MAGNETIC ANISOTROPY IN NICKEL AND IRON ;
THE EFFECT OF PRESSURE

J. J. M. FRANSE

Natuurkundig Laboratorium der Universiteit van Amsterdam, The Netherlands

Résumé. — On discute les travaux expérimentaux et théoriques sur l'anisotropie magnétique et sur la magnétostriction du nickel et du fer, accordant beaucoup d'attention aux expériences sous haute pression. On essaye d'élucider plus spécialement la situation expérimentale en ce qui concerne l'anisotropie magnétique du nickel. Des arguments sont allégués qui soutiennent un calcul dans le modèle des bandes, dans lequel les contributions principales à l'anisotropie magnétique ont leur origine dans les régions de la zone de Brillouin, où les bandes, en l'absence de l'interaction spin-orbite, sont dégénérées.

Abstract. — The experimental and theoretical work on the magnetic anisotropy energy and the magnetostriction of nickel and iron is reviewed with special attention to the experiments under high pressure. It is attempted to clear up the experimental situation around the magnetic anisotropy of nickel in particular. Arguments are collected that support a band theoretical calculation in which the main contributions to the anisotropy energy of nickel arise from regions in the Brillouin zone, where bands are degenerate in absence of the spin-orbit interaction.

I. **Introduction.** — Discussions on the magnetic anisotropy energy and the magnetostriction of nickel and iron remain often restricted to an interpretation of the temperature dependence of these phenomena. Relations have been deduced by Zener [1] and by Kittel and Van Vleck [2] in which the temperature dependence of the magnetic anisotropy and the magneto-elastic constants are related to that of the magnetization. These relations describe with reasonably good success the magnetic anisotropy energy of iron, see for instance the work of Klein and Kneller [3], and the magnetostriction of nickel, as has been pointed out by Lee and Birss [4]. For the magnetostriction of iron, where Tatsumoto and Okamoto [5] found a maximum in one of the magnetostriction constants not far from the Curie temperature, a different model has been proposed by Callen and Callen [6]. It presents arguments to understand this maximum in iron and the absence of such a maximum in nickel. In case of the magnetic anisotropy energy of nickel any simple model fails and one has to look for the basic interactions that are responsible for the magnetic anisotropy.

The spin-orbit interaction is generally assumed to be the origin of the magnetic anisotropy in nickel and iron. To get from this insight to an explicit value for the anisotropy and magnetostriction constants is not a very easy task. Several calculations on this subject have been reported in the literature. In the earlier work a localized spin model has been used. Van Vleck [7] has shown that there is for instance a pseudo dipolar coupling, due to the spin-orbit interaction, that may lead in second order perturbation calculation to an anisotropy in the energy with cubic symmetry. The higher order anisotropy constants follow in this theory from higher order perturbation calculations and have to converge rapidly, which is not in accordance with the experimental data for nickel. The temperature dependence of the magnetic anisotropy energy can be related also in this model to that of the magnetization. For nickel and iron it means that the first anisotropy constant K_1 has to change with a power of the magnetization in the order of ten [8], whereas the experimental value for this power varies for nickel between 50 and 100 in the low temperature region.

A theory for the magnetic anisotropy, based on the itinerant electron model, has been developed by Brooks [9] and it has been used by Brooks, Fletcher [10], Asdente and Delitala [11], Furey [12] and Mori [13] to calculate the anisotropy energy of nickel and iron. The spin-orbit interaction may be considered in this model as a small perturbation for the ferromagnetic band structure. The energy shifts of the bands, due to the spin-orbit interaction, can be calculated in general by perturbation theory. It turns out that these perturbation calculations have to be carried out to the fourth order before getting a non-vanishing contribution to the anisotropy in the energy for these metals. A perturbation calculation, in which contributions to the anisotropy energy arise from energy states over a large region of the Brillouin zone, can not yield the proper description of the experimental facts for nickel by nearly the same arguments as given before. Furey solved these problems by identifying the anisotropy energy of nickel with the energy shifts of a few bands in a small region of the zone. These bands are degenerate without the spin-orbit interaction and are situated close to the Fermi level. Higher order constants, obtained from contributions of these degenerate states, are not *a priori* small in comparison with the first anisotropy constant ; a series expansion of the anisotropy energy is even questionable in this model. Moreover large effects can be expected from a change in the location of the Fermi level.

