

The heat-capacity data for both tantalum and niobium have also been compared with the empirical relation,

$$C_{el}/\gamma T_e = A e^{-bT_e/T},$$

of Corak, Goodman, Satterthwaite, and Wexler.<sup>10</sup>

The data for niobium and tantalum appear to be on

<sup>10</sup> Corak, Goodman, Satterthwaite, and Wexler, *Phys. Rev.* **102** 656 (1956).

the same curve, Fig. 6. However, it is evident that there is considerable deviation from the straight line predicted, particularly at the lowest temperatures.

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## Remarks on the Volume Magnetostriction and the Anisotropic Forced Magnetostriction

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The manner in which the anisotropic forced magnetostriction contributes to the magnetostriction of ferromagnetic substances at high-magnetic fields is discussed theoretically. In the past, the isotropic forced volume magnetostriction has been considered to be the only cause of the forced magnetostriction. This is probably one reason for the discrepancies between the experimental value of the volume magnetostriction and the theoretical value expected from the pressure dependence of the Curie point, etc. In addition to this, the limitations of the present theory concerning the relation between the isotropic volume magnetostriction and the related properties mentioned above are briefly examined in connection with the discrepancies between experiment and theory.

### I. INTRODUCTION

IT is generally accepted that there are two completely different kinds of magnetostriction corresponding to the two magnetization processes. The first is one corresponding to the magnetization process of alignment of domain magnetization vectors while the second involves an increase in the spontaneous magnetization of the domain itself.

In the alignment process the domain magnetization vectors change their direction; the crystallographic orientation remaining fixed. It should therefore be possible to explain the magnetostriction curves by assuming that below the Curie temperature all the domains are spontaneously strained by the spontaneous magnetization within each domain. The strain within each domain is assumed to vary with the direction of the domain magnetization. Therefore, as the domain distribution changes with the magnetization, magnetostriction occurs and this should saturate when the magnetization saturates. Furthermore, there is only a very small volume change when the domain magnetization changes from one crystallographic direction to the other. This type of dependence of the spontaneous lattice strain can be calculated from considerations of lattice symmetry and has been treated in detail by Becker and others.<sup>1-3</sup> Since this part of the magneto-

striction derives from the spontaneous lattice strain and a volume change is not involved, we shall call this part the "morphic term"<sup>4</sup> of the magnetostriction.

After the magnetization is saturated, one can still observe a change in size of a ferromagnetic sample which depends linearly upon the magnetic field. This differs from the low-field magnetostriction in that whereas the latter takes place without change of volume, the magnetostriction in a high field is primarily a volume effect, the expansion being the same in all directions. It is natural to associate this volume strain with the field-induced increase in the spontaneous magnetization. In this sense, the effect is called forced magnetostriction. Since this effect is related to the nature of the special internal forces in a ferromagnetic crystal (about which information is not obtainable by other means) it has been the object of an intense study and also is the main concern of this note.

As a matter of fact, the volume magnetostriction is usually considered to consist of three different terms which arise from different sources.<sup>1,2</sup> These are the form effect, the crystal effect, and the forced magnetostriction mentioned above.

The form effect arises purely from sample geometry. When the sample is magnetized, because of its finite demagnetizing factor, it has a certain amount of magnetostatic energy and magnetostriction occurs in order

<sup>1</sup> R. Becker and W. Döring, in *Ferromagnetismus* (Verlag Julius Springer, Berlin, 1938), pp. 270-311.

<sup>2</sup> E. W. Lee, in *Report on Progress in Physics* (Physical Society, London, 1955), Vol. 18, p. 184.

<sup>3</sup> W. P. Mason, *Phys. Rev.* **82**, 715 (1951).

<sup>4</sup> This word has been used in a different sense to describe the change in the elastic constants due to the deformation of the crystal lattice accompanying spontaneous magnetostriction.

to decrease this energy. This term,  $\omega_f$ , can be expressed in terms of the demagnetizing factor  $N$  (a definite constant in the case of an ellipsoid) and the magnetization  $M$  (referred to a unit volume) and the bulk modulus  $\kappa$ .

$$\omega_f = \frac{1}{2}NM^2/\kappa. \quad (1)$$

This reaches a limiting value when  $M = M_s$ , the saturation magnetization, and is usually the dominant term in low fields.

