PSEUDO-DIPOLAR AND QUADRUPOLAR SPIN COUPLING AND MAGNETICALLY PREFERRED DIRECTIONS IN TETRAGONAL FERROMAGNETS

By H. Pfeiffer

Institute of Theoretical Physics, University of Wrocław*

And J. Ulner

Institute for Low Temperature and Structure Research, Polish Academy of Sciences, Wrocław**

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By minimizing the saturation-state energy of a spin system with pseudo-dipolar and quadrupolar short-range interactions in the nereast-neighbour approximation, the magnetically preferred directions of a ferromagnet with a simple or body-centred tetragonal crystal lattice are determined and theii dependence on the lattice deformation and on the sign and magnitude of the coupling constants is examined.

1. *Introduction*

The use of short-range multipolar spin couplings in describing the anisotropic properties of ferromagnets was first proposed by Van Vleck [1]. Since then, this type of semi-phenomenological interactions has been successfully applied to many problems of ferromagnetism [2–11], usually by restricting the coupling to dipolar or, at the most, to quadrupolar terms. Notwithstanding the fact that multipolar spin couplings are mathematically hard to handle, their clear advantage compared to simpler phenomenological anisotropy forms is that they need not be specified for each particular crystal symmetry. In other words, multipolar spin couplings are believed to lead automatically to correct anisotropy directions when applied to a specific crystal lattice.

However, proof that this is true has thus far been established only for the cubic and hexagonal crystal lattices and is given in [5]. The aim of the present paper is to extend the proof of [5] to tetragonal crystal structures (*s. t.* and *b. c. t.*), and to examine the influence of the lattice deformation and the sign and magnitude of the pseudo-dipolar and quadrupolar

* Address: Instytut Fizyki Teoretycznej, Uniwersytet Wrocławski, Wrocław, Cybulskiego 36, Poland.

** Address: Instytut Niskich Temperatur i Badań Strukturalnych, Polska Akademia Nauk, Wrocław, Plac Katedralny 1, Poland.

coupling constants on the magnetic anisotropy of the crystal. As we are interested in determining the magnetically preferred directions only, we shall disregard surface effects (magnetic surface poles, demagnetizing field, and shape anisotropy) and work in the saturation-state approximation (cp. [12, 13]), *i. e.*, we shall minimize the spin system's energy in the saturation state with respect to the direction of spin alignment. This is precisely the same approximation as was made in [5] and corresponds, strictly speaking, to the case of small (*i.e.*, below the critical dimensions; see, *e. g.*, [14]), spherical, single-domain ferromagnetic particles in which the magnetocrystalline anisotropy is the sole factor that determines the direction of spontaneous magnetization.

Except for the first two Sections where the general minimum conditions are derived, we restrict ourselves to the nearest-neighbour approximation. For the *b. c. t.* case, we extend the considerations to include next-nearest-neighbour interactions in the tetragonal plane. The influence of long-range dipole-dipole coupling will be considered in a subsequent paper.

2. *The minimization*

We start with the Hamiltonian

$$H = \sum_{\alpha,\beta} P^{\alpha\beta}_{\mu_1\mu_2} S^{\alpha}_{\mu_1} S^{\beta}_{\mu_2} + \sum_{\alpha,\beta} Q^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4} S^{\alpha}_{\mu_1} S^{\alpha}_{\mu_2} S^{\beta}_{\mu_3} S^{\beta}_{\mu_4} \tag{1}$$

where the tensor

$$P^{\alpha\beta}_{\mu_1\mu_2} = [C(r^{\alpha\beta}) - D(r^{\alpha\beta}) - J(r^{\alpha\beta})]\,\delta^{\mu_1\mu_2} - 3\,[(C(r^{\alpha\beta}) - D(r^{\alpha\beta})]\,\frac{r^{\alpha\beta}_{\mu_1} r^{\alpha\beta}_{\mu_2}}{(r^{\alpha\beta})^2} \tag{2}$$

