Scaling Theory for Critical Phenomena in Coupled Magnetic Systems – Critical Exponents of πd Antiferromagnets –

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(Received January 20, 2020)

The scaling theory for critical phenomena is extended to coupled magnetic systems that consist of two subsystems, and some extended relations of critical exponents are derived. It is shown that the extended theory practically reduces to the conventional scaling theory in ferromagnets and in non-ferromagnetic systems with $\beta_1 = \beta_2$; however, the extended form of the theory can be relevant otherwise, where β_1 and β_2 are exponents of the order parameters in subsystems 1 and 2, respectively. The theory is applied to a model of the organic πd antiferromagnet λ-$(BETS)_2FeCl_4$, which contains π- and d-spin subsystems, where BETS stands for bis(ethylenedithio)tetraselenafulvalene. It is shown that an effective Hamiltonian for the π spins is reduced to the two-dimensional Ising model in the vicinity of the critical temperature T_c. This supports a conjecture from a recent experimental observation. Consequently, $\beta_1 = 1/8$ is obtained, where subsystems 1 and 2 correspond to the π- and d-spin systems, respectively. Additional relations $\alpha = 1 - \beta_1 - \beta_2$ and $\beta_1 = \beta_2 \equiv \beta$ are derived from specific features of the λ-$(BETS)_2FeCl_4$ system. These relations result in $\alpha = 1 - 2\beta$, which was previously obtained in a free-energy functional model. Critical exponents below T_c are obtained as $\alpha = 3/4$, $\beta = 1/8$, $\gamma = 1$, $\delta = 9$, $\psi = 1/5$, and $\nu = 5/8$. The value of α is close to a recent experimental result of $\alpha = 0.77$ in λ-$(BETS)_2FeCl_4$.

The phase transition and critical phenomena in coupled magnetic systems have been examined experimentally[1–5] and theoretically[6–9] in connection with the organic πd antiferromagnet λ-$(BETS)_2FeCl_4$,[10–17] where BETS stands for bis(ethylenedithio)tetraselenafulvalene. In this compound, the long-range order is principally sustained in a π-electron system[2] despite the large length of d spins. This phenomenon originates from the fact that $J_1 \gg J_2 \approx 0$,[12,13] where J_1 and J_2 denote the coupling constants of the interactions between π electrons and between d spins, respectively. Despite its large value, J_1 does not induce the transition by itself, because the π-electron system is two-dimensional and isotropic in the spin space.[2,6,18,19] Instead, an effective interaction between π-electrons via d-spins[6–9] introduces anisotropy in the spin space and induces the transition at a temperature T_c. Consequently, $J_1 \gg T_c \gg J_2$ is satisfied, where we use units in which $k_B = 1$.

The phenomena in the vicinity of T_c are also intriguing; for example, the magnetic specific heat $C(T)$ exhibits an extremely sharp peak around T_c.[1–5] This phenomenon can be understood[9] as a consequence of the mismatch between the energy scales of T_c and J_1, which is a unique feature of this compound as mentioned above. Furthermore, it was predicted[9] that the system undergoes a pseudo second-order phase transition with a small drop in the order parameter at T_c (the transition finally becomes a first-order transition), which originates from a reduction of the effective interaction between the π electrons[7] due to a logarithmic divergence of the fluctuations at T_c. The small drop in the order parameter agrees with a small hysteresis in the specific heat, which was observed recently.[5] When the logarithmic divergence is ignored, the first-order transition does not occur; instead, the system undergoes a second-order transition with a transition temperature $T_c^{(2nd)}$ that is slightly higher ($\lesssim 0.1$ %) than the true T_c.

The critical exponents can be defined in the temperature region where the influence of the first-order transition is negligible. The width of the temperature region in which the first-order transition is significant must be of the same order as $T_c^{(2nd)} - T_c \lesssim 10^{-3} T_c$, which is extremely small. In the previous study,[9] the Gaussian approximation was adopted to examine the fluctuation near T_c, and the conventional results for the critical exponents in the Gaussian model, such as $\alpha = 1/2$, were obtained; however, those values are artifacts due to the approximation. In this Letter, we examine critical exponents using the scaling theory,[20–25] in which the Gaussian approximation is not used. In the first part, we extend the theory to a generic coupled magnetic system. In the second part, we apply it to a model of the πd antiferromagnet λ-$(BETS)_2FeCl_4$, taking into account specific features of this compound. We consider the system below T_c.