The second term, the crystal effect  $\omega_k$ , is the small volume change which occurs with the morphic effect. As mentioned before, a change in the direction of the domain magnetization causes a change in the symmetry of the crystal lattice which is almost without a volume change. However, a small volume change which is different for different directions of magnetization with respect to the crystal lattice does occur and contributes to the observed volume magnetostriction. This term should be included in the morphic term mentioned before and is related to the volume dependence of the ferromagnetic anisotropy constant.

The third term, the forced magnetostriction, comes from the volume dependence of the saturation magnetization. If we can assume this dependence, the change of volume at high magnetic field can be explained. Since the increase of  $M_s$  by an external field is small and can be said to be linear in the field, this volume change is also linear in the external field.

As is easily understood from the foregoing discussion, the morphic effect as well as the first two effects of the volume magnetostriction,  $\omega_f$  and  $\omega_k$ , saturate as the magnetization saturates. (As a matter of fact, the magnetization never saturates except for the case of a single crystal in certain special directions but approaches saturation asymptotically by increasing the external field. However, the contribution to the magnetostriction from this fact can be easily shown to be negligible if the external field is appropriately high.) At high fields, therefore, the forced volume magnetostriction is the only term which contributes to the change of size of the sample and this term is an isotropic volume change in the case of a cubic crystal, if only the volume dependence of the spontaneous magnetization is considered.

These ideas represent the usual interpretation of the phenomena of magnetostriction. We shall hereafter treat only the forced magnetostriction of cubic ferromagnetic substances.

Since the forced magnetostriction arises from the volume dependence of the saturation magnetization, this effect should be connected directly to the pressure dependence of the saturation magnetization and also, less directly, to the pressure dependence of the magnetic Curie point, the anomalous thermal expansion near the Curie point, etc. However, experiments so far show that there is no satisfactory agreement between the experimental results and the theoretical prediction of these

quantities using the above relations. Also, a recent experiment<sup>5</sup> shows that the forced linear magnetostriction is not isotropic as the theory would demand.

The existence of an anisotropic forced magnetostriction indicates that there must necessarily be a discrepancy between the measured forced magnetostriction and that which can be deduced from the related effects mentioned above. It is an aim of this note to investigate the origin of such an anisotropic forced magnetostriction. At the same time, the limitations involved in the deduction of the relations between the isotropic forced volume magnetostriction and the related phenomena will be discussed. These relations are of fundamental importance for obtaining information about the nature and behavior of the internal force in a ferromagnetic crystal.

## II. ANISOTROPIC FORCED MAGNETOSTRICTION

Since the phenomenological treatment so far advanced is insufficient, we must start from a more fundamental point of view in order to understand the origin of anisotropic forced magnetostriction. First, we neglect all kinds of contributions to the magnetostriction which depend upon the geometrical shape of the sample, for example, the form effect, and look into the properties which are characteristic only of the substance. In this case, a knowledge of the free energy of the system (referred to a definite quantity of substance) will be sufficient to treat the whole problem. We shall treat the problem with a localized model such as the Weiss-Heisenberg model of ferromagnetism. With this assumption, we can conveniently divide the free energy  $F$  of a ferromagnetic substance into three terms:

$$F = F_n + F_m + F_k. \quad (2)$$

Here,  $F_n$  is the part of the free energy of nonmagnetic origin, similar to the state above the Curie point. In other words, all free-energy terms which do not depend upon the alignment of the constituent magnetic moments are included here. We then divide the free energy of magnetic origin into two parts,  $F_m$  and  $F_k$ , the isotropic part and the anisotropic part. That is, we separate out a part of the free energy of magnetic origin which depends only on the direction of the aligned magnetic moments with respect to the crystallographic axes. This term is  $F_k$  and just corresponds to the so-called ferromagnetic anisotropy energy as is easily understood. The meaning of the separation in the above manner can be realized in the following discussion. The interaction energy  $E$  between magnetic moments in metals, which is essentially an exchange interaction, is actually not isotropic because of the spin-orbit interaction. It can then be expanded in terms of pseudomulti-

