05,08

Magnetocaloric effect in a ferromagnet/antiferromagnet structure with exchange coupling

© *M.A. Kuznetsov*

Institute for Physics of Microstructures, Russian Academy of Sciences,
Nizhny Novgorod, Russia

E-mail: kuznetsovm@ipmras.ru

Received March 6, 2025
Revised March 6, 2025
Accepted May 5, 2025

Within the framework of the Landau theory of second-order phase transitions, a ferromagnet/antiferromagnet structure is considered, in which the ferromagnetic and antiferromagnetic layers are connected by exchange interaction. In the vicinity of the Neel temperature, such an interaction leads to a modification of the order parameter of the antiferromagnet. The magnetocaloric effect (isothermal change in magnetic entropy) of the system under consideration is calculated when the magnetization direction of the ferromagnet changes relative to the easy anisotropy axis of the antiferromagnet. The cases of compensated and uncompensated antiferromagnet boundaries are considered. The results obtained suggest the possibility of using ferromagnet/antiferromagnet multilayer for cooling micro- and nanoelectronic devices.

Keywords: magnetic cooling, magnetic proximity effect, antiferromagnets, Neel temperature.

DOI: 10.61011/PSS.2025.06.61706.6HH-25

1. Introduction

Magnetocaloric effect (MCE) is a change of magnetic material temperature during adiabatic magnetization or demagnetization [1]. This effect may be used to arrange a magnetic cooling cycle that will result in heat removal from the load into atmosphere, in particular at room temperature [2]. Such coolers are expected to be environmentally friendly, compact and energy-saving [3,4]. Note that even when MCE is irreversible, a magnetic cooling cycle still can be arranged [5]. The need for applying very strong magnetic fields in order to achieve the required MCE is one of the reasons why the magnetic cooling technique hasn't been widely used yet. Thus, to change the temperature of a reference magnetocaloric material, Gd, only by 1 K, a field of about 10 kOe shall be applied, and to change the temperature by 10 K, a field of about 50 kOe will be necessary [6].

The problem of very strong fields may be avoided by using nanostructured magnetocaloric materials instead of bulk ones. The former may be actually used for cooling microelectronic and nanoelectronic devices. In [7], a focus was made on a ferromagnet/paramagnet multilayer, where exchange fields occurring at the interfaces magnetize the paramagnetic layers acting as magnetocaloric materials. Application of a weak ($10-100$ Oe) external field induces reorientation of exchange fields (ferromagnetic layer magnetizations) leading to a magnetic entropy (temperature) change of such system. A number of later theoretical [8–10] and experimental [8,11–16] works demonstrated a significant exchange enhancement of MCE. Recently [17], the idea of using exchange fields for MCE enhancement has been transferred to a ferromagnet|antiferromagnet structure, where antiferromagnetic layers served as magnetocaloric materials. Since the critical field between the antiferromagnetic and paramagnetic phases of a bulk antiferromagnetic material depends very much on the external magnetic field orientation with respect to an easy axis of anisotropy [18], then simple rotation of this field leads to a change of magnetic state and,consequently, to MCE in the antiferromagnetic material. In [17], focus was made on an antiferromagnetic material with a compensated interface, i. e. where this interface contained equal amounts of atoms of both sublattices, and the exchange field induced at the ferromagnetic and antiferromagnetic interfaces played a role of a magnetic field.

This study investigated MCE in a ferromagnet/antiferromagnet exchange-coupled structure in the uncompensated interface case, i. e. when the interface contains atoms of only one of two antiferromagnetic sublattices. Such situation takes place when a structure is sputtered in external magnetic field and is identified by the presence exchange bias effect [19,20]. Exchange field that occurs at the interface modifies the order parameter of the antiferromagnetic material, and application of a weak (~ 100 Oe) external field leads to a change of direction of this exchange field and, consequently, to MCE. As we see below, magnetocaloric properties of the ferromagnet|antiferromagnet system depend considerably on the type of interface. The findings may turn out to be useful for the development of microscale and nanoscale coolers operating in weak magnetic fields.

2. Compensated interface case

Let's consider a two-layer ferromagnet/antiferromagnet structure, where the ferromagnetic and antiferromagnetic layers have thicknesses h and d, respectively, and the interface coincides with the plane $z = 0$ (see Figure 1, a). Suppose the antiferromagnetic material has two sublattices with magnetizations $\mathbf{m}_1$ and $\mathbf{m}_2$ ($m_1 = m_2$), and its magnetic anisotropy easy axis is oriented along the x axis. The system temperature T is assumed to be close to the Neel temperature T_N of the antiferromagnetic material, so $T \le T_\mathrm{N}$ and the Curie temperature of the ferromagnetic material is much higher than T_N. The latter makes it possible to neglect the ferromagnetic state change caused by the exchange coupling with the antiferromagnetic material and to consider the ferromagnetic magnetization $\mathbf{M}$ to be uniform.

Let's first consider the compensated interface case (Figure 1, b). According to the Landau theory of second-order phase transitions [18], free energy per unit area of the given system F can be represented as an expansion in powers of the Neel vector (order parameter) $\mathbf{L} = \mathbf{m}_1 - \mathbf{m}_2$ and magnetization $\mathbf{m} = \mathbf{m}_1 + \mathbf{m}_2$ of the antiferromagnetic material, and of combinations and derivatives thereof:

$$F = F_v + F_s, \tag{1}$$

$$F_v = \int_0^\infty \left(\frac{\gamma}{2}\left(\frac{d\mathbf{L}}{dz}\right)^2 - \frac{\alpha\tau}{2}\mathbf{L}^2 + \frac{\beta}{4}\mathbf{L}^4 + \frac{K_a}{2}(L_y^2 + L_z^2)\right) dz, \tag{2}$$

$$F_s = \int_0^\infty \left(\frac{\gamma_0}{2}\left(\frac{d\mathbf{m}}{dz}\right)^2 + \frac{1}{2\chi}\mathbf{m}^2 + \frac{\Delta}{2}(\mathbf{mL})^2 + \frac{\delta}{2}\mathbf{m}^2\mathbf{L}^2\right) dz$$

