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Anomalies in the multicritical behavior of staggered magnetic and direct magnetic susceptibilities of iron group dihalides

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ABSTRACT

The temperature dependencies of magnetic response functions of the anhydrous dihalides of iron-group elements are examined in the neighborhood of the multicritical points (tricritical point, critical end point, and double critical end point) within molecular field approximation. Our findings reveal the fact that the spin -$\frac{1}{2}$ metamagnetic Ising system exhibits anomalies in the temperature dependence of the magnetic response functions for $r<0.3$. In addition, we extensively investigated how an inter- and intra-layer exchange interaction ratio influence the magnetic response properties of these systems. Finally, a comparison has been made with related works.

1. Introduction

Intensive theoretical and experimental efforts have been devoted to investigating the multicritical phenomena for more than half a century. The tricritical point (TCP) is one of the first multicritical points which can be roughly viewed as a point separating a second-order transition line from a first-order transition line and at which the three coexisting phases simultaneously become critical. Itinerant ferromagnets [1], multicomponent fluid mixtures [2], pentenary micro-emulsions [3], ammonium chloride [4], and ^{3}He–^{4}He mixtures [5] are other systems that represent tricritical behavior. In addition, it is shown that there exist TCPs in an experimentally accessible three-dimensional space of the electric field, temperature, and pressure in ferroelectrics [6]. For an extensive review on the critical behavior of the ferroelectrics at Lifshitz points, tricritical points, and tricritical Lifshitz points, see Ref. [7].

On the other hand, a critical end point (CEP) appears when a line of second-order phase transitions terminates at a first-order phase boundary delimiting a new noncritical phase. At this multicritical point, a line of second-order phase transitions intersects with a first-order phase boundary beyond which a new noncritical phase is formed. Binary alloys [8], relaxor ferroelectrics [9], binary fluid mixtures [10], ferromagnets [11], the random-field Ising model [12], and metamagnets [13,14] are the physical systems in which the CEP is common. In 1997, an extensive Monte Carlo (MC) simulation [15] presented the singular behavior of the first-order transition line close to CEP in a classical binary fluid [16–18].

In addition to TCP and CEP, the double critical (bicritical) end point (DCP) appears where two critical lines end simultaneously at a first-order phase boundary. DCPs have been observed in binary and quasi-binary mixtures [19], and there is also some indication of the existence of a DCP in the metamagnet $FeBr_2$ [20,21]. According to mean-field approximation (MFA), the next-nearest-neighbor Ising anti-ferromagnetic model, the layered metamagnet and the random-field Ising model have DCPs [12,13,22]. In addition, MC simulations exhibited the decomposition of the TCP into a DCP and a CEP in three dimensional spin-1 Blume–Capel (BC) model [23] whereas, in $d=2$ only a fully stable TCP is observed [24]. Recently, Plascak and Landau studied the behavior of the $d=2$ spin -$\frac{3}{2}$ BC model near the DCP via extensive MC simulations [25].

The behavior of the staggered and direct susceptibilities in the neighborhood of phase transitions has been a subject of experimental and theoretical research for quite a long time: In 1975, a two lattice model of anti-ferromagnetic phase transitions is discussed in detail using the Gell–Mann–Low formulation of renormalization group methods and Wilson's ϵ expansion [26]. In this study, Alessandrini et al. have obtained the disordering and the staggered susceptibilities in terms of two-point function at zero magnetic field and zero momentum [26]. Later, Landau has obtained MC data for a simple cubic anti-ferromagnet with nearest- and next-nearest-neighbor interactions which reveal asymptotic tricritical behavior of the order parameter and high-temperature susceptibilities which are mean-field-like without

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corrections and in agreement with renormalization-group calculations [27,28]. Using the high-temperature series expansion for the extended Hubbard model, Barkowiak et al. have obtained the series to the sixth order for the staggered magnetic and the charge-ordered susceptibilities [29]. Recently, Li et al. studied the susceptibility of the two-dimensional Ising model on a distorted Kagome lattice by means of the exact solutions and the tensor renormalization-group method [30]. In addition, magnetic behaviors of the β-$Cu_2V_2O_7$ single crystals are investigated by means of magnetic susceptibility measurements [31]. Millis et al. reported the measurements of the magnetization and the susceptibilities of a series of samples of two different variants of the molecular magnet Mn12-ac: the usual, much studied form referred to as Mn_{12}-ac and a new form abbreviated as Mn_{12}-ac-MeOH [32].