<sup>5</sup> B. A. Calhoun and W. J. Carr, Proceedings of the Pittsburgh Conference on Magnetism and Magnetic Materials, AIEE (1955), p. 107.

pole interactions;

$$E = \alpha(\mathbf{S}_i \cdot \mathbf{S}_j) + \beta \left[ \mathbf{S}_i \cdot \mathbf{S}_j - 3 \frac{(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{r_{ij}^2} \right] + \dots \quad (3)$$

Here,  $\mathbf{S}_i$  and  $\mathbf{S}_j$  denote vectors indicating the direction of the magnetic moments  $i$  and  $j$ , and  $\mathbf{r}_{ij}$  is a radius vector connecting the two magnetic moments. The quantities  $\alpha$ ,  $\beta$ , etc. are constants which include a radial dependence.  $F_m$  is then the sum of the isotropic terms (including the entropy term which depends on the relative alignments of magnetic moments with one another) and  $F_k$  is the sum of such pseudomultipole interaction terms which have an angular dependence. The magnitude of the sums depends upon the degree of alignment of the magnetic moments. Therefore there will also be a constant term corresponding to the free energy of the state of random spin orientation which will be considered as part of the  $F_n$  term.

Now, consider the condition of equilibrium,  $\partial F / \partial A_{ik} = 0$ , where  $A_{ik}$  is a component of the strain tensor. Above the Curie point, this can be replaced by  $\partial F_m / \partial A_{ik} = 0$ , because  $F_m$  and  $F_k$  do not exist. Below the Curie point, the magnetic moments become spontaneously parallel to each other and  $F_m$  and  $F_k$  appear. Since both  $\partial F_m / \partial A_{ik}$  and  $\partial F_k / \partial A_{ik}$  are not usually zero, the lattice deforms in order to satisfy the new equilibrium condition  $\partial F / \partial A_{ik} = 0$ . This is the origin of spontaneous magnetostriction. Thus we can clearly separate two different types of contributions to the spontaneous magnetostriction with this model. This deformation is usually very small and can usually be treated as a perturbation.

The second term  $F_m$  comes from the isotropic part of the exchange energy. Therefore, the spontaneous magnetostriction arising from this term does not depend upon the direction of magnetization but does depend only upon the degree of relative alignment of spins, i.e., on the magnitude of the spontaneous magnetization  $M_s$ . Therefore, this is an isotropic volume effect and we can observe this effect as an anomalous change in the volume or in the lattice constant below the Curie point. Since this is an isotropic volume change, this term does not contribute to the magnetostriction during the process of magnetization by alignment of domain magnetization vectors. However, at high magnetic fields, the saturation magnetization  $M_s$  is raised by an external field and then a volume change will occur. Therefore, this term contributes to the isotropic forced volume magnetostriction discussed in the previous section.

The third term gives rise to a spontaneous magnetostriction that comes from the strain dependence of the ferromagnetic anisotropy energy. Since this term depends upon the direction of the magnetization of the domain with respect to the crystal lattice, the spontaneous magnetostriction arising from this term is an anisotropic deformation of the lattice with respect to the direction of the aligned magnetic moments. This is

then the "morphic effect" mentioned in the previous section including the "crystal effect" of the volume magnetostriction  $\omega_k$ . Although this effect was treated in detail from symmetry arguments, Kittel<sup>6</sup> seems to have pointed out for the first time the importance of relating it to the anisotropy energy. This is important, because only this kind of treatment allows us to judge whether or not the magnetostriction coming from the  $F_k$  term gives rise to a forced magnetostriction in addition to that from the term  $F_m$ . If this effect exists, then this is the anisotropic forced magnetostriction.

