ ). (b) Scaling of the magnetization curve. The dashed line is the difference  $(4\pi M_s - 4\pi m_n)$  and solid line is scaled magnetization curve  $(4\pi M_s - 4\pi m_n)/(1 + 4\pi\chi)$ . solid curve is compared with nonmagnetic case with  $\kappa' = \kappa/\sqrt{1 + 4\pi\chi}$ .

one with  $\kappa'_B$  scaled as

$$\kappa'_B = \frac{\kappa_B}{(1 + 4\pi\chi_0)^{1/2}} \quad (3.47)$$

In Fig. 3, we present the numerically calculated magnetization curves for the parameters used in Fig. 1. Figure 3(a) is for magnetic superconductor and Fig. 3(b) is obtained from the result of Fig. 3(a) by the scaling (3.34). Figure 3(c) is for the nonmagnetic case with scaled  $\kappa'_B = \kappa_B/(1 + 4\pi\chi_0)^{1/2}$ . Figures 3(b) and 3(c) show good agreement.

This result indicates that effective  $\kappa'_B$  changes with temperature, since  $\chi_0$  changes ( $\kappa_B$  is a temperature-independent parameter). With decreasing temperature,  $\chi_0$  increases when  $T$  approaches  $T_m$ , the Curie temperature of normal ferromagnet. Accordingly,  $\kappa'_B$  decreases and as a result, the transition from type II/2 to type II/1 or from type II to type I is induced with decreasing temperature, which was predicted in the previous paper<sup>6</sup> and is confirmed by the experiment.<sup>18</sup>

The practical usefulness of our formulation lies in

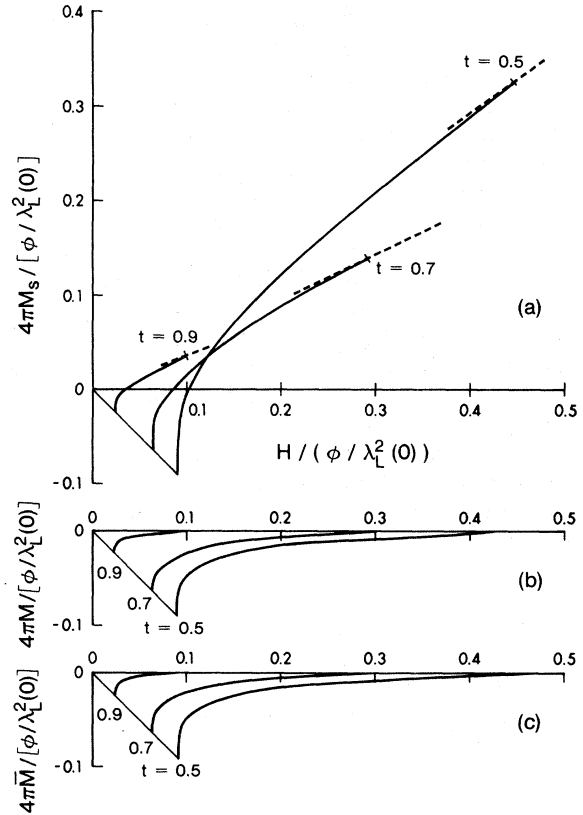


FIG. 3. Numerical results of magnetization curves (a)  $4\pi M_s$ , (b)  $4\pi M$  [ $= 4\pi(M_s - m_n)/(1 + 4\pi\chi)$ ], and (c)  $4\pi \bar{M}$  obtained from scaling of  $\kappa_B$  ( $\kappa'_B = \kappa_B/\sqrt{1 + 4\pi\chi}$ ) in the nonmagnetic case.

the fact that it presents a simple relation between the magnetization curves of the magnetic superconductors and those of nonmagnetic one. This was explicitly shown in Figs. 1, 2, and 3.

In the Appendix, we also show that some of the present results are also reproduced in the GL theory.