Metamagnetic materials are of great interest since it is possible to induce novel kinds of critical behavior by forcing competition between ferromagnetic and anti-ferromagnetic couplings existing in them, in particular by applying an external magnetic field. Magnetic materials that exhibit field-induced transitions can generally be divided in two classes; (i) highly anisotropic, (ii) weakly anisotropic or isotropic. The phase transitions in anisotropic materials (class (i)) are usually characterized by simple reveals of the spin directions which are in contrast with transitions in class (ii). The field-induced transitions in class (ii) materials are related to a rotation of the local spin directions [33]. Iron group dihalides; compounds such as $FeCl_2$, $FeBr_2$, $FeCl_22H_2O$, $FeMgBr_2$, $CoCl_2$ and $NiCl_2$ fall in the first class [34,35]. Some theoretical Hamiltonian models describing the behavior of iron group dihalides have been proposed. MC [36,37] and high-temperature series expansion calculations [38,39] have been performed on the simple cubic lattice Ising model with in-plane ferromagnetic coupling and anti-ferromagnetic coupling between adjacent planes (the metamodel) and on the next-nearest-neighbor (nnn) model with anti-ferromagnetic nearest-neighbor (nn) and ferromagnetic nnn interactions. Recently, a MC simulation has been performed on a quite realistic model of $FeCl_2$ in a magnetic field [40] and this typical metamagnet has also been treated by the high-density expansion method on the two-sublattice collinear Heisenberg–Ising ($S=1$) metamagnet with uniaxial three- and four-ion anisotropies [41,42]. In addition, metamagnetism has also been discussed for magneto-caloric applications: Mukherjee et al. discussed the concept of magnetic cooling by utilizing multi-layers. They provided an experimental evidence of principle, and explained the involved thermodynamics by a two-sublattice mean-field model [43]. Gulpinar and Vatansever have presented a study which investigates the critical behavior of the AC anti-ferromagnetic and ferromagnetic susceptibilities of nnn model with anti-ferromagnetic nn and ferromagnetic nnn interactions [44]. In this paper, the temperature variations of the equilibrium and the non-equilibrium anti-ferromagnetic and ferromagnetic susceptibilities of a metamagnetic system are examined near the critical point.

Harbus and Stanley investigated the spin -$\frac{1}{2}$ Ising metamagnet via high temperature expansions and shown that the staggered susceptibility has an exponent $\frac{5}{4}$ at the critical line, while at the TCP, the direct magnetic susceptibility shows a tricritical exponent of $\frac{1}{2}$ [45]. A study on a dilute hexagonal anti-ferromagnet ($Fe_{0.85}Mg_{0.15}Br_2$) which is under the effect of an axial external field has illustrated the existence of a spin-flop phase line ending at a multicritical point [46]. Katori et al. reported that the diamagnetic impurities in the diluted anti-ferromagnet $Fe_{0.95}Mg_{0.05}$ Br_2 have given rise to random-field criticality along the second-order phase line between $T_N = 13.1$ K and a multicritical point at $T_M = 5$ K, and to a spin-flop transition line between T_M and $T_{CEP} = 3.5$ K [47]. In that study, field variances of the field derivatives of the total magnetization ($\partial m/\partial H$ versus H), and temperature, field dependencies of the complex direct susceptibilities which have been obtained by means of the super-conducting quantum interference device and Faraday rotation techniques have been investigated in detail. In addition, it is discussed by Azevedo et al. that regions of strong noncritical fluctuations are encountered above the multicritical point, apart from the critical phase line $H_C(T)$ [48]. Recently, Chou and Pleimling have investigated the equilibrium behavior of the Ising metamagnets in thin film geometry and shown that the phase diagram of the thin film Ising metamagnets includes an additional intermediate phase in which one of the surface layers has aligned itself with the direction of the external magnetic field. This additional phase transition is of first-order and the first-order transition line ends in a CEP [49]. Although much effort devoted the critical and multicritical behavior of the metamagnetic systems, to the best of our knowledge, there has been no studies investigating the temperature and field dependencies of direct magnetic and staggered magnetic susceptibilities of iron group dihalides by making use of the spin -$\frac{1}{2}$ metamagnetic Ising model in the neighborhood of its multicritical critical points such as CEP, DCP, and TCP.

The layout of this paper is as follows: the derivation of the expressions describing the mean field staggered magnetic and magnetic susceptibilities is represented in Section 1. The results describing the temperature and field dependencies of the direct and staggered magnetic response functions are given in Section 3, and finally Section 4 contains the conclusions and discussions.

