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## MAGNETIZATION AND MAGNETOCALORIC EFFECT IN ANTIFERROMAGNETS WITH COMPETING ISING EXCHANGE AND SINGLE-ION ANISOTROPIES

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*The magnetization of a two-sublattice Ising antiferromagnet with easy-plane single-ion anisotropy, which is accompanied by two phase transitions, has been studied. The both phase transitions are induced by the magnetic field. One of them is isostructural, i.e., the system symmetry remains unchanged and a transition between two antiferromagnetic states with different sublattice magnetizations takes place. The other phase transition occurs when the antiferromagnetic state transforms into the ferromagnetic one. At both phase transitions, the field dependence of the system entropy has two successive positive jumps, which is not typical of ordinary antiferromagnets. On the other hand, if the temperature of the system is higher than the tricritical temperature of the isostructural phase transition, there appears a continuous maximum in the field dependence of the entropy.*

*Keywords:* phase transitions of the first kind, antiferromagnet, Ising model, easy-plane single-ion anisotropy, magnetocaloric effect.

### 1. Introduction

It is known that the magnetization of a strongly anisotropic antiferromagnet (AFM) can occur in a jump-like manner as a metamagnetic phase transition of the first kind (PT1) [1–8]. Such PTs induced by an external magnetic field were detected for the first time in those AFMs for which the magnitude of the magnetic anisotropy field was found to be larger than the effective intersublattice exchange field, which is responsible for the intensity of the intersublattice spin-spin exchange interaction. A substantial contri-

but ion to the study of the magnetic properties of the antiferromagnetic iron-group dihalides with weak intersublattice exchange, magnetostriction, magnetic PTs, and other phenomena was made by the famous Ukrainian experimental physicist S.M. Ryabchenko and his disciples (see, e.g., works [10–15]). This publication is dedicated to his jubilee.

The process of AFM magnetization can sometimes be accompanied by several metamagnetic PTs [16–20], with each of them having a jump-like character. This situation is typical, in particular, of multisublattice AFMs [21, 22]. In work [16], a physically simpler case was considered, when two meta-

magnetic PTs take place even in a two-sublattice AFM. Such a situation can occur, if an AFM with a more complicated character of interionic interactions is magnetized; for example, if the intersublattice interaction has an Ising-like limit and orients the spins in the sublattices along the easy axis; at the same time, the single-ion magnetic anisotropy has the easy-plane character rather than the easy-axis one, as was in work [23]. The easy-plane single-ion magnetic anisotropy can stabilize the singlet state or, in a more general case, the triplet state [16, 23] with the spin projection magnitude less than the nominal value in either of the sublattices.

In this work, we made an attempt to study the sequence of two metamagnetic PTs in the Ising AFM [24–26] with the easy-plane single-ion magnetic anisotropy [27–29] in the case of ionic spins  $S = 1$  and at finite temperatures,  $T \neq 0$ . The problem of magnetic ordering of an Ising AFM with spins  $S = 1/2$  has been exhaustively studied in work [30], where it was shown that only one PT1 – either metamagnetic or magnetic isostructural – can take place in such a model system. In the cited work, the free energy was used in the form of a power series expansion of the Landau potential in the magnitude of the antiferromagnetism vector (assuming that this parameter is small) up to the eighth order of magnitude inclusive.

However, this approximation of the Landau theory cannot be used in the case of two metamagnetic PTs with finite (not small) magnetization jumps, even if the invariants with large power exponents of the order parameter are taken into consideration. Moreover, the results can only be obtained, if the free energy is analyzed numerically. Furthermore, the magnetic isostructural PT1 in AFMs with  $S = 1/2$  exists only in a narrow temperature interval near the tricritical point [30]. The same result was obtained for AFMs with easy-axis single-ion anisotropy and  $S = 1$  [31]. However, it should be noted that, as follows from the results of work [23], in the case of easy-plane single-ion anisotropy and ion spins  $S = 1$ , the isostructural magnetic PT must take place within the whole temperature interval extending from  $T = 0$  to the tricritical point.

