Various methods for analyzing data on anisotropic scalar properties in cubic symmetry: Application to magnetic anisotropy energy of nickel

G. Aubert
Laboratoire de Magnétisme, Centre National de la Recherche Scientifique, Boîte Postale 166, Centre de tri, 38042 Grenoble Cedex, France

Y. Ayant, E. Belorizky, and R. Casalegno
Laboratoire de Spectrométrie Physique, associé au Centre National de la Recherche Scientifique, Université Scientifique et Médicale de Grenoble, Boîte Postale 53, Centre de tri, 38041 Grenoble Cedex, France

(Received 12 April 1976)

In cubic symmetry, the description of a scalar anisotropic physical quantity E relative to a static property of a given material can be made with the help of various expansions in terms of symmetrical polynomials or cubic harmonics. A general method for generating the relevant symmetrical polynomials up to an arbitrary order is presented and a new set of cubic harmonics particularly suitable to this problem is tabulated up to order $l = 36$. The relations of the latter with the symmetrical polynomials are set up. A careful study of the very important case where E can be accurately measured only in the planes {100} and {110} is made. From the knowledge of the Fourier expansions of E in these planes the conditions for deriving E in a unique way for arbitrary directions are given. It is also shown that some of the coefficients appearing in the former expansions of E may always be determined unambiguously whatever the order up to which these expansions have to be performed. Practical applications of these results are developed with specific reference to the magnetic anisotropy energy of nickel at low temperatures. This study demonstrates the advantages of this method of analysis over the usual procedures.

I. INTRODUCTION

The physical properties of crystals are defined by relations, usually of tensorial type, between several measurable quantities which are associated with this crystal. Many properties may be specified by a relation between a scalar quantity and a polar or axial vector. Some examples are given by the free energy of a single-domain ferro- or ferrimagnetic crystal in equilibrium as a function of the magnetization vector $E(\vec{M})$ or by the radius of the Fermi surface for a metal as a function of the direction $\vec{u}$ of the wave vector $k_F(\vec{u})$.

In a general way, a scalar quantity E may be expressed as a function of a vector $\vec{V}$ through an expansion of the form

$$E = a_0 + \sum_i a_i \alpha_i + \sum_{i,j} a_{ij}\alpha_i\alpha_j + \sum_{i,j,k} a_{ijk}\alpha_i\alpha_j\alpha_k + \cdots, \tag{1}$$

where the α_i's ($i = 1, 2, 3$) are the direction cosines of $\vec{V}$ in an orthogonal coordinate system xyz.

The form of expansion (1) is such that the dependence of the $\underline{a}$ tensors on $\vec{V}$ is only through its modulus $|\vec{V}|$. These tensors, which may be dependent on other parameters (temperature or hydrostatic pressure, for example), are characteristic of the physical property under consideration.

The point-group symmetry of the crystal, the parity and the time-reversal symmetry of $\vec{V}$ imply some restrictions on the existence and the form of the $\underline{a}$ tensors. Introduction of the time-reversal operation requires a distinction between time-odd and time-even tensors according to whether or not their components change sign on time reversal.[1] Here we shall restrict ourselves to the case where the property under consideration is static, so that there is no preferred direction in time, E being a pure scalar time-even quantity and $\vec{V}$ a polar or axial time-odd vector. With the above conditions all the odd-order tensors in (1) are time-odd and so must vanish; E may be rewritten

$$E = a_0 + \sum_{i,j} a_{ij}\alpha_i\alpha_j + \sum_{i,j,k,l} a_{ijkl}\alpha_i\alpha_j\alpha_k\alpha_l + \cdots. \tag{2}$$

When further restrictions imposed by spatial symmetry are expressed, the appropriate expansion of E in terms of increasing powers of the direction cosines of $\vec{V}$ is obtained.

For some applications, such an expansion is hardly tractable and the expansion (3) in terms of the spherical harmonics $Y_{l,m}(\theta, \phi)$ of the direction (θ, ϕ) of $\vec{V}$ is more appropriate

$$E = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \lambda_{l,m} Y_{l,m}(\theta, \phi). \tag{3}$$

The main advantage of the spherical harmonics is that they form a complete orthonormal set of square integrable functions on the unit sphere.

The convergence of expansions (2) and (3) is not necessarily rapid and the order to which one may limit them is essentially dependent on the kind of experiment and the accuracy of the measurements. It will be seen that to describe correctly the variation of the anisotropy energy in nickel at liquid-helium temperature, an expansion up to the 18th order at least is required, while for iron at the same temperature there are no significant terms beyond the 8th order.

The usefulness of such expansions up to such high orders may be questionable since it is almost impossible in the frame of available theories to calculate from first principles the components of the $\underline{a}$ tensors or the $\lambda_{l,m}$ coefficients. But, as we shall see later, these expansions are absolutely necessary for correct analysis of the experimental data.

Furthermore the evolution of the scalar quantity E as a function of various parameters, such as temperature in the case of anisotropy energy, or any comparison of the properties of different materials is most easily described using the coefficients of the above expansions.

Our study will be limited to crystals with cubic symmetry for which the relevant point group is O_h. The expansion in terms of direction cosines is relatively simpler in that case; on the other hand the corresponding expansion in terms of spherical harmonics is more complicated because one of the three fourfold axes plays a privileged role as z axis. Although this problem has already been extensively studied by several authors (references will be found in the papers by Altmann, and Cracknell,[2] Mueller and Priestley,[3] and Birss and Keeler[4]), important questions remain unclear for some practical applications.

In Sec. II the explicit forms of the expansions appropriate to O_h symmetry are recalled: expansion (2) is preferably expressed in terms of suitable symmetrical polynomials and expansion (3) is written in terms of normalized linear combinations of spherical harmonics invariant in cubic symmetry[5] (cubic harmonics). Special attention is paid to the number of independent coefficients to be determined up to a given order. From the symmetrical polynomials, it is possible to derive a set of cubic harmonics the coefficients of which are square roots of exact rational ratios. The relations between the symmetrical polynomials and these cubic harmonics are tabulated and thus we avoid any fruitless discussion about the respective advantages of either type of expansion.

Section III is mainly devoted to the methods of extracting from the experimental data the coefficients of the above expansions. In principle, n measurements of E for n arbitrary directions of $\vec{V}$ provide n equations for determining n coefficients in the expansion of E; but the experimental difficulties usually do not allow the possibility of getting reliable enough results for directions of $\vec{V}$ outside the symmetry planes $\{100\}$ and $\{110\}$ for a cubic crystal. The consequences of these limitations on the number of independent coefficients in the expansions which can be obtained, are discussed. General methods are proposed for the calculation of the coefficients from the experimental data according to the convergence of the expansions.

In Sec. IV, the methods outlined in Sec. III are used and the unknown coefficients of the expansions of E in terms of symmetrical polynomials are explicitly determined from the results of Fourier analysis of the experimental data in the symmetry planes.

Some experimental results for the magnetic anisotropy energy of nickel at low temperatures are analyzed in Sec. V. This reference to a typical experimental situation will make evident the usefulness of our method which allows the determination of the first coefficients of our expansions without having to invert any system of equations.

II. EXPLICIT FORMS OF THE EXPANSIONS FOR O_h SYMMETRY

A. Number of independent coefficients

The appropriate form of the expansion (3) in terms of spherical harmonics may be written

$$E=\sum_{i,l} \mathcal{A}_{i,l} K_{i,l}, \qquad (4)$$

where the $K_{i,l}$, called the cubic harmonics,[5] are linear combinations of the spherical harmonics normalized on the unit sphere and invariant under the operations of the O_h group. Generally, we have

$$K_{i,l}=\sum_{m} {}_i a_{l,m} C_{l,m}, \qquad (5)$$

where the $C_{l,m}$ are real normalized spherical harmonics.[3] The index i labels the g_l different independent cubic harmonics for a given l when $g_l > 1$. g_l is simply given by the number of unit irreducible representations Γ_1^+ of the O_h group contained in the reduction of the representation D_l^+ of the full rotation group. Using the character table of the O_h group one easily gets

$$g_l = 1+[\tfrac{1}{4}l]-[\tfrac{1}{6}(l+4)] \qquad (6)$$

for l even, where the symbol $[x]$ denotes the largest integer $\leqslant x$, and, of course, $g_l = 0$ for l odd.