2. Equilibrium magnetic response functions of iron group dihalides

In iron group dihalides, there exists two competing interactions which characterize the metamagnetic feature of the materials. The nn interactions in the Hamiltonian of the spin -$\frac{1}{2}$ metamagnetic Ising model should be anti-ferromagnetic $J<0$, whereas the nnn interactions should be ferromagnetic $J'>0$. Because of the existence of the anti-ferromagnetic coupling, for the sake of the analysis, it is convenient to divide the system into two sublattices. Under these conditions, the total Hamiltonian can be written as below:

$$\hat{H} = -\sum_{i,j\epsilon a,k\epsilon b} \sigma_i(J\sigma_j + J'\sigma_k + H + H_s) - \sum_{i,j\epsilon b,k\epsilon a} \sigma_i(J\sigma_j + J'\sigma_k + H - H_s), \quad (1)$$

where $\sigma_i = \pm 1$ is the spin variable and H and H_s are the physical and staggered external magnetic fields.

The mean-field Helmholtz free energy per spin is

$$f = \frac{1}{2}Jz_1 m_a m_b - \frac{1}{4}J'z_2(m_a^2 + m_b^2) - \frac{1}{2}\mu H(m_a + m_b) - \frac{1}{2}N\mu H_s(m_a - m_b)$$
$$-\frac{kT}{4}(4\ln 2 - (1+m_a)\ln(1+m_a) - (1-m_a)\ln(1-m_a)$$
$$-(1+m_b)\ln(1+m_b) - (1-m_b)\ln(1-m_b)). \quad (2)$$

In the constant magnetic field distribution, the sublattice magnetization m_a and m_b are functions of the independent variables T, H, and H_s so that free energy per spin represented by Eq. (2) is a non-equilibrium thermodynamic potential which depends on several order variables [50]. The equilibrium state corresponds to the minimum of f with respect to m_a and m_b. In order to investigate the behavior of the metamagnetic system in the neighborhood of phase transition points, it is more convenient to formulate the system in terms of total and staggered magnetization which are given as follows:

$$m_t = \frac{m_a + m_b}{2}, \quad m_s = \frac{m_a - m_b}{2}. \quad (3)$$

Inserting m_a and m_b in Eq. (3), one obtains the following mean-field equations of state for the spin -$\frac{1}{2}$ metamagnetic Ising model on a cubic lattice as below

$$m_t = \frac{1}{2}\left(\tanh\left(\frac{z_2 r m_a - z_1 m_b + H_r + H_{s_r}}{T_r}\right) + \tanh\left(\frac{z_2 r m_b - z_1 m_a + H_r - H_{s_r}}{T_r}\right)\right),$$
$$m_s = \frac{1}{2}\left(\tanh\left(\frac{z_2 r m_a - z_1 m_b + H_r + H_{s_r}}{T_r}\right) - \tanh\left(\frac{z_2 r m_b - z_1 m_a + H_r - H_{s_r}}{T_r}\right)\right), \tag{4}$$

here $H_r = \mu H/J'$, $H_{s_r} = \mu H_s/J'$, $T_r = k_B T/J'$, $r = J/J'$, $z_1 = 2$, and $z_2 = 4$. The spin -$\frac{1}{2}$ metamagnetic Ising model exhibits field-induced phase transitions. Kincaid and Cohen has shown in their extensive review that metamagnetic Ising model exhibits different types of phase boundaries [13]. In this study, a Landau expansion of the free energy is performed and the possibility of different phase diagrams has been revealed by a careful analysis of the signs of the coefficients. Moreira et al. have extended this analysis considering terms up to 12th order [51]. The Landau expansion consists in expressing the mean-field free energy given by Eq. (2) in a power series of the order parameter (m_s) which vanishes near the critical point:

$$\Psi(T,H,m_s) = \sum_{k=0}^{n} \psi_{2k}(T,H) m_s^{2k}. \tag{5}$$

The spin -$\frac{1}{2}$ metamagnetic Ising model exhibits different phase diagram topologies according to the values and signs of the expansion coefficients as well as the value of the ratio of the exchange interactions ($\eta = z_2 J/z_1 J'$). For $\eta > 0.6$, the phase diagram contains a TCP. In this case there are three types of phase transition points:

(i) If $\psi_2 = 0$ and $\psi_4 > 0$, an ordinary critical point takes place.
(ii) If $\psi_2 = 0$, $\psi_4 = 0$, and $\psi_6 > 0$, one observe a TCP whose location in the field-temperature plane (h_{TCP},t_{TCP}) depends on η

$$h_{TCP} = \frac{t_{TCP}}{2} \ln \frac{1+\sqrt{1-t_{TCP}}}{1-\sqrt{1-t_{TCP}}} + \frac{1-\eta}{1+\eta}\sqrt{1-t_{TCP}},$$
$$t_{TCP} = \left(1 - \frac{1}{3\eta}\right), \tag{6}$$

here $h = \mu H/(z_2 J + z_1 J')$, $t = k_B T/(z_2 J + z_1 J')$ and $H_s = 0$.
(iii) If $\psi_2 = 0$, $\psi_4 < 0$, and $\psi_6 > 0$, a first-order transition appears.