Another important factor for the study of a system with several metamagnetic PTs is the possibility to analyze its magnetic entropy. It will be shown below that the behavior of such systems and, hence, the

behavior of the magnetocaloric effect differ from the same effect in AFMs with one metamagnetic PT [32].

## 2. Model Hamiltonian

Let us present the Hamiltonian of a two-sublattice system with competing Ising and single-ion anisotropies in the form

$$\hat{H} = \frac{1}{2} \sum_{f_\alpha, g_\beta} I_{f_\alpha, g_\beta} \hat{S}_{f_\alpha}^z \hat{S}_{g_\beta}^z + D \sum_{f_\alpha} (\hat{S}_{f_\alpha}^z)^2 - H_z \sum_{f_\alpha} \hat{S}_{f_\alpha}^z. \quad (1)$$

Here,  $I_{f_\alpha, g_\beta}$  are the constants of the exchange (Ising) interaction between magnetic ions with spins  $S = 1$  (their locations are described by the numbers  $f_\alpha$  and  $g_\beta$ , where the subscripts  $\alpha$  and  $\beta$  mark the sublattices ( $\alpha, \beta = 1, 2$ ); the intrasublattice exchange constant is negative, if  $\alpha = \beta$ , and positive, if  $\alpha \neq \beta$ ; the exchange has an interlattice character, thus providing the initial AFM structure);  $\hat{S}_{f_\alpha}^z$  are spin-projection operators on the  $Z$  axis;  $D$  is the constant of the positive (i.e., “perpendicular” to the  $Z$  axis) easy-plane anisotropy ( $D > 0$ ); and the external magnetic field is assumed to be parallel to the Ising axis ( $H_z \parallel Z$ ).

On the basis of Hamiltonian (1), the interaction energy between two ions from different sublattices can be written as follows:

$$E = \frac{1}{2} \sum_{\alpha\beta} I_{\alpha\beta} s_\alpha s_\beta + D \sum_{\alpha} q_\alpha - H_z \sum_{\alpha} s_\alpha. \quad (2)$$

Here,

$$I_{\alpha\alpha} = p_{f_\alpha g_\alpha} I_{f_\alpha g_\alpha}, \quad I_{\alpha\neq\beta} = p_{f_\alpha g_\beta} I_{f_\alpha g_\beta};$$

$p_{f_\alpha g_\beta}$  and  $p_{f_\alpha g_\alpha}$  are the numbers of the nearest neighbors of an ion in its “native” and “foreign”, respectively, sublattices;  $s_\alpha$  are the thermodynamic averages of the operators  $\hat{S}_{f_\alpha}^z$ ; and  $q_\alpha$  are the thermodynamic averages of the operators  $(\hat{S}_{f_\alpha}^z)^2$  (they correspond to the quadrupole spin moments).

At  $T = 0$ , the averages in Eq. (2) are determined as the quantum-mechanical ones with the use of the ionic wave function  $|\psi_\alpha\rangle = \sum_{m_\alpha} C_{m_\alpha} |m_\alpha\rangle$ , where  $m_\alpha = 0$  or  $1$ , and  $C_{m_\alpha}$  are variational parameters (see below). The resulting evident equalities read

$$s_\alpha = |C_{+1_\alpha}|^2 - |C_{-1_\alpha}|^2, \quad q_\alpha = |C_{+1_\alpha}|^2 + |C_{-1_\alpha}|^2. \quad (3)$$

When determining the states, PT points, and phase stability boundaries, we can use the Lagrange function written as a functional of the parameters  $C_{m_\alpha}$  [33, 34]. It looks like

$$L = \frac{1}{2} \sum_{\alpha\beta} I_{\alpha\beta} (|C_{+1_\alpha}|^2 - |C_{-1_\alpha}|^2) (|C_{+1_\beta}|^2 - |C_{-1_\beta}|^2) + D \sum_{\alpha} (|C_{+1_\alpha}|^2 + |C_{-1_\alpha}|^2) - H_z \sum_{\alpha} (|C_{+1_\alpha}|^2 - |C_{-1_\alpha}|^2) + \sum_{\alpha} \lambda_{\alpha} (1 - |C_{+1_\alpha}|^2 - |C_{0_\alpha}|^2 - |C_{-1_\alpha}|^2), \quad (4)$$

where  $\lambda_{\alpha}$  are the Lagrange factors, and the normalization condition  $\sum_{m_\alpha} |C_{m_\alpha}|^2 = 1$  was taken into account.