Mueller and Priestley[3] with the choice of a particular set of cubic harmonics, give numerical values of the coefficients ${}_i a_{l,m}$ with 8 significant figures up to $l = 30$.

B. Symmetrical polynomials

Let α_1, α_2, α_3 be the direction cosines of $\vec{V}$ with respect to the three fourfold axes of the cube. The equivalence of these three axes leads immediately to an expansion for E of the form

$$E = \sum_{l=0}^{\infty} \sum_{n,m} A^l_{n,m} S^n P^m , \qquad (7)$$

with l even and $n, m \geq 0$; S and P are defined by

$$S = 3(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) , \qquad (8)$$

$$P = 27\alpha_1^2\alpha_2^2\alpha_3^2 , \qquad (9)$$

where the coefficients 3 and 27 are such that the above expressions both vary from the minimum value 0 for direction ⟨100⟩ to the maximum value 1 for direction ⟨111⟩. The $A^l_{n,m}$ are independent coefficients to be determined from experiment. The number g_l of coefficients $A^l_{n,m}$ for a fixed value of l is equal to the number of integer solutions of the equation

$$4n + 6m = l . \qquad (10)$$

It is easy to check the agreement with the previous determination of g_l from group theory. For example, in the case of $l = 24$, we have three coefficients: $A^{24}_{6,0}$, $A^{24}_{3,2}$, and $A^{24}_{0,4}$.

As we shall see later in this paper, the particularly simple form of expansion (7) is certainly the most convenient for analyzing experimental data. Its only disadvantage comes from the fact that the different monomials S^nP^m are neither orthogonal nor normalized.

For those who would prefer, however, to use an expansion in terms of spherical harmonics, we have been led to build a set of cubic harmonics related with the above polynomials in a more straightforward way than the cubic harmonics of Mueller and Priestley from which they differ for $l \geq 12$.

C. Cubic harmonics

A monomial such as S^nP^m, of order $l = 4n + 6m$, being invariant under the operations of the cubic group, may be expressed as a linear combination of spherical harmonics associated with the Γ_1^+ representation of the O_h group. The maximum order of these spherical harmonics is obviously l.

A cubic harmonic of order l is then defined by normalizing on the unit sphere the part of order l of the above linear combination. Each monomial S^nP^m thus allows one to define a cubic harmonic $K^l_{n,m}$ which may be written

$$K^l_{n,m} = \sum_{q=0}^{n+m} a^{(q)}_{n,m} C_{l,4q} . \qquad (11)$$

The explicit method for obtaining these cubic harmonics is given in Appendix A, but the following remarks must be made: (i) The coefficients $a^{(q)}_{n,m}$ appearing in (11) have the advantage to be known exactly; their square in rational form and their numerical values with eight exact figures up to $l = 36$ are available.[6] (ii) When $g_l = 1$ our cubic harmonics are of course identical with those given by other authors, as they are unique: $K_l = K^l_{n,m}$. (iii) When $g_l > 1$, we obtain g_l independent cubic harmonics $K^l_{n,m}$ derived from the g_l independent monomials S^nP^m with $4n + 6m = l$. The cubic harmonics involve coefficients $a^{(q)}_{n,m}$ which are still square roots of fractional ratios, but they are no longer orthogonal within each subset of order l. Their main advantage is that they may be expressed quite simply in terms of the symmetrical polynomials. If necessary, each subset of cubic harmonics of order l may of course be orthogonalized by the usual procedures.

D. Relations between the two kinds of expansion

With the previously defined cubic harmonics the anisotropic scalar quantity E may be written

$$E = \sum_{l=0}^{\infty} \sum_{n,m} \mathfrak{A}^l_{n,m} K^l_{n,m} \quad (l = 4n + 6m) . \qquad (12)$$

Of course expansions (7) and (12) involve the same number of independent coefficients which depend only on the required precision to fit the experimental data. In order to obtain the relations between the two independent sets of coefficients $A^l_{n,m}$ and $\mathfrak{A}^l_{n,m}$ the S^nP^m will be expressed in terms of the cubic harmonics $K^l_{n,m}$. For convenience we introduce the functions $H^l_{n,m}$ related to the cubic harmonics by

$$K^l_{n,m} = a^{(0)}_{n,m} \, [(2l+1)/4\pi]^{1/2} H^l_{n,m} . \qquad (13)$$

We can then write

$$S^nP^m = \sum_{\lambda=0}^{l} \sum_{\nu,\mu} \beta^{n;m}_{\nu,\mu} H^\lambda_{\nu,\mu} , \qquad (14)$$

with, for the integers λ, ν, μ, $\lambda = 4\nu + 6\mu$.

According to the way we have derived the cubic harmonics we note that in Eq. (14), for $\lambda = l$, $\beta^{n;m}_{n,m}$ is the only nonvanishing coefficient.

The numerical values of the $\beta^{n;m}_{\nu,\mu}$ in rational form are also available[6] for $l \leq 36$. In Appendix B the method used to get these results is summarized.

III. ANALYSIS OF THE DATA

A. Fourier expansions in the symmetry planes

As has already been stated, in most cases the anisotropic scalar quantity E can be measured conveniently in the symmetry planes of the crystal only, i.e., in cubic symmetry, the $\{100\}$ and the $\{110\}$ planes. The main reason for these restrictions is particularly clear in the specific example of the magnetic anisotropy energy of ferromagnetic crystals: the external magnetic field $\vec{H}$ must be rotated in a symmetry plane in order that the magnetization vector $\vec{M}$ remains in the same plane as $\vec{H}$, and thus has its direction accurately calculable from the measured torque, for instance.[7]

The experimental variations of $E(\theta, \phi)$ in the planes $\{100\}$ or $\{110\}$ can be given as expansions in terms of the angle θ between the measuring direction and the [001] direction:

$$E_{100} = E(\theta, 0) = A(\theta) = \sum_{k=0}^{K_a} a_{4k} \cos 4k\theta \ , \tag{15a}$$

$$E_{110} = E(\theta, \tfrac{1}{4}\pi) = B(\theta) = \sum_{k=0}^{K_b} b_{2k} \cos 2k\theta \, . \tag{15b}$$

The orders $4K_a$ or $2K_b$ up to which the above expansions are limited depend on the accuracy of the experiment. In these conditions a given number of coefficients a_{4k} or(and) b_{2k} may be obtained from the experimental data. By projecting Eq. (7) in the two planes $\{100\}$ and $\{110\}$, the coefficients a_{4k} and b_{2k} are expressed as linear combinations of the $A^l_{n,m}$. In what follows we discuss in detail the the possibility of inverting in a unique way the previous linear system in order to obtain the unknown coefficients $A^l_{n,m}$; this last point is of fundamental importance for the knowledge of E for an arbitrary direction, i.e., for a complete description of the investigated anisotropic property.

B. Uniqueness of the inversion

We shall discuss successively three different cases depending on whether experimental data are taken in a single or in both symmetry planes.

1. $\{100\}$ plane only

Projection of Eq. (7) involves only the coefficients $A^l_{n,0}$ because $P \equiv 0$ in this plane. The experimental expansion (15a) limited to an order $L = 4K_a$ according to the accuracy of the measurements, is identified with the above projection limited to the same order ($n \leq K_a$). We obviously get a linear system of $(K_a + 1)$-independent equations with $K_a + 1$ unknown $A^l_{n,0}$. Therefore, experiments in the $\{100\}$ planes only, allow the determination of all the coefficients $A^l_{n,0}$ for which $l \leq L = 4K_a$.

2. $\{110\}$ plane only

As E is extremal for the symmetry direction [111], we must have

$$\left(\frac{dB}{d\theta}\right)_{\theta=\theta_0} = 0 \, , \quad \cos\theta_0 = 1/\sqrt{3} \ , \tag{16a}$$

thus the $K_b + 1$ experimental coefficients b_{2k} are not independent and they must obey the following relation:

$$\sum_{k=1}^{K_b} k b_{2k} \sin 2k\theta_0 = 0 \, . \tag{16b}$$

Projection of Eq. (7) limited to the order $L = 2k_b$ involves all the coefficients $A^l_{n,m}$ up to this order. The number N_L of these unknown coefficients is given by

$$N_L = \sum_{l=0}^{L} g_l \, , \tag{17}$$

where the values of g_l are determined by Eq. (6).