At $\eta = 0.6$ different type of critical behavior is observed. Kincaid et al. named this point as a higher order point (T^*). At this specific point, $\psi_2 = \psi_4 = \psi_6 = 0$ and the coexistence m^*-T^* curve intersects the λ line in a manner similar to this which happens when $\eta > 0.6$ [13,51]. The difference arises from the fact that the lower branch of the coexistence curve approaches the critical point parabolically. The TCP point decomposes into the DCP and CEP for $\eta < 0.6$ with a line of first-order transitions in between separating two anti-ferromagnetic phases [13,14].

The staggered magnetic susceptibility of a metamagnetic system is

$$\chi_s = \lim_{H_{s_r} \to 0} \frac{\partial m_s}{\partial H_{s_r}}. \tag{7}$$

If one uses this definition and the equations of state given in Eq. (9), the staggered magnetic susceptibility can be written as below:

$$\chi_s = \lim_{H_{s_r} \to 0} \left(\frac{c_2 a_{12} - a_{22} c_1}{a_{21} a_{12} - a_{22} a_{11}}\right). \tag{8}$$

The relations for a_{11}, a_{12}, a_{21}, a_{22}, c_1, and c_2 are given in Appendix A.

The direct magnetic susceptibility (χ_t) corresponds to the response function of a system due to the variance of physical field and it can be expressed as

$$\chi_t = \lim_{H_{s_r} \to 0} \frac{\partial m_t}{\partial H_r}. \tag{9}$$

Following the similar steps we have used in obtaining staggered magnetic susceptibility, one obtains χ_t as

$$\chi_t = \lim_{H_{s_r} \to 0} \left(\frac{b_{22} d_1 - b_{12} d_2}{b_{11} b_{22} - b_{12} b_{21}}\right), \tag{10}$$

b_{11}, b_{12}, b_{21}, b_{22}, d_1, and d_2 are given in Appendix B. Before discussing the findings of mean field analysis done in this paper one should note that the role of fluctuations, consequently the self-consistency of mean field theory can be assessed by applying a real space version the Ginzburg criterion: Nielsen and Birgenau shown that the concept of marginal dimensionality d^* emerges in a natural way [52]. As it is discussed in Ref. [52], the marginal dimensionality is given by the following expression:

$$d^* = (\gamma + 2\beta)/(\nu - m), \tag{11}$$

where m is a parameter which takes different values for the systems with short range interactions, TCPs and percolation phenomena. For $d > d^*$, MFA represents a self-consistent picture at least insofar as the critical exponents are concerned. Ising model with only nn interactions, which is a paradigmatic model of cooperative phenomena with short range interactions, corresponds to the case $m=0$ and $d^* = 4$. Consequently, MFA is not self-consistent for the $d=3$ Ising model. If d is further away from d^*, i.e. for two dimensional Ising model and $d=2$ anti-ferromagnet K_2CoF_4, one should expect even more larger deviations from the mean-field exponents [53,54]. On the other hand, the systems such as the Ising type metamagnet $FeCl_2$ and ^3He–^4He mixtures which have TCPs are characterized with $d^*_{tcp} = 3$. Thus, we should note that the analysis and critical exponents given in this study are in accordance with conventional there-dimensional systems with logarithmic correction terms. The experimental results reveal the fact that the phase diagrams of the $FeCl_2$ [55,56] and ^3He–^4He mixtures [57–59] are well depicted by MFA with marginal dimensionality corrections [52].

