TO THE THEORY OF MAGNETOSTRICTION AT QUANTUM PHASE TRANSITIONS IN VAN VLECK FERROMAGNETS

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A stimulated magnetostriction of an easy-plane singlet one-sublattice magnet in a magnetic field directed along its axis of hard magnetization has been considered. It has been shown that the magnetostriction in such a magnetic system is governed by a quantum phase transition belonging to displacement magnetic phase transitions induced by an external field. The amplitude of the stimulated magnetostriction has been demonstrated proportional to the field strength.

1. Introduction

A transition to a magnetically ordered state in van Vleck magnets can occur in the form of a displacement magnetic phase transition [1–3] classified by the modern terminology as a typical quantum phase transition (see, e.g., work [4]). The order parameter describing such phase transitions is the spin polarization of the ground ion singlet state. This state is not polarized in the paramagnetic phase, while the magnetic polarization of the initial singlet state becomes non-zero in the magnetically ordered state, arising self-consistently at the phase transition point. Contrary to the order–disorder phase transitions, which involve the reconstruction of the ion state spectrum at the phase transition point and where the exchange field spontaneously abolishes the degeneration of ion levels in the ordered phase, the quantum magnetic phase transition of the displacement type is not accompanied by such a rearrangement of ion levels, so that the ground state remains non-degenerate.

This quantum phase transition has a purely magnetic origin and, in principle, does not demand that ion displacements should be taken into consideration; its mechanism is defined by a competition of various spin interactions different by their nature. However, this phase transition, as well as any other that results in the emergence of the magnetic order, should invoke the reaction of the lattice, which is known to manifest itself as the magnetostriction phenomenon. Taking into account that the magnetization has a critical character at the displacement magnetic phase transition, one may also expect such a behavior of the magnetostriction which arises as soon as a certain finite magnetization begins to appear in the crystal.

In so doing, one should distinguish [5] between the striction related to the establishment of the magnetic order (the spontaneous magnetostriction) and the striction stimulated by an external magnetic field (the stimulated magnetostriction). At the displacement magnetic phase transitions, however, pertinent to the spontaneous magnetostriction is only the part of the striction of a magnetic singlet (i.e., in essence, non-magnetized) state that is brought about by the contribution of the basic singlet population, which varies in accordance with the temperature growth or reduction, to the linear thermal expansion of the magnet. At the same time, the striction related to the establishment of the magnetic order under the action of the magnetic field has to be attributed to the stimulated magnetostriction studied below.

In addition to the indicated difference, quantitative distinctions are also to be expected. First, at temperatures $T \ll T_{\mathrm{C}}$, where T_{C} is the Curie temperature, when the magnetization is almost saturated, ordinary ferromagnets demonstrate only the stimulated striction depending on the direction of the magnetization vector they acquire after the magnetic field having been introduced. The striction of the exchange nature must reveal itself at quantum magnetic phase transitions of the displacement type, so that a phenomenological approach which considers the vector of magnetization to be constant is unacceptable here, even at $T \to 0$.

The displacement magnetic phase transition is governed by the action of a single-ion anisotropy. Therefore, while considering the corresponding magnetostriction, interionic magnetoelastic interactions should be taken into account together with magnetoelastic ones of the single-ion nature [6]. It was shown in work [7] that the latter can result in the emergence of a rather high stimulated magnetostriction in the paramagnetic phase which depends quadratically on the magnetic field strength [7]. Such a behavior seems the most reasonable because the influence of the exchange interaction is absent or very weak when the paramagnetic phase is magnetized. In the range of the displacement magnetic phase transition, the ordering is brought about by a competition between the single-ion anisotropy and the exchange interaction. Therefore, the appearance of a finite magnetization has to affect the character of the field dependence of magnetostriction, which is connected with both single-ion and ion-ion interactions.