At  $T = 0$ , the states are determined by minimizing the Lagrange function with respect to the wave function parameters,  $L(C_{m_\alpha})$  [33, 34]. In so doing, it is easy to verify that the following states can be stable:

1. the AFM state with  $s_1 = 1, s_2 = -1$ , and the energy  $E_{AFM1} = I_{11} + I_{12} + 2D$ ;

2. the AFM state with  $s_1 = 1, s_2 = 0$ , and the energy  $E_{AFM2} = \frac{1}{2}I_{11} + D - H_z$  (in this state, if  $H_z \neq 0$ , the ions in either of the sublattices are in the van Fleck nonmagnetic ground state  $|0\rangle$ ; if this spin configuration is ground at  $H_z = 0$ , this state should be defined as ferrimagnetic);

3. the FM state with  $s_1 = 1, s_2 = 0$ , and the energy  $E_{FM} = I_{11} + I_{12} + 2D - 2H_z$ .

It is evident that the energy  $E_{AFM1}$  does not depend on the magnetic field, whereas the energy  $E_{AFM2}$  does. The FM state energy also depends on the magnetic field magnitude. Such peculiarities in the field behavior of the ground state energy are responsible for the appearance of two sequential PTs in the course of magnetization. The critical field of the first PT1 between two AFM states equals  $H_I = -\frac{1}{2}I_{11} + I_{12} - D$ . The critical field of the second PT1 between the AFM phase with the nonmagnetic sublattice and the FM state equals  $H_{II} = \frac{1}{2}I_{11} + I_{12} + D$ . As was shown in work [23], the inequality  $H_I < H_{II}$  may hold true. The field of transition from the non-magnetized AFM state to the FM state equals  $H_c = I_{12}$ . Hence, for two PTs to take place, there must be  $H_I < H_c$ , i.e., the inequality  $-\frac{1}{2}I_{11} - D < 0$  must be satisfied, which is only possible in the case of easy-plane single-ion anisotropy.

It should be noted that Hamiltonian (1) with  $D < 0$ , i.e., in the case of easy-axis single-ion anisotropy, was analyzed in works [17, 31].

### 3. Free Energy of the System

In the case  $T \neq 0$ , the equations of states and the boundaries of the phase stability regions are determined from the minimum of the free energy  $F$ . The latter is defined in the standard way by the formula  $F = E - T\sigma$ , which includes the entropy

$$\sigma = - \sum_{m_\alpha} p_{m_\alpha} \ln p_{m_\alpha},$$

where  $p_{m_\alpha}$  is the thermodynamic probability for an ion to be in the state with  $m_\alpha = (0, \pm 1)$ . The required normalization condition follows from the equality for the total probability  $p_{1_\alpha} + p_{-1_\alpha} = 1$ . Then the thermodynamic averages can be written in the simple form

$$s_\alpha = p_{1_\alpha} - p_{-1_\alpha}, \quad q_\alpha = p_{1_\alpha} + p_{-1_\alpha}, \quad (5)$$

and the expression for the free energy can be easily represented as a functional of the thermodynamic averages  $s_\alpha$  and  $q_\alpha$ ,

$$F = \frac{1}{2} \sum_{\alpha\beta} I_{\alpha\beta} s_\alpha s_\beta + D \sum_{\alpha} q_\alpha - H_z \sum_{\alpha} s_\alpha + T \sum_{\alpha} \left( \frac{q_\alpha + s_\alpha}{2} \ln \frac{q_\alpha + s_\alpha}{2} + \frac{q_\alpha - s_\alpha}{2} \times \ln \frac{q_\alpha - s_\alpha}{2} + (1 - q_\alpha) \ln(1 - q_\alpha) \right). \quad (6)$$

The equilibrium states and the boundaries of their stability can be found by minimizing the free energy  $F(s_\alpha, q_\alpha)$  [33]. In the general case, the problem of finding the sequence of PTs is nonlinear and has to be solved numerically.