$$- J(\mathbf{mM})|_{z=0}, \tag{3}$$

where $\alpha, \beta, \gamma, \gamma_0, \Delta$ and δ are the positive phenomenological parameters, $K_a > 0$ is the magnetic anisotropy constant, $\tau = (T_\mathrm{N} - T)/T_\mathrm{N}$, χ is the susceptibility, J is the interlayer -exchange interaction constant. Suppose the antiferromagnetic layer is quite thick so the magnetic state at its opposite interface (at $z = d$) may be considered as unperturbed. Therefore integration in equations (2) and (3) is taken over the half-space $z > 0$. Note that interface compensation is reflected in the term — $J(\mathbf{mM})|_{z=0} = -J(\mathbf{m}_1\mathbf{M})|_{z=0} - J(\mathbf{m}_2\mathbf{M})|_{z=0}$, i.e. the exchange interaction links the ferromagnetic magnetization to the magnetizations of both antiferromagnetic sublattices. We assume that the spatial scale of $\mathbf{L}$ is much larger than that of $\mathbf{m}$, which may be always fulfilled in the vicinity of T_N. Then after calculation of $\mathbf{m}$ and substitution in Equation (3), we get (see the Appendix)

$$F_s \approx \frac{\chi^{3/2}J^2}{4\gamma_0^{1/2}}(\delta\mathbf{M}^2\mathbf{L}^2 + \Delta(\mathbf{ML})^2)|_{z=0}, \tag{4}$$

where nonessential constant term was truncated. As can be seen, in case of compensated interface, exchange interaction between the ferromagnetic and antiferromagnetic materials, on the one hand, leads to antiferromagnetic order suppression ($\delta\mathbf{M}^2\mathbf{L}^2$), on the other hand, to the surface spin-flop effect, i.e. to the deviation of vector $\mathbf{L}$ from the easy axis ($\Delta(\mathbf{ML})^2$). Let

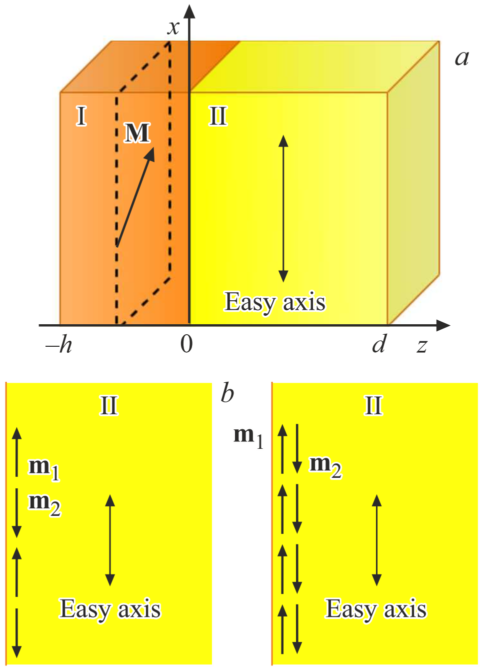

Figure 1. Schematic diagrams a) of the planar ferromagnet|antiferromagnet (I|II) structure and antiferromagnetic interfaces: b) compensated and c) uncompensated. Double arrows denote the anisotropy easy axis of the antiferromagnetic material.

$$\mathbf{M} = M(\hat{\mathbf{x}}\cos\phi + \hat{\mathbf{y}}\sin\phi)$$

and

$$\mathbf{L}(z) = L(z)\big(\hat{\mathbf{x}}\cos\theta(z) + \hat{\mathbf{y}}\sin\theta(z)\big).$$

Then, minimizing the free energy F with respect to the modulus L and θ, we get the following equations and boundary conditions:

$$\frac{d^2L}{dz^2} + \left(\frac{1}{l_c^2} - \left(\frac{d\theta}{dz}\right)^2 - \frac{1}{l_d^2}\sin^2\theta\right)L - \frac{\beta}{\gamma}L^3 = 0, \tag{5}$$

$$L^2\frac{d^2\theta}{dz^2} + 2L\frac{dL}{dz}\frac{d\theta}{dz} - \frac{1}{l_d^2}L^2\sin\theta\cos\theta = 0, \tag{6}$$

$$\left.\frac{dL}{dz}\right|_{z=0} = \frac{\chi^{3/2}J^2M^2}{2\gamma\gamma_0^{1/2}}L\big(\delta + \Delta\cos^2(\theta - \phi)\big)\big|_{z=0}, \tag{7}$$

$$\left.\frac{d\theta}{dz}\right|_{z=0} = -\frac{\chi^{3/2}J^2M^2\Delta}{2\gamma\gamma_0^{1/2}}\cos(\theta - \phi)\sin(\theta - \phi)\big|_{z=0}, \tag{8}$$

where $l_c = \sqrt{\gamma/(\alpha\tau)}$ and $l_d = \sqrt{\gamma/K_a}$ are the correlation length and domain wall thickness of the antiferromagnetic material. With $z \to \infty$, θ and modulus L shall reach unperturbed values, i.e. $\theta(z \to \infty) = 0$ and $L(z \to \infty) = L_0 = \sqrt{\alpha\tau/\beta}$. By differentiating the free energy with respect to temperature taking into account (5)–(8), we find the volume density of magnetic entropy $s = -(\partial F/\partial T)/d$:

$$s = -\frac{\alpha}{2T_{\mathrm{N}}}\overline{L^2} = -\frac{\alpha}{2dT_{\mathrm{N}}}\int\limits_0^\infty L^2(z)dz. \qquad (9)$$

It can be seen that magnetic entropy is defined by the average square of the order parameter over the thickness of the antiferromagnet $\overline{L^2}$, the magnitude of which depends, according to boundary conditions (7) and (8), on the direction of vector $\mathbf{M}$. It can be seen from the energy symmetry (4) that the largest isothermal change of magnetic entropy Δs is provide by changing the direction of $\mathbf{M}$ from perpendicular ($\perp$) to parallel ($\|$) with respect to the antiferromagnetic easy axis.