Coupled magnetic systems – We consider generic coupled systems that consist of two subsystems $\ell = 1, 2$ on lattices L_ℓ in d dimensions. We refer to the spins on sites $i \in L_1$ and $i' \in L_2$ as $\boldsymbol{s}_i$ and $\boldsymbol{S}_{i'}$, respectively, where the spin-quantization axis z for the spin on each site is taken in the direction of each magnetization below T_c so that the order parameters are defined by $m = \langle s_i^z \rangle$ and $M = \langle S_{i'}^z \rangle$. We refer to the magnitudes of $\boldsymbol{s}_i$ and $\boldsymbol{S}_{i'}$ as s and S, respectively. We define $\tau = T - T_c$ and symmetry-breaking fields $h_1 > 0$ and $h_2 > 0$, which are coupled with s_i^z and S_i^z and stabilize $m > 0$ and $M > 0$, respectively. We define the free energy per site $f(\tau, h_1, h_2)$ that gives

$$m(\tau, h_1, h_2) \equiv -\frac{\partial f}{\partial h_1}, \qquad M(\tau, h_1, h_2) \equiv -\frac{\partial f}{\partial h_2}.$$

Here, h_1 and h_2 are formally distinguished; however, they are equated as $h_1 = h_2 \equiv h$ in the arguments of physical quantities at the field h. For example, the order parameters of subsystems 1 and 2 at the field h are expressed as $m(\tau, h) = m(\tau, h, h)$ and $M(\tau, h) = M(\tau, h, h)$, respectively. We refer to the numbers of spins in subsystems ℓ as N_ℓ.

Block spins and scaling hypothesis – In the vicinity of the critical point, we consider the scale transformation by block spins of size b, where the coherence length $\xi \gg b \gg 1$. We refer to the block (the cell) that contains b^d spins around a spatial position labeled by j as $B_j(b)$. We define

$$\bar{\boldsymbol{s}}_j = \frac{1}{b^d}\sum_{i\in B_j(b)} \boldsymbol{s}_i, \quad \bar{\boldsymbol{S}}_{j'} = \frac{1}{b^d}\sum_{i'\in B_{j'}(b)} \boldsymbol{S}_{i'},$$

and the block spins $\tilde{s}^z_j = \bar{s}^z_j b^{\psi_1}$ and $\tilde{S}^z_{j'} = \bar{S}^z_{j'} b^{\psi_2}$, where ψ_1 and ψ_2 denote the scaling dimensions of the block spins so that the magnitudes of the block spins coincide with s and S, respectively.

The scaling hypothesis[20–25] can be extended as

$$f(\tilde{\tau}, \tilde{h}_1, \tilde{h}_2) = b^d f(\tau, h_1, h_2) \tag{1}$$

for coupled systems, where $\tilde{\tau} = \tau b^{1/\nu}$ and $\tilde{h}_\ell = h_\ell b^{d-\psi_\ell}$. The definition of $\tilde{\tau}$ is consistent with the definition of the exponent ν, i.e., $\xi \sim |\tau|^{-\nu}$. The free energy can be written in the form

$$f(\tau, h_1, h_2) = f_{\rm s}(h_1, h_2)|\tau|^p + f_{\rm l}(h_1, h_2)\frac{\tau^2}{2}\ln|\tau| + \Delta f(\tau, h_1, h_2)$$

with appropriate factors $f_{\rm s}$ and $f_{\rm l}$ and a positive and noninteger exponent p, where Δf denotes the remainder term, which is negligible near the critical point.