In this work, we attempted to determine the behavior of the stimulated magnetostriction in a simplest ferromagnetic crystal provided the displacement magnetic phase transition. The total energy of the system is presented as the sum

$$E = E_{\rm exch} + E_{\rm an} + E_{\rm h} + E_{\rm m-e} + E_{\rm el}, \qquad (1)$$

where $E_{\rm exch}$ is the exchange energy, $E_{\rm an}$ the energy of magnetic anisotropy, $E_{\rm h}$ the Zeeman energy of spins, $E_{\rm el}$ the elastic energy, and $E_{\rm m-e}$ the magnetoelastic energy. We assume that magnetoelastic interactions are much weaker than exchange ones, do not affect the magnetic ordering, and, hence, cannot induce a structural transition. The assumptions made allow the calculations to be carried out in the elementary case confining the account by only the terms in the magnetoelastic and elastic energies which are, respectively, linear and quadratic by deformation. In addition, under those conditions, one can consider the problem of the type of the magnetic ordering at first and then use the derived solutions for finding the field dependences of the striction.

2. The Ground State of an Easy-Plane Singlet Ferromagnet with $S = 1$ in a Longitudinal Magnetic Field

While analyzing the type of the magnetic ordering, let us confine ourselves to the consideration of bilinear isotropic exchange interactions, a single-ion anisotropy, and the Zeeman term. The corresponding Hamiltonian of a ferromagnet looks like

$$H = -\frac{J}{2}\sum_{\mathbf{n},\rho} \mathbf{S_n S_{n+\rho}} + D\sum_{\mathbf{n}} (S^Z_{\mathbf{n}})^2 - \mathbf{h}\sum_{\mathbf{n}} \mathbf{S_n} \qquad (2)$$

in the crystallographic coordinate system. Here, $J > 0$ is the exchange interaction between the nearest spins located at the $\mathbf{n}$ and $\mathbf{n}+\rho$ sites; $D > 0$ is the constant of a single-ion magnetic anisotropy of the "easy"-plane (EP) type; the vector of the magnetic field strength $\mathbf{h}$ is determined in terms of energy units, $\mathbf{h} = \mu_{\rm B} g \mathbf{H}$, and directed along the axis of "hard" magnetization Z; $\mu_{\rm B}$ is the Bohr magneton; and g is the g-factor.

Since the first three terms in Eq. (1) are admitted dominating, the energy $E_{\rm gr}$ of the ground state of a ferromagnet will be determined, considering it equal to the energy of the ground state with Hamiltonian (2). In this case, the expression for the energy per one spin looks like

$$E_{\rm gr} = -\frac{1}{2} Jz\mathbf{s}^2 + DQ - \mathbf{hs}, \qquad (3)$$

where z is the number of the nearest neighbors, $\mathbf{s}$ the vector of the average spin of ions, and Q the quantum-mechanical average of the squared Z-projections of the spin operators; these projections are usually referred to as the components of the quadrupole spin moment [8—10].

Let us define a proper coordinate system, where the average ion spin is directed along the axis of quantization. The coordinate axes are selected in a way so that the angle between the ζ axis of spin quantization and the Z axis be equal to θ and the ξ axis lie in the plane $Z\zeta$. In this system, as was shown in work [11], the wave function of the ground state of every ion can be presented by the linear combination

$$\Psi_{\rm gr} = \cos\phi|1\rangle + \sin\phi|-1\rangle, \qquad (4)$$

where the angle of "mixing" ϕ (see below) is determined by the condition that the energy of the ground state should be minimal.

According to the form of function (4), we find that the components of projections of both the average spin vector and the spin quadrupole moment, which are distinct from zero in the proper coordinate system, are

$$s = \cos 2\phi, \quad Q^{\zeta\zeta} = 1, \quad Q^{\xi\xi} = \frac{1}{2}(1 + \sin 2\phi),$$

$$Q^{\eta\eta} = \frac{1}{2}(1 - \sin 2\phi). \tag{5}$$

Making use of them allows energy (3) to be displayed in the form

$$E_{\rm gr} = -\frac{1}{2}Jz\cos^2 2\phi +$$

$$+D\left[\cos^2\theta + \frac{\sin^2\theta}{2}(1+\sin 2\phi)\right] - h\cos\theta\cos 2\phi, \tag{6}$$

where the magnetic field strength $\mathbf{h}$ is presented by its longitudinal component only ($\mathbf{h} \parallel Z$).