comprises the isotropic Heisenberg exchange interaction $J(r^{\alpha\beta})$, the classical dipole-dipole interaction

$$D(r^{\alpha\beta}) = \frac{g^2\gamma^2}{(r^{\alpha\beta})^3} \tag{3}$$

with the Landé factor g and Bohr's magneton γ, and the short-range pseudo-dipolar anisotropy $C(r^{\alpha\beta})$. The tensor

$$Q^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4} = \frac{Q(r^{\alpha\beta})}{(r^{\alpha\beta})^4}\, r^{\alpha\beta}_{\mu_1} r^{\alpha\beta}_{\mu_2} r^{\alpha\beta}_{\mu_3} r^{\alpha\beta}_{\mu_4} \tag{4}$$

describes short-range pseudo-quadrupolar anisotropic coupling. The indices α, β refer to lattice sites, and $\mu_1, \mu_2, \ldots$ denote tensor (vector) components to which Einstein's summation rule applies. The components of the lattice vector between sites α and β are denoted by $r^{\alpha\beta}_{\mu}$, and the distance by $r^{\alpha\beta}$. The spin operators S^{α}_{μ} satisfy the conventional commutation rules.

With the familiar spin raising and lowering operators $S^{\alpha}_{\pm} = S^{\alpha}_1 \pm i S^{\alpha}_2$ the saturation state $|-S>$ along the negative z-asix is defined as follows:

$$S^{\alpha}_3|-S> = -S|-S>, \quad S^{\alpha}_-|-S> = 0. \tag{5}$$

Following [15], we introduce the unitary transformation $U(\vartheta, \varphi)$ which first rotates the spins around the y-axis by the angle ϑ and afterwards around the z-axis by the angle φ. Hence, the state

$$|\vartheta, \varphi > = U^{+}(\vartheta, \varphi)|-S> \tag{6}$$

represents saturation in the direction given by the angles ϑ, φ. Since

$$US_{\mu}^{\alpha}U^{+} = R_{\mu\nu}S_{\nu}^{\alpha} \tag{7}$$

where

$$R_{\mu\nu} = \begin{pmatrix} \cos\varphi\cos\vartheta & -\sin\varphi & \cos\varphi\sin\vartheta \\ \sin\varphi\cos\vartheta & \cos\varphi & \sin\varphi\sin\vartheta \\ -\sin\vartheta & 0 & \cos\vartheta \end{pmatrix} \tag{8}$$

the quantity to be minimized with respect to ϑ, φ is

$$h = \langle\vartheta, \varphi\,|H|\,\vartheta, \varphi\rangle = \langle -S\,|\tilde{H}|-S\rangle \tag{9}$$

where

$$\tilde{H} = UHU^{+} = \sum_{\alpha,\beta} \tilde{P}_{\mu_1\mu_2}^{\alpha\beta} S_{\mu_1}^{\alpha} S_{\mu_2}^{\beta} + \sum_{\alpha,\beta} \tilde{Q}_{\mu_1\mu_2\mu_3\mu_4}^{\alpha\beta} S_{\mu_1}^{\alpha} S_{\mu_2}^{\alpha} S_{\mu_3}^{\beta} S_{\mu_4}^{\beta}$$

$$\tilde{P}_{\mu_1\mu_2}^{\alpha\beta} = P_{\nu_1\nu_2}^{\alpha\beta} R_{\nu_1\mu_1} R_{\nu_2\mu_2} \tag{10}$$

$$\tilde{Q}_{\mu_1\mu_2\mu_3\mu_4}^{\alpha\beta} = Q_{\nu_1\nu_2\nu_3\nu_4}^{\alpha\beta} R_{\nu_1\mu_1} R_{\nu_2\mu_2} R_{\nu_3\mu_3} R_{\nu_4\mu_4}.$$