### 4. Field Dependences of Magnetization

The field dependences of magnetization can be calculated using the functional expressions (4) and (6). For convenience, the model parameters should be made dimensionless. Then the dimensionless temperature is  $t = T/T_N$ , where  $T_N = |I_{11}| + I_{12}$ , is the Néel temperature of the AFM without taking its easy-plane anisotropy into account. The intrasublattice exchange constant, single-ion anisotropy constant, magnetic field, and free energy are normalized by the

same quantity, namely,  $k = I_{12}/T_N$ ,  $d = D/T_N$ , and  $h = H_z/T_N$ .

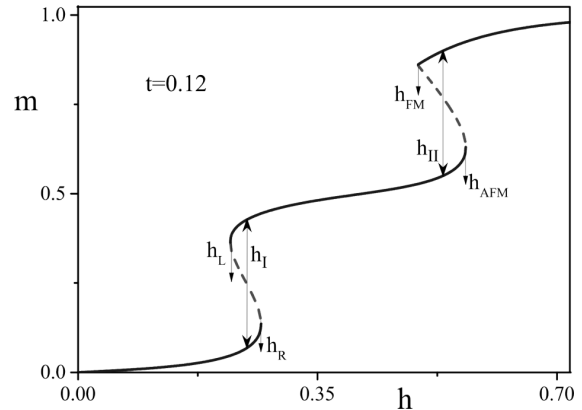
As an example, Fig. 1 demonstrates the field dependence of the magnetization  $m = (s_1 + s_2)/2$  calculated at  $t = 0.12$  in the case where the dimensionless anisotropy constant  $d = 0.45$  and the intra-sublattice exchange constant  $k = 0.6$ . At this parameter combination, a sequence of two metamagnetic PT1s takes place.

The first PT1 is determined by the equality between the free energies. Its critical field equals  $h_I = 0.245$  at  $t = 0.12$  and  $h_I = 0.25$  at  $t = 0$ . At the point  $h_I(t = 0.12)$ , there arises a magnetization jump, which is indicated by a double arrow. The point  $h_I$  is a point of the metamagnetic PT1 from the AFM state with  $m \approx 0$  and  $s_1 \approx -s_2$  (the spin in sublattice 1 is directed along the field,  $s_1 \uparrow \uparrow h$ , and the spin in sublattice 2 against the field,  $s_2 \downarrow \downarrow h$ ) into the AFM state with  $m = 1/2$ , for which  $s_1/|s_2| \gg 1$ . Therefore, the first metamagnetic PT occurs between two AFM states, which are different in the magnitude of the average spin in sublattice 2, although being characterized by almost the same spin value in sublattice 1.

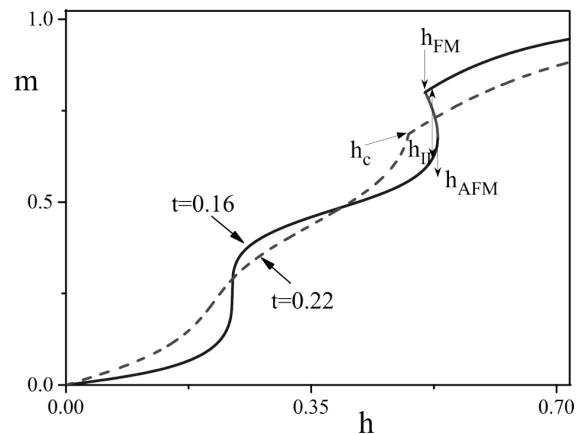
The critical field of the second transition equals  $h_{II} = 0.54$  at  $t = 0.12$  and  $h_{II} = 0.55$  at  $t = 0$ . At the point  $h_{II}(t = 0.12)$ , there arises a magnetization jump, which corresponds to a metamagnetic PT1 from the AFM state into the FM one with  $m \approx s_1 = s_2$ . This jump is also indicated by a double arrow in Fig. 1. As one can see, the values of both critical fields corresponding to the PT1s weakly depend on the temperature.