3. Results

Fig. 1(a) and (b) represents the behavior of staggered and direct magnetic susceptibilities of the spin -$\frac{1}{2}$ metamagnetic Ising model in the neighborhood of TCP for $r=1.0$. One can see from the figure that the staggered susceptibility (χ_s) increases rapidly with increasing temperature and diverges at the TCP. Meanwhile, there is a discontinuity in the direct magnetic susceptibility (χ_t) at the TCP. We should note that Ţukovic et al. presented a study on the dilute metamagnetic Ising Model within effective field theory (EFT) which takes account of the spin correlations [60,61]. Comparing Fig. 12 of Ref. [60] with Fig. 1(b) of the present paper, one can see that our results are in accordance with the results of EFT. We should stress that the discontinuity in the direct susceptibility (χ_t) is an artifact of MFA. It is discussed in detail in the fourth chapter of Ref. [50] that the discontinuity in the non-ordering parameter's response function is a characteristics of a second-order transition. Here we observe that same behavior is valid also for the TCP. In addition, Ţukovic et al. shown the existence of a finite jump in the inverse direct susceptibility of the dilute metamagnetic Ising model at T_C as well as T_{TCP} (see Ref. [60, Fig. 9 and Fig. 12]). Fig. 2 illustrates the temperature variation of the tricritical direct magnetic susceptibility for various values of the ratio of the exchange interactions (r). One can see from this

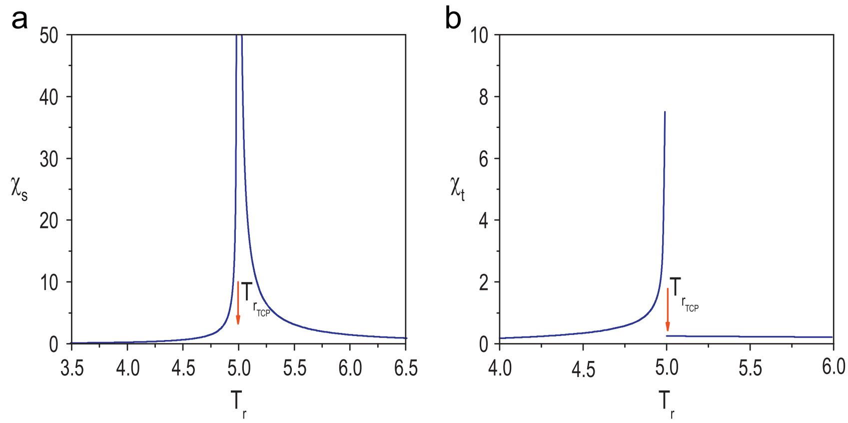

Fig. 1. Behaviors of staggered (a) and magnetic (b) susceptibilities each as a function of reduced temperature for $H_r = \mu H/J' = 1.354$ and $r = J/J' = 1.0$. For these values, the system has a TCP. And also, staggered susceptibility increases rapidly with increasing temperature and diverges as the temperature approaches to the TCP.

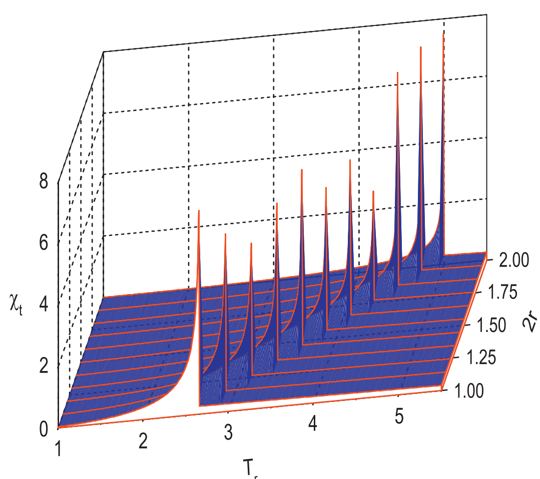

Fig. 2. The temperature variation of the tricritical direct magnetic susceptibility for various values of the ratio of the exchange interactions. Here the arrows illustrate the phase transition temperatures.

figure that the amplitude of the ferromagnetic susceptibility grows considerably high values for $r > 1.78$.

One of the characteristic behavior of the spin -$\frac{1}{2}$ metamagnetic Ising model for strong anti-ferromagnetic case is the existence of the re-entrance phenomena. One can see this fact in the phase diagram of the system for $r < 0.3$. For high values of the magnetic field, the spin -$\frac{1}{2}$ metamagnetic Ising model is in a disordered state for $T_r \to 0$ and there is a transition from disorder to order at a finite temperature. In addition, the system undergoes another second-order phase transition between ordered and disordered phases in high temperature regime (see Fig. 6(a) and (b) of the present paper).