The strong temperature dependence and the higher order contributions are not the only remarkable facts in the magnetic anisotropy of nickel. Looking for values for the anisotropy and magnetostriction constants of nickel in the literature one can find a large spread in the experimental data. Before going into the details of Furey's model we shall review the experimental situation around the magnetic anisotropy and try to bring some order in the experimental results.

II. **Phenomenological theory and experimental methods.** — The free energy F of a ferromagnetic single crystal is in general a function of the orientation $\boldsymbol{\alpha}$ of the magnetization $\mathbf{M}$ and of the state of strain, denoted by the strain components e_{ij}. Follo-

wing Becker and Döring [14] one can represent this functional relationship in contributions to the free energy, in which the strain components enter with ascending powers :

$$F(\alpha, e_{ij}) = F_A(\alpha) + F_M(\alpha, e_{ij}) + F_E(e_{ij}) \qquad (1)$$

where F_A, F_M and F_E are the magnetic anisotropy, the magneto-elastic and the elastic energies, respectively. Symmetry arguments dictate the special forms in which these energies can be expressed in the direction cosines α_i and in the strain components.

Taking the cube axes as reference directions one can write for crystals with cubic symmetry :

$$F_A = K_1 s + K_2 p + K_3 s^2 + \cdots \qquad (2)$$

with K_1, K_2, etc. the magnetic anisotropy constants, s equal to $\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2$, and p equal to $\alpha_1^2 \alpha_2^2 \alpha_3^2$. This expansion, that is commonly used, is less useful for a detailed description of the magnetic anisotropy energy of nickel at low temperature [15]. A development of this energy in the cubic harmonics H_l will be used as an alternative to represent the experimental data for nickel in this temperature region :

$$F_A = \sum_{l=4,6,\ldots} k_l H_l \qquad (3)$$

where the k_l are the corresponding anisotropy constants.

Restricting the expansion of the magneto-elastic energy to the fourth power in the direction cosines α_i, one can express the spontaneous deformation upon rotation of **M** in the magnetostriction constants $h_1, \ldots, h_5$ [16].

Two experimental methods are suited to study the magnetic anisotropy energy in detail. Recent torque [15] as well as magnetization [17] experiments on the magnetic anisotropy energy of nickel report the same features for the constants k_4, k_6 and k_8 at low temperature. An advantage of the magnetization method is the rapid and simple registration of the experimental results. The analysis of the experiments is simpler in torque experiments. The accuracy of ferromagnetic resonance experiments on the magnetic anisotropy energy of nickel and iron is below that of the other methods, given before. The relatively large value of the linewidth, in particular in experiments on bulk material of nickel, makes an accurate determination of the resonance fields impossible. For a detailed study of the higher order contributions to the magnetic anisotropy energy of nickel this method, that requires a non-linear least squares adjustment in evaluating the anisotropy constants, can not be applied [18].

The first two methods can also be used for an investigation of the anisotropy energy under pressure. Pressure experiments by the magnetization process have as an advantage that the experimental situation is not disturbed by the mechanical connection of the high pressure vessel with the pressure generating system [19, 20]. In torque measurements under pressure some specific problems have to be solved. In experiments where the whole torque equipment is placed inside the high pressure vessel the information about the magnetic torque must be brought outside the high pressure vessel by electrical or optical means [21].