Relations of critical exponents – The exponents that are not directly related to the order parameters satisfy the same relations as those in conventional systems that consist of a single kind of magnetic degrees of freedom (hereafter called the conventional systems). The critical exponent α of the specific heat $c_{\rm heat}$ is defined by $c_{\rm heat} \sim |\tau|^{-\alpha}$ and obtained in the same manner as that for the conventional systems. When $f_{\rm s}(0,0) \neq 0$ and $p > 2$, the most singular term of the specific heat satisfies $c_{\rm heat} \sim |\tau|^{p-2}$; hence, we obtain $p = 2 - \alpha$. The scaling hypothesis results in $p = \nu d$ and

$$\alpha = 2 - \nu d. \tag{2}$$

In contrast, when $f_{\rm s}(0,0) = 0$ or $p \leq 2$, the most singular term becomes $c_{\rm heat} \sim \ln\tau$, which is expressed as $\alpha = +0$.

When the order parameters in two subsystems are considered, the scaling hypothesis needs to be extended as presented in Eq. (1), which results in the scaling relations for the order parameters

$$\begin{aligned} m(\tilde{\tau}, \tilde{h}_1, \tilde{h}_2) &= b^{\psi_1} m(\tau, h_1, h_2), \\ M(\tilde{\tau}, \tilde{h}_1, \tilde{h}_2) &= b^{\psi_2} M(\tau, h_1, h_2). \end{aligned} \tag{3}$$

For $h = 0$ and $T < T_{\rm c}$, we define exponents β_ℓ by

$$m(\tau, 0) \sim |\tau|^{\beta_1}, \quad M(\tau, 0) \sim |\tau|^{\beta_2},$$

and hence, from Eq. (3), we obtain

$$\beta_\ell = \nu\psi_\ell. \tag{4}$$

For $T = T_{\rm c}$ and $h > 0$, we define exponents $\delta_\ell^{(\ell')}$ by

$$\begin{aligned} m(0, h_1, 0) &\sim h_1^{1/\delta_1^{(1)}}, \quad m(0, 0, h_2) \sim h_2^{1/\delta_1^{(2)}}, \\ M(0, h_1, 0) &\sim h_1^{1/\delta_2^{(1)}}, \quad M(0, 0, h_2) \sim h_2^{1/\delta_2^{(2)}}. \end{aligned}$$

From Eq. (3), it follows that $d - \psi_{\ell'} = \psi_\ell \delta_\ell^{(\ell')}$. We also define exponents δ_ℓ of the order parameters of subsystems ℓ (= 1, 2) by

$$m(0, h) \sim h^{1/\delta_1}, \quad M(0, h) \sim h^{1/\delta_2}.$$

Because the leading terms are retained in these definitions, we obtain $\delta_\ell = \max(\delta_\ell^{(1)}, \delta_\ell^{(2)})$. Hence, using Eq. (2) and (4), we obtain

$$\begin{aligned} \psi_1\delta_1 &= \psi_2\delta_2 = d - \psi_0, \\ \beta_1\delta_1 &= \beta_2\delta_2 = \nu d - \beta_0 = 2 - \alpha - \beta_0, \end{aligned} \tag{5}$$

where we define $\psi_0 \equiv \min(\psi_1, \psi_2)$ and $\beta_0 \equiv \min(\beta_1, \beta_2)$.

Before examining the susceptibilities, we define the functions $\chi_{\ell\ell'}$ and their exponents $\gamma_{\ell\ell'}$ as

$$\begin{aligned} \chi_{1\ell} &\equiv \left[\frac{\partial m(\tau, h_1, h_2)}{\partial h_\ell}\right]_{h_1=h_2=0} \sim |\tau|^{-\gamma_{1\ell}}, \\ \chi_{2\ell} &\equiv \left[\frac{\partial M(\tau, h_1, h_2)}{\partial h_\ell}\right]_{h_1=h_2=0} \sim |\tau|^{-\gamma_{2\ell}}. \end{aligned}$$

It follows from Eq. (3) that

$$\gamma_{\ell\ell'} = (d - \psi_\ell - \psi_{\ell'})\nu, \tag{6}$$

and hence, Eqs. (2) and (4) result in

$$\alpha + \beta_\ell + \beta_{\ell'} + \gamma_{\ell\ell'} = 2, \tag{7}$$

which is an extension of the famous scaling relation $\alpha + 2\beta + \gamma = 2$.