The linear system of equations obtained by identification of the experimental expansion (15b) with this projection is then a system of K_b-independent equations for $K_b \geq 2$. It is easy to check that, up to $L = 2K_b \leq 10$, we have $K_b = N_L$ and so, experiments in the $\{110\}$ planes only allow a unique determination of E. On the other hand, for $L \geq 12$, as $N_L > K_b$ we have an indeterminate system, but we shall see later that the coefficient $A^4_{1,0}$ (and of course $A^0_{0,0}$) can still be determined in a unique way.

3. $\{100\}$ and $\{110\}$ planes

We now use both experimental expansions (15a) and (15b) with $K_a = [\frac{1}{2}K_b]$, and projections of Eq. (7) in both planes limited to the order $L = 2K_b$. The number N_L of unknown coefficients $A^l_{n,m}$ is still given by Eq. (17). In order to have the number of independent Fourier coefficients, we must take into account the following relations required by the symmetry of the problem:

$$A(0) = B(0) \, , \tag{18a}$$

$$A(\tfrac{1}{4}\pi) = B(\tfrac{1}{2}\pi) \, , \tag{19a}$$

$$\left(\frac{d^2A}{d\theta^2}\right)_{\theta=0} = \left(\frac{d^2B}{d\theta^2}\right)_{\theta=0} \, . \tag{20a}$$

The experimental coefficients a_{4k} and b_{2k} must then satisfy

$$\sum_{k=0}^{K_a} a_{4k} = \sum_{k=0}^{K_b} b_{2k} = A^0_{0,0} \, , \tag{18b}$$

$$\sum_{k=0}^{K_a}(-1)^k a_{4k} = \sum_{k=0}^{K_b}(-1)^k b_{2k}\ , \qquad (19b)$$

$$4\sum_{k=1}^{K_a} k^2 a_{4k} = \sum_{k=1}^{K_b} k^2 b_{2k}\ . \qquad (20b)$$

We express by Eqs. (20) that near the fourfold axis [001], $(\partial^2 E/\partial\theta^2)_{\theta=0}$ is independent of ϕ; this property results directly from the fact that when $\theta\to 0, E(\theta,\phi)=\alpha+\beta\theta^2+\ldots$, where α and β are constants.

We then have K_a+K_b+2 coefficients a_{4k} and b_{2k} which are subject to the four Eqs. (16b), (18b), (19b), and (20b). One may easily see that these four relations are independent for $L=2K_b \geqslant 8$, but, if we expect the trivial case where $L=0$, only three of them are independent for $L=4$ and $L=6$. Therefore, experiments in both symmetry planes lead, after checking their reliability through the previous relations, to $(N_C=K_a+K_b-1)$- or $(N_C=K_a+K_b-2)$-independent Fourier coefficients according to whether $4\leqslant L\leqslant 6$ or $L\geqslant 8$, respectively. The values of N_C and N_L are compared in Table I and we see immediately that $N_C=N_L$ for $L\leqslant 16$. So a unique determination of E is possible up to this order. But for $L\geqslant 18$, we have $N_L > N_C$ which means that there is an infinity of functions $E(\theta,\phi)$ compatible with $A(\theta)$ and $B(\theta)$.

It must be noted that with a particular choice of cubic harmonics Mueller and Priestley[3] have already shown that a unique expansion of the anisotropic quantity is possible for $L<18$ from data in two planes.

The reason for the indeterminacy appearing for $L\geqslant 18$ will be discussed below together with a possible process of inversion for this case. However, we shall show first that all the coefficients $A^l_{n,m}$ up to the order $l=10$, that is, the first five coefficients of expansion (7), can always be unambiguously obtained. This is a particularly interesting feature of the expansion of E in terms of symmetrical polynomials which does not exist for an expansion in terms of cubic harmonics.

TABLE I. Comparison, for each value of L, between the number N_C of independent Fourier coefficients in the symmetry planes and the number N_L of unknown coefficients in the expansions (7) or (12) of E.

L	4	6	8	10	12	14	16	18	20	22	24	⋯
K_a	1	1	2	2	3	3	4	4	5	5	6	⋯
K_b	2	3	4	5	6	7	8	9	10	11	12	⋯
N_C	2	3	4	5	7	8	10	11	13	14	16	⋯
N_L	2	3	4	5	7	8	10	12	14	16	19	⋯

C. Direct determination of the first coefficients

$A^0_{0,0}$ is immediately given in either of the two planes by Eq. (18b). In the same way, using the projections of Eq. (7) in the two symmetry planes, we get from relations (20)

$$\left(\frac{d^2A}{d\theta^2}\right)_{\theta=0} = \left(\frac{d^2B}{d\theta^2}\right)_{\theta=0} = 6A^4_{1,0} \qquad (21)$$

and

$$A^4_{1,0} = -\frac{8}{3}\sum_{k=1}^{K_a} k^2 a_{4k} = -\frac{2}{3}\sum_{k=1}^{K_b} k^2 b_{2k}\ . \qquad (22)$$

This result shows that the coefficient $A^4_{1,0}$ may be determined independently in the {100} and {110} planes and separately from the other coefficients $A^l_{n,m}$. Besides the intrinsic importance of obtaining directly $A^4_{1,0}$, we can in this way check the reliability of the data taken in two different planes which may have been affected by slight variations in the experimental conditions. This is particularly important in the case of anisotropy energy measurements because of the very strong variation with temperature of the anisotropy constants.[7-9]

Pursuing the procedure initiated by Eq. (21), we calculate higher-order derivatives of $B(\theta)$ for $\theta=0$. We thus get

$$\left(\frac{d^4B}{d\theta^4}\right)_{\theta=0} = 16\sum_{k=1}^{K_b} k^4 b_{2k} = -78A^4_{1,0}+162A^6_{0,1}+216A^8_{2,0}\ , \qquad (23)$$

$$\left(\frac{d^6B}{d\theta^6}\right)_{\theta=0} = -64\sum_{k=1}^{K_b} k^6 b_{2k} = 1176A^4_{1,0}-8100A^6_{0,1} - 14\,040A^8_{2,0}+14\,580A^{10}_{1,1}+19\,440A^{12}_{3,0}\ . \qquad (24)$$

As all the $A^l_{n,0}$ are known from data in the {100} plane, Eq. (23) provides $A^6_{0,1}$ and then Eq. (24) provides $A^{10}_{1,1}$. The following unknown coefficients cannot be derived from higher-order derivatives because, at each step, at least two new indetermined $A^l_{n,m}$ appear.

This possibility of obtaining uniquely the five first $A^l_{n,m}$ in the expansion of E, irrespective of the total number of coefficients necessary to describe the property, is of fundamental importance and, to our knowledge, has never yet been used.

The complete inversion of the systems involved in the three cases of Sec. III B will be discussed in Sec. IV.

D. Origin of the indeterminacy for $L\geqslant 18$

The indeterminacy appearing at order 18 results directly from the fact that the experiments provide the values of E only in nine planes: the three equivalent $\{100\}$ planes and the six equivalent $\{110\}$ planes. Indeed, let us build the polynomial ψ of lowest order with respect to α_1, α_2, α_3 invariant in cubic symmetry (O_h group) and vanishing in the nine symmetry planes. This polynomial must be divisible by the ninth-order polynomial Q_9 defined by

$$Q_9 = \alpha_1\alpha_2\alpha_3(\alpha_1^2 - \alpha_2^2)(\alpha_2^2 - \alpha_3^2)(\alpha_3^2 - \alpha_1^2) . \quad (25)$$

It may be noted that Q_9, which is odd under inversion vanishes in the nine planes and is invariant in the O group but not in the O_h group; Q_9 is proportional to the odd cubic harmonic of lowest order K_9. The ratio between our unknown polynomial ψ and Q_9 must be odd and invariant in O group and is thus proportional to K_9. Consequently ψ is proportional to $(K_9)^2$ and is of order 18. More precisely, in what follows we will define ψ_{18} as

$$\psi_{18} = (Q_9)^2 . \quad (26)$$

Thus if $A(\theta)$ and $B(\theta)$ contain nonvanishing terms up to order $L=18$, for instance, it is of course possible to find a set of $A^l_{n,m}$ in the expansion (7) of E for arbitrary directions by choosing an additional condition such as $A^{18}_{0,3}=0$; but any other solution of the type $E-\lambda\psi_{18}$, where λ is an arbitrary parameter, will be equally valid.