Moreover, the effect of pressure on the torsion rigidity of the torsion wire has to be known. Another solution for these problems has been found by using the high pressure tubing itself as a torsion wire in the torque experiments [22]. A limit is set in this case to the sensitivity of the torque measurements, since high pressure tubing is not available in all desired dimensions. In both cases small changes in the maximum torque, due to variations in the pressure, can be observed. Using the second method the effect of pressure on the magnetic anisotropy energy can be followed over a large temperature region.

The application of the strain gauge technique in the magnetostriction problem turns out to be successful. The temperature dependence of the magnetostriction constants and the higher order contributions to the magnetostriction have been studied in this way. The use of this technique in experiments under pressure is not straightforward. The problems of cementing the strain gauges on the sample surface are very serious by a penetration of the gas under the backing material. Besides these problems, that could be solved by using strain gauges with a paper base, one has to measure the gauge factor of the strain gauges as a function of pressure. It was possible to obtain with this technique reproducible results for the effect of pressure on the magnetostriction constants of nickel and iron [22].

III. **Experimental data.** — This discussion will be restricted mainly to the case of nickel where a large spread in the literature data exists.

In the earlier work two constants K_1 and K_2 have been used to describe the experimental results for nickel. The data for K_1 vary at 77 °K gradually from $- 54$ to $- 84 \times 10^4$ erg/cm³ [23, 24, 25, 26, 27, 15, 17]. Analysing these experimental results we concluded that trivial circumstances could not be responsible for this large spread and that a physical interpretation had to be sought [18]. In order to clear up this question the influence of small amounts of Fe, Co and Cu on the magnetic anisotropy energy of nickel has been investigated [28]. Some of the results with impurity percentages in the order of 0.1 and 1 percent are given in table I. It turns out that the large spread in

TABLE I

Values of the first magnetic anisotropy constant k_4 for nickel and some nickel alloys at 4.2 °K in 10^4 erg/cm³.

Ni	$+ .1\% $ Cu	$+ .14\% $ Fe	$+ .9\% $ Cu	$+ 1.0\% $ Co
37.3	37.0	35.3	33.9	27.5

the literature results for K_1 of nickel can very well be ascribed to small differences in the purities of the different samples. Literature values for K_2 of nickel not only differ in absolute value but even in sign. Most of these values have been obtained from experiments in the (111) plane, in which plane the anisotropy is determined in principle by K_2 only. Additional torques, partly field dependent, make the torque experiments in this plane very complicated and ask a careful analysis of the experimental data in order to obtain a reliable value for K_2. A Fourier analysis of these additional torques can be helpful in determining

the crystallographic directions in this plane since certain relations between the Fourier coefficients have to be satisfied [15]. A reinterpretation of some of the literature data on the anisotropy in this plane brings the results for K_2 into coincidence over a large temperature region [29]. Values for K_2 from experiments in the (110) plane are in good agreement with these results. The same agreement can be found with the results of Rodbell's ferromagnetic resonance experiments [30] after modification of the resonance condition in the [110] direction in the (111) plane. This implies that there is no experimental evidence that static and dynamic methods yield different values for the anisotropy constants of nickel.

In the past few years it has become clear that a third and even higher anisotropy constants are needed for a description of the experimental results at low temperature [31, 15, 17]. The complex character of the magnetic anisotropy energy at low temperature can be demonstrated by a special plot in which the torque data in the (100) plane are divided by the factor sin $(2 \theta \times \cos 2 \theta)$, in order to separate the contributions of K_1 to the angle dependence from those of the higher constants, as follows from the expression :

$$L_A/(\sin 2 \theta \times \cos 2 \theta) = B_1 + B_2 \sin^2 2 \theta + + B_3 \sin^4 2 \theta + \cdots \quad (4)$$