The susceptibilities of subsystems 1 and 2 for the field $h = h_1 = h_2$ are expressed as

$$\begin{aligned} \chi_1 &= \left[\frac{\partial m(\tau, h)}{\partial h}\right]_{h=0} = \chi_{11} + \chi_{12}, \\ \chi_2 &= \left[\frac{\partial M(\tau, h)}{\partial h}\right]_{h=0} = \chi_{21} + \chi_{22}, \end{aligned}$$

and the exponents are defined by $\chi_\ell \sim |\tau|^{-\gamma_\ell}$. Because the most singular terms are retained in these definitions, $\gamma_\ell = \max(\gamma_{\ell 1}, \gamma_{\ell 2})$; hence,

$$\gamma_\ell = (d - \psi_\ell - \psi_0)\,\nu, \tag{8}$$

$$\alpha + \beta_\ell + \beta_0 + \gamma_\ell = 2, \tag{9}$$

where we have used Eqs. (6) and (7). Equations (8) and (9) are also extensions of the conventional scaling relations, and Eq. (9) leads to $\beta_1 + \gamma_1 = \beta_2 + \gamma_2$.

Ferromagnets – In ferromagnets, the total magnetization $M_{\rm total}$ and the total magnetic susceptibility χ are defined by $M_{\rm total} = N_1 m + N_2 M$ and $\chi \equiv N_1\chi_1 + N_2\chi_2$, respectively, and their exponents are defined by $M_{\rm total}(\tau, 0) \sim |\tau|^\beta$, $M_{\rm total}(0, h) \sim h^{1/\delta}$, and $\chi \sim |\tau|^{-\gamma}$. These definitions result in $\beta = \beta_0$, $\gamma = \max(\gamma_1, \gamma_2)$, and $\delta = \max(\delta_1, \delta_2)$; hence, from Eqs. (4), (5), (8), and (9), we obtain

$$\begin{aligned} &\alpha + 2\beta + \gamma = 2, \quad \beta = \beta_0 = \nu\psi_0, \\ &\gamma = (d - 2\psi_0)\nu, \quad \delta = d/\psi_0 - 1. \end{aligned} \tag{10}$$

Therefore, in ferromagnets, the relations for conventional systems are recovered, where ψ_0 is regarded as ψ.

Non-ferromagnetic systems – In non-ferromagnetic systems in which summations $N_1 m + N_2 M$ and $N_1\chi_1 + N_2\chi_2$ do not have a clear physical meaning, we cannot define β, δ, and γ in the same manner as those in ferromagnetic systems, and the relations in Eq. (10) are not necessarily satisfied. In fact, when

$\psi_1 \neq \psi_2$, the free energy $f(\tau, h) = f(\tau, h, h)$ does not satisfy the scaling hypothesis. Therefore, the extended relations, such as Eqs. (4)–(9), can be relevant to non-ferromagnetic systems when $\psi_1 \neq \psi_2$. In contrast, when $\psi_1 = \psi_2 \equiv \psi$, Eq. (1) is reduced to the conventional scaling hypothesis

$$f(\tilde{\tau}, \tilde{h}) = b^d f(\tau, h),$$

where $\tilde{h} = hb^{d-\psi}$. In this case, Eqs. (4), (5), (8), and (9) are reduced to the conventional relations as

$$\begin{aligned} &\beta \equiv \beta_1 = \beta_2 = \nu\psi, \\ &\gamma \equiv \gamma_{\ell\ell'} = \gamma_\ell = (d - 2\psi)\nu, \\ &\delta = \delta_\ell^{(\ell')} = \delta_\ell = d/\psi - 1, \\ &\alpha + 2\beta + \gamma = 2 \end{aligned} \tag{11}$$

for arbitrary (ℓ, ℓ'). The relation $\beta_1 = \beta_2$ implies that $M \propto m$ is satisfied for $h = 0$.