The strain dependence of the term  $F_k$  can be obtained by expanding it in a Taylor series in the strain:

$$F_k = F_k^0 + \sum_{i \geq j} \left( \frac{\partial F_k}{\partial A_{ij}} \right)_0 A_{ij} + \dots \quad (4)$$

In this expression,  $F_k^0$  refers to the undistorted lattice and must satisfy cubic symmetry. The terms  $(\partial F_k / \partial A_{ij})_0 A_{ij}$  may have lower symmetry because these terms refer to the deformed lattice. Considering only the lowest order of expansion in terms of orientation, we may take from symmetry considerations,

$$\partial F_k / \partial A_{ii} = B_1 \alpha_i^2, \quad \partial F_k / \partial A_{ij} = B_2 \alpha_i \alpha_j, \quad (5)$$

where the  $\alpha_i$ 's are the direction cosines of the magnetization with respect to the crystallographic axes. According to the previous discussion, these terms should give the morphic effect of the magnetostriction and  $B_1$  and  $B_2$  are called magnetoelastic coupling constants. By comparison with the treatment from the symmetry argument,<sup>1</sup>  $B_1$  and  $B_2$  are related to the magnetostriction constants ( $h$ 's) in Becker's expression or to the usual expression  $\lambda_{100}$  and  $\lambda_{111}$  in the following manner:

$$\begin{aligned} \lambda_{100} &= \frac{2}{3} h_1 = -\frac{2}{3} B_1 / (C_{11} - C_{12}), \\ \lambda_{111} &= \frac{2}{3} h_2 = -\frac{1}{3} B_2 / C_{44}, \end{aligned} \quad (6)$$

where  $C_{11}$ ,  $C_{12}$ , and  $C_{44}$  are the elastic constants. By taking the higher symmetry terms, we can easily get more relations involving more  $B$ 's and  $h$ 's.<sup>3</sup>

In addition to its strain dependence, the anisotropy energy also depends strongly on the temperature. Since it arises from the relative alignment of the magnetic moments, the temperature dependence of the anisotropy energy should be related to the temperature dependence of the spontaneous magnetization. This idea was first expressed by Zener<sup>7</sup> in a clear-cut way and later treated in more detail by Keffer.<sup>8</sup> According to this idea, the temperature dependence of the anisotropy constant  $K$  is given by

$$K / K^0 = c (M_s / M_s^0)^n. \quad (7)$$

Here  $K^0$  and  $M_s^0$  are the values of  $K$  and  $M_s$  at absolute zero temperature, respectively, and  $c$  is a proportionality

<sup>6</sup> C. Kittel, *Revs. Modern Phys.* **21**, 555 (1949).

<sup>7</sup> C. Zener, *Phys. Rev.* **96**, 1335 (1954).

<sup>8</sup> F. Keffer, *Phys. Rev.* **100**, 1692 (1955).

constant. The value of  $n$  depends upon the mode of the spin alignment and, therefore, is not necessarily constant throughout the whole temperature range.<sup>8</sup> In the case of  $K_1$ , the first anisotropy constant, the behavior is well reproduced by  $n=10$  for iron and  $n=20$  for Ni. Although the  $K$ 's are used here instead of  $F_k$  in Eq. (7), the relation between  $F_k$  and the  $K$ 's can be understood easily and  $K_1$  is the most important term in  $F_k$ .

At high fields, the saturation magnetization is raised by the external field and, thus, a change of the anisotropy constant should occur through Eq. (7):

$$\Delta K/K^0 = n(M_s/M_s^0)^{n-1} \Delta M_s/M_s^0. \quad (8)$$

Therefore  $K$  and hence  $F_k$  depends on the magnetic field as well as  $F_m$ . Since the factor  $n$  is big, the change of  $K$  is not small and the constants  $B_1$ ,  $B_2$ , etc., as well as the magnetostriction constants  $h_1$ ,  $h_2$ , etc., depend rather strongly on the magnetic field. The forced magnetostriction, therefore, not only consists of the isotropic volume effect as mentioned before, but also has an anisotropic linear magnetostriction which comes from the term  $F_k$ . Actually, the phenomenon corresponding to this fact was observed by Vautier<sup>9</sup> in the case of iron and by Calhoun and Carr<sup>5</sup> in the case of Si-Fe. Although the magnitude of this contribution is small, it is not correct to calculate the forced volume magnetostriction by taking three times a linear forced magnetostriction. At the same time, the crystal effect of the volume magnetostriction  $\omega_k [= 3h_3s; s$  is given by Eq. (10)], being of the same origin, also gives rise to a forced volume magnetostriction which is sometimes not negligible as compared to the isotropic forced volume magnetostriction. In the case of nickel, for example, the actual forced volume magnetostriction is very small on the one hand, and  $n$  is large ( $\sim 20$ ) on the other hand. In such a case, the effect of the contribution of such an anisotropic forced volume magnetostriction would be serious. This might be the reason for the discrepancies even in the sign which exist in the measured values of the volume magnetostriction.<sup>10</sup>