with $B_1 = K_1$, $B_2 = \frac{1}{2} K_3$, etc. and with θ the angle between the direction of **M** and the [001] direction, see figure 1.
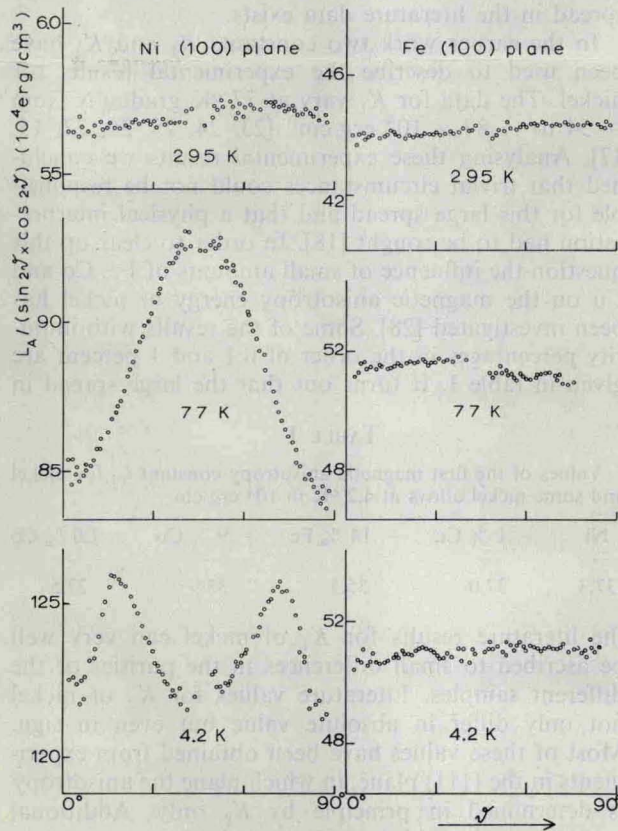
FIG. 1. — Higher order contributions to the torque curves of Ni and Fe in the (100) plane. In order to eliminate contributions from K_1 to the angle dependence, the experimental data have been multiplied with the factor $(\sin 2 \theta_i \times \cos 2 \theta_i)^{-1}$.
(After ref. [15] and [18]).

This plot turns out to be very helpful to show the complicated structure of the anisotropy energy at low temperature. It clearly demonstrates that it is hard to describe the results at 4.2 °K with a small number of the K_i. The anisotropy constants K_1, K_2 and K_3, given in figure 2, have been obtained from
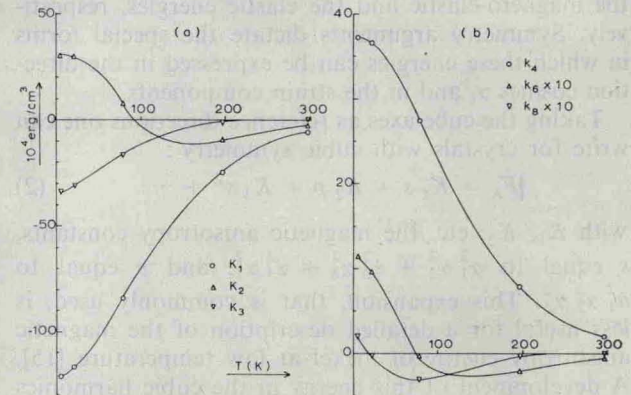
FIG. 2. — Temperature dependence of the magnetic anisotropy constants of nickel.
(a) in a description of the energy with eq. (2) ;
(b) with eq. (5).

(Data obtained from ref. [15].)

an analysis of the experimental results in the neighbourhood of the [001] direction in the (110) and the (100) plane. A least squares determination of the coefficients in the expansion of the anisotropy energy in cubic harmonics results in a more reliable representation of the experimental results over the full range of orientations. Values for the first three constants in this expansion are also presented in figure 2. It should be noted that these three constants are far from capable to describe the experiments at 4.2 °K within the experimental accuracy. Even a description with six constants does not reveal some specific fine details in the torque curve [15].

The procedure, that for nickel leads to a clear demonstration of higher order contributions to the magnetic torque, results for iron in a flat curve, indicating that only one anisotropy constant has to be used in the (100) plane below room temperature.