Concerning only with the change of the order parameter modulus, we find the conditions under which $\theta = 0$. Then we assume that θ is small. Then in the first approximation in equation (5), $\theta = 0$ may be assumed. In this case the spatial scale of the modulus L will be defined by l_c. Suppose $l_d/l_c \ll 1$, which can be always satisfied in the vicinity of T_{N}. Then, in equation (6), the term proportional to dL/dz may be neglected, after which the equation will be written as:

$$\frac{d^2\theta}{dz^2} - \frac{1}{l_d^2}\sin\theta\cos\theta = 0, \qquad (10)$$

and solution to it with $\phi = 0, \pi(\|)$ may be written as

$$\cos\theta_{\|}(z) = \mathrm{th}\,\frac{z + z_\theta^{\|}}{l_d}, \qquad (11)$$

where $z_\theta^{\|}$ may be determined from

$$\cos\theta_{\|}(0) = \left(\frac{J_\theta}{J}\right)^2. \qquad (12)$$

Here

$$J_\theta^2 = 2\gamma\gamma_0^{1/2}/(\chi^{3/2}M^2 l_d\Delta).$$

The obtained solution is valid with $J \geq J_\theta$. With $J \leq J_\theta$, $\theta_{\|}(z) = 0$ is the solution to equation (10). Note that in case of $\phi = \pm\pi/2(\perp)$ according to (4), we have $\theta_\perp(z) = 0$ with any J. With $\theta(z) = 0$ from (5) and (7) for $\phi = \pm\pi/2$ and $\phi = 0, \pi$, we get the following dependences of the order parameter modulus on the z coordinate:

$$L_{\perp(\|)}(z) = L_0\mathrm{th}\,\frac{z + z_{\perp(\|)}}{\sqrt{2}l_c}. \qquad (13)$$

$z_{\perp(\|)}$ is determined by

$$\frac{L_{\perp(\|)}(0)}{L_0} = \sqrt{1 + \left(\frac{J}{J_{\perp(\|)}}\right)^4} - \left(\frac{J}{J_{\perp(\|)}}\right)^2, \qquad (14)$$

where

$$J_\perp^2 = 2^{3/2}\gamma\gamma_0^{1/2}/(\chi^{3/2}M^2 l_c\delta)$$

and

$$J_{\|}^2 = 2^{3/2}\gamma\gamma_0^{1/2}/\big(\chi^{3/2}M^2 l_c(\Delta + \delta)\big).$$

It can be seen that the antiferromagnetic order is suppressed as J grows. In case of weak interlayer exchange ($J \ll J_{\perp(\|)}$), the order parameter is not so different from the volume quantity, i. e. $L_{\perp(\|)}(0)/L_0 \approx 1 - (J/J_{\perp(\|)})^2$. In the opposite case ($J \gg J_{\perp(\|)}$) we have $L_{\perp(\|)}(0)/L_0 \approx (J_{\perp(\|)}/J)^2/2 \ll 1$.

Calculation of the isothermal entropy change $\Delta s = s_{\|} - s_\perp$ according to (9) brings us to the following result:

$$\Delta s = \Delta s_{\max}\left(\frac{L_\perp(0)}{L_0} - \frac{L_{\|}(0)}{L_0}\right), \qquad (15)$$

$$\Delta s_{\max} = \frac{\alpha}{2T_{\mathrm{N}}}\frac{\sqrt{2}l_c L_0^2}{d} = \frac{(2\gamma\tau)^{1/2}\alpha^{3/2}}{2T_{\mathrm{N}}\beta d}. \qquad (16)$$

Thus, Δs is defined by the order parameters at the interface. Then to reach $\Delta s_{\max}$, it is necessary that $L_\perp(0) \approx L_0$ and $L_{\|}(0) \approx 0$. This situation occurs when condition $J_{\|} \ll J \ll J_\perp$ is fulfilled. Since $(J_{\|}/J_\perp)^2 = \delta/(\Delta + \delta)$, then $\Delta \gg \delta$ shall be in any case. The latter is satisfied at least for most antiferromagnetic materials [21]. Note that, since $(J_{\|}/J_\theta)^2 \propto l_d/l_c \ll 1$, then as J increases, significant decrease in the order parameter modulus at the interface takes place before it deviates from the easy axis direction. Then, when $l_d/l_c \ll 1$ is satisfied, $\theta(z) = 0$ may be assumed at any J.

Let's estimate the Landau theory parameters for an adequately studied antiferromagnet, MnF_2. Comparison with experiment [22–25] gives

$$T_{\mathrm{N}} \approx 67.33\,\mathrm{K},\ \alpha/T_{\mathrm{N}} = 1\,\mathrm{K}^{-1},$$

$$\beta \approx 9.50\cdot10^{-7}\,\mathrm{erg}^{-2}\cdot\mathrm{G}^2\cdot\mathrm{cm}^6,$$

$$\Delta \approx 1.55\cdot10^{-4}\,\mathrm{erg}^{-2}\cdot\mathrm{G}^2\cdot\mathrm{cm}^6,$$

$$\delta \approx 5.40\cdot10^{-6}\,\mathrm{erg}^{-2}\cdot\mathrm{G}^2\cdot\mathrm{cm}^6,$$

$$\chi \approx 1\cdot10^{-3},\ K_a \approx 2.33.$$

γ may be estimated from the equality of energy $k_{\mathrm{B}}T$ and exchange energy in the point of phase transition, i. e. $\gamma \approx k_{\mathrm{B}}T_{\mathrm{N}}/(am_s^2)$ [18], where a is the lattice parameter, m_s is the antiferromagnet saturation magnetization. For MnF_2, we find $\gamma^{1/2} \approx 4\,\mathrm{nm}$. γ_0 will be assumed equal to γ. Figures 2 and 3, a show the dependences of $L_{\perp(\|)}(0)$ and Δs on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet|MnF_2 structure at $h = 20\,\mathrm{nm}$. To satisfy the boundary condition at $z = d$, antiferromagnet thickness d was set to $5l_c$. Then the order parameter at $z = d$ almost reaches the unperturbed L_0 at any temperature. It can be seen from Figure 3, a that Δs grows with distance from T_{N}. However, it is impossible to determine the temperature at which Δs will take the maximum value within the developed model, because $l_d/l_c \ll 1$ was assumed to be satisfied.