Before discussing the magnitude of this effect, it might be appropriate to remark here that the anisotropy constant will increase with field through the mechanism (7). In torque measurements of the constant  $K$ , for example, the values are actually observed to depend upon the magnetic field. This is usually attributed to the inhomogeneity in magnetization inside the sample because its shape is different from that of an ellipsoid, which changes with the field.<sup>11,12</sup> However, from the above reasoning, a part of this effect should be the real increase in  $K$  with field which, in some cases, may not be negligible, especially when an extrapolation to infinite field is involved as the usual procedure requires.

<sup>9</sup> R. Vautier, Ann. phys. **9**, 322 (1954).

<sup>10</sup> K. Azumi and J. E. Goldman, Phys. Rev. **93**, 630 (1954).

<sup>11</sup> L. P. Tarasov, Phys. Rev. **56**, 1224 (1939).

<sup>12</sup> J. S. Kouvel and C. D. Graham, Jr.; Proceedings of the Boston Conference on Magnetism and Magnetic Materials, AIEE (1956), p. 85.

Let us now estimate the size of the volume effect contributed by the morphic term (crystal effect). According to Becker's calculation,<sup>1</sup>  $\omega_k$  is given as follows:

$$\omega_k = (\nu K/\kappa)s. \quad (9)$$

Here  $\kappa$  is the bulk modulus,  $K$  is the anisotropy constant,  $\nu$  is the volume dependence of the anisotropy constant,  $(1/K) (\partial K/\partial V)$ , and

$$s = \alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2, \quad (10)$$

where the  $\alpha$ 's are the direction cosines of the magnetization vector with respect to the crystal axes. Thus the forced volume magnetostriction arising from the morphic term is

$$d\omega_k/dH = (\nu/\kappa)(dK/dH)s. \quad (11)$$

Because of the relations (9) and (11), we can measure separately  $\omega_k$  and  $d\omega_k/dH$  and thus separate them from the total effect by using a single crystal. So far  $\omega_k$  has been only estimated indirectly from measurements of the field dependence of the volume magnetostriction of a polycrystalline sample, by estimating the form effect according to Eq. (1) and by extracting the forced magnetostriction by a linear extrapolation from high fields. However, a single-crystal measurement should give more reliable information concerning  $\omega_k$  and  $d\omega_k/dH$  which may not be obtained by other means.

Now for a polycrystal

$$dK/dH = n(K/M)(dM/dH),$$

and

$$d\omega_k/dH = -(\nu K/\kappa)\bar{s}n(1/M)(dM/dH), \quad (12)$$

where  $\bar{s}$  is the average of  $s$ . The quantity  $-(\nu K/\kappa)\bar{s}$  has been estimated, from the field dependence of the volume magnetostriction, as mentioned above, to be of the order of  $10^{-6}$  in the case of iron. If we take  $n=10$  and  $(1/M_s)(dM_s/dH)$  to be  $10^{-6}$  to  $10^{-7}$ , the value of  $\partial\omega_k/\partial H$  would be of the order of  $10^{-11}$  to  $10^{-12}$ . The forced volume magnetostriction of iron is of the order of  $10^{-10}$ . Therefore, the contribution of such an effect can be of the order of 10% of the total forced volume magnetostriction. In the case of single crystal, the magnitude of the effect will, of course, depend upon the direction of the field.

As for the forced linear effect  $\partial h/\partial H$ , its order of magnitude can be estimated from the ratio of  $\omega_k$  to  $\partial\omega_k/\partial H$ , because this ratio is nearly the same as the ratio of  $h$  to  $\partial h/\partial H$ . This shows that the contribution of the linear effect to the forced magnetostriction is more serious. Phenomenologically, the magnitude of the anisotropic forced magnetostriction can also be estimated from the temperature dependence of the magnetostriction constants which may be more direct, (i.e., the dependence of the magnetostriction constants on the saturation magnetization  $M_s$ .)