In Fig. 1, the magnetization of steady states is indicated by solid curves. The AFM phase is stable within the field interval  $[0, h_{AFM}]$ , and the equilibrium phase within the field interval  $[0, h_{II}]$ . The less magnetized AFM phase is stable within the field interval  $[0, h_R]$ , and the more magnetized AFM phase remains stable within the field interval  $[h_L, h_{AFM}]$ . The FM phase is stable at the fields  $h > h_{FM}$  and equilibrium at  $h > h_{II}$ .

Figure 2 illustrates the field dependence of the magnetization at the temperatures  $t = 0.16$  (solid curve) and  $0.22$  (dashed curve). Only one metamagnetic PT1 takes place at the temperature  $t = 0.16$ . This is a PT between the AFM and FM phases. The magnetization jump is also indicated in the figure by a double arrow, and the critical field equals  $h_{II}$ . The AFM phase is stable at the fields  $h < h_{AFM}$  and equi-



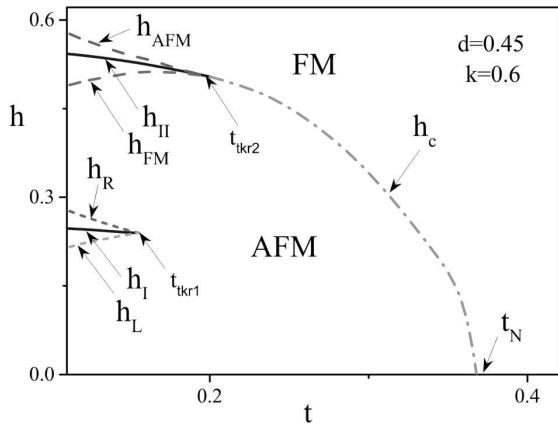
**Fig. 1.** Field dependence of the magnetization,  $m(h)$ . Notations:  $h_I$  and  $h_{II}$  are critical fields of PT1s,  $h_{AFM}$  is the stability boundary of the AFM state,  $h_{FM}$  is the stability boundary of the FM state, and  $h_L$  and  $h_R$  are the stability fields of the AFM phase with different spin values for sublattice 2



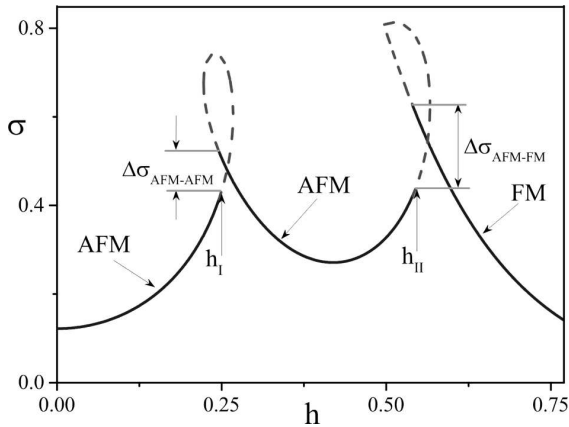
**Fig. 2.** Field dependence of the magnetization,  $m(h)$ . Notations:  $h_{II}$  is the critical field of PT1 between the AFM and FM phases,  $h_{AFM}$  the stability boundary of the AFM state,  $h_{FM}$  the stability boundary of the FM state, and  $h_c$  the critical field of the PT2 between the AFM and FM phases

librium at  $h < h_{II}$ . The FM phase remains stable at the fields  $h > h_{FM}$  and is equilibrium at  $h > h_{II}$ . At the same time, if  $t = 0.22$ , the transition from the AFM phase into the FM one occurs continuously, being a PT of the second kind (PT2) at the point  $h_c$ . In this case, the AFM state exists at  $h < h_c$  and is continuously transformed into the FM state at the point  $h = h_c$ .