Switching between states with $\phi = \pm\pi/2$ and $\phi = 0, \pi$ may be carried out by applying the external magnetic

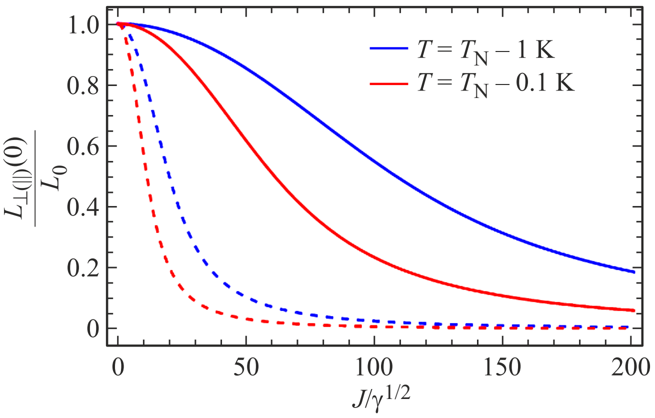

Figure 2. Dependence of the order parameter modulus at the compensated interface $L_{\perp(\|)}(0)$ on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet|MnF_2 structure. Solid lines correspond to the perpendicular direction ($\perp$) of the ferromagnet magnetization $\mathbf{M}$ with respect to the anisotropy easy axis of the antiferromagnet, dashed lines correspond to the parallel direction ($\|$).

field $\mathbf{H}$. Let's find the magnitude of such switching field H_{sw}. For this, note first that, taking into account (5)−(8), F may be rewritten as

$$F = -\frac{\beta}{4}\int_0^\infty L^4(z)dz. \tag{17}$$

In view of $J_\| \ll J_\perp$, we have $L_\|(z) \le L_\perp(z)$ and the state with $\phi = \pm\pi/2$ has the lowest free energy. Then the volume density of effective free energy of the given system in the presence of magnetic field may be written as

$$f_{\text{eff}} = \frac{1}{2}K_{\text{eff}}(\mathbf{M}\cdot\hat{\mathbf{x}})^2 - (\mathbf{MH}), \tag{18}$$

where $K_{\text{eff}} = 2\Delta F/(M^2h) > 0$ is the effective anisotropy constant (easy-plane), $\Delta F = F_\| - F_\perp$ is the free energy difference (17). For switching between the states, $\mathbf{H}$ shall be applied along the easy axis of the antiferromagnet. Minimizing f_{eff} with respect to ϕ, we find $H_{sw} = K_{\text{eff}}M$. Figure 3, b shows the dependences of H_{sw} on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet|MnF_2 structure at $h = 20$ nm and $d = 5l_c$. It can be seen that switching between the states takes place in a quite small field, the highest value of which is about 150 Oe. Δs corresponding to this field is comparable with Δs in bulk Gd in the 10 kOe field [26].

3. Uncompensated interface case

We assume the antiferromagnet interface to be uncompensated (Figure 1, c). Then in the contribution of F_s (equation (3)) instead of $-J(\mathbf{mM})|_{z=0}$ take the magnetization interaction $\mathbf{M}$ with the magnetization of only one of two sublattices (for definiteness, $\mathbf{m}_1$), i. e.

$$F_s = \int_0^\infty \left(\frac{\gamma_0}{2}\left(\frac{d\mathbf{m}}{dz}\right)^2 + \frac{1}{2\chi}\mathbf{m}^2 + \frac{\Delta}{2}(\mathbf{mL})^2 + \frac{\delta}{2}\mathbf{m}^2\mathbf{L}^2\right)dz - \frac{J}{2}(\mathbf{mM})|_{z=0} - \frac{J}{2}(\mathbf{ML})|_{z=0}, \tag{19}$$

where $\mathbf{m}_1 = (\mathbf{m}+\mathbf{L})/2$ was used. Then, after the calculation of $\mathbf{m}$ and substitution into F_s (see the Appendix), we get

$$F_s \approx \frac{\chi^{3/2}J^2}{16\gamma_0^{1/2}}\left(\delta\mathbf{M}^2\mathbf{L}^2 + \Delta(\mathbf{ML})^2\right)|_{z=0} - \frac{J}{2}(\mathbf{ML})|_{z=0}, \tag{20}$$

Equation (20) differs from equation (4), that is valid for the compensated interface, on the one hand, by the coefficient of the first summand and, on the other hand, by the additional summand that is linear in J and $\mathbf{L}$. As can be seen below, appearance of this summand will affect considerably the magnetocaloric properties of the given

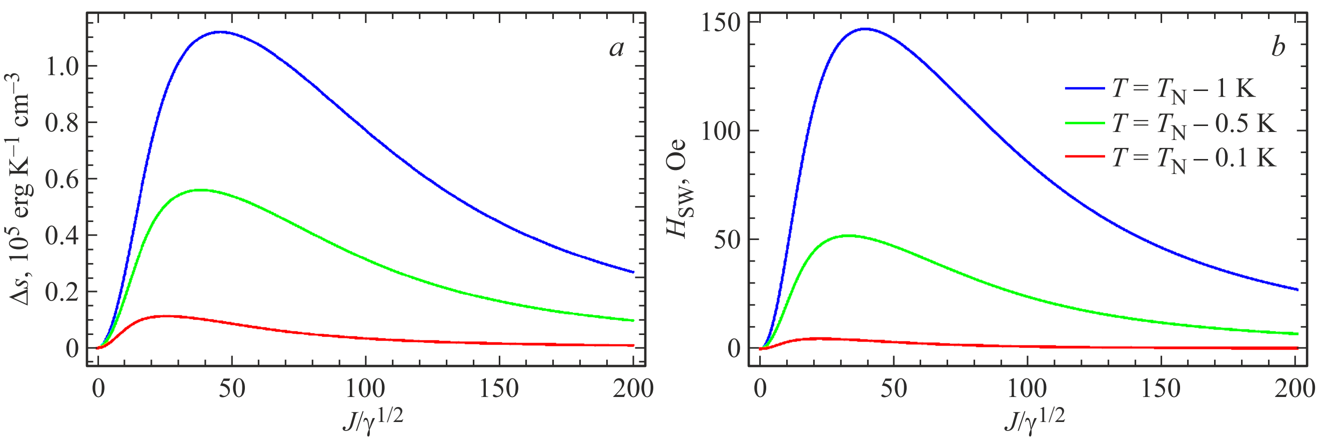

Figure 3. Dependence of a) the isotermal entropy change Δs and b) switching field H_{sw} on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet|MnF_2 structure for the compensated interface.