In the same way, other polynomials $\psi_{22}, \psi_{24}, \ldots$ invariant in O_h group and vanishing in the nine symmetry planes may be obtained by multiplying ψ_{18} by the successive even cubic invariants proportional to the cubic harmonics $K_4, K_6, \ldots$. This is in agreement with the fact that $N_L - N_C = 1, 2, 3, \ldots$ for $18 \leq L \leq 20$, $L=22$, $L=24$, ..., respectively.

It is easy to express ψ_{18} defined by Eqs. (25) and (26) in terms of S and P, as given in (8) and (9), as

$$\psi_{18} = 27^{-2}(-4P^2 + 3S^2P + 6SP^2 - P^3 - 4S^3P) . \quad (27)$$

E. Attempts at optimisation for $L \geq 18$

We have just seen that when $L \geq 18$ it is necessary to introduce additional arbitrary conditions to achieve a complete inversion of the data. When $L=18$, for example, there are $12\,A^l_{n,m}$ to be determined which are related by 11 equations only. One additional arbitrary condition is then required, but the results of the inversion vary from one condition to the other. The aim of this section is to find which could be the best condition. In fact, of the 12 unknown $A^l_{n,m}$, $A^0_{0,0}$, $A^4_{1,0}$, $A^6_{0,1}$, $A^8_{2,0}$, $A^{10}_{1,1}$, $A^{12}_{3,0}$, $A^{16}_{4,0}$ may be obtained directly as explained in Sec. III C and only five of them, i.e., $A^{12}_{0,2}$, $A^{14}_{2,1}$, $A^{16}_{1,2}$, $A^{18}_{3,1}$, $A^{18}_{0,3}$, truly depend on our particular choice. Thus, the part of expansion (7) limited to the order $l=18$ which remains unknown involves the same monomials S^nP^m as the function ψ_{18} expressed by Eq. (27). This point results directly from the fact that two different solutions of the problem $E(\theta,\phi)$ and $F(\theta,\phi)$ are necessarily related by

$$F(\theta,\phi) = E(\theta,\phi) - \lambda\phi_{18} , \quad (28)$$

where λ is a real constant and ϕ_{18} is the normalized expression for ψ_{18}. In what follows we designate $D(\theta,\phi)$ as the common part of E and F involving the seven known $A^l_{n,m}$, and $e(\theta,\phi)$ and $f(\theta,\phi)$ as the remaining parts; we have then

$$f(\theta,\phi) = e(\theta,\phi) - \lambda\phi_{18} . \quad (29)$$

The simplest way of inverting our whole system is to set $A^{18}_{3,1}=0$ or $A^{18}_{0,3}=0$. As there is no special reason for setting any one of the five unknown coefficients equal to zero more than an other we have tried to build a function $F(\theta,\phi)$ describing our anisotropic physical property which would be less arbitrary.

It was initially proposed to make the function $F(\theta,\phi)$ orthogonal to ϕ_{18}. Starting from a function $E(\theta,\phi)$ obtained from one of the above arbitrary choices, the condition

$$\langle F|\phi_{18}\rangle = \int F\phi_{18}\,d\Omega = 0 \quad (30)$$

leads to a unique function $F(\theta,\phi)$ with $\lambda = \langle E|\phi_{18}\rangle$. It is easy to check that Eq. (30) is equivalent to minimizing $\langle F|F\rangle$ on the unit sphere. The value of λ obtained in this way is of the order of the constant term $A^0_{0,0}$ and the coefficient of P^2 for instance, which is $A^{12}_{0,2}$ in $E(\theta,\phi)$, becomes of the order of $300\,A^0_{0,0}$ in $F(\theta,\phi)$. Thus the function $F(\theta,\phi)$ is strongly affected by this condition for any direction outside the symmetry planes. The procedure is thus inadequate. An improvement is obtained if, instead of orthogonalizing $F(\theta,\phi)$ to ϕ_{18}, we orthogonalize only the unknown part $f(\theta,\phi)$ to ϕ_{18} and thus take $\lambda = \langle e|\phi_{18}\rangle$. In this way λ is of the order of $0.2\,A^{12}_{0,2}$, and a term such as $A^{12}_{0,2}P^2$ in $E(\theta,\phi)$ is replaced by roughly $-26A^{12}_{0,2}P^2$ in $F(\theta,\phi)$, a result which however is still not very satisfactory.

It seemed to us more reasonable, in order to have as smooth as possible a surface $r=F(\theta,\phi)$, to build a function $f(\theta,\phi)$ in such a way that $\langle f^2\rangle - \langle f\rangle^2$ is minimum ($\langle f\rangle$ and $\langle f^2\rangle$ are, respectively, the mean values of f and f^2 over all the space directions; we must note that making $\langle f|f\rangle$ minimum is equivalent to the condition $\langle f^2\rangle$ minimum). In this way $A^{12}_{0,2}P^2$ in $E(\theta,\phi)$ is replaced by $35\,A^{12}_{0,2}P^2$ in $F(\theta,\phi)$, this result is not more satisfactory.

The reason for this failure comes from the fact that the function ϕ_{18} is strongly anisotropic with very sharp peaks in 48 directions. In the octant where α_1, α_2, α_3 are positive there are six such directions given by

$$\alpha_i = 0.835\,992\,09, \quad \alpha_j = 0.504\,896\,07,$$
$$\alpha_k = 0.214\,935\,27\ ,$$

where (i, j, k) is one of the permutations of $(1, 2, 3)$. Thus all the surfaces $F(\theta, \phi)$ previously built differ considerably from the surface $E(\theta, \phi)$ because of the existence of these peaks (or holes). The best condition is probably to choose λ in such a way that the area of the surface $F(\theta, \phi)$ is minimum, but this requirement is very hardly tractable.

To conclude this section, we think that for $L = 18$ the most reasonable arbitrary condition is still to set $A^{18}_{3,1} = 0$ or $A^{18}_{0,3} = 0$. The same procedure may be pursued for higher values of L: (i) for $L = 20$, we shall take $A^{20}_{2,2} = 0$; (ii) for $L = 22$, two additional conditions are necessary and we can take $A^{20}_{2,2} = 0$ and $A^{22}_{1,3} = 0$.

IV. INVERSION OF THE SYSTEMS

A. {100} plane

The projection of Eq. (7) in the {100} plane is

$$E_{100} = \sum_{n=0}^{\infty} A^{l}_{n,0} S^n\ , \tag{31}$$

with

$$S^n = (3^n / 2^{2n}) \sin^{2n} 2\theta\ .$$

From the relation

$$\sin^{2n} 2\theta = \frac{1}{2^{2n}} C^n_{2n} + \frac{1}{2^{2n-1}} \sum_{p=1}^{n} (-1)^p C^{n+p}_{2n} \cos 4p\theta\ , \tag{32}$$

expansion (31) can be put in the form (15a) and we get, by identification of the coefficients

$$a_0 = \sum_{n=0}^{K_a} \frac{3^n}{2^{4n}} C^n_{2n} A^l_{n,0}\ ,$$
$$a_{4k} = \sum_{n=k}^{K_a} (-1)^k \frac{3^n}{2^{4n-1}} C^{n+k}_{2n} A^l_{n,0}\ . \tag{33}$$

We shall leave aside the determination of $A^0_{0,0}$ which is easily achieved by the relation (18b). However, in some cases $A^0_{0,0}$ cannot be obtained: for instance, the most accurate studies of magnetic anisotropy energy, through torque measurements,[7,9] provide only the first derivative with respect to θ of the projection of Eq. (7) in the plane of measurement. The inversion of the linear system (33) limited to $L = 4K_a = 18$ gives in matrix form

$$\begin{bmatrix} 3/2^3 & A^4_{1,0} \\ 3^2/2^7 & A^8_{2,0} \\ 3^3/2^{11} & A^{12}_{3,0} \\ 3^4/2^{15} & A^{16}_{4,0} \end{bmatrix} = \begin{bmatrix} -1 & -4 & -9 & -16 \\ & 1 & 6 & 20 \\ & & -1 & -8 \\ & & & 1 \end{bmatrix} \begin{bmatrix} a_4 \\ a_8 \\ a_{12} \\ a_{16} \end{bmatrix} \tag{34}$$

The corresponding calculations when $L = 36$ have also been performed.[6]

It is to be noticed that the order of magnitude of the factors $3^n/2^{4n-1}$ makes the contribution of high-order coefficients $A^l_{n,0}$ to the experimental coefficients a_{4k} decrease very fast with n. The reliability of the determination of the former is thus very sensitive to the accuracy of the experiment.