Fig. 3(a) illustrates the temperature dependence of anti-ferromagnetic susceptibility for $H = H_{r_{DCP}} = 1.994$ which corresponds to the field value of the DCP of the spin -$\frac{1}{2}$ Ising model for $r = 0.2$. For $r = 0.2$ and $H = H_{r_{DCP}} = 1.994$, firstly, the system undergoes a second-order transition from paramagnetic phase to anti-ferromagnetic phase at $T_{r_{N_1}} < T_{r_{DCP}}$. The staggered susceptibility diverges as $T \to T_{r_{N_1}}$ and $T \to T_{r_{DCP}}$. In addition, one can clearly see from Fig. 3(a) that there exist a non-critical maximum at the ordered phase. This maximum corresponds to an anomaly in the multicritical behavior of iron group dihalides. It is important to emphasize that Selke has reported the existence of the two lines of anomalies in the MFA phase diagram of the spin -$\frac{1}{2}$ Ising model at which the temperature derivative of the total magnetization exhibits an isomagnetic maximum below the transition point (see Ref. [14, Fig. 3]). In that study, the anomalies are related to the competing ordering tendencies of the external field and the interlayer couplings in a metamagnetic crystal. Pleimling and Selke have investigated the anomalies of the specific heat and the total magnetization in the ordered phase of related spin models to $FeBr_2$ by extensive numerical simulations [62]. Their results suggest that the anomalies usually do not correspond to a sharp phase transition [62]. We should also note that, there are experimental data which emphasizes the anomalies for quite some time [13]. There have been various experimental studies on the field-induced Griffiths phase in Ising-type metamagnets such as $FeBr_2$, $FeCl_2$ and $Fe_{1-x}Zn_xF_2$ [63,64]. Katori et al. commented the phase line due to anomalies is probably due to symmetric non-diagonal exchange in $Fe_{0.95}Mg_{0.05}Br_2$ [47]. Fig. 3(b) shows the temperature variation of ferromagnetic susceptibility for $H = H_{r_{DCP}}$. In this case the signature of the second-order transition from paramagnetic phase to anti-ferromagnetic phase is a discontinuity in the direct magnetic susceptibility which is in accordance with the literature [44,50]. In addition, there exists a special multicritical point which separates the two different anti-ferromagnetic phases (AFI and AFII). This special continuous phase transition is of fourth-order and the direct magnetic susceptibility represents a discontinuity at the DCP. In addition, direct susceptibility has a finite jump at $T_{r_{N_2}}$, the regular second-order transition temperature from anti-ferromagnetic phase to paramagnetic phase. Fig. 4(a) and (b) shows the temperature dependencies of staggered and direct susceptibilities of the spin -$\frac{1}{2}$ metamagnetic system for $r = 0.2$ and $H = H_{r_{CEP}} = 1.99176$. At this value of the reduced physical magnetic field, the system undergoes two phase transitions of different character. The first transition which occurs at the CEP which is of fourth-order [13]. This transition is between the disordered phase at lower temperatures and the anti-ferromagnetic phase at higher temperature regime. One can easily observe from Fig. 4(a) and (b) that the staggered susceptibility diverges at CEP and the direct magnetic susceptibility shows a discontinuity. Similar to the anomaly at $H = H_{r_{DCP}}$, both χ_t and χ_s make non-critical maximums in the anti-ferromagnetic phase. In Fig. 5(a) and (b) we have given the temperature variations of the magnetic response functions of the system for different constant reduced physical field values. One can see from these figures that the broad maximum in the ordered phase declines with decreasing the amplitude of the physical external magnetic field. Finally, the line of anomalies in the staggered and direct susceptibilities is depicted in Fig. 6. Here $(T-H)_\chi$ denotes the field and temperature

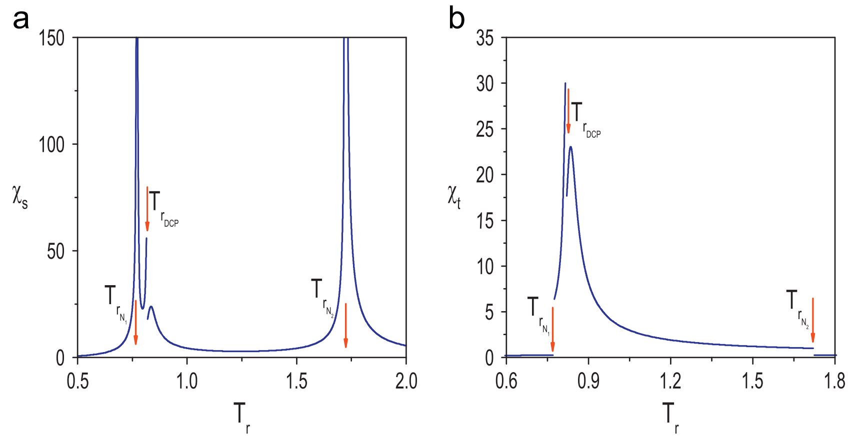

Fig. 3. The temperature dependencies of staggered and total susceptibilities in the neighborhood of the DCP and second-order phase transition point which takes place for the value of the reduced magnetic field $H_{r_{DCP}} = 1.994$ for r=0.2. Here the arrows illustrate the phase transition temperatures.