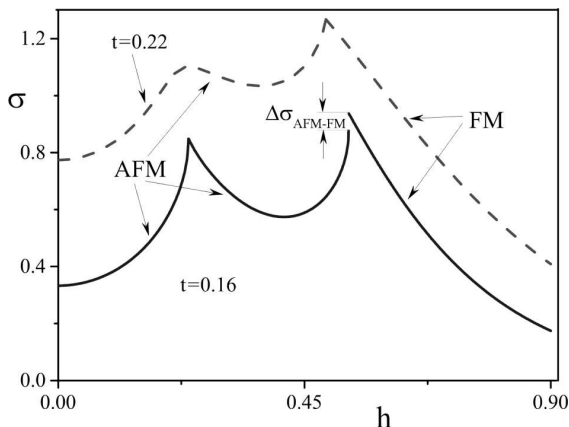
By calculating the values of the critical fields that are indicated by arrows in Figs. 1 and 2, it is possible to determine their temperature dependences.



**Fig. 3.**  $h-t$  phase diagram. Solid curves denote PT1s, dashed curves are phase stability boundaries, and the dash-dotted curve corresponds to the PT2



**Fig. 4.** Field dependence of the magnetic entropy,  $\sigma(h)$ , at two metamagnetic PTs. The parameter values  $t = 0.12$ ,  $d = 0.45$ , and  $k = 0.6$



**Fig. 5.** Field dependences of the magnetic entropy,  $\sigma(h)$ , at  $t = 0.16$  (solid curve) and  $0.22$  (dashed curve)

### 5. $h-t$ Phase Diagram

By plotting the temperature dependences of the fields  $h_I(t)$ ,  $h_{II}(t)$ ,  $h_{AFM}(t)$ ,  $h_{FM}(t)$ ,  $h_L(t)$ ,  $h_R(t)$ , and  $h_c(t)$ , we can find the corresponding  $h-t$  phase diagram. For the magnetization types considered in Figs. 1 and 2, this diagram is shown in Fig. 3.

The curves corresponding to the PT1s and the phase stability fields converge at the tricritical points  $t_{tkr1}$  and  $t_{tkr2}$ . One can see that  $t_{tkr2} < t_{tkr1}$ . The tricritical point  $t_{tkr2}$  is also an endpoint for the curve  $h_c(t)$  corresponding to the PT2 between the AFM and FM phases. At high temperatures, the curve  $h_c(t)$  tends to the Néel point.

### 6. Magnetic Entropy

Figure 4 demonstrates the field dependence of the magnetic entropy  $\sigma(h)$  at  $t = 0.12 < t_{tkr1}$  and for the model parameters used when calculating the magnetization dependences in Figs. 1 and 2. The solid and dashed sections correspond to the entropy in the equilibrium and nonequilibrium, respectively, states.

At the critical fields  $h_I$  and  $h_{II}$  corresponding to the PT1s, the magnetic entropy has jumps: the jump  $\Delta\sigma_{AFM-AFM}$  at the isostructural PT and the jump  $\Delta\sigma_{AFM-FM}$  at the AFM-FM phase transition. The behavior of the entropy near the critical fields  $h_I$  and  $h_{II}$  differs little qualitatively and even quantitatively. Therefore, in the course of magnetization, the maximum value of the magnetocaloric effect will actually be reached at the lower critical field  $h_I$ . The field dependence of the entropy plotted in Fig. 4 and corresponding to the two-lattice Ising AFM with easy-plane single-ion anisotropy differs substantially from its counterpart for the AFM with one metamagnetic PT [32]. The curve in Fig. 4 has two maxima and two jumps of the entropy. In the absence of a single-ion anisotropy, there is only one maximum and one jump of the entropy at the PT point between the AFM and FM phases.