In experiments under pressure too the magnetic anisotropy energies of nickel and iron behave differently. The influence of pressure on K_1 of iron is at room temperature nearly the same as at 77 °K. The effect of pressure on the absolute value of K_1 for nickel increases with a factor of about four going from room temperature to 77 °K. The literature data on the pressure effect in the magnetic anisotropy energy, obtained by torque and magnetization methods, agree very well [20, 21, 22].

The information about the magnetostriction constants of iron and nickel is less extensive than that on the magnetic anisotropy constants. Irrespective of a number of details there is uniformity about the main points in the magnetostriction of nickel [33, 34, 35] and iron [5, 32] : the first two constants of nickel and the second constant of iron decrease gradually with increasing temperature ; the first constant of iron shows

a maximum not far from the Curie temperature. For the higher magnetostriction constants h_3, h_4 and h_5 contradictory results have been reported [34, 35, 16, 32, 36]. Some of these results must be ascribed to errors in the analysis of the experimental data, introduced by an incorrect application of the angle correction between **M** and **H** [37, 22]. Such incorrect procedures become manifest by a field dependence of these constants like $1/H$ and a temperature dependence equal to that of K_1. Recent data on the magnetostriction constants of nickel report values for the higher magnetostriction constants that are small at room temperature and at 77 °K and that are independent of the external magnetic field [38]. The values for the third magnetostriction constant h_3 are at the two temperatures in agreement with the results for the pressure dependence of the first magnetic anisotropy constant [22]. In case of iron a similar agreement has not yet been obtained.

The effect of pressure on the magnetostriction constants of nickel and iron is clearly present in the first magnetostriction constant of iron. For the other magnetostriction constant no change with pressure could be found [22].

IV. **Recent theoretical models.** — At this point we return to the origin of the magnetic anisotropy energy in nickel and iron. Furey has treated the anisotropy problem for nickel using improved energy bands, provided by Hodges e.a. [39], in which hybridization between the 3 d-bands and the conduction bands also has been considered. Since there are remarkable points of similarity between Furey's work and the experimental data, we briefly deal here with some of the keypoints of this work.

1. The main contributions to the magnetic anisotropy energy arise from special points in the Brillouin zone where energy bands are degenerate in absence of the spin-orbit interaction. These points are situated on the $\Gamma - X$ and $\Gamma - L$ directions.

2. The full eigenvalue problem has been solved in the neighbourhood of these directions in **k** space for different orientations of the magnetization. The energy shifts, due to a rotation of the magnetization, turn out to be largest near the point X and do not have the same sign for all bands. Some typical d-bands in this important region are shown in figure 3. In calculating the anisotropy energy the summation of the energy shifts has been performed over the unoccupied states.

3. The partial cancellation of the contributions of the different bands to the magnetic anisotropy energy is strongly affected by the occupation of the relevant states. A description of these occupations in terms of Fermi distribution functions results in a rapid variation of the magnetic anisotropy energy with temperature.