Application to the πd antiferromagnet λ-(BETS)$_2$FeCl$_4$ – For this compound below T_c, because the system is insulating, simplification by localized spin models would be useful. According to previous studies,[1–13] we can assume that the π spin system is isotropic in the spin space, and the π–d interaction is anisotropic. Assuming that subsystems 1 and 2 correspond to the π- and d-spin systems, respectively, we consider the model of coupled antiferromagnets[8,13] expressed by the Hamiltonian $H = H_1 + H_2 + H_{12}$ with

$$\begin{aligned} &H_1 = \sum_{(i,j)} J_1 \boldsymbol{s}_i \cdot \boldsymbol{s}_j - \sum_i h_1^{(i)} s_i^z, \\ &H_2 = -\sum_i h_2^{(i)} S_i^z, \quad H_{12} = \sum_i J_{12} s_i^z S_i^z. \end{aligned} \tag{12}$$

The magnitudes of d spins are $S = 5/2$ because each $FeCl_4$ anion has five d electrons. For the π spins, we must take into account the dimerization of BETS molecules, and thus, we assume that $N_1 = N_2 \equiv N$ and $s = 1/2$.[26] We set the lattice constants as unity for convenience. We split the lattice of subsystem ℓ into two sublattices A_ℓ and B_ℓ. The site index i in subsystem 1 is defined so that i is an odd and even integer when $i \in A_1$ and $i \in B_1$, respectively. Because $J_{12} > 0$, $i \in B_2$ when $i \in A_1$, whereas $i \in A_2$ when $i \in B_1$. Therefore, the symmetry-breaking fields are expressed as $h_\ell^{(i)} = h_\ell(-1)^{i+\ell}$. In the π–d interaction H_{12}, each d spin is coupled with only a single π spin.[27] Because they are labeled with the same index as defined above, the interaction can be regarded as being "on-site."

In order to apply the scaling theory presented above, we redefine the spin-quantization axis and spin operators so that the (redefined) up spin is favored on every site. The resultant Hamiltonians are

$$\begin{aligned} &H_1 = -\sum_{(i,j),\mu} J_1^\mu s_i^\mu s_j^\mu - \sum_i h_1 s_i^z, \\ &H_2 = -\sum_i h_2 S_i^z, \quad H_{12} = -\sum_i J_{12} s_i^z S_i^z, \end{aligned} \tag{13}$$

where $J_1^z = J_1^x = J_1$ and $J_1^y = -J_1$.

Block Hamiltonian and Ising fixed point – For the compound λ-$(BETS)_2FeCl_4$ near the critical temperature, it has been conjectured from the specific heat that the sublattice magnetization in the π-electron system exhibits a temperature dependence similar to that in the two-dimensional Ising model.[1,5,28] In the following, using the block-spin analysis, we show that this conjecture is true.

Let us rewrite the Hamiltonians in Eq. (13) in terms of the block spins $\tilde{s}_i^z$ and $\tilde{S}_i^z$. Because $\xi \gg b \gg 1$, the majority of the spins in the same block behave coherently. Hence, we replace s_i^z and S_i^z on site $i \in B_j(b)$ with $\bar{s}_j^z = b^{-\psi_1}\tilde{s}_j^z$ and $\bar{S}_j^z = b^{-\psi_2}\tilde{S}_j^z$, respectively. This results in

$$H_{12} \approx -\sum_j^{(\mathrm{b})} \tilde{J}_{12} \tilde{s}_j^z \tilde{S}_j^z, \tag{14}$$

where the summation $\sum_j^{(\mathrm{b})}$ is taken over all $b^{-d}N$ block spins and

$$\tilde{J}_{12} \equiv J_{12} b^{d-\psi_1-\psi_2} = J_{12} b^{\gamma_{12}/\nu}, \tag{15}$$

because each d spin is coupled with only a single π spin. The last equation follows from Eq. (6). Because it is plausible that $\gamma_{12} > 0$, which will be confirmed later, we obtain $\tilde{J}_{12} \gg k_B T$ when $\xi \gg b \gg 1$. Therefore, in this limit, the block spins can take the values $\tilde{s}_j^z = \pm s$ and