#### IV. CONCLUDING REMARKS

We have presented a generalization of  $\kappa_1$ ,  $\kappa_2$  in the case of magnetic superconductors in such a way that the effect of the average spin polarization is subtracted. When the magnetization of the localized spins is approximately proportional to the magnetic field and when the  $\bar{k}$  dependence of the staggered susceptibility is neglected,  $\kappa_1$ ,  $\kappa_2$  have the simple scaling relations (3.44) with the  $\kappa^0$  functional form of the nonmagnetic superconductors. It was shown that the temperature behavior of  $n_c\phi$  is not greatly affected by the presence of the magnetic spins, though  $H_{c2}$  may show a very different behavior from the nonmagnetic

case. This result comes from the assumption that  $H_c$  is not much modified because the  $s$ - $f$  interaction is assumed to be very weak.

It is also shown that, if the electromagnetic interplay is the main mechanism, the magnetization curve becomes that of nonmagnetic superconductors with  $\kappa' = \kappa/(1+4\pi\chi_0)^{1/2}$  by a suitable scaling in the region where  $t \sim 1$  and the approximation  $m_n(H) \sim \chi_0 H$  is valid. Conversely, if experiments show a drastic modification of the temperature behavior of  $n_c \phi$  due to the magnetic effect and if the simple scaling rule needs large modifications, this would be an indication that the  $s$ - $f$  interaction effect or others is not negligible. Those give us more information about the magnetic superconductors.

As was seen in the text, the statement that the phase transition at  $H = H_{c2}$  is of the second order plays a significant role in the derivation of many of the results in Sec. III.

#### APPENDIX

We show that some of the results similar to the ones obtained in Secs. II and III can be derived from the GL theory also.

Let us start from the GL equations

$$-(\bar{\nabla} - i\kappa\bar{\mathbf{A}})^2\phi = \kappa^2\phi(1 - |\phi|^2), \quad (\text{A1})$$

$$\bar{\nabla} \times \bar{\mathbf{B}} = \frac{1}{2i\kappa} [\phi^*(\bar{\nabla} - i\kappa\bar{\mathbf{A}})\phi - (\bar{\nabla} + i\kappa\bar{\mathbf{A}})\phi^*\phi] + 4\pi\bar{\nabla} \times \bar{\mathbf{M}}. \quad (\text{A2})$$

Here we assumed that  $\phi$ ,  $\bar{\mathbf{A}}$ ,  $\bar{\mathbf{B}}$ , and  $\bar{\mathbf{M}}$  are suitably normalized. Reflecting the assumption that  $s$ - $f$  interaction is neglected, no terms exist which include  $\bar{\mathbf{M}}$  in (A1). We assume that, at  $H_{c2}$ ,  $B$  is homogeneous and is given by  $n_c$  [so that  $\bar{\mathbf{A}}_c = (0, n_c x, 0)$ ] and  $\phi$  vanishes. When  $H$  is in the vicinity of  $H_{c2}$ ,  $\phi$  must satisfy the linear equation

$$-(\bar{\nabla} - i\kappa\bar{\mathbf{A}}_c)^2\phi = \kappa^2\phi, \quad (\text{A3})$$

which determines

$$n_c = \kappa \quad (\text{A4})$$

as the eigenvalue.

When the applied field is slightly below  $H_{c2}$ , we expand  $B$  and  $M$  in terms of  $H_{c2} - H$ :  $B = n_c + b(x)$ ,  $M = M(H_{c2}) + m(x)$ . From the second GL equation we have<sup>20</sup>

$$\frac{\partial}{\partial x}(b - 4\pi m) = -\frac{\partial}{\partial x}\left(\frac{1}{2\kappa}\phi^*\phi\right), \quad (\text{A5a})$$

$$\frac{\partial}{\partial y}(b - 4\pi m) = -\frac{\partial}{\partial y}\left(\frac{1}{2\kappa}\phi^*\phi\right). \quad (\text{A5b})$$