### III. LIMITATIONS OF THE ARGUMENT CONCERNING THE VOLUME MAGNETOSTRICTION

The basic assumption in deriving the origin of the anisotropic forced magnetostriction is Eq. (2). This assumption is rather general if we can justify adopting a localized model in treating ferromagnetism. Actually, with a localized model many fruitful results have been obtained concerning the phenomena of ferromagnetism. However, if we start from a band model, the division of the free energy in a manner similar to that in Eq. (2) is no longer clear-cut. The actual situation lies between these two points of view and thus the limitation of the present treatment is that we do not have a sufficiently exact knowledge of ferromagnetism. It seems futile, in the present stage, to go further.

In the discussions of the previous section, it was pointed out that one possible reason for the discrepancies between the experimental values of the forced magnetostriction and the values which are calculated from the pressure dependence of the saturation magnetization or of the Curie point, etc., is the contribution of the anisotropic forced magnetostriction (linear and volume effect) to the forced magnetostriction. Since we can separate experimentally the anisotropic forced magnetostriction by using a single crystal, we should then expect good agreement between experiment and theory. However, there still remain several points in the theory to be examined. These arise mainly from the fact that we must adopt some kind of a model of ferromagnetism which can be treated in a quantitative manner in order to derive the relationships between these various measured quantities.

It is somewhat easier to investigate experimentally the pressure dependence of the Curie point than that of the saturation moment, and as a result the data for the former are much more complete than for the latter. Since the present status of the theory of ferromagnetism is such that any direct calculation of the change of Curie temperature with pressure is out of the question, the relation between the volume magnetostriction and the pressure dependence of the Curie point is more frequently required than others. As has been pointed out before, since this relation and others like it are of fundamental importance for obtaining information concerning the interaction between magnetic moments in a ferromagnetic crystal, the limitations of a treatment of this kind will be examined here.

The earliest attempt to correlate these quantities is that by Kornetzki.<sup>13</sup> He assumes the same basic viewpoint as Weiss and further assumes that the absolute saturation referred to a fixed quantity of substance does not change. Therefore, the change in the saturation magnetization by a change in volume at a definite temperature is assumed to occur only through the volume dependence of the Curie point, in other words,

through the volume dependence of the exchange integral or molecular field constant (these quantities being directly related to the Curie temperature). By doing this he could connect the pressure dependence of the Curie point to the volume magnetostriction only by assuming  $M_s = f(T/\theta)$  (where  $\theta$  is the Curie point and is a function of the volume), without any exact knowledge of the functional form of  $f$ . It was found, by this assumption, that

$$\frac{1}{\theta} \frac{\partial \theta}{\partial p} = \frac{1}{T} \frac{\partial \omega / \partial H}{(\partial M_s / \partial T)_p - 3\alpha\kappa \partial \omega / \partial H}, \quad (13)$$

in which  $p$  is the pressure,  $\partial \omega / \partial H$  the isotropic forced volume magnetostriction,  $\alpha$  the coefficient of thermal expansion, and  $\kappa$  the bulk modulus. Usually the second term in the denominator is much smaller than the first and since  $(\partial M_s / \partial T)_p$  is always negative, a positive  $\partial \theta / \partial p$  is associated with a negative  $\partial \omega / \partial H$  and vice versa. This relation is still a basis for interpretation of results. Since the exchange interaction is known to depend on interatomic distance, the pressure dependence of the Curie point tells us the dependence of the interaction energy on the interatomic distance. In this sense the volume magnetostriction occupies an important place in the investigation of the nature of the fundamental interaction between the magnetic moments. However, Kornetzki's assumptions are especially doubtful in the case of metals, because, with a change of volume, the degree to which the electrons distribute among  $s$  and  $d$  states may differ and also the relative shift of bands for the electrons with plus and minus spins may change because of the change in the kinetic energy of free electrons. Especially in the case of iron, for example, the latter effect might be rather important, because both plus and minus  $d$  bands are assumed to be incompletely filled.