In Fig. 5, the field dependences of the magnetic entropy  $\sigma(h)$  are shown for  $t = 0.16$  and  $0.22$ . In the former case,  $t_{tkr1} < t < t_{tkr2}$  and there is only one PT1 in the course of magnetization, namely, from the AFM phase to the FM one, at which a finite jump  $\Delta\sigma_{AFM-FM}$  of the entropy takes place. It is of interest that this jump is preceded by a maximum in the dependence  $\sigma(h)$ , where the latter changes continuously. This maximum is a result of the rapid magne-

tization, as one can see from Fig. 2. At the same time, the magnitude of the entropy jump  $\Delta\sigma_{\text{AFM-FM}}$  turns out insignificant. At the higher temperature  $t = 0.22$  corresponding to the condition  $t > t_{tkr2}$ , there are no entropy jumps, but the dependence  $\sigma(h)$  has a cusp point, which testifies to the presence of the PT2 from the AFM phase into the FM one. In low fields, the dependence  $\sigma(h)$  may have a wide maximum, which disappears at temperatures much higher than  $t_{tkr2}$ .

## 7. Conclusions

In this work, it was shown that two PT1s may take place at the magnetization of an Ising AFM with easy-plane single-ion anisotropy and ionic spins  $S = 1$ . Owing to the easy-plane magnetic anisotropy, the first PT is isostructural. It is a transition between two AFM phases with different lattice magnetization values. At  $T = 0$ , it is a magnetic quantum PT [33–38], at which the ground state of ions in sublattice 2 becomes a singlet one,  $| -1 \rangle \rightarrow | 0 \rangle$ , and the single-ion anisotropy energy becomes lower. At  $T \neq 0$ , the minimization of the single-ion anisotropy energy at the isostructural PT is associated with a jump-like change in the ionic states: the population is maximum for ions in sublattice 2 with a spin projection of  $-1$  before the transition point and with the zero spin projection after it. Therefore, the isostructural PT can be considered as a magnetic quantum PT even at  $T \neq 0$ . An inhomogeneous multidomain AFM state, the domains of which differ from one another by the sublattice spin magnitudes, can exist in the interval of stability fields from  $h_L(t)$  to  $h_R(t)$ .

The PTs induced by the magnetic field lead to the appearance of jumps in the magnetic entropy. The entropy jumps are positive for both PT1s, and the field dependence of the magnetic entropy at the isostructural PT differs little from its counterpart at the PT into the FM phase. At higher temperatures, when the isostructural PT disappears, a rather narrow maximum can be observed in the field dependence of the magnetic entropy. It becomes more acute when the temperature approaches the tricritical temperature of the isostructural PT. It is important that, because of the competition between the Ising exchange interaction and the easy-plane single-ion anisotropy, the isostructural PT1 occurs at a magnetic field that is much lower than the critical field of the phase transition from the AFM phase into the FM one. Therefore, owing to such a competition between the interactions,

the appearance of a substantial magnetocaloric effect can be expected at lower external magnetic fields.

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НАМАГНІЧУВАННЯ І МАГНІТОКАЛОРИЧНИЙ ЕФЕКТ В АНТИФЕРОМАГНЕТИКУ З КОНКУРУЮЧИМИ ІЗІНГІВСЬКОЮ ОБМІННОЮ ТА ОДНОІОННОЮ АНІЗОТРОПІЯМИ

Резюме

Досліджено намагнічування двопідґраткового ізінгівського антиферомагнетика з легкоплощинною одноіонною анізотропією, яке може супроводжуватися двома фазовими перетвореннями 1-го роду. Перше, індукване магнітним полем, є ізоструктурним, коли симетрія системи не змінюється і відбувається перехід між двома антиферомагнітними станами з різними величинами намагніченості підґраток. Друге, також індукване магнітним полем, перетворення має місце при зміні стану системи з антиферомагнітного на феромагнітний. При обох цих фазових перетвореннях поведінка ентропії в залежності від поля містить два послідовних і додатних за величиною стрибки її величини, що не є типовим для класичних антиферомагнетиків. З іншого боку, коли температура системи перевищує трикритичну температуру ізоструктурного фазового переходу, в залежності ентропії від поля виникає неперервний максимум.