$$\tilde{S}_j^z = \mathrm{sign}(\tilde{s}_j^z) S \tag{16}$$

only, for which H_{12} falls to the lower limit $-\tilde{N}\tilde{J}_{12} sS \equiv \bar{E}_{12}$, where $\tilde{N} \equiv N/b^d$. This implies that the block spins behave like Ising spins $\tilde{\boldsymbol{s}}_i = (0, 0, \pm s)$ near the critical point. Hence, we obtain the effective Hamiltonian

$$H_1^{\mathrm{eff}} = H_{\mathrm{I}}(\{\tilde{s}_j^z/s\}; \tilde{N}, \tilde{J}_1) - \sum_j^{(\mathrm{b})} \tilde{h}_1 \tilde{s}_j^z + \bar{E}_{12} \tag{17}$$

for subsystem 1 near the critical point, where $\tilde{h}_{\mathrm{I}} = \tilde{h}_1 + \tilde{h}_2 S/s$ and H_{I} is the Ising Hamiltonian defined by

$$H_{\mathrm{I}}(\{\sigma_j\}; \tilde{N}, J) = -\sum_{(i,j)}^{(\mathrm{b})} J\sigma_i\sigma_j.$$

It is difficult to find a form of the coupling constant $\tilde{J}_1$ of the interactions between the block spins in contrast to $\tilde{J}_{12}$; however, it is reasonable to assume that they exist only between the nearest-neighbor block spins.[20] This is because the range of the original interactions does not exceed the lattice constant. Note that the effective Hamiltonian in Eq. (17) for subsystem 1 is not suitable for calculation of physical quantities to which fluctuations of the spins in H_{12} are relevant.

Exponents β_1 and β_2 of the πd antiferromagnet – From Eq. (17), it follows[29] that

$$m(\tau, 0) = b^{-\psi_1} m_{\mathrm{I}}(\tilde{\tau}, 0), \tag{18}$$

where m_{I} denotes the order parameter in the Ising model, which satisfies $m_{\mathrm{I}}(\tau, 0) \sim |\tau|^{\beta_{\mathrm{I}}}$ with β_{I} being the exponent in the Ising model. Hence, Eq. (18) leads to

$$|\tau|^{\beta_1} \sim b^{-\psi_1}|\tilde{\tau}|^{\beta_{\mathrm{I}}} = b^{-\psi_1}|\tau|^{\beta_{\mathrm{I}}} b^{\beta_{\mathrm{I}}/\nu},$$

which results in $\beta_1 = \beta_{\mathrm{I}}$. For the d spins, it follows[30] from Eq. (16) that

$$M(\tau, 0) = b^{-\psi_2} s^{-1} S\, m_{\mathrm{I}}(\tilde{\tau}, 0), \tag{19}$$

which results in $\beta_2 = \beta_{\mathrm{I}}$. Because it is reasonable to assume that $d = 2$ in the present system,[1–13] we can use the result $\beta_{\mathrm{I}} = 1/8$ obtained in a previous theory;[31] hence, we obtain

$$\beta = \beta_1 = \beta_2 = \frac{1}{8}. \tag{20}$$

Note that as shown in the above derivations, $\beta_1 = \beta_2$ (i.e., $\psi_1 = \psi_2$) is a consequence of the on-site π–d interaction.[27] Because $\beta_1 = \beta_2$, the conventional scaling theory is recovered as explained above.

The above argument implies neither that the system is completely reduced to the Ising system nor that all the critical exponents coincide with those of the Ising model. For example, α does not necessarily coincide with the exponent $\alpha_{\rm I} = +0$ of the specific heat of the two-dimensional Ising model, because the principal contribution to the specific heat is from the interaction term H_{12},[9,13] which does not appear in the Ising model.[32] This paradox can be understood by considering the difference in the relevance of $\Delta H_{12} \equiv H_{12} - \bar{E}_{12}$ to the specific heat and order parameter. The probability that $|\tilde{s}^z_j|$ significantly deviates from s due to ΔH_{12} is proportional to $e^{-\beta\tilde{J}_{12}} \ll 1$, which is negligible as long as $\tilde{J}_{12} \gg k_{\rm B}T$ is satisfied. For this reason, in the effective Hamiltonian $H_1^{\rm eff}$ in Eq. (17), the interaction term H_{12} has been replaced with the constant $\bar{E}_{12}$, which results in $\beta_1 = \beta_{\rm I}$. In contrast, ΔH_{12} is relevant to the specific heat because the constant term $\bar{E}_{12}$ does not contribute to it at all. For a similar reason, the effective Hamiltonian $H_1^{\rm eff}$ is not suitable for the calculation of χ_1. If H_{12} were replaced with the constant $\bar{E}_{12}$, the influence of the fluctuations of the d spins on χ_1 would be completely excluded from the consideration. In calculation of physical quantities to which the fluctuations are relevant, such as the specific heat and spin susceptibility, H_{12} cannot be replaced with the constant $\bar{E}_{12}$, even when $b \gg 1$.