system. Minimizing F with respect to L and θ, we again get equations (5) and (6), however, now the boundary conditions with $z = 0$ will be written as:

$$\left.\frac{dL}{dz}\right|_{z=0} = \frac{\chi^{3/2}J^2M^2}{8\gamma\gamma_0^{1/2}} L\big(\delta + \Delta\cos^2(\theta-\phi)\big)\big|_{z=0} - \frac{JM}{2\gamma}\cos(\theta-\phi)\big|_{z=0}, \tag{21}$$

$$L^2\left.\frac{d\theta}{dz}\right|_{z=0} = -\frac{\chi^{3/2}J^2M^2\Delta}{8\gamma\gamma_0^{1/2}} L^2\cos(\theta-\phi)\sin(\theta-\phi)|_{z=0} + \frac{JM}{2\gamma}L\sin(\theta-\phi)\big|_{z=0}. \tag{22}$$

With $z \to \infty$, θ and modulus L reach unperturbed values, i. e. $\theta(z \to \infty) = 0$ and $L(z \to \infty) = L_0$. Differentiating the free energy with respect to temperature, taking into account (5), (6), (21) and (22), we will again get equation (9) for the volume density of the system's magnetic entropy. As for the compensated interface, the magnetic entropy is defined by the average square of the order parameter over the thickness of the antiferromagnet $\overline{L^2}$, the magnitude of which depends on the direction of vector $\mathbf{M}$.

Let's first consider the parallel orientation of $\mathbf{M}$ with respect to the easy axis of antiferromagnet anisotropy. Unlike the compensated interface, the states with $\phi = 0$ and $\phi = \pi$ differ from each other due the appearance of the linear summand in equation (20). We restrict ourselves only to the case with $\phi = 0$. As before, we will first assume that θ is small. Then in the first approximation in equation (5), $\theta = 0$ may be assumed. Taking into account the new boundary conditions (21), we have

$$L_{\parallel}(z) = \begin{cases} L_0\,\mathrm{cth}\,\frac{z+z_{\parallel}}{\sqrt{2}l_c}, & J \le J_0, \\ L_0\mathrm{th}\frac{z+z_{\parallel}}{\sqrt{2}l_c}, & J \ge J_0, \end{cases} \tag{23}$$

where $J_0 = 2J_{\parallel}^2/J_c$, $J_c = \sqrt{2}\gamma L_0/(Ml_c)$. $z_{\parallel}$ may be determined from

$$\frac{L_{\parallel}(0)}{L_0} = \sqrt{1 + \left(\frac{J}{2J_{\parallel}}\right)^4 + \frac{J}{J_c}} - \left(\frac{J}{2J_{\parallel}}\right)^2. \tag{24}$$

Sufficiently close to the Neel temperature, $J_c \ll J_{\parallel} \ll J_0$ is true. With low interlayer exchange ($J \ll J_c$), we have $L_{\parallel}(0)/L_0 \approx 1 + J/(2J_c)$, i.e. the order parameter at the interface exceeds the volume value of L_0, which was not observed for the compensated interface. Maximum $L_{\parallel}(0)/L_0$ was about $0.8(J_{\parallel}/J_c)^{2/3}$ and is reached at $J \approx 2^{-1/3}J_0^{2/3}J_c^{1/3}$. With $J = J_0$, we have $L_{\parallel}(0)/L_0 = 1$, and with $J > J_0$, antiferromagnetic order suppression takes place. Finally, with high interlayer exchange ($J \gg J_0$) we have $L_{\parallel}(0)/L_0 \approx J_0/J$. We show that, in case of $\phi = 0$, $\mathbf{L}$ is parallel to $\mathbf{M}$, i. e. $\theta_{\parallel}(z) = 0$ at any J. Suppose again $l_d/l_c \ll 1$, which can be always satisfied in the vicinity of T_{N}. Then in equation (6), the summand proportional to dL/dz may be neglected, then we come to equation (10), solution to which is written as (11), where instead of (12) we have

$$\cos\theta_{\parallel}(0) = \left(\frac{2J_\theta}{J}\right)^2\left(1 + \frac{JMl_d}{2\gamma}\frac{1}{L_{\parallel}(0)}\right). \tag{25}$$

With the growth of J, $\cos\theta_{\parallel}(0)$ decreases monotonously. Within $J \to \infty$, we have $\cos\theta_{\parallel}(0) = 1$. Thus, $\theta_{\parallel}(z) = 0$ is the solution to equation (10).

Let's consider the perpendicular orientation of $\mathbf{M}$ with respect to the easy axis of antiferromagnet anisotropy ($\phi = \pm\pi/2$). Unlike the compensated interface, $\mathbf{L}$ will deviate from the easy axis direction and this deviation may be significant. However, since $l_d/l_c \ll 1$, then in the first approximation, to which we restrict ourselves, equation (5) may neglect the summands containing θ, and equation (6) may neglect the summand containing dL/dz. Then equations for L and θ will be separated again, and the link between L and θ will be included in boundary conditions (21) and (22). Solutions to the derived equations are written as

$$L_{\perp}(z) = \begin{cases} L_0\,\mathrm{cth}\,\frac{z+z_{\perp}}{\sqrt{2}l_c}, & J \le J_0, \\ L_0\mathrm{th}\frac{z+z_{\perp}}{\sqrt{2}l_c}, & J \ge J_0, \end{cases} \tag{26}$$

$$\cos\theta_{\perp}(z) = \mathrm{th}\,\frac{z+z_\theta^{\perp}}{l_d}, \tag{27}$$