4. The calculated value for the magnetic anisotropy energy is very sensitive to the position of the Fermi level with respect to the minority spin d-bands. A second calculation, in which the Fermi level was lowered with an amount of 0.006 eV, resulted in a 25 percent lower value for the first magnetic anisotropy constant.
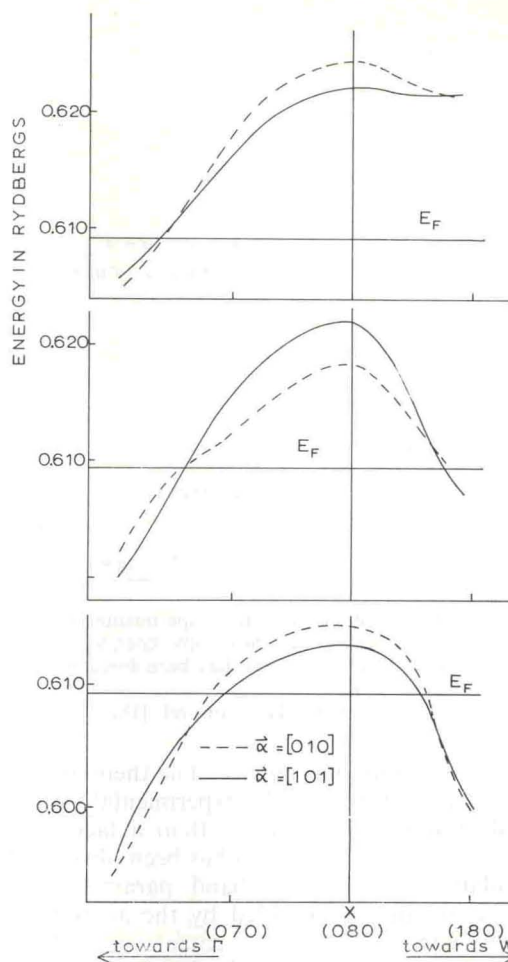

FIG. 3. — Energy bands of nickel near the point X in the Brillouin zone for two orientations of the magnetization. (After Furey ref. [12]).

By mentioning these four points we intend to call attention for this outstanding work which also deals with the orbital contribution to the magnetization and with the magnetostriction of nickel.

The energy shifts in the bands near the point X upon rotation of the magnetization have been revealed by the De Haas-Van Alphen studies on the hole pockets near this point. The X_5 levels that are responsible for these hole pockets, that vary in size upon rotation of the magnetization, turn out to yield essential contributions to the anisotropy in the energy. For a discussion of these De Haas-Van Alphen measurements we may refer to the review article by Gold [40].

To compare the results of Furey's work on the temperature dependence of the magnetic anisotropy energy with the experimental data, the differences in the free energy F_A have been determined between two orientations of the magnetization, parallel to the [100] and [110] directions, respectively, by integrating the experimental torque curves and applying a correction for the magneto-elastic energies and for the anisotropy in the magnetization. For nickel, where an expansion of the anisotropy energy in the direction cosines of the magnetization is questionable at low temperature from experimental as well as from theoretical points of view, this procedure must be preferred. Figure 4 shows the experimental and theoretical data
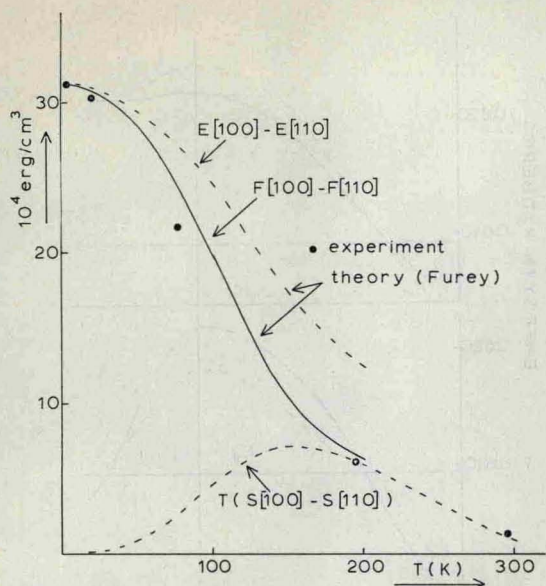
FIG. 4. — Comparison between the experimental temperature dependence of the magnetic anisotropy energy and Furey's calculations. The theoretical curve has been fitted to the experimental value at 0 °K.
(Experimental data from ref. [18]).

on the temperature dependence. The theoretical curve has been fitted at 0 °K to the experimental value. This value does not differ by more than a factor of two from the theoretical value that has been obtained from a calculation with use of band parameters and a spin-orbit parameter provided by the analysis of the De Haas-Van Alphen measurements near the point X [41]. An appreciable contribution to the anisotropy energy has been found to arise from the entropy term in the free energy. These entropy data have been taken from Furey's work directly and have not been fitted to the experiments. It should be emphasized that various contributions of different sign built up the magnetic anisotropy energy. A change in sign for K_1 at high temperature is quite reasonable within this theory. The experimental situation around this change in sign is not fully clear [30, 42, 43] ; it requires an accurate knowledge of the magneto-elastic contribution to the experimental value of K_1 [18].