In order to calculate the striction values, it is necessary to find the field dependences of the average spin and the variations of its orientation in the field and to calculate the field dependences of the components of the quadrupole spin moment of ions. As was shown in works [1, 3], to obtain a solution of the problem concerning the determination of a spin configuration in a magnetic field, expression (6) has to be minimized over its variables – the geometric angle θ and the "mixing" angle of states ϕ. In so doing, the following equations are obtained:

$$Jz\sin 4\phi + D\sin^2\theta\cos 2\phi + 2h\cos\theta\sin 2\phi = 0, \tag{7}$$

$$-D\sin 2\theta(1-\sin 2\phi) + 2h\sin\theta\cos 2\phi = 0. \tag{8}$$

It is known [1] that, provided the external magnetic field is absent, the system of equations (7) and (8) possesses two solutions: a zero-magnetization one with $s = 0$, which is realized at $D > 2Jz$; and a nonzero-magnetization solution with

$$s_0 = \sqrt{1-(D/2Jz)^2}, \tag{9}$$

at $D < 2Jz$.

In order that a quantum phase transition from a singlet state to a magnetically ordered one be observed in a magnetic field $h \neq 0$, a singlet with $s = 0$ has to be the ground state of the system. Therefore, in what follows, we assume that the inequality $D > 2Jz$ evident in this case is satisfied. Provided such a relationship between the model parameters and the absence of the magnetic field, the magnetic order cannot be established at any temperature [2], which defines, in fact, a condition for the ground state of a magnet to be in the singlet state; it is an attribute of the van Vleck magnet. The solution with $s = 0$ satisfies Eq. (7), including the case of a certain finite range of the magnetic field.

Making use of this equation at $s \neq 0$, we obtained the expression for the orientation of the average spin with respect to the crystallographic axis

$$\cos\theta = \frac{h_{\parallel}\cos 2\phi}{D(1-\sin 2\phi)}. \tag{10}$$

Equations (7) and (10) testify to that the state with the spin directed along the hard axis ($\theta = 0$) can be realized in high fields, where $h \geq D$. In this case, the projection of the average spin onto the direction of the external field is utmost and equals $s = 1$. For lower fields, $h < D$, the average spin is canted with respect to the hard magnetization axis.

Using expression (10), let us expand the energy of the ground state in a power series of the ground state polarization s, assuming the latter small. Confining the expansion to the fourth degree, the energy $E_{\rm gr}$ reads

$$E_{\rm gr} = \frac{1}{2}\left(-Jz + \frac{1}{2}(D - \frac{h^2}{D})\right)s^2 + \frac{1}{16}\left(D - \frac{h^2}{D}\right)s^4. \tag{11}$$

Minimizing this expression over the parameter s, both a singlet state and a magnetized one can be obtained. It is easy to show that the transition from the former to the latter occurs at the magnetic field

$$h = h_{\rm OP} = \sqrt{D(D-2Jz)}. \tag{12}$$

Thus, in the full agreement with the Landau theory of phase transitions, the quantum transition of the second kind induced by the external field from the non-magnetic singlet state to the ferromagnetic one with the magnetization directed at a certain angle with respect to the crystallographic axis takes place at the point $h = h_{\rm OP}$. The modulus of the average spin vector also depends on the field and, in the regarded approximation of the self-consistent field, has the standard form

$$s = \frac{2\sqrt{h_{\rm OP}}}{D}\sqrt{h - h_{\rm OP}}. \tag{13}$$

Taking into account Eqs. (10) and (13), we get the following expressions for the dependences of the average spin projections onto the crystallographic axis (the axis Z) and onto the plane on the field h within the range $h_{\rm OP} < h < D$:

$$s_Z = 2\frac{h_{\rm OP}^2}{D^3}(h - h_{\rm OP}); \tag{14}$$

$$s_X = 2\frac{\sqrt{h_{\rm OP}}}{D}\left[1 - \frac{h_{\rm OP}^3}{2D^4}(h - h_{\rm OP})\right]\sqrt{h - h_{\rm OP}}, \qquad (15)$$

where the axis X lies in the easy plane and coincides with the intersection of this plane by the plane $Z\zeta$. In the same manner, one can also find the field dependences of the components of the quadrupole spin moment.