According to [15] one has

$$h = S^2 \sum_{\alpha,\beta} (\tilde{P}_{33}^{\alpha\beta} + \tilde{Q}^{\alpha\beta}) \tag{11}$$

with the abbreviation

$$\tilde{Q}^{\alpha\beta} = \frac{1}{4}(\tilde{Q}_{1111}^{\alpha\beta} + \tilde{Q}_{1122}^{\alpha\beta} + \tilde{Q}_{2211}^{\alpha\beta} + \tilde{Q}_{2222}^{\alpha\beta}) +$$

$$+ \frac{1}{2} S(\tilde{Q}_{1133}^{\alpha\beta} + \tilde{Q}_{3311}^{\alpha\beta} + \tilde{Q}_{2233}^{\alpha\beta} + \tilde{Q}_{3322}^{\alpha\beta}) + S^2\tilde{Q}_{3333}^{\alpha\beta}. \tag{12}$$

The necessary minimum conditions are

$$\frac{\partial h}{\partial\vartheta} = \frac{\partial h}{\partial\varphi} = 0 \tag{13}$$

and the sufficient ones read

$$\Delta = \frac{\partial^2 h}{\partial\varphi^2}\cdot\frac{\partial^2 h}{\partial\vartheta^2} - \left(\frac{\partial^2 h}{\partial\varphi\partial\vartheta}\right)^2 > 0, \qquad \frac{\partial^2 h}{\partial\varphi^2} > 0. \tag{14}$$

The explicit form of $h = h(\vartheta, \varphi)$ in the general case is given in Appendix I. In the following, we shall assume tetragonal symmetry and consider the simple (*s. t.*) and body-centred (*b. c. t.*) crystal lattice in the nearest-neighbour approximation. The extention of the considerations to the case of next-nearest-neighbour coupling is demonstrated for the *b. c. t.* lattice with $b > a$ where b is the lattice constant in the tetragonal direction.

3. *The tetragonal crystal lattices*

It is easily proved that in the case of tetragonal crystal symmetry one has

$$P_{\mu_1\mu_2} \equiv \sum_{\alpha,\beta} P^{\alpha\beta}_{\mu_1\mu_2} = 0 \quad \text{if} \quad \mu_1 \neq \mu_2 \tag{15}$$

and

$$Q_{\mu_1\mu_2\mu_3\mu_4} \equiv \sum_{\alpha\beta} Q^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4} = 0 \tag{16}$$

if there are three different or three identical indices among $\mu_1 \ldots \mu_4$. For the remaining quantities one obtains the relations

$$P_{11} = P_{22}, \qquad Q_{1111} = Q_{2222}, \tag{17}$$

$$Q_{1133} = Q_{2233} = Q_{P[1133]} = Q_{P[2233]}$$

where the symbol $P[\;]$ denotes an arbitrary permutation of the indices in the brackets.

Due to Eqs (15)–(17) the conditions (13) take the form

$$\sin 4\varphi \sin^4 \vartheta = 0, \tag{18a}$$

$$\sin 2\vartheta\{\tfrac{1}{2}(4Q_{1133}-Q_{1111}-Q_{1122})+2S^2(3Q_{1133}-Q_{3333})+$$

$$+S(Q_{3333}-7Q_{1133}+Q_{1111}+Q_{1122})+(S-\tfrac{1}{2})^2 \sin^2 \vartheta \times$$

$$\times [2(Q_{1111}+Q_{3333}-6Q_{1133})+\sin^2 2\varphi\,(3Q_{1122}-Q_{1111})]+P_{11}-P_{33}\} = 0 \tag{18b}$$

and the solutions are

I. $\sin \vartheta = 0$, φ — arbitrary

II. $\cos \vartheta = 0$ (20)
- a) $\sin \varphi = 0$
- b) $\cos \varphi = 0$
- c) $\cos 2\varphi = 0$

III. $\sin^2 \vartheta = [\tfrac{1}{2}(4Q_{1133}-Q_{1111}-Q_{1122})+2S^2(3Q_{1133}-Q_{3333})+S(Q_{3333}-7Q_{1133}+$
$+Q_{1111}+Q_{1122})+P_{11}-P_{33}] \div [2(S-\tfrac{1}{2})^2(6Q_{1133}-Q_{1111}-Q_{3333})]$ (21)
- a) $\sin \varphi = 0$
- b) $\cos \varphi = 0$