Exponent α of the πd antiferromagnet – When almost the entire specific heat comes from $E_{12}(\tau) \equiv \langle H_{12}\rangle$, it holds that

$$c_{\rm heat} \sim N^{-1}\frac{dE_{12}}{dT} \sim |\tau|^{-\alpha},$$

and hence, $E_{12}(\tau) \sim NJ_{12}|\tau|^{1-\alpha}$. From Eq. (14), $E_{12}(\tau)$ satisfies

$$b^d E_{12}(\tau) = -\sum_j \tilde{J}_{12}\langle \tilde{s}^z_j \tilde{S}^z_j\rangle \equiv \tilde{E}_{12}(\tilde{\tau}).$$

If the system is scale invariant, the function $\tilde{E}_{12}$ should have the same form as that of E_{12}, which results in $E_{12}(\tilde{\tau}, \tilde{J}_{12}) = b^d E_{12}(\tau, J_{12})$ and $\psi_1 + \psi_2 = (1-\alpha)/\nu$, where we have used Eq. (15) and $\tilde{\tau} = \tau b^{1/\nu}$. Therefore, from Eq. (4), we obtain the relation

$$\alpha = 1 - \beta_1 - \beta_2. \tag{21}$$

Using Eq. (20), we obtain

$$\alpha = 1 - 2\beta = \frac{3}{4}. \tag{22}$$

The first equation coincides with the relation obtained in a free-energy functional model.[9] The theoretical value of α in Eq. (22) agrees with the experimental value $\alpha = 0.77 \equiv \alpha^{(\rm obs)}$ obtained by Nishio et al.[5] As shown in the previous Letter,[9] due to the two-dimensionality of the system, the specific heat has a logarithmic term in the vicinity of $T_{\rm c}$; however, this term is less singular than the term proportional to $|\tau|^{-\alpha} = |\tau|^{-3/4}$.

Other exponents of the πd antiferromagnet – As mentioned above, we can use Eq. (11) because $\beta_1 = \beta_2$; hence, using Eqs. (2), (11), and (22), we obtain

$$\gamma = 1, \quad \delta = 9, \quad \nu = \frac{5}{8}, \quad \psi = \frac{1}{5}. \tag{23}$$

It also follows from Eq. (11) that $\gamma_{\ell\ell'} = 1$ and $\delta_\ell = \delta_\ell^{(\ell')} = 9$ for arbitrary ℓ and ℓ'.

Possible reason for the error – The difference between the theoretical prediction $\alpha = 0.75$ and the experimental result $\alpha^{(\rm obs)} = 0.77$ can be due to an experimental error and/or theoretical assumptions. The latter includes the localized spin model of the π-electron system and the neglect of the interlayer coupling. If the equation $\beta_1 = \beta_{\rm I}$ has an error, where $\beta_1 = \beta_2 \equiv \beta$ is satisfied, the relation $\alpha^{(\rm obs)} = 1 - 2\beta$ leads to $\beta = 0.115$, which is slightly smaller than $\beta_{\rm I} = 1/8 = 0.125$.

As mentioned above, the localized spin model might not be precise for the π electrons in the λ-$(BETS)_2FeCl_4$ system, which are metallic above $T_{\rm c}$. Even when the π electrons are mobile, the form of H_{12} in Eq. (12) has been considered to be appropriate,[10,33–35] and as long as the system is ordered and $\xi \gg b \gg 1$ is satisfied, the approximation given by Eq. (14) and $\tilde{J}_{12} \gg k_{\rm B}T$ would also remain correct. Hence, the effective Hamiltonian may be expressed in a form of the Ising model; however, in this case, the length of the effective Ising spins for the π electrons might be smaller than $s = 1/2$ because of the Fermi degeneracy.