$z_{\perp}$ and $z_\theta^{\perp}$ may be derived from the following equations:

$$\frac{L_{\perp}(0)}{L_0} = \sqrt{1+\left(\frac{J}{2J_{\perp}}\right)^4\left(1+\frac{\Delta}{\delta}\sin^2\theta_{\perp}(0)\right)^2 + \frac{J}{J_c}\sin\theta_{\perp}(0)} - \left(\frac{J}{2J_{\perp}}\right)^2\left(1+\frac{\Delta}{\delta}\sin^2\theta_{\perp}(0)\right), \tag{28}$$

$$1 + \frac{J}{J_c}\,\frac{\left(\frac{2J_\theta}{J}\right)^2\sin\theta_{\perp}(0)\,\mathrm{tg}\,\theta_{\perp}(0) - \frac{\delta}{\Delta}}{\sin\theta_{\perp}(0) + \left(\frac{2J_\theta}{J}\right)^2\mathrm{tg}\,\theta_{\perp}(0)} - \frac{\left(\frac{2J_\theta}{J}\right)^2\left(\frac{2J_\theta}{J_d}\right)^2}{\left(\sin\theta_{\perp}(0) + \left(\frac{2J_\theta}{J}\right)^2\mathrm{tg}\,\theta_{\perp}(0)\right)^2}, \tag{29}$$

where $J_d = 2\gamma L_0/(Ml_d)$. Sufficiently close to the Neel temperature, $J_c \ll J_d \ll J_{\perp}(J_\theta) \ll J_0$ is satisfied. With weak layer exchange interaction ($J \ll \sqrt{J_cJ_d}$), from equations (28) and (29) we get $\theta_{\perp}(0) \approx J/J_d$ and $L_{\perp}(0)/L_0 \approx 1 + J^2/(2J_cJ_d)$. Unlike $\phi = 0$, as J increases, the order parameter grows slower. With $J \gg 2(\Delta/\delta)^{1/2}J_\theta$, $\theta_{\perp}(0)$ approaches $\pi/2$, so that $\mathrm{tg}\,\theta_{\perp}(0) \approx (\delta/\Delta)\big(J/(2J_\theta)\big)^2$. Since $\Delta \gg \delta$, then $L_{\perp} \to L_{\parallel}$. With $J > J_0$, antiferromagnetic order suppression takes place. Finally, with high interlayer exchange ($J \gg J_0$) we have $L_{\perp}(0)/L_0 \approx J_0/J$. Figure 4, a and b show the dependences of $L_{\perp(\parallel)}(0)$ and $L_{\parallel}(0) - L_{\perp}(0)$ on the interlayer exchange constant J at various temperatures T calculated

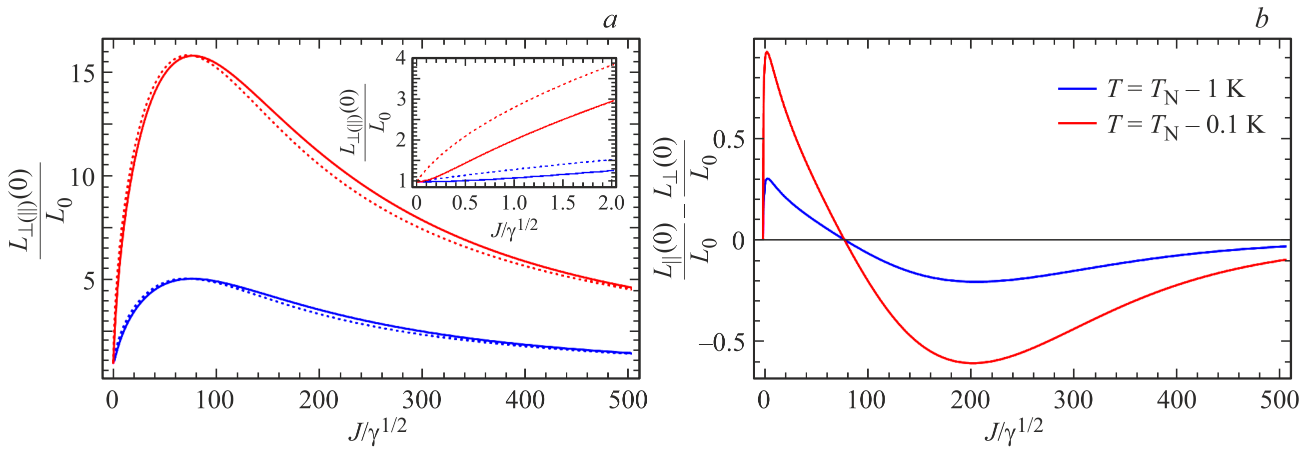

Figure 4. Dependences of *a*) the order parameter modulus at the uncompensated interface $L_{\perp(\parallel)}(0)$ and *b*) of $L_{\parallel}(0){-}L_{\perp}(0)$ on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet$|MnF_2$ structure. Solid lines in Figure 4,*a* correspond to the perpendicular direction ($\perp$) of the ferromagnet magnetization $\mathbf{M}$ with respect to the anisotropy easy axis of the antiferromagnet, dashed lines correspond to the parallel direction ($\parallel$, $\phi = 0$). The inset shows the same dependences for low values of J.

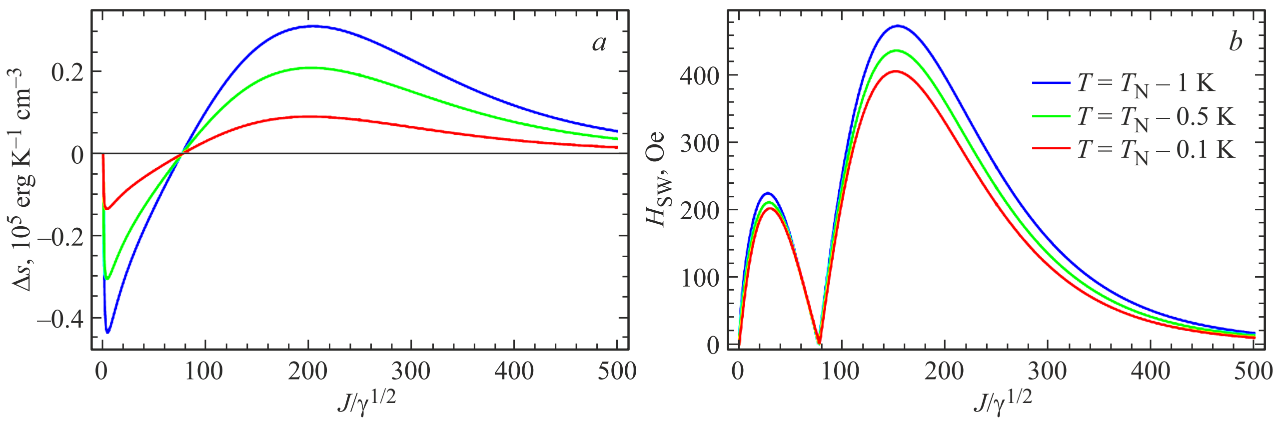

Figure 5. Dependence of *a*) the isotermal entropy change Δs and *b*) switching field H_{sw} on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet$|MnF_2$ structure for the uncompensated interface.