B. {110} plane

The projection of Eq. (7) in the {110} plane is obtained by replacing S and P by the expressions

$$S = 3\sin^2\theta - \tfrac{9}{4}\sin^4\theta\ ,$$
$$P = \tfrac{27}{4}\sin^4\theta - \tfrac{27}{4}\sin^6\theta\ . \tag{35}$$

As already pointed out, identification of this projection as a function of θ with the experimental expansion (15b) leads to a linear system of equations which is indeterminate for $L = 2K_b \geq 12$. Thus the inversion is not so straightforward as in the previous case. Nevertheless, we shall show below that it is possible to define linear combinations of the coefficients $A^l_{n,m}$ which are related to the coefficients b_{2k} by an inversible system of equations. This possibility is of great interest when it is necessary to use data taken in both symmetry planes, that is when $L \geq 12$, because it simplifies considerably the inversion problem.

In the {110} plane we get from Eqs. (35) the identity

$$S^3 = -P - \tfrac{3}{4}S^2 + \tfrac{3}{2}SP - \tfrac{1}{4}P^2\ , \tag{36}$$

which simply expresses that the function ψ_{18} vanishes in the plane. Using this identity, the projection of Eq. (7) may be written

$$E_{110} = A^0_{0,0} + \sum_{m=0}^{\infty} (C_{4+6m}S + C_{6+6m}P + C_{8+6m}S^2P)P^m\ . \tag{37}$$

The coefficients C_{2p} are linear combinations of the $A^l_{n,m}$; their derivation is developed in Appendix C. The first C_{2p} limited to the order $L = 18$ are

$$C_4 = A^4_{1,0}\,,$$
$$C_6 = A^6_{0,1} - A^{12}_{3,0} - \tfrac{3}{4}A^{16}_{4,0} + \cdots\,,$$
$$C_8 = A^8_{2,0} + \tfrac{3}{4}A^{12}_{3,0} + \tfrac{9}{16}A^{16}_{4,0} + \cdots\,, \tag{38}$$
$$C_{10} = A^{10}_{1,1} + \tfrac{3}{2}A^{12}_{3,0} + \tfrac{1}{8}A^{16}_{4,0} + \cdots\,,$$
$$C_{12} = A^{12}_{0,2} - \tfrac{1}{4}A^{12}_{3,0} - \tfrac{3}{16}A^{16}_{4,0} - A^{18}_{3,1} + \cdots\,.$$

In order to identify expansions (37) and (15b) we write the monomials S^nP^m in the form

$$S^nP^m = \sum_{p=0}^{2n+3m} \gamma^p_{n,m}\cos 2p\theta\,. \tag{39}$$

The derivation of the coefficients $\gamma^p_{n,m}$ is described in Appendix D.

Before proceeding further in the identification we recall that the coefficient $A^0_{0,0}$ can be immediately calculated by Eq. (18b), and that the $K_b + 1$ experimental coefficients b_{2k} are not independent, but must satisfy the relation (16b) which takes the explicit form

$$b_2 = \tfrac{4}{3}b_4 + \tfrac{5}{3}b_6 - \tfrac{112}{27}b_8 + \tfrac{55}{81}b_{10} + \tfrac{460}{81}b_{12} - \tfrac{3913}{729}b_{14} - \tfrac{7616}{2187}b_{16} + \tfrac{6935}{729}b_{18} + \cdots \tag{40}$$

This is a good preliminary test of the reliability of the experimental data.

Using Eqs. (37), (39), and Appendix D, we get the following results in matrix form:

$$\begin{bmatrix} 3/2^5 & C_4 \\ 3^3/2^7 & C_6 \\ 3^2/2^{11} & C_8 \\ 3^4/2^{13} & C_{10} \\ 3^6/2^{15} & C_{12} \end{bmatrix} = \begin{bmatrix} -\frac{1}{3} & -\frac{2}{3} & -\frac{20}{27} & -\frac{130}{81} & -\frac{211}{81} \\ & 1 & -\frac{8}{3} & \frac{59}{9} & \frac{176}{9} \\ & & \frac{1}{9} & \frac{2}{27} & \frac{2}{27} \\ & & & -\frac{1}{3} & -\frac{4}{3} \\ & & & & 1 \end{bmatrix} \begin{bmatrix} b_4 \\ b_6 \\ b_8 \\ b_{10} \\ b_{12} \end{bmatrix} \tag{41}$$

[See the Ref. 6 for the expressions of Eqs. (40) and (41) up to $L = 36$.]

As in the previous case, the order of magnitude of the multiplicative factors of the coefficients C_{2p} ($3^{3m+1}/2^{8m+5}$, $3^{3m+3}/2^{8m+7}$, $3^{3m+2}/2^{8m+11}$ for C_{4+6m}, C_{6+6m}, C_{8+6m}, respectively) causes their contributions to the coefficients b_{2k} to decrease very fast with their order $2p$, leading to the same difficulties in their determination.

The relations (38) together with the inverted system (41) confirm our previous conclusions: (i) the coefficient $A^4_{1,0} = C_4$ can always be determined whatever the order $L = 2K_b$ of the expansion (15b) is; (ii) when $L \leq 10$, C_6, C_8, and C_{10} are simply equal to $A^6_{0,1}$, $A^8_{2,0}$, and $A^{10}_{1,1}$, respectively, and a unique determination of E is possible using data taken in the $\{110\}$ plane only.

C. $\{100\}$ and $\{110\}$ planes

When $L \geq 12$, it is necessary to use data taken in both $\{100\}$ and $\{110\}$ planes. The practical process of inversion follows directly from the preceding discussions, and we note successively that: (i) $A^0_{0,0}$ can be determined by Eq. (18b); (ii) $A^4_{1,0}$ can be determined either by Eq. (34) or Eq. (41). The formula coming from Eq. (41) differs from the formula (22) because it does not involve the coefficient b_2. The former is easily obtained from the latter by using Eq. (40). As already mentioned, this double determination of $A^4_{1,0}$ is a good test of the reliability of the experiments and can allow some corrections to be made to the data before going on with the process: (i) Eqs. (34) give all the coefficients $A^l_{n,0}$ up to $L = 4n = 4K_a$. (ii) Eqs. (41) give all the coefficients C_{2p} up to $L = 2p = 2K_b$. It is then obvious by inspection of relations (38) that if $L = 2K_b \leq 16$ the determination of the coefficients of E is completed simply by replacing in these relations the values of the coefficients $A^l_{n,0}$ given by Eqs. (34). Note that C_8 provide us with a supplementary test of the reliability of the experiments. It is also clear that for $L = 18$ we meet an indeterminacy for the $A^l_{n,m}$ because we cannot separate $A^{18}_{0,3}$ and $A^{18}_{3,1}$.

We recall now that, for any order $L = 2K_b$, the relations (23) and (24) enable us to determine $A^6_{0,1}$ and $A^{10}_{1,1}$ from data taken in the $\{110\}$ plane, using only the coefficients $A^4_{1,0}$, $A^8_{2,0}$, and $A^{12}_{3,0}$ known from data taken in the $\{100\}$ plane. It is much easier to derive $A^6_{0,1}$ and $A^{10}_{1,1}$ from these relations than from Eqs. (38) which require the knowledge of all the coefficients $A^l_{n,0}$ up to $L = 4n = 4K_a$.

The applicability of this method of inversion was tested[6] on an entirely calculable "experimental" situation, i.e., the surface obtained by geometrical inversion of a cube with respect to its center. It appeared that small numerical errors in the highest Fourier coefficients affect considerably the values of many coefficients $A^l_{n,m}$ of lower order.