We note that the energy of the ground state (10) does not contain terms that are linear in s or h. In such a case, the field counteracts the single-ion anisotropy, and, as is seen from Eq. (15), a polarization caused by the field and the exchange interaction appears spontaneously in the easy plane, with the derivative $\partial s_X/\partial h \to \infty$ at $h \to h_{\rm OP}$. The finite value of $\partial s_Z/\partial h$ at $h = h_{\rm OP}$ indicates that, after the spontaneous emergence of the average spin, its cant (rotation) with respect to the hard axis decreases as h grows.

3. Stimulated Magnetostriction of a van Vleck Ferromagnet in a Longitudinal Magnetic Field

For the sake of definiteness, when considering the magnetoelastic and elastic energies, we assume that the ferromagnet has a hexagonal structure.

In the case of $E_{\rm m-e}$ (see Eq. (1)), we confine ourselves to the spin-spin magnetoelastic interactions, which involve only the second powers of the average spin projections [12]. We also take into account the contribution of the single-particle magnetoelastic energy which is described by the terms containing the average values of components of the tensor of the spin quadrupole moment [5]. Therefore, the expressions for the magnetoelastic and elastic components of the energy in Eq. (1) look like

$$E_{\rm m-e} = B_{11}^{(1)}(Q^{XX}U_{XX} + Q^{YY}U_{YY}) +$$

$$+B_{33}^{(1)}Q^{ZZ}U_{ZZ} + B_{12}^{(1)}(Q^{XX}U_{YY} + Q^{YY}U_{XX}) +$$

$$+4B_{44}^{(1)}(Q^{YZ}U_{YZ} + Q^{XZ}U_{XZ}) + 4B_{66}^{(1)}Q^{XY}U_{XY} +$$

$$+B_{11}^{(2)}(s_X^2U_{XX} + s_Y^2U_{YY}) +$$

$$+B_{33}^{(2)}s_Z^2U_{ZZ} + B_{12}^{(2)}(s_X^2U_{YY} + s_Y^2U_{XX}) +$$

$$+4B_{44}^{(2)}(s_Ys_ZU_{YZ} + s_Xs_ZU_{XZ}) + 4B_{66}^{(2)}s_Xs_YU_{XY}, \qquad (16)$$

$$E_{\rm el} = \frac{1}{2}C_{11}(U_{XX}^2 + U_{YY}^2) + \frac{1}{2}C_{33}U_{ZZ}^2 +$$

$$+C_{12}U_{XX}U_{YY} + C_{13}(U_{XX} + U_{YY})U_{ZZ} +$$

$$+2C_{44}(U_{XZ}^2 + U_{YZ}^2) + 2C_{66}U_{XY}^2, \qquad (17)$$

where $B_{11}^{(1)}, \ldots, B_{66}^{(1)}, B_{11}^{(2)}, \ldots, B_{66}^{(2)}$ are the parameters of magnetoelastic interactions [superscripts (1) and (2) indicate the single-ion and the interionic origin of corresponding magnetoelastic interactions, respectively]; $U_{XX}, \ldots$ are the components of the strain tensor; and $C_{11}, \ldots$ are the elastic moduli. Note that single-ion magnetoelastic interactions in Eq. (17) are written down in the crystallographic coordinate system. That is why, contrary to Eq. (5), the components of the quadrupole moment are defined as $Q^{jk} = \frac{1}{2}\langle s_js_k + s_ks_j\rangle$, where the indices j and k designate the crystallographic axes: $j,k = X,Y,Z$.