Summary and conclusion – The scaling theory was extended to coupled magnetic systems, and some relations of the critical exponents were derived. This extension can be relevant to non-ferromagnetic systems in which $\psi_1 \neq \psi_2$, i.e., $\beta_1 \neq \beta_2$; however, the extended theory is practically reduced to the conventional scaling theory for ferromagnets and for systems with $\psi_1 = \psi_2$.

In the application to the πd system in λ-$(BETS)_2FeCl_4$ below $T_{\rm c}$, we obtained the following results in the vicinity of $T_{\rm c}$. First, the π-spin system is reduced to the two-dimensional Ising system near $T_{\rm c}$, which results in $\beta_1 = \beta_{\rm I} = 1/8$. However, this does not imply that the system is completely reduced to the two-dimensional Ising model. Second, $\beta_1 = \beta_2$, which is a consequence of the fact that each d spin principally interacts with only a single π spin. Hence, the conventional scaling hypothesis is recovered. Third, $\alpha = 1 - \beta_1 - \beta_2$, which follows from the fact that H_{12} makes the principal contribution to the specific heat. Fourth, the exponents α, γ, δ, ν, and ψ were obtained as presented in Eqs. (22) and (23). The exponent $\alpha = 3/4$ explains the experimental value $\alpha^{(\rm obs)} = 0.77$.

In conclusion, the block-spin picture explains the observed critical behavior of the specific heat below $T_{\rm c}$. In future studies, the metallic nature of the π-electron system above $T_{\rm c}$ and the metal–insulator transition should be incorporated.

Acknowledgments The author would like to thank Y. Nishio for useful discussions and experimental data.

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28) In the analysis in Ref. 1, the authors evaluated the entropy $S_{\rm exp}$ from experimental values of the specific heat $C_{\rm exp}$, and they evaluated $H_{\rm int} \propto m_{\rm exp}$ from the condition $S_{\rm Sch}(T, H_{\rm int}) = S_{\rm exp}(T)$ at each temperature, where $H_{\rm int}$ and $m_{\rm exp}$ denote the internal field on d spins created by m and the experimental value of m, respectively. $S_{\rm Sch}(T, H_{\rm int})$ is the expression for the entropy of the Schottky model, in which it is assumed that $H_{\rm int}$ does not depend on T. In this procedure, the assumption of the model that $H_{\rm int}$ does not depend on T may appear to contradict the result that $H_{\rm int}$ depends on T. However, it can be easily verified that the relation between $m_{\rm exp}(T)$ obtained by this procedure and $C_{\rm exp}(T)$ coincides with the relation between $m(T)$ and the correct expression for the specific heat of the d-spin system[9]
$$\tilde{c}_2(T) = -bm\frac{dM}{dT},$$
which gives the correct saturation value $k_{\rm B}N\ln 6$ in the high-temperature limit.
29) Equation (18) is derived as follows. Because $e^{-\beta N f} = {\rm Tr}[e^{-\beta H}]$, the order parameter is formally expressed as
$$m(\tau, 0) = \left[\frac{1}{\beta N}\frac{\partial}{\partial h_1}\ln {\rm Tr}[e^{-\beta H}]\right]_{h_1=0},$$
which results in
$$m(\tau, 0) = b^{-\psi_1}\frac{\sum_{\{\tilde{s}_j^z\}}\tilde{s}_l^z e^{-\beta H_1^{\rm eff}}}{\sum_{\{\tilde{s}_j^z\}} e^{-\beta H_1^{\rm eff}}} = b^{-\psi_1} m_{\rm I}(\tilde{\tau}, 0).$$
30) Equation (19) follows from the expression
$$M(\tau, 0) = \left[\frac{1}{\beta N}\frac{\partial}{\partial h_2}\ln {\rm Tr}[e^{-\beta H}]\right]_{h_2=0}.$$
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