V. APPLICATION TO THE MAGNETIC ANISOTROPY ENERGY OF NICKEL

The magnetic anisotropy energy of a ferromagnetic crystal is a function[7,9] $E_a(T, H_{iM}, \vec{\alpha})$ of the temperature T, the projection H_{iM} of the internal field along the direction of the magnetization and the direction $\vec{\alpha}$ of the magnetization. Nickel crystallizes in the fcc system, so E_a can be expanded in terms of symmetrical polynomials of the direction cosines of the magnetization in the form (7). The coefficients $A^l_{n,m}$ of this expansion which depend on T and H_{iM} are called in this case "anisotropy constants." In order to determine these constants, the most powerful experimental method is to measure the mechanical torque Γ which must be applied to hold in place a spherical sample, in a uniform magnetic field high enough to have a single domain. For given field and temperature, Γ is measured as a function of ϕ, the angle of the applied field $\vec{H}$ with the [001] direction either in a {100} or a {110} plane. This situation is necessary in order to deduce from this angle ϕ the angle θ of the magnetization $\vec{M}$ with the same origin direction because, when $\vec{H}$ lies in a symmetry plane, the sample being a single domain, $\vec{M}$ remains in the same plane and we simply have

$$\Gamma = HM \sin(\phi - \theta) . \quad (42)$$

With the help of Eq. (42), it is thus possible, from the Fourier expansions of Γ_{100} and Γ_{110} measured as functions of ϕ, to derive the same expansions as functions of θ. Γ_{100} and Γ_{110} are then related to the anisotropy energy E_a by

$$\Gamma_{100} = \frac{\partial E_{a\,100}}{\partial \theta} , \quad \Gamma_{110} = \frac{\partial E_{a\,110}}{\partial \theta} . \quad (43)$$

It is of course impossible to determine $A^0_{0,0}$ by this method, but this coefficient is not relevant to this problem. From Eqs. (15a) and (15b) we get

$$\Gamma_{100} = \sum_{k=1}^{K_a} (-4ka_{4k}) \sin 4k\theta = \sum_{k=1}^{K_a} a'_{4k} \sin 4k\theta , \quad (44a)$$

$$\Gamma_{110} = \sum_{k=1}^{K_b} (-2kb_{2k}) \sin 2k\theta = \sum_{k=1}^{K_b} b'_{2k} \sin 2k\theta . \quad (44b)$$

We can use the coefficients a'_{4k} and b'_{2k} directly in the inversion procedure providing a straightforward change in the formulas. As an example we give, in Table II, the coefficients a'_{4k} and b'_{2k} measured on the same spherical sample of nickel for $T = 4.2$ K and $H = 19\,179$ Oe ($H_{iM} = 16\,981$ Oe). All the Fourier coefficients and the anisotropy constants of this section, except in Table IV, are given in experimental units further denoted

TABLE II. Fourier coefficients of the expansions of Γ_{100} and Γ_{110} relative to a spherical sample of nickel for $T = 4.2$ K and $H = 19\,179$ Oe. The a'_{4k} and b'_{2k} are given in experimental units (1 e.u. = 58.68 ergs/cm³).

		b'_2	−5194.78
a'_4	−10 700.48	b'_4	−7485.58
		b'_6	−458.48
a'_8	−46.00	b'_8	−29.85
		b'_{10}	38.33
a'_{12}	73.95	b'_{12}	37.30
		b'_{14}	27.44
a'_{16}	33.11	b'_{16}	16.68
		b'_{18}	7.77
a'_{20}	3.75	b'_{20}	1.40
		b'_{22}	−1.93
a'_{24}	−5.52	b'_{24}	−2.83
		b'_{26}	−2.53
a'_{28}	−3.02	b'_{28}	−1.69
		b'_{30}	−0.49
a'_{32}	2.28	b'_{32}	1.23
		b'_{34}	1.49

as e.u. with 1 e.u. = 58.68 ergs/cm³. In Table II, the coefficient b'_2 has been calculated as a function of the other b'_{2k} using Eqs. (40) and (44b) and compared with the measured value −5190.50. The small difference is easily explainable by parasitic torques[7,9] which mainly affect this term. The fact that we never use the measured value of b'_2 in the inversion procedure, except to compare it to the calculated one, is of a great interest for minimizing the effects of these parasitic torques.

Our experimental procedure enables us to determine the coefficients a'_{4k} and b'_{2k} up to $K_a = [\frac{1}{2}K_b] = 8$ and $K_b = 17$. The convergence of the expansions (44a) and (44b) appears to be sufficient at this order and the high-order terms can be considered as an experimental "noise."

We first determine $A^4_{1,0}$ independently from the two experiments by formulas (22) which may be rewritten here as

$$A^4_{1,0} = \tfrac{2}{3} \sum_{k=1}^{K_a} ka'_{4k} = \tfrac{1}{3} \sum_{k=1}^{K_b} kb'_{2k} . \quad (45)$$

We get $A^4_{1,0} = -6970.31$ and -6969.99 in the {100} and {110} planes, respectively. The agreement is very good and allows simultaneous use of the two experiments when necessary. We then use Eqs. (34) in order to get the coefficients $A^l_{n,0}$ the values of which are given in Table III. Although

TABLE III. Anisotropy constants $A^l_{n,0}$ (in experimental units) of nickel for $T = 4.2$ K and $H = 19\,179$ Oe, which may be deduced from the values of the a'_{4k} given in Table II.

$A^4_{1,0}$	−6970.31
$A^8_{2,0}$	−862.40
$A^{12}_{3,0}$	1493.52
$A^{16}_{4,0}$	−6431.98
$A^{20}_{5,0}$	30 642.80
$A^{24}_{6,0}$	−65 244.73
$A^{28}_{7,0}$	63 343.33
$A^{32}_{8,0}$	−23 320.87

the torques were measured with a relative accuracy of 10^{-5}, this is not sufficient to allow for the determination of the higher-order coefficients as might be expected after the study of a test example which showed that the precision of a standard computer calculation was itself inadequate.

It is therefore absolutely necessary to truncate the experimental expansions at an order above which the coefficients seem to become unsignificant, i.e., in this case, at $K_b = 9$ and $K_a = 4$ (between upper and lower part of Table II). From (45) we recalculate $A^4_{1,0}$ obtaining $A^4_{1,0} = -6958.79$ and −6949.96 in the {100} and {110} planes, respectively. The agreement is not as good as before, but yet acceptable. We then get by Eqs. (34)

$$A^4_{1,0} = -6958.79, \quad A^{12}_{3,0} = 1723.16\,, \qquad A^8_{2,0} = -1032.71, \quad A^{16}_{4,0} = -837.15\,, \tag{46}$$

There are two ways of proceeding further by using either Eqs. (41) or relations (23) and (24) which may be rewritten here as

$$-8 \sum_{k=1}^{K_b} k^3 b'_{2k} = -78A^4_{1,0} + 162A^6_{0,1} + 216A^8_{2,0}\,, \tag{47}$$

$$32 \sum_{k=1}^{K_b} k^5 b'_{2k} = 1176A^4_{1,0} - 8100A^6_{0,1} - 14\,040A^8_{2,0} + 14\,580A^{10}_{1,1} + 19\,440A^{12}_{3,0}\,. \tag{48}$$

From the value of $A^4_{1,0}$ obtained in the {110} plane and relations (46)–(48), we get

$$A^6_{0,1} = 149.40\,, \quad A^{10}_{1,1} = 621.10\,. \tag{49}$$

Besides, inversion (41) provides

$$C_4 = -6949.61, \quad C_{12} = -902.91, \qquad C_6 = -938.50, \quad C_{14} = -1860.70, \qquad C_8 = -216.70, \quad C_{16} = 1161.09, \qquad C_{10} = 3100.02, \quad C_{18} = -183.97\,. \tag{50}$$

The value of C_4 differs slightly from that calculated from Eq. (45) because the coefficient b'_2 entering this formula was calculated before truncating the expansion at order 18. Using formulas (38) and the values (46), we deduce from C_6 and C_{10}, respectively, $A^6_{0,1} = 156.80$ and $A^{10}_{1,1} = 619.92$. These values are in good agreement with those of (49). We can calculate $C_8 = -211.24$ which, on comparison with its value in (50), confirms the agreement. It is impossible to go further in the inversion using C_{12} to C_{18} because of the indeterminacy arising at $L = 18$.

We draw attention again to the advantage offered by the possibility of determining $A^4_{1,0}$ without the complicated inversion of the experimental data. For this purpose, we have analyzed the data taken at various temperatures in the {110} plane for nickel (values extrapolated to $H_{iM} = 0^{7,9}$) assuming that only the three coefficients $A^4_{1,0}$, $A^6_{0,1}$, and $A^8_{2,0}$ were sufficient to describe the anisotropy energy of the material. These coefficients were calculated from b'_4, b'_6, and b'_8; the resulting value of $A^4_{1,0}$ is compared in Table IV with its value obtained without any assumption from formula (45). We see immediately from this comparison that more than three coefficients are required to describe correctly the anisotropy energy of nickel at temperatures lower than about 100 K. A similar test with two coefficients only shows, contrary to the usual practice described in the literature, that it is never possible to describe this anisotropy with the two constants $K_1 = 3A^4_{1,0}$ and $K_2 = 27A^6_{0,1}$. In contrast to the case of nickel, two constants are sufficient to represent the anisotropy energy

TABLE IV. Comparative values of the anisotropy constants $A^4_{1,0}$ of nickel at various temperatures obtained from data in the {110} plane, (I) using only three Fourier coefficients and, (II) using all the available coefficients with Eq. (45).