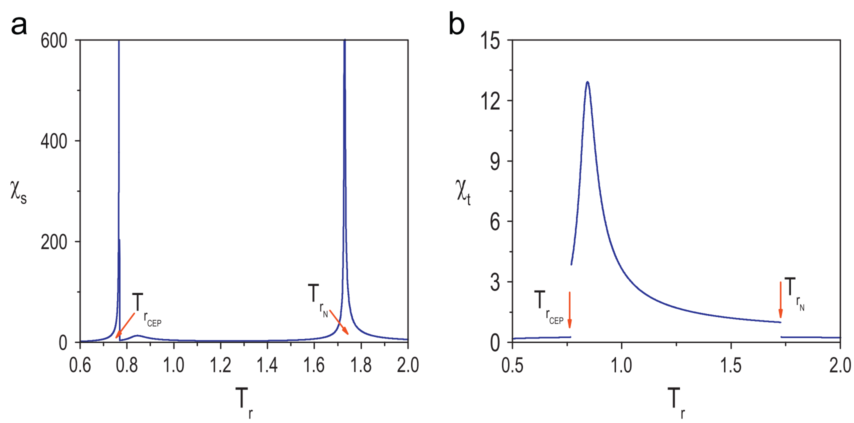

Fig. 4. The temperature dependencies of staggered and total susceptibilities in the neighborhood of the CEP and second-order phase transition point which takes place for the value of the reduced magnetic field $H_{r_{CEP}} = 1.99176$ for r=0.2. Here the arrows illustrate the phase transition temperatures.

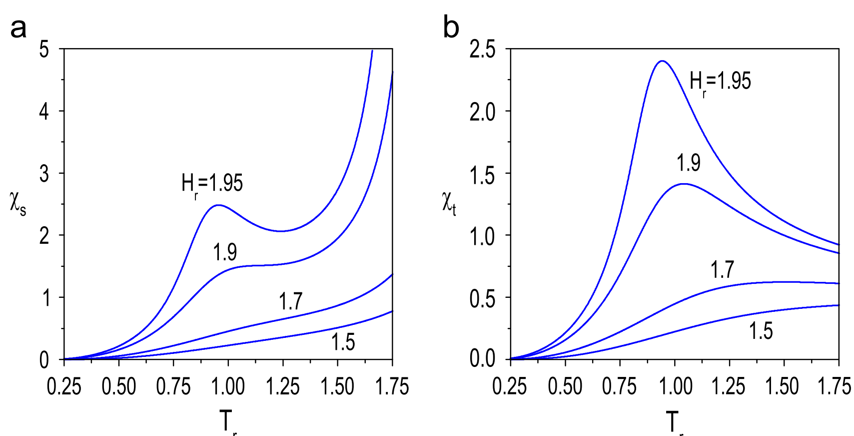

Fig. 5. The behavior of (a) the staggered susceptibility χ_s and (b) the direct magnetic susceptibility χ_t as a function of the reduced temperature, where $T_r = k_B T/J'$ is for several values of reduced field, $H_r = \mu H/J'$.

values at which both the staggered and direct susceptibilities exhibit a broad maximum in the ordered phase as exemplified in Figs. 3 and 4.Unlike the anomalies discussed by Selke, the broad maximum does not diverges as one approaches the double critical endpoint. Further, the anomalies the magnetic response functions of the metamagnetic Ising system disappear for the case $r \geq 0.3$ where the critical end point and the double critical end points emerge to a TCP. We should note that there is no re-entrance in the phase diagram for $r \geq 0.3$.

4. Conclusions and discussions

In this paper, the temperature dependencies of the magnetic response functions of spin $-\frac{1}{2}$ Ising model are studied in the neighborhood of multicritical points. The expressions that describe the staggered (anti-ferromagnetic) and direct (ferromagnetic) susceptibilities are derived by making use of the MFA. The findings of this study can be summarized as follows: the direct susceptibility exhibits discontinuity not only at the second-order transition point but also at