IV. $\sin^2\vartheta = [\tfrac{1}{2}(4Q_{1133}-Q_{1111}-Q_{1122})+2S^2(3Q_{1133}-Q_{3333})+S(Q_{3333}-7Q_{1133}+$
$+Q_{1111}+Q_{1122})+P_{11}-P_{33}] \div [(S-\tfrac{1}{2})^2(12Q_{1133}-Q_{1111}-2Q_{3333}-3Q_{1122})]$ (22)
$\cos 2\varphi = 0.$

The conditions following from (14) under which the solutions (19)—(22) represent a minimum of h are given in Table I. The last two conditions for the solutions III and IV follow from the obvious restriction

$$0 \leqslant \sin^2 \vartheta \leqslant 1.$$

For the special case of cubic symmetry we have in addition to (15)-(17) the relations

$$P_{11} = P_{33}, \quad Q_{1111} = Q_{3333}, \quad Q_{1122} = Q_{1133} \tag{23}$$

and the minimum conditions from Table I reduce simply to the sign of the difference $3Q_{1122} - Q_{1111}$ as shown in Table II. In the nearest-neighbour approximation with the quadrupolar coupling constant Q the minimum condition reduces to the sign of Q as shown in the last column of Table II. The correspondence to [5] is thus evident.

TABLE I

Solution	Minimum conditions
I	$Q_{1111} + Q_{1122} + 2(3S-2)Q_{1133} - 2SQ_{3333} + \dfrac{P_{11}-P_{33}}{S-\frac{1}{2}} > 0$
IIa, b	$3Q_{1122} - Q_{1111} > 0$ $2SQ_{1111} + Q_{1122} - 2\,(3S-1)\,Q_{1133} - Q_{3333} + \dfrac{P_{11}-P_{33}}{S-\frac{1}{2}} < 0$
IIc	$3Q_{1122} - Q_{1111} > 0$ $(2S+1)Q_{1111} + (6S-1)Q_{1122} - 4(3S-1)Q_{1133} - 2Q_{3333} + 2 \cdot \dfrac{P_{11}-P_{33}}{S-\frac{1}{2}} < 0$
IIIa, b	$3Q_{1122} - Q_{1111} > 0$ $Q_{1111} + Q_{3333} - 6Q_{1133} > 0$ $2SQ_{1111} + Q_{1122} - 2(3S-1)Q_{1133} - Q_{3333} + \dfrac{P_{11}-P_{33}}{S-\frac{1}{2}} > 0$ $Q_{1111} + Q_{1122} + 2(3S-2)Q_{1133} - 2SQ_{3333} + \dfrac{P_{11}-P_{33}}{S-\frac{1}{2}} < 0$
IV	$3Q_{1122} - Q_{1111} < 0$ $Q_{1111} + 3Q_{1122} - 12Q_{1133} + 2Q_{3333} > 0$ $Q_{1111} + Q_{1122} + 2(3S-2)Q_{1133} - 2SQ_{3333} + \dfrac{P_{11}-P_{33}}{S-\frac{1}{2}} < 0$ $(2S+1)Q_{1111} + (6S-1)Q_{1122} - 4(3S-1)Q_{1133} -$ $-2Q_{3333} + 2\,\dfrac{P_{11}-P_{33}}{S-\frac{1}{2}} > 0$

TABLE II

Solution	*	Minimum condition	Sign Q		
			Simple cubic lattice	Body centered cubic lattice	Face centered cubic lattice
I $\sin \vartheta$ — 0, φ — arbitrary	[001]	$3Q_{1122} - Q_{1111} > 0$	−	+	+
IIa $\cos \vartheta = 0, \sin \varphi = 0$	[100]	$3Q_{1122} - Q_{1111} > 0$	−	+	+
IIb $\cos \vartheta = 0, \cos \varphi = 0$	[010]	$3Q_{1122} - Q_{1111} > 0$	−	+	+
IIc $\cos \vartheta = 0, \cos 2\varphi = 0$		Not fulfilled			
IIIa $\sin^2 \vartheta = \frac{1}{2}, \sin \varphi = 0$		Not fulfilled			
IIIb $\sin^2 \vartheta = \frac{1}{2}, \cos \varphi = 0$		Not fulfilled			
IV $\sin^2 \vartheta = \frac{2}{3}, \cos 2\varphi = 0$	[111]	$3Q_{1122} - Q_{1111} < 0$	+	−	−