T (K)	$-A^4_{1,0}$ (10^5 ergs/cm^3) I	II
4.20	4.212	4.049
21.50	4.006	3.895
35.00	3.711	3.646
50.55	3.333	3.304
78.35	2.786	2.780
95.15	2.406	2.404
109.15	2.109	2.110
125.15	1.778	1.780
155.75	1.288	1.289
185.55	0.898	0.898
212.90	0.628	0.628
245.10	0.397	0.397
273.15	0.258	0.258
294.60	0.183	0.183

of iron at all temperatures.[8]

The different tests which were used in this section, such as the determination of $A^4_{1,0}$ and C_8 from data in both symmetry planes, allow an estimation of the reliability of the coefficients $A^l_{n,m}$ obtained by inversion of the experimental data; this reliability would be almost impossible to predict from the initial accuracy of the measurements.

VI. CONCLUSION

The scalar quantity $E(\theta,\phi)$ may be expanded in terms of symmetrical polynomials as well as in terms of cubic harmonics. All the coefficients of these expansions may be obtained in a unique way from measurements taken in a $\{110\}$ plane only when the order of these expansions is $L\leqslant 10$, and from measurements in both the $\{110\}$ and $\{100\}$ planes when $12\leqslant L\leqslant 16$; for $L\geqslant 18$, there is no longer a unique solution of the problem. However, it should be emphasized that the first coefficients $A^l_{n,m}$ of the expansion in terms of symmetrical polynomials may be obtained in a unique way from the experimental data in the symmetry planes whatever the value of L. Furthermore, the first two coefficients $A^0_{0,0}$ and $A^4_{1,0}$, the latter being of fundamental importance for describing magnetic anisotropy properties for instance, may be obtained from data taken in one symmetry plane only. These features are specific to an expansion of E in terms of symmetrical polynomials and there is no equivalent for an expansion in terms of cubic harmonics.

ACKNOWLEDGMENTS

We are particularly grateful to Dr. Y. Siret and to M. Geynet for their assistance in solving many computational difficulties. We are also indebted to Dr. E. Geissler for reading the manuscript.

APPENDIX A: DERIVATION OF THE CUBIC HARMONICS

It is easy to express S and P in terms of K_4 and K_6, the cubic harmonics of order 4 and 6, as

$$S=\tfrac{3}{5}-\tfrac{4}{5}\left(\tfrac{3}{7}\pi\right)^{1/2}K_4\,,$$
$$P=\tfrac{9}{35}-\tfrac{36}{55}\left(\tfrac{3}{7}\pi\right)^{1/2}K_4+\tfrac{72}{77}\left(\tfrac{2}{13}\pi\right)^{1/2}K_6\,, \tag{A1}$$

with

$$K_4=\left(\tfrac{7}{12}\right)^{1/2}C_{4,0}+\left(\tfrac{5}{12}\right)^{1/2}C_{4,4}\,,$$
$$K_6=\left(\tfrac{1}{8}\right)^{1/2}C_{6,0}-\left(\tfrac{7}{8}\right)^{1/2}C_{6,4}\,. \tag{A2}$$

Let us consider the terms of highest order $l=4n+6m$ in the expansion of S^nP^m in spherical harmonics. These terms come necessarily from the expansion of $(-1)^n(K_4)^n(K_6)^m$. In order to obtain their explicit expression, we consider the term with highest power of $\cos\theta$ which, apart a multiplicative constant is given by

$$(-1)^n(\cos\theta)^{4n+6m}(7+\cos4\phi)^n(1-\cos4\phi)^m\,, \tag{A3}$$

and may be rewritten (still neglecting a constant factor) as

$$(\cos\theta)^{4n+6m}\left(\tfrac{1}{2}\,\alpha^{(0)}_{n,m}+\sum_{q=1}^{n+m}\alpha^{(q)}_{n,m}\cos4q\phi\right), \tag{A4}$$

where $\alpha^{(q)}_{n,m}$ is the coefficient of x^{n+m+q} in the polynomial $\mathcal{P}(x)$ given by

$$\mathcal{P}(x)=(-1)^{n+m}(x^2+14x+1)^n(x^2-2x+1)^m\,. \tag{A5}$$

A cubic harmonic of order l is necessarily of the form

$$K^l_{n,m}=\sum_{q=0}^{[l/4]}a^{(q)}_{n,m}C_{l,4q}\,. \tag{A6}$$

Setting

$$\eta^l_{4q}=(2l)!/(l-4q)!(l+4q)!\,, \tag{A7}$$

i.e., the coefficient of y^{l+4q} in the polynomial

$$\mathcal{Q}(y)=(y+1)^{2l}\,, \tag{A8}$$

it is easy to check that the coefficient of the $(\cos\theta)^l$ term in (A6) is

$$A\left(a^{(0)}_{n,m}(\eta^l_0)^{1/2}+\sum_{q=0}^{[l/4]}a^{(q)}_{n,m}(2\eta^l_{4q})^{1/2}\cos4q\phi\right), \tag{A9}$$

where A is a constant factor.

Comparison of (A9) with the term in large parentheses in (A4) shows that it is possible to define a unique cubic harmonic for each pair (n,m) by limiting the summation over q in (A6) to $q=n+m$ and by taking

$$a^{(0)}_{n,m}=k\,\alpha^{(0)}_{n,m}/(2\eta^l_0)^{1/2}\,,\quad a^{(q)}_{n,m}=k\alpha^{(q)}_{n,m})/(\eta^l_{4q})^{1/2}\,. \tag{A10}$$

The constant k is obtained from the normalization condition for $K^l_{n,m}$

$$\sum_{q=0}^{n+m}[a^{(q)}_{n,m}]^2=1\,. \tag{A11}$$

The numerical coefficients $a^{(q)}_{n,m}$ are very easily obtained from the integer coefficients of the polynomials $\mathcal{P}(x)$ and $\mathcal{Q}(y)$, and as can be seen from (A10) and (A11), they are square roots of rational ratios. When l is small, this calculation is immediate. For an arbitrary value of l, the calculation is straightforward and was obtained with a computer by using the PL/1-FORMAC language[10] allowing formal multiplication of polynomials.

APPENDIX B: EXPANSION OF THE S^nP^m IN TERMS OF THE CUBIC HARMONICS

As pointed out in Sec. II C, the cubic harmonics $K^l_{n,m}$ or the related functions $H^l_{n,m}$ given by (13) do not form a complete orthonormal set. Indeed, when several functions belong to the same subset of order l ($g_l > 1$), they are not orthogonal to each other. Nevertheless, these harmonics were built in close connection with the symmetrical polynomials and this lack of orthogonality does not introduce any major difficulty in the calculation of the coefficients $\beta_{\nu,\mu}^{n,m}$ of expression (14).