for the ferromagnet$|MnF_2$ structure. $\theta_{\perp}(0)$ was calculated numerically from equation (29). As can be seen, at some value of J, the sign of $L_{\parallel}(0) - L_{\perp}(0)$ is changed. Actually, with $J \ll J_c$, we have $L_{\parallel}(0)/L_0 - L_{\perp}(0)/L_0 \approx J/(2J_c)$,, and with $J \gg J_0$, we have $L_{\parallel}(0)/L_0 - L_{\perp}(0)/L_0 \propto -1/J^5$. Calculation of the isothermal entropy change $\Delta s = s_{\parallel} - s_{\perp}$ according to (9) again brings us to equations (15) and (16). Figure 5,*a* shows the dependences of Δs on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet$|MnF_2$ structure at $h = 20$ nm and $d = 5l_c$. Pairs of maxima of these dependences are linked by the above-mentioned features of $L_{\perp}(0) - L_{\parallel}(0)$ depending on J. As with the compensated interface, Δs grows with distance from T_N. However, it is impossible to determine the temperature at which Δs will take the maximum value within the developed model, because $l_d/l_c \ll 1$ shall be satisfied.

Let's find now the magnitude of switching field H_{sw}. For this, taking into account (5), (6), (21) and (22), we rewrite F as

$$F = -\frac{\beta}{4}\int\limits_0^{\infty} L^4(z)dz - \frac{J}{4}ML\cos(\theta-\phi)\big|_{z=0}. \qquad (30)$$

With $J \ll J_c$, we have $\Delta F = F_{\parallel} - F_{\perp} \propto -J$, and with $J \gg J_0$, we have $\Delta F \propto 1/J^4$. Thus, with large interlayer exchange, a state with $\phi = \pm\pi/2$ has the lowest free energy, and the volume density of effective free energy of the given system in the presence of a magnetic field may be written as (18). In this case $H_{sw} = K_{eff}M = 2\Delta F/(Mh)$. In the opposite case, a state with $\phi = 0$ has the free energy, and f_{eff} may be written as

$$f_{eff} = -\mathbf{M}(\mathbf{H} + \mathbf{H}_{eb}), \qquad (31)$$

where $\mathbf{H}_{eb}$ is the exchange bias field oriented along the x axis. Minimizing f_{eff} with respect to ϕ, we find $H_{sw} \approx H_{eb} = -\Delta F/(Mh)$. Figure 5,*b* shows the dependences of H_{sw} on the interlayer exchange constant J at various temperatures T calculated for the ferromagnet|MnF_2 structure at $h = 20$ nm. Thus, switching between the states with $\phi = 0$ and $\phi = \pm\pi/2$ is carried out in a quite weak field, which is about 100 Oe with the maximum isothermal magnetic entropy change $\Delta s \approx -0.4 \cdot 10^5\,\mathrm{erg \cdot K^{-1} \cdot cm^{-3}}$.

4. Conclusion

Within the Landau theory, of second-order phase transitions, the isothermal magnetic entropy change Δs was calculated for the ferromagnet|MnF_2 structure for the compensated and uncompensated MnF_2 interfaces. It is shown that the behavior of the dependence of Δs on J depends considerably on the type of interface: for the compensated interface, Δs has only one peak (Figure 3, *a*), and for the uncompensated interface, it has two peaks with unlike signs (Figure 5, *a*). Isothermal magnetic entropy change may reach $1 \cdot 10^5\,\mathrm{erg \cdot K^{-1} \cdot cm^{-3}}$ in the 150 Oe field in the compensated interface case, and $\Delta s \approx -0.4 \cdot 10^5\,\mathrm{erg \cdot K^{-1} \cdot cm^{-3}}$ in the 100 Oe field in the uncompensated interface case. In the given structure, the interlayer exchange interaction leads to MCE enhancement. Actually, Δs in an individual MnF_2 layer in the 150 Oe field is just $2\,\mathrm{erg \cdot K^{-1} \cdot cm^{-3}}$ [17], which is lower by 4 to 5 orders of magnitude. The findings may turn out to be useful for the development of microscale and nanoscale coolers operating in weak magnetic fields.

The developed model is used to formulate criteria to be satisfied by an antiferromagnetic material in the ferromagnet|antiferromagnet structure in order to achieve a significant MCE. First, the desired material shall have a strong magnetic anisotropy (K_a) and, second, Δ/δ shall be as high as possible. Satisfying these criteria provides the maximum change of the order parameter modulus of the antiferromagnetic material during exchange field rotation. Thus, antiferromagnets with strong spin-flop-transition field and strong dependence of the antiferromagnet-paramagnet phase transition fields on the external field direction shall be selected [18,21].

Funding

The study was supported by the grant provided by the Foundation for the Advancement of Theoretical Physics and Mathematics „BASIS“, and State Contract for 2024–2026 (No. FFUF-2024-0021).

Conflict of interest

The author declares that he has no conflict of interest.

Appendix

Let's calculate $\mathbf{m}$ induced in the antiferromagnetic material with the compensated interface due to the exchange coupling with the ferromagnetic material. Minimization of the F_s contribution with respect to $\mathbf{m}$ (equation (3)) gives the following equation and boundary condition:

$$\gamma_0 \frac{d^2\mathbf{m}}{dz^2} - \frac{1}{\chi}\mathbf{m} - \Delta(\mathbf{mL})\mathbf{L} - \delta\mathbf{L}^2\mathbf{m} = 0, \qquad \text{(A1)}$$

$$\frac{d\mathbf{m}}{dz}\Big|_{z=0} = -\frac{J}{\gamma_0}\mathbf{M}. \qquad \text{(A2)}$$

With distance from the interface, magnetization shall decrease, so the additional boundary condition $\mathbf{m}(z \to \infty) = 0$ shall be introduced. We assume that the spatial scale of $\mathbf{L}$ is much larger than that of $\mathbf{m}$. In this case, in equation (A1) $\mathbf{L}(z) \approx \mathbf{L}(0)$, and the solution may be written as

$$\mathbf{m}(z) = \frac{J}{\gamma_0 q}\bigg(\mathbf{M}\exp(-qz) + \Big(\frac{q}{p}\exp(-pz) - \exp(-qz)\Big)(\mathbf{M}\hat{\mathbf{L}})\hat{\mathbf{L}}\bigg), \qquad \text{(A3)}$$

where

$$\hat{\mathbf{L}} = \mathbf{L}/L, \quad q^2 = (1 + \chi\delta\mathbf{L}^2)/(\chi\gamma_0)$$

and

$$p^2 = \big(1 + \chi(\Delta + \delta)\mathbf{L}^2\big)/(\chi\gamma_0).$$

Using equation (A1) and boundary condition (A2), F_s is written as

$$F_s = -\frac{J}{2}(\mathbf{mM})\big|_{z=0}. \qquad \text{(A4)}$$

Finally, after substitution of $\mathbf{m}(0)$ and expansion of F_s to $\mathbf{L}^2$, equation (4) is derived.