* Corresponding crystallographic direction

4. *Nearest- and next-nearest-neighbour approximation*

For a more quantitative analysis of the results listed in Table I further approximations and specifications are necessary. First of all, we shall henceforth disregard the long-range dipole-dipole interactions. Furthermore, we shall restrict the short-range interactions to the nearest or next-nearest neighbourhood and thus confine the tetragonal deformation of the crystal lattice as follows:
for the *s. t.* lattice (nearest-neighbour approximation):

$$\frac{1}{2} < \frac{b}{a} < \sqrt{2},$$

and for the *b. c. t.* lattice:

$$\sqrt{2/3} < \frac{b}{a} < \sqrt{2}$$

in the nearest-neighbour approximation, and

$$1 < \frac{b}{a} < \sqrt{6}$$

when taking into account next-nearest-neighbour interactions in the tetragonal plane. Here, b and a denote respectively the lattice constants in the tetragonal direction and plane. Let J, C and Q be respectively the exchange integral and the pseudodipolar and quadrupolar coupling constant for the nearest neighbours in the tetragonal plane, and J', C' and Q' the respective quantities for the neighbouring atoms above and below this plane. Then, we assume after Van Vleck [16] .

$$|C| \approx |J|(g-2)^2, \qquad |Q| \approx \frac{1}{4}|J|(g-2)^4 \tag{24}$$

and the same for the primed quantities. Hence, in this approximation we have

$$\frac{Q'}{Q} = \frac{C'}{C} = \frac{J'}{J} \equiv q, \qquad \frac{C}{Q} = \frac{C'}{Q'} = \pm 4(g-2)^{-2} \equiv r \tag{25}$$

$$\frac{C'}{Q} = r \cdot q$$

Finally, we specify $S = 1$ and $g = 2.1$, *i.e.*, $r = \pm 400$.

a) Simple tetragonal lattice

With the above assumptions we have, according to (15)-(17),

$$\begin{aligned} &Q_{1111} = 2NQ \\ &Q_{3333} = 2NQ' \qquad P_{11} - P_{33} = 6N(C' - C) \\ &Q_{1122} = Q_{1133} = 0 \end{aligned} \tag{26}$$

where N is the number of lattice sites. Now we examine the minimum conditions listed in Table I. For the solution I we obtain

$$Q\{1 - 2q + 6r(q-1)\} > 0 \tag{27}$$

Depending on the sign of C and Q, the above inequality leads to different conclusions.
1) If $Q > 0$ and $C > 0$ one has

$$q > \frac{1}{2}\,\frac{6r-1}{3r-1} > 1 \tag{28}$$

which implies $b < a$, *i.e.*, the lattice must be constracted in the tetragonal direction if this direction is to be magnetically preferred.
2) If $Q < 0$ and $C < 0$, one easily proves that again $q > 1$, *i. e.*, the above conclusion holds.
3) If $Q < 0$ and $C > 0$, it follows from (27) that $q < 1$ which implies $b > a$, *i.e.*, the lattice must be expanded in the tetragonal direction if the latter is to be magnetically preferred.
4) If $Q < 0$ and $C < 0$, again $q < 1$ and $b > a$.