Some of these difficulties were removed by Smoluchowski,<sup>14</sup> although he still used a molecular-field treatment. He admitted a change in the absolute saturation with the change of volume, but instead he had to assume, at constant volume, a definite functional form of  $f(T/\theta)$ . In his case, this dependence was represented by that calculated from a Brillouin function. This treatment is certainly a compromise between the Ising-type localized treatment and the band model to cover the defects mentioned above. A drawback of this treatment is, however, that the theoretical results are very sensitive to the form of the  $M_s-T$  curve. This situation causes trouble in two ways. The determination of the internal quantum number of the atomic spin system,  $j$ , in order to specify the Brillouin function, and the approximational method of calculating the Brillouin function itself. It is very difficult to give a reasonable value of  $j$  in the case of metals, because the number of Bohr magnetons per atom is not an integral number. The

<sup>13</sup> M. Kornetzki, Z. Physik **98**, 289 (1935).

<sup>14</sup> R. Smoluchowski, Phys. Rev. **59**, 309 (1941); **60**, 249 (1941).

value of  $j$  probably differs from atom to atom and the system is a kind of combination of several different states. Furthermore, a change in the absolute saturation by a volume change means a change in the number  $j$ , and we can not specify this. The shape of the Brillouin function depends sensitively on  $j$ . Second, the shape of the Brillouin function is also very sensitive to the approximations in the calculation. For example, the Brillouin function which is usually used for the comparison of theory and experiment, using the shape of the temperature dependence of the saturation value and susceptibility above the Curie point, etc., to determine  $j$  is that which is calculated on the basis of a zeroth approximation in the statistical treatment (Weiss, Bragg-Williams). If we adopt the Bethe approximation to this problem, for example, the  $M_s$  versus  $T/\theta$  curve for  $j=1$  is almost the same as for  $j=\frac{1}{2}$  in the Bragg-Williams approximation for both body-centered and face-centered cubic cases.<sup>15</sup> Furthermore, in the case of alloys, the  $M_s$  versus  $T/\theta$  curve seems to deviate from that of the pure element appreciably but in a systematic way which cannot be ascribed to the change of  $j$  in any way.<sup>16</sup> For these reasons, Smoluchowski's improvement of the treatment has no practical advantage over Kornetzki's treatment.

In connecting the volume magnetostriction with other properties, the change of the spontaneous magnetization by an external field appears implicitly. To obtain the result quantitatively, it is therefore necessary to know the form of the  $M_s$  versus  $T/\theta$  curve under an external field because the volume magnetostriction is measured under a high external field. As is well known, this is definitely different from that under no field. The deviation is especially large near the Curie point. In deriving the necessary relations, this effect has been completely neglected. In principle, we can derive this effect theoretically, if we can assume, for example, the validity of the Brillouin function for this problem. Then the external field can be included in the expression analytically and an  $M_s$  versus  $T/\theta$  curve can be derived for a definite external field. However, this brings up the same problems pointed out before, i.e., the applicability of the Brillouin function and the selection of  $j$ . In any event, at the present stage of the theory, we can not rely too much on a quantitative relationship between

the intrinsic isotropic forced volume magnetostriction and the pressure dependence of the Curie point, the form of the so-called Bethe-Slater curve, etc., though we can expect an improved qualitative agreement by subtracting the anisotropic forced magnetostriction from the total forced magnetostriction.

#### IV. SUMMARY

The origin of the anisotropic forced magnetostriction is discussed. The main points of this discussion can be summarized as follows:

(a) Crystal anisotropy is shown to depend upon the external magnetic field. In some cases, this may affect the result of the measurements of the anisotropy.

(b) As a result, the morphic term of the magnetostriction depends upon the field strength and this gives rise to an anisotropic forced magnetostriction. Therefore it is not correct to interpret the observed linear forced magnetostriction as coming from the isotropic volume effect only.

(c) The morphic term also includes a volume effect. The field dependence of this anisotropic forced volume magnetostriction can be estimated to be of the order of  $10^{-11}$ – $10^{-12}$  in the case of iron. This corresponds to, at most, 10% of the total volume effect. The anisotropic linear forced magnetostriction can be much bigger