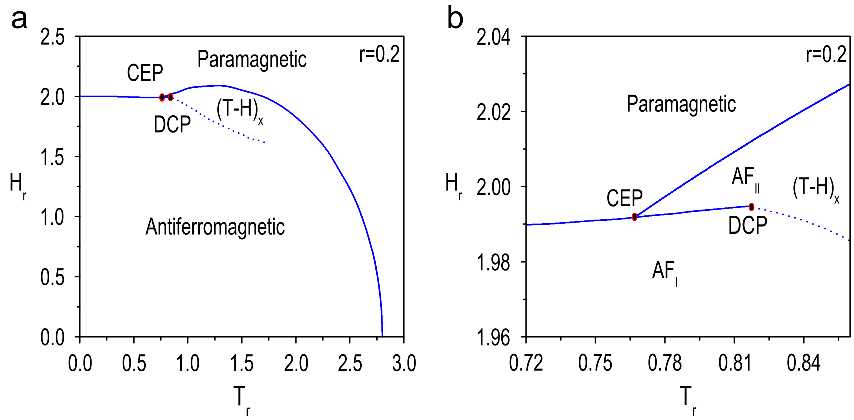

Fig. 6. (a) The calculated MFA phase diagram of the metamagnetic Ising model for $r=J/J'=0.2$ in the temperature-field plane. (b) Detailed phase diagram in the neighborhood of the CEP and the DCP. The dashed lines denote the anomalies in the staggered and direct susceptibilities.

multicritical points such as TCP, CEP, and DCP. In addition, the both magnetic response functions of the metamagnetic Ising model exhibit non-critical maximums in the ordered phase at the region of the $H_r - T_r$ where the system shows re-entrance phenomena.

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Appendix A

The coefficients $a_{11}, a_{12}, a_{21}, a_{22}, c_1$, and c_2 which are used in Eq. (9) are defined as follows:

$$a_{11} = 1 - \left(1 - \tanh\left(\frac{-2(m_t - m_s) + 4r(m_t + m_s) + H_r + H_{s_r}}{T_r}\right)^2\right)(2+4r)T_r^{-1},$$

$$a_{12} = 1 - \left(1 - \tanh\left(\frac{-2(m_t - m_s) + 4r(m_t + m_s) + H_r + H_{s_r}}{T_r}\right)^2\right)(-2+4r)T_r^{-1},$$

$$a_{21} = -1 + \left(1 - \tanh\left(\frac{-2(m_t + m_s) + 4r(m_t - m_s) + H_r - H_{s_r}}{T_r}\right)^2\right)(2+4r)T_r^{-1},$$

$$a_{22} = 1 - \left(1 - \tanh\left(\frac{-2(m_t + m_s) + 4r(m_t - m_s) + H_r - H_{s_r}}{T_r}\right)^2\right)(-2+4r)T_r^{-1},$$

$$c_1 = \left(1 - \tanh\left(\frac{-2(m_t - m_s) + 4r(m_t + m_s) + H_r + H_{s_r}}{T_r}\right)^2\right)T_r^{-1},$$

$$c_2 = \left(-1 + \tanh\left(\frac{-2(m_t + m_s) + 4r(m_t - m_s) + H_r - H_{s_r}}{T_r}\right)^2\right)T_r^{-1}. \quad (12)$$

Appendix B

The coefficients $b_{11}, b_{12}, b_{21}, b_{22}, d_1$, and d_2 in Eq. (10) are defined as follows:

$$b_{11} = 1 - \left(1 - \tanh\left(\frac{-2(m_t - m_s) + 4r(m_t + m_s) + H_r + H_{s_r}}{T_r}\right)^2\right)(2+4r)T_r^{-1},$$

$$b_{12} = 1 - \left(1 - \tanh\left(\frac{-2(m_t - m_s) + 4r(m_t + m_s) + H_r + H_{s_r}}{T_r}\right)^2\right)(-2+4r)T_r^{-1},$$

$$b_{21} = -1 + \left(1 - \tanh\left(\frac{-2(m_t + m_s) + 4r(m_t - m_s) + H_r - H_{s_r}}{T_r}\right)^2\right)(2+4r)T_r^{-1},$$

$$b_{22} = 1 - \left(1 - \tanh\left(\frac{-2(m_t + m_s) + 4r(m_t - m_s) + H_r - H_{s_r}}{T_r}\right)^2\right)(-2+4r)T_r^{-1},$$

$$d_1 = \left(1 - \tanh\left(\frac{(-2(m_t - m_s) + 4r(m_t + m_s) + H_r + H_{s_r})}{T_r}\right)^2\right)T_r^{-1},$$

$$d_2 = \left(1 - \tanh\left(\frac{(-2(m_t + m_s) + 4r(m_t - m_s) + H_r - H_{s_r})}{T_r}\right)^2\right)T_r^{-1}. \quad (13)$$

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