TABLE III

Solution	Sign Q	Sign C	Condition for a and b
I	+	+	$b < a$
	+	−	$b > a$
	−	+	$b < a$
	−	−	$b > a$
IIa, b	−	−	$b < a$
	−	+	$b > a$
IIc	+	+	$b > a$
	+	−	$b < a$

For the remaining solutions in Table I the analysis is analogous though a bit more complicated, as there are two or four inequalities to be examined. The results are listed in Table III. For the solutions IIIa, b and IV (except for the cubic case) the minimum conditions are not satisfied.

b) Body-centred tetragonal lattice; nearest-neighbour approximation

In this case we have, according to (15)–(17),

$$Q_{1111} = \frac{8Na^4Q}{(2a^2+b^2)^2} = Q_{1122}, \qquad Q_{3333} = \frac{8Nb^4Q}{(2a^2+b^2)^2},$$

$$Q_{1133} = \frac{8Na^2b^2Q}{(2a^2+b^2)^2}, \qquad P_{11}-P_{33} = \frac{24C(b^2-a^2)N}{2a^2+b^2}. \tag{29}$$

The analysis of the conditions from Table I is analogous to the preceding case, except that now the lattice deformation enters explicitly the inequalities through a and b, according to (29). It leads to the results given in Table IV. Again, for the solutions IIIa, b IV (except for the cubic case) the minimum conditions are not satisfied.

TABLE IV

Solution	Sign Q	Sign C	Condition for a and b
I	+	+	$b > a$
	−	−	$b < a$
	+	−	$b < a$
	−	+	$b > a$
IIa, b	+	+	$b > a$
	+	−	$b < a$
IIc	−	+	$b < a$
	−	−	$b > a$

TABLE V

Solution	Sign Q	Sign C	Conditions for q	Conditions for $\frac{b}{a}$
I	$\pm$	$+$	$0 < q < \frac{1}{4}$	not fulfilled
			$\frac{1}{4} < q < 1$	$\frac{b}{a} > A$
			$q > 1$	$\frac{b}{a} > \sqrt{2}$
	$\pm$	$-$	$0 < q < \frac{1}{4}$	none
			$\frac{1}{4} < q < 1$	$\sqrt{2} < \frac{b}{a} < A$
			$q > 1$	$\frac{b}{a} < A$
IIa, b	$+$	$+$	$0 < q < \frac{1}{4}$	none
			$\frac{1}{4} < q < 1$	$\sqrt{2} < \frac{b}{a} < A$
			$1 < q < 1.6$	$B < \frac{b}{a} < A$
	$+$	$-$	$1.6 < q < \infty$	not fulfilled
			$0 < q < \frac{1}{4}$	not fulfilled
			$\frac{1}{4} < q < 1.6$	$A < \frac{b}{a} < \sqrt{2}$
			$1.6 < q < 2$	$B < \frac{b}{a} < \sqrt{2}$
	$-$	$+$	$2 < q < \infty$	not fulfilled
			$0 < q < \frac{1}{2}$	not fulfilled
			$\frac{1}{2} < q < 1.6$	$\sqrt{2} < \frac{b}{a} < B$
			$1.6 < q < \infty$	$\sqrt{2} < \frac{b}{a} < A$
	$-$	$-$	$0 < q < 1.6$	not fulfilled
			$1.6 < q < \infty$	$A < \frac{b}{a} < \sqrt{2}$

TABLE V — continued

Solution	Sign Q	Sign C	Conditions for q	Conditions for $\frac{b}{a}$
	+	+	$0 < q < \frac{1}{2}$	not fulfilled
			$\frac{1}{2} < q < 1.6$	$\sqrt{2} < \frac{b}{a} < B$
			$1.6 < q < \infty$	$\sqrt{2} < \frac{b}{a} < A$
	+	−	$0 < q < 1.6$	not fulfilled
			$1.6 < q < \infty$	$A < \frac{b}{a} < \sqrt{2}$
	−	+	$0 < q < \frac{1}{4}$	none
IIc			$\frac{1}{4} < q < 1$	$\sqrt{2} < \frac{b}{a} < A$
			$1 < q < 1.6$	$B < \frac{b}{a} < A$
			$1.6 < q < \infty$	not fulfilled
	−	−	$0 < q < \frac{1}{4}$	not fulfilled
			$\frac{1}{4} < q < 1.6$	$A < \frac{b}{a} < \sqrt{2}$
			$1.6 < q < 2$	$B < \frac{b}{a} < \sqrt{2}$
			$2 < q < \infty$	not fulfilled

$$\sqrt{\frac{4q+2}{4q-1}} \equiv A$$
$$\sqrt{2(\sqrt{2q}-1)} \equiv B$$

c) Body-centred tetragonal lattice; next-nearest-neighbour approximation in the tetragonal plane