The deformation amplitudes which correspond to the spin configurations calculated above are determined by minimizing the sum of energies (16) and (17) over the components of the strain tensor. As a result, the deformations turn out proportional to the average values of s_j and Q^{jk}

$$U_{XX} - U_{YY} = \frac{-1}{C_{11} - C_{12}} \times$$

$$\times\Big[(B_{11}^{(1)} - B_{12}^{(1)})(Q^{XX} - Q^{YY}) +$$

$$+(B_{11}^{(1)} - B_{12}^{(1)})(s_X^2 - s_Y^2)\Big], \qquad (18)$$

$$U_{XX} + U_{YY} = \frac{-1}{C_{11} + C_{12} - 2C_{13}^2/C_{33}} \times$$

$$\times\Big[(B_{11}^{(1)} + B_{12}^{(1)})(Q^{XX} + Q^{YY}) - 2B_{33}^{(2)}Q^{ZZ}C_{13}/C_{33} +$$

$$+(B_{11}^{(2)} + B_{12}^{(2)})(s_X^2 + s_Y^2) - 2B_{33}^{(2)}s_Z^2C_{13}/C_{33}\Big], \qquad (19)$$

$$U_{ZZ} = \frac{C_{13}}{C_{33}(C_{11} + C_{12} - 2C_{13}^2/C_{33})} \times$$

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$$\times\Big[(B_{11}^{(1)}+B_{12}^{(1)})(Q^{XX}+Q^{YY})+$$

$$+(B_{11}^{(2)}+B_{12}^{(2)})(s_X^2+s_Y^2)-$$

$$-2(B_{33}^{(1)}Q^{ZZ}+B_{33}^{(2)}s_Z^2)C_{13}/C_{33}\Big]-$$

$$-\frac{[B_{33}^{(1)}Q^{ZZ}+B_{33}^{(2)}s_Z^2]}{C_{33}}, \quad (20)$$

$$U_{XY}=\frac{-1}{C_{66}}\Big[B_{66}^{(1)}Q^{XY}+B_{66}^{(2)}s_Xs_Y\Big], \quad (21)$$

$$U_{XZ}=\frac{-1}{C_{44}}\Big[B_{44}^{(1)}Q^{XZ}+B_{44}^{(2)}s_Xs_Z\Big], \quad (22)$$

$$U_{XY}=\frac{-1}{C_{44}}\Big[B_{44}^{(1)}Q^{YZ}+B_{44}^{(2)}s_Ys_Z\Big]. \quad (23)$$

Expression (18) determines the anisotropic striction in the easy plane, expression (19) describes the expansion or squeezing (depending on the signs of magnetoelastic constants) in this plane, and expression (20) the stretching or squeezing along the hard axis. The last three expressions (21)–(23) determine shear strains.

Let us write down the corresponding expressions for the case of spontaneous deformation in the singlet phase by substituting $s=0$, $Q^{ZZ}=0$, and $Q^{XX}=Q^{YY}=1$ into Eqs. (18)–(23). Different from zero will be only those deformations which are connected with the isotropic expansion or squeezing of the easy plane and the stretching or squeezing along the hard axis:

$$U_{XX}^{(0)}+U_{YY}^{(0)}=-2\frac{B_{11}^{(1)}+B_{12}^{(1)}}{C_{11}+C_{12}-2C_{13}^2/C_{33}}, \quad (24)$$

$$U_{ZZ}^{(0)}=2\frac{C_{13}(B_{11}^{(1)}+B_{12}^{(1)})}{C_{33}(C_{11}+C_{12}-2C_{13}^2/C_{33})}. \quad (25)$$

Here, superscript 0 denotes the spontaneous magnetostriction. One can see that spontaneous deformations in the singlet phase satisfy the relations $U_{XX}^{(0)}=U_{YY}^{(0)}=-U_{ZZ}^{(0)}C_{33}/2C_{13}$. Expressions (24) and (25) are valid also in the range of magnetic fields $h<h_{\rm OP}$, where the singlet phase exists.