In the very recent work of Mori also band calculations on the anisotropy energy of nickel and iron are presented. This work contradicts that of Furey in so far that for nickel contributions of degenerate states to the anisotropy energy are found to be less important. The experimental facts, present in the complicated low temperature structure, support the work of Furey.

The behaviour of the anisotropy energy of iron indicates that states very close to the Fermi level are not dominating and that degeneracy plays a less important role here. An actual calculation of K_1 by perturbation theory has to deal with the uncertain values for the bandwidth and the spin-orbit parameter, as has been stressed by Asdente and Delitala [11].

A quantitative interpretation of the effects of pressure on the anisotropy energy lies beyond the possibilities of the present theories and we have to restrict the discussion on this point to some qualitative

remarks. The anisotropy energy usually follows in the band model from fourth order perturbation calculation and it is determined by the spin-orbit parameter and the energy differences between the relevant bands. Assuming an increase of the bandwidth with increasing pressure as a general rule, one finds the energy denominators in the fourth order perturbation calculation to become larger, resulting in lower values for the fourth order energy corrections. This is of course a very poor model since we have no precise knowledge of the energy bands and their shifts under pressure. An interpretation of the pressure effect is perhaps most successful for nickel, where only a few bands in a small region of the zone have to be known. It might be argued that the degeneracy in this small region of the zone is lifted to some extent under pressure. From these arguments a decrease of the anisotropy energy with pressure may be expected, which is in accordance with the experimental results. The increase in the pressure effect for nickel to lower temperatures may be attributed to an increasing sharpness of the Fermi distribution functions. The cancellation of the different contributions to the anisotropy energy is more sensitive to a small shift in the energy bands, introduced by pressure, if the Fermi level is sharper. In a case where states near the Fermi level are not of special importance for the anisotropy energy, like for iron, a strong temperature dependence of the pressure effect in the low temperature region is not to be expected.

Only a few words will be devoted to the magnetostriction problem. It is remarkable that for nickel two phenomena that both are related with the spin-orbit interaction, the anisotropy energy and the magnetostriction, are so different in character. No spectacular effects are present in the magnetostriction at low temperature. This suggests that the above discussed contributions from degenerate states are not dominant in the magnetostriction problem. This conclusion follows from Furey's work indeed. A meaningful calculation of the magneto-elastic energy could not be given since there are large fluctuations in the contributions to this energy over the Brillouin zone.

V. **Further developments.** — Finally we deal with some features of the magnetic anisotropy energy of nickel that ask for further investigations.

In order to calculate the magnetic anisotropy energy of nickel drastic assumptions had to be made and several contributions, one order of magnitude smaller than those from the energy shifts near the point X, have been neglected. This occurred for instance with contributions from states near the point L and with the effects, due to a change in the occupation of the energy bands upon rotation of the magnetization. This change in the occupation of states, caused by the spin-orbit interaction, has been discussed by Slonczewski [44] in a comment on Fletcher's calculation of the magnetic anisotropy energy of nickel. Furey pointed out that for the improved band structure he used this surface correction is at least one order smaller than the other contributions. These corrections can be important, however, for a discussion on the fine structure in the anisotropy energy. In particular a band, passing the Fermi level at the zone

boundary upon rotation of the magnetization, can cause a dramatic change in these fine structures. Perhaps an interpretation for the sharp peak in the curve $L_A/(\sin 2\theta \times \cos 2\theta)$ at 4.2 °K, given in figure 1, has to be sought in such effects. Measurements on the magnetic anisotropy energy could be helpful in this way to understand some of the fine details in the band structure of nickel.