For the quantities (15)-(19) we have

$$Q_{1111} = N\left[Q + \frac{4a^4Q'}{(a^2+b^2)^2}\right],$$
$$Q_{1122} = NQ$$
$$Q_{3333} = \frac{8b^4Q'N}{(a^2+b^2)^2},$$

$$Q_{1133} = \frac{4a^2b^2Q'N}{(a^2+b^2)^2}, \qquad P_{11}-P_{33} = 6\left(C'\,\frac{4b^2-2a^2}{a^2+b^2}-C\right) \tag{30}$$

and now the lattice deformation manifests itself implicitly (trough q) as well as explicitly (trough a and b) in the minimum conditions from Table I. This complicates the analysis insofar as there is the additional obvious condition to be accounted for, namely, $q < 1$ if $(b/a) > 1$ (and *vice versa*). The results are presented in Table V which lacks the solutions IIIa, b and IV as the corresponding minimum conditions are not fulfilled (except for IV in the cubic case).

5. *Final remarks*

The method applied here in determining the magnetically preferred directions in tetragonal ferromagnets is based on the minimization of the system's energy in the class of saturation states (6). Inasmuch as there are assumed anisotropic interactions of dipolar and quadrupolar type, this procedure may raise objections because the saturation state is not an eigenstate of the system. In fact, it is yet to be proved that the saturation state is at least a reasonable approximation of the system's exact ground state. None the less, there is a strong experimental justification for our approach, as small single-domain ferromagnetic particles of spherical shape are indeed spontaneously magnetized to saturation in magnetically preferred directions. Furthermore, even in larger single crystals with domain structures the domains themselves are also magnetized in magnetically preferred directions. We therefore believe that the saturation-state approximation should in this case work well. A further argument in favour of this conviction is provided by the fact that the inclusion of long-range dipole-dipole interactions in our considerations leads to restrictive conditions for the dimensions of the single-domain crystal and, for crystals of ellipsoidal shape, to shape-dependent magnetically preferred directions — quite like in the phenomenological theory. This will be shown in a subsequent paper.

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APPENDIX

The explicit form of the average energy $h(\vartheta, \varphi)$, Eq. (11), in the saturation state (6) is as follows:

$$\begin{aligned} h = S^2 \sum_{\alpha,\beta} \Big\{ & P_{33}^{\alpha\beta} + \tfrac{1}{2}(Q_{1111}^{\alpha\beta}+Q_{1122}^{\alpha\beta}) + 2SQ_{1133}^{\alpha\beta} + S\,Q_{3333}^{\alpha\beta} + \\ & +(S-\tfrac{1}{2})\sin^2\vartheta \left[\frac{P_{11}^{\alpha\beta}-P_{33}^{\alpha\beta}}{S-\frac{1}{2}} + Q_{1111}^{\alpha\beta}+Q_{1122}^{\alpha\beta}+2(3S-2)Q_{1133}^{\alpha\beta}-2SQ_{3333}^{\alpha\beta}\right] + \\ & +\tfrac{1}{2}(S-\tfrac{1}{2})^2\sin^4\vartheta\,(Q_{1111}^{\alpha\beta}-6Q_{1133}^{\alpha\beta}+Q_{3333}^{\alpha\beta}) + \\ & + \tfrac{1}{2}(S-\tfrac{1}{2})^2\sin^4\vartheta\,\sin^2\varphi\,(3Q_{1122}^{\alpha\beta}-Q_{1111}^{\alpha\beta})\Big\}. \end{aligned}$$

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