The first GL equation conditions the space average of quantities  $b\phi^*\phi$  and  $(\phi^*\phi)^2$ :

$$\langle b(x)\phi^*(x)\phi(x) \rangle = -\kappa \langle [\phi^*(x)\phi(x)]^2 \rangle, \quad (\text{A6})$$

where  $\langle \rangle$  means the space average. The internal magnetic field  $H(x) = H_{c2} + h(x)$  with  $h(x) = b(x) - 4\pi m(x)$  is obtained from (A5) as

$$h(x) = H_{c2} - H - \frac{1}{2\kappa}\phi^*\phi. \quad (\text{A7})$$

Here it is assumed that for  $\phi = 0$ ,  $h(x) = H_{c2} - H$ . Since  $m(x)$  is the variable part of  $M(x)$ , it is related to  $h(x)$  linearly in a reasonable approximation:

$$m(x) = \chi h(x). \quad (\text{A8})$$

Then we have

$$\begin{aligned} b(x) &= (1 + 4\pi\chi)h(x) \\ &= (1 + 4\pi\chi)(H_{c2} - H) - \frac{1 + 4\pi\chi}{2\kappa}\phi^*\phi. \end{aligned} \quad (\text{A9})$$

Taking the space average of (A9), we have

$$n - n_c = (1 + 4\pi\chi)(H_{c2} - H) - \frac{1 + 4\pi\chi}{2\kappa} \langle \phi^*\phi \rangle. \quad (\text{A10})$$

The condition (A6) together with (A9) leads to

$$(1 + 4\pi\chi)(H_{c2} - H) \langle \phi^*\phi \rangle = \frac{1 + 4\pi\chi - 2\kappa^2}{2\kappa} \langle (\phi^*\phi)^2 \rangle \quad (\text{A11})$$

which gives

$$\frac{1}{2\kappa} \langle \phi^*\phi \rangle = \frac{(1 + 4\pi\chi)(H_{c2} - H)}{1 + 4\pi\chi - 2\kappa^2} \frac{\langle (\phi^*\phi)^2 \rangle}{\langle (\phi^*\phi) \rangle^2}. \quad (\text{A12})$$

Therefore we have

$$\begin{aligned} n - n_c &= -(1 + 4\pi\chi)(H - H_{c2}) \\ &\quad - \frac{(1 + 4\pi\chi)^2(H - H_{c2})}{\beta_A(1 + 4\pi\chi - 2\kappa^2)}, \end{aligned} \quad (\text{A13})$$

with

$$\beta_A = \frac{\langle (\phi^*\phi)^2 \rangle}{\langle \phi^*\phi \rangle^2}.$$

Then

$$\begin{aligned} \frac{\partial 4\pi M_S}{\partial H} &= -1 - (1 + 4\pi\chi) \frac{1 + 4\pi\chi - \beta_A(1 + 4\pi\chi - 2\kappa^2)}{\beta_A(1 + 4\pi\chi - 2\kappa^2)}, \end{aligned} \quad (\text{A14})$$

which leads to

$$4\pi(\chi_S - \chi) = \frac{1}{\beta_A} (1 + 4\pi\chi) \frac{1}{2\kappa^2/[1 + 4\pi\chi] - 1} \quad (\text{A15})$$

The result is identical to (2.50).

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- <sup>24</sup>Even when the linear approximation (3.32) does not hold, one can consider the behavior around  $H_{c2}$ . In this case
- $$m(n) \sim m(n_c) + \frac{\chi(H_{c2})}{1 + 4\pi\chi(H_{c2})} (n - n_c)\phi.$$
- Then (3.33) is modified as
- $$H(n)/[\phi/\lambda_L^2(t)] \sim \bar{H}(\bar{n}') + \left[ H_{c2} - \frac{n_c\phi}{1 + 4\pi\chi(H_{c2})} \right] / [\phi/\lambda_L^2(t)],$$
- inducing only a constant shift between  $H(n)/[\phi/\lambda_L^2(t)]$  and  $\bar{H}(\bar{n}')$ . Equation (3.34) is not modified.
- <sup>25</sup>In the present method, this statement is also satisfied approximately.