From Eqs. (A1) and (A2), and setting $z = \cos\theta$, we have

$$S = a - b\cos 4\phi \,, \tag{B1}$$

$$P = c(1-\cos 4\phi)\,, \tag{B2}$$

with

$$a = \tfrac{3}{8}(1-z^2)^2 + 3z^2(1-z^2)\,,$$

$$b = \tfrac{3}{8}(1-z^2)^2,$$

$$c = \tfrac{27}{8} z^2(1-z^2)^2 .$$

From (B1) and (B2) it is possible to expand $S^n P^m$ in the form

$$S^n P^m = d_0 + \sum_{q=1}^{n+m} d_q \cos 4q\phi \,, \tag{B3}$$

where d_0 and $\frac{1}{2} d_q$ are the coefficients of x^{n+m} and x^{n+m+q}, respectively, in the expansion of the polynomial

$$(-\tfrac{1}{2})^{n+m} c^m (bx^2 - 2ax + b)^n (x^2 - 2x + 1)^m .$$

Introducing the Legendre polynomials $P_\lambda(z)$ and the associated Legendre polynomials, we have, from Eqs. (11), (13), and (A10),

$$H_{\nu,\mu}^{\lambda} = P_\lambda(z) + \sum_{q=1}^{\nu+\mu} 2\frac{(\lambda-4q)!}{\lambda!}\frac{\alpha_{\nu,\mu}^{(q)}}{\alpha_{\nu,\mu}^{(0)}}\cos(4q\phi) P_\lambda^{4q}(z)\,. \tag{B4}$$

Using (B4), the identification of expansions (14) and (B3) leads to

$$d_0 = \sum_{\lambda=0}^{l}\sum_{\nu,\mu}\beta_{\nu,\mu}^{n,m} P_\lambda(z),$$

with $\lambda = 4\nu + 6\mu$;

$$d_q = \sum_{\lambda'=4q}^{l}\sum_{\nu',\mu'}\beta_{\nu',\mu'}^{n,m}\, 2\frac{(\lambda'-4q)!}{\lambda'!}\frac{\alpha_{\nu',\mu'}^{(q)}}{\alpha_{\nu',\mu'}^{(0)}} P_{\lambda'}^{4q}(z)\,,$$

with $\lambda' = 4\nu' + 6\mu'$ and $q \leq \nu' + \mu' \leq n+m$; furthermore, when $\nu' + \mu' > n+m$, we have $\beta_{\nu',\mu'}^{n,m} = 0$.

The expression of d_0 allows one to calculate $\beta_{0,0}^{n,m}$ simply by

$$\beta_{0,0}^{n,m} = \int_0^1 d_0\, dz \;.$$

Using the expression of d_q and the orthogonality properties of the $P_\lambda^{4q}(z)$, we get

$$\gamma_{\nu,\mu}^{n,m} = \sum_{\nu',\mu'}\frac{\alpha_{\nu',\mu'}^{(\nu+\mu)}}{\alpha_{\nu',\mu'}^{(0)}}\beta_{\nu',\mu'}^{n,m}$$

$$= \frac{(2\lambda+1)\lambda!}{2[\lambda+4(\nu+\mu)]!}\int_0^1 d_{\nu+\mu} P_\lambda^{4(\nu+\mu)}(z)\, dz\,, \tag{B5}$$

with $4\nu' + 6\mu' = 4\nu + 6\mu = \lambda$ and $\nu' + \mu' \geq \nu + \mu$.

The coefficients $\beta_{0,0}^{n,m}$ and $\gamma_{\nu,\mu}^{n,m}$ are obtained with a computer using the PL/1-FORMAC language[10] which gives the results in exact rational form. When $g_\lambda = 1$, there is only one possibility $\nu' = \nu$, $\mu' = \mu$ and

$$\beta_{\nu,\mu}^{n,m} = [\alpha_{\nu,\mu}^{(0)}/\alpha_{\nu,\mu}^{(\nu+\mu)}]\,\gamma_{\nu,\mu}^{n,m} .$$

When $g_\lambda > 1$, (B5) is a linear system of g_λ equations which allows the calculation of the g_λ unknown coefficients $\beta_{\nu',\mu'}^{n,m}$ in a straightforward way. The numerical values of the $\beta_{\nu,\mu}^{n,m}$ in rational form have been calculated[6] up to $l = 36$. These calculations have been checked by comparing the numerical values of the $S^n P^m$ obtained by direct calculation and by using the $\beta_{\nu,\mu}^{n,m}$ and the numerical values of the $H_{\nu,\mu}^{\lambda}$ for the three directions $\langle 100\rangle$, $\langle 110\rangle$, and $\langle 111\rangle$. For instance, for the direction $\langle 100\rangle$ we have

$$\sum_{\lambda=0}^{l}\sum_{\nu,\mu}\beta_{\nu,\mu}^{n,m} = 0\,.$$

APPENDIX C: EXPRESSIONS OF THE COEFFICIENTS C_{2p} IN TERMS OF THE $A_{n,m}^{l}$

Starting from Eq. (36), it is possible to express S^n in the $\{110\}$ plane as

$$S^n = \sum_{i=0}^{[2n/3]} a_{n,i} P^i + \sum_{j=0}^{[(2n-2)/3]} b_{n,j} SP^j + \sum_{k=0}^{[(2n-4)/3]} c_{n,k} S^2 P^k \,, \tag{C1}$$

with the recurrence relations between the coefficients,

$$a_{n+1,i} = -c_{n,i-1} - \tfrac{1}{4} c_{n,i-2}\,,$$

$$b_{n+1,i} = \tfrac{3}{2} c_{n,i-1} - c_{n-1,i-1} - \tfrac{1}{4} c_{n-1,i-2}\,, \tag{C2}$$

$$c_{n+1,i} = \tfrac{3}{4} c_{n,i} + \tfrac{3}{2} c_{n-1,i-1} - c_{n-2,i-1} - \tfrac{1}{4} c_{n-2,i-2}\,.$$

Replacing the S^n by their expressions (C1), we get E_{110} in the form (37). If we limit our expansions to the order 18, application of the recurrence relations (C2) leads to the following result, in matrix form:

$$\begin{pmatrix} S^3 \\ S^4 \end{pmatrix} = \begin{pmatrix} -1 & -\frac{1}{4} \\ -\frac{3}{4} & -\frac{3}{16} \end{pmatrix} \begin{pmatrix} P \\ P^2 \end{pmatrix} + \begin{pmatrix} \frac{3}{2} & \\ \frac{1}{8} & -\frac{1}{4} \end{pmatrix} \begin{pmatrix} SP \\ SP^2 \end{pmatrix} + \begin{pmatrix} \frac{3}{4} & \\ \frac{9}{16} & \frac{3}{2} \end{pmatrix} \begin{pmatrix} S^2 \\ S^2P \end{pmatrix} . \quad \text{(C3)}$$

It is then straightforward to deduce Eqs. (38) from (C1) and (C3). The expressions of Eqs. (C3) and (38) completed up to order 36 are also available.[6]

APPENDIX D: EXPANSION OF THE MONOMIALS S^nP^m IN THE $\{110\}$ PLANE

In the $\{110\}$ plane S and P are given by expressions (35) as functions of the angle θ between the measuring direction and the [001] direction of the plane. Let $x = e^{2i\theta}$, so

$$S^nP^m = (3^{n+3m}/2^{6n+8m})(-x^2+2x-1)^{n+2m} \times (3x^2+10x+3)^n(x^2+2x+1)^m/x^{2n+3m} . \quad \text{(D1)}$$

Noting that $\cos 2p\theta = \frac{1}{2}(x^p + x^{-p})$, we get Eq. (39) of the main text:

$$S^nP^m = \sum_{p=0}^{2n+3m} \gamma^p_{n,m} \cos 2p\theta ,$$

where $2\gamma^0_{n,m}$ and $\gamma^p_{n,m}$ are the coefficients of x^{2n+3m} and $x^{2n+3m+p}$, respectively, in the expansion of the polynomial

$$y = (3^{n+3m}/2^{6n+8m-1})(-x^2+2x-1)^{n+2m} \times (3x^2+10x+3)^n(x^2+2x+1)^m . \quad \text{(D2)}$$

This calculation has been carried out with a computer by using PL/1-FORMAC language[10] which allows for formal multiplication of polynomials.

[1]R. R. Birss, *Symmetry and Magnetism: Series of Monographs on Related Topics in Solid State Physics*, 2nd ed. (North-Holland, Amsterdam, 1966), Vol. III.

[2]S. L. Altmann and A. P. Cracknell, Rev. Mod. Phys. 37, 19 (1965).

[3]F. M. Mueller and M. G. Priestley, Phys. Rev. 148, 638 (1966).

[4]R. R. Birss and G. J. Keeler, Phys. Status Solidi 64, 357 (1974).

[5]F. C. Von der Lage and H. A. Bethe, Phys. Rev. 71, 612 (1947).

[6]G. Aubert, E. Belorizky, and R. Casalegno, Internal Report, Grenoble (1975) (unpublished).

[7]G. Aubert, J. Appl. Phys. 39, 2, 504 (1968).

[8]G. Aubert and P. Escudier, *Proceedings of the International Congress of Magnetism* (Nauka, Moscow, 1973), Vol. I, p. 215.

[9]P. Escudier, thèse (Grenoble, 1974) (unpublished): and Ann. Phys. 9, 125 (1975).

[10]A. Laplace, thèse (Grenoble, 1973) (unpublished).