For the uncompensated interface, J shall be replaced with $J/2$ in equations (A2) and (A3), and instead of (A4) we have

$$F_s = -\frac{J}{4}(\mathbf{mM})\big|_{z=0} - \frac{J}{2}(\mathbf{ML})\big|_{z=0}. \qquad \text{(A5)}$$

Then, instead of (4) we get equation (20).

References

[1] P. Weiss, A. Piccard. J. Phys. Theor. Appl. **7**, *1*, 103 (1917).
[2] A.M. Tishin, Y.I. Spichkin. The Magnetocaloric Effect and Its Applications. CRC Press, Boca Raton (2016). 475 p.
[3] V. Franco, J.S. Blázquez, J.J. Ipus, J.Y. Law, L.M. Moreno-Ramírez, A. Conde. Prog. Mater. Sci. **93**, 112 (2018).
[4] N.R. Ram, M. Prakash, U. Naresh, N. Suresh Kumar, T.S. Sarmash, T. Subbarao, R. Jeevan Kumar, G. Ranjith Kumar, K.C. Babu Naidu. J. Supercond. Nov. Magn. **31**, *7*, 1971 (2018).
[5] A.P. Kamantsev, Yu.S. Koshkidko, E.O. Bykov, T. Gottschall, A.G. Gamzatov, A.M. Aliev, A.G. Varzaneh, P. Kameli. Appl. Phys. Lett. **123**, *20*, 202405 (2023).
[6] Yu.S. Koshkid'ko, J. Ćwik, T.I. Ivanova, S.A. Nikitin, M. Miller, K. Rogacki. J. Magn. Magn. Mater. **433**, 234 (2017).
[7] A.A. Fraerman, A. Shereshevskii. JETP Lett. **101**, *9*, 618 (2015).

[8] M.A. Kuznetsov, I.Y. Pashenkin, N.I. Polushkin, M.V. Sapozhnikov, A.A. Fraerman. J. Appl. Phys. **127**, *18*, 183904 (2020).
[9] M.A. Kuznetsov, A.B. Drovosekov, A.A. Fraerman. JETP **132**, *1*, 79 (2021).
[10] M. Persson, M.M. Kulyk, A.F. Kravets, V. Korenivski. J. Phys.: Condens. Matter **35**, *7*, 075801 (2023).
[11] S.N. Vdovichev, N.I. Polushkin, I.D. Rodionov, V.N. Prudnikov, J. Chang, A.A. Fraerman. Phys. Rev. B **98**, *1*, 014428 (2018).
[12] D.M. Polishchuk, Y.O. Tykhonenko-Polishchuk, E. Holmgren, A.F. Kravets, A.I. Tovstolytkin, V. Korenivski. Phys. Rev. Mater. **2**, *11*, 114402 (2018).
[13] N.I. Polushkin, I.Y. Pashenkin, E. Fadeev, E. Lähderanta, A. Fraerman. J. Magn. Magn. Mater. **491**, 165601 (2019).
[14] I.Y. Pashenkin, N.I. Polushkin, M.V. Sapozhnikov, E.S. Demidov, E.A. Kravtsov, A.A. Fraerman. Phys. Solid State **64**, *10*, 1343 (2022).
[15] M. Kulyk, M. Persson, D. Polishchuk, V. Korenivski. J. Phys. D: Appl. Phys. **56**, *2*, 025002 (2023).
[16] I.Y. Pashenkin, R.V. Gorev, M.A. Kuznetsov, D.A. Tatarskiy, S.A. Churin, P.A. Yunin, M.N. Drozdov, D.O. Krivulin, M.V. Sapozhnikov, E.S. Demidov, V.A. Skuratov, E.V. Kudyukov, G.V. Kurlyandskaya, A.A. Fraerman, N.I. Polushkin. J. Phys. Chem. C **128**, *21*, 8853 (2024).
[17] M.A. Kuznetsov, E.A. Karashtin. Phys. Rev. B **109**, *22*, 224432 (2024).
[18] L.D. Landau, E.M. Lifshitz. Teoreticheskaya fizika. Ucheb. posob.: Dlya vuzov. V 10 t. T. VIII. Elektrodinamika sploshnykh sred. Fizmatlit, M. (2005). p. 652. (in Russian).
[19] W.H. Meiklejohn, C.P. Bean. Phys. Rev. **102**, *5*, 1413 (1956).
[20] W.H. Meiklejohn, C.P. Bean. Phys. Rev. **105**, *3*, 904 (1957).
[21] A.S. Borovik-Romanov. Lektsii po nizkotemperaturnomu magnetizmu. Magnitnaya simmetriya antiferromagnetikov. Institut fizicheskikh problem im. P.L. Kapitsy RAN, M. (2010). p. 56. (in Russian).
[22] H. Bizette, B. Tsai. Compt. Rend. Acad. Sci. Ser. **238**, 1575 (1954).
[23] Y. Shapira, S. Foner. Phys. Rev. B **1**, *7*, 3083 (1970).
[24] P. Nordblad, L. Lundgren, E. Figueroa, O. Beckman. J. Magn. Magn. Mater. **23**, *3*, 333 (1981).
[25] J. Barak, V. Jaccarino, S.M. Rezende. J. Magn. Magn. Mater. **9**, *4*, 323 (1978).
[26] R. Bjørk, C.R.H. Bahl, A. Smith, N. Pryds. Int. J. Refrig. **33**, *3*, 437 (2010).

Translated by E.Ilinskaya