Now, let us write down the expressions for crystal deformations after the phase transition induced by an external longitudinal field $h>h_{\rm OP}$, provided $(h-h_{\rm OP})/h_{\rm OP}\ll 1$. We now substitute the dependences of the average spin projections and the components of the quadrupole spin moment tensor Q^{jk} on the field into Eqs. (18)—(23) and get the required strain tensor components as

$$U_{XX}-U_{YY}=$$

$$=\frac{(B_{11}^{(1)}-B_{12}^{(1)})-4(B_{11}^{(2)}-B_{12}^{(2)})}{C_{11}-C_{12}}\frac{h_{\rm OP}}{D^2}(h-h_{\rm OP}), \quad (26)$$

$$U_{XX}+U_{YY}=$$

$$=U_{XX}^{(0)}+U_{YY}^{(0)}+\frac{h_{\rm OP}}{D^2}\frac{h-h_{\rm OP}}{C_{11}+C_{12}-2C_{13}^2/C_{33}}\times$$

$$\times\Big[B_{11}^{(1)}+B_{12}^{(1)}-4(B_{11}^{(2)}+B_{12}^{(2)})+2B_{33}^{(1)}C_{13}/C_{33}\Big], \quad (27)$$

$$U_{ZZ}=U_{ZZ}^{(0)}+\frac{h_{\rm OP}}{D^2}\frac{h-h_{\rm OP}}{C_{11}+C_{12}-2C_{13}^2/C_{33}}\frac{C_{13}}{C_{33}}\times$$

$$\times\Big[4(B_{11}^{(2)}+B_{12}^{(2)})-B_{11}^{(1)}-B_{12}^{(1)}-2B_{33}^{(1)}C_{13}/C_{33}\Big]-$$

$$-\frac{B_{33}^{(1)}}{C_{33}}\frac{h_{\rm OP}}{D^2}(h-h_{\rm OP}), \quad (28)$$

$$U_{XZ}=-\frac{h_{\rm OP}^{3/2}(h-h_{\rm OP})^{1/2}B_{44}^{(1)}}{D^2C_{44}}. \quad (29)$$

From Eq. (26), it follows that the shear strain turns out to be proportional to the field strength. The values of striction (27) and (28), which describe the expansion or squeezing in the easy plane and the stretching or squeezing of the crystal along its hard axis, are also linear in the field. The emergence of the interlayer shear strain (29) is connected with the noncollinearity of the average spin and the field, and its field dependence has a critical index equal to 1/2.

We note that, in the case of a classical easy-plane ferromagnet with the saturated magnetic moment in the field directed along the hard axis, the magnetostriction, provided magnetoelastic interactions of type (16), would have a square-law field dependence. But if the value of the ratio $h_{\rm OP}/D$ is small, one may neglect the terms related to the spin rotation in expressions (27) and (28).

4. Conclusion

Thus, the analysis of the dependences of the stimulated magnetostriction of a singlet magnet on the field showed that, in the case of low fields, when the singlet phase is stable, the deformations do not depend on the applied field and remain equal to their initial values. After the quantum phase transition, a stimulated striction, whose value turns out mainly to be proportional to the first power of the applied magnetic field strength, emerges in the magnetically ordered state. Contrary to classical ferromagnets, where the stimulated striction at temperatures $T \ll T_{\mathrm{C}}$ is connected to the rotation of magnetization only, in the case of a van Vleck magnet where the induced quantum phase transition of the displacement type takes place, the character of the field dependence of the striction is determined, primarily, by the polarization of ionic states.

The linear dependence of the stimulated magnetostriction on the field evidences for the constant derivative of the striction with respect to the field. This property of the striction manifests itself only in fields above the field of the phase transition. In this case, experimental observations of a linear magnetostriction in magnets with a low value of the critical field determined by the condition $h_{\mathrm{OP}}/D \ll 1$ do not require the introduction of high magnetic fields.

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ДО ТЕОРІЇ МАГНІТОСТРИКЦІЇ ПРИ КВАНТОВИХ ФАЗОВИХ ПЕРЕХОДАХ У ВАН-ФЛЕКІВСЬКИХ ФЕРОМАГНЕТИКАХ

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Р е з ю м е

Розглянуто вимушену магнітострикцію легкоплощинного синглетного однопідграткового магнетика в магнітному полі, орієнтованому вздовж важкої осі. Показано, що поведінка магнітострикції такої магнітної системи пов'язана з квантовим фазовим переходом, який належить до магнітних фазових переходів типу зміщення, індукованим зовнішнім полем. Показано, що величини вимушеної магнітострикції прямо пропорційні величині напруженості цього поля.