A second point that asks for further experimental studies is the sensitivity of the anisotropy energy to the position of the Fermi level with respect to the 3 d-bands. By alloying nickel with cobalt or copper the Fermi level can be influenced and a comparison can be made between the calculated and the experimental change in the anisotropy energy. Using a rigid band approximation and taking values for the density of states and its derivative at the Fermi level from the work of Hodges e.a. [39], we find that a concentration of one percent cobalt in nickel causes a shift in the Fermi level downwards over 0.006 eV. Furey calculated for this shift a 25 percent lower value for the absolute value of K_1 ; the experiments result in nearly the same percentage. As a consequence one would expect to find an increase in the anisotropy energy by adding small percentages of copper to nickel. The experiments, however, show a decrease in this case too, though less pronounced than with cobalt.

It is not so difficult to construct situations in Furey's model in which an increase as well as a decrease in the Fermi energy leads to a decrease in the anisotropy energy. The position of the Fermi level at a cross point of the same band at two orientations of the magnetization is such a situation that is not excluded by the data of figure 3. The influence of the impurities on the fine structures in the anisotropy energy is even larger. These fine structures for nickel and some nickel alloys are presented in figure 5, where the energy in the (110) plane, reduced by the contribution of k_4, is plotted as a function of the orientation of the magnetization at 4.2 °K. Results for pure nickel (purity at least 99.99 %) at higher temperatures are also collected in this figure in order to direct the attention to the fact that the effects of alloying or raising temperature on the magnetic anisotropy energy are closely related. The magnetic anisotropy energy of nickel with one percent cobalt at 4.2 °K is nearly identical to that of pure nickel at 77 °K.

The identification of the anisotropy energy of nickel with the energy states near the point X turns out to be successful : it results in a value for the first anisotropy constant in the right order of magnitude ; it explains the rapid decrease of this energy in the low temperature region ; it predicts a large influence of impurities ; it presents a way of understanding complicated structures in this energy and it can account
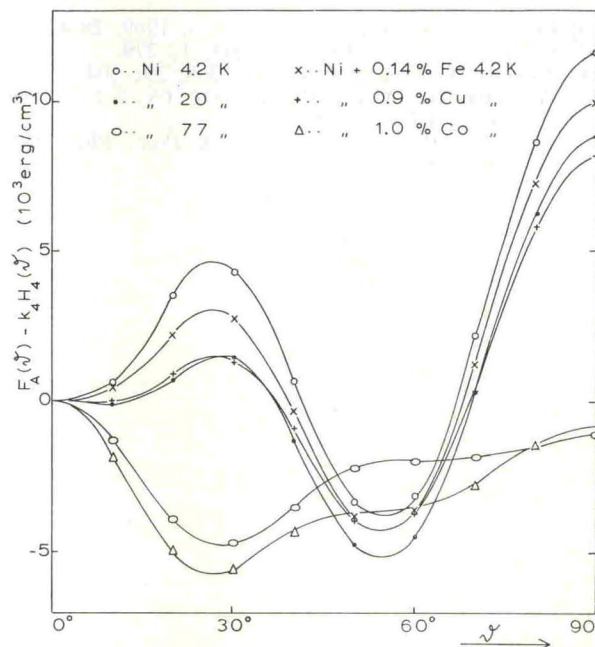

FIG. 5. — Higher order contributions to the magnetic anisotropy energy in the (110) plane for Ni at 4.2, 20 and 77 °K and for some Ni-alloys at 4.2 °K (ref. [28]).

qualitatively for the effects under pressure. By this identification we arrive for nickel in a relatively simple situation since in order to explain the properties of the anisotropy energy we have to investigate only a few bands in a small region of the zone. In the observation of these bands we have considerable help from other experimental methods, in particular from the study of the De Haas-Van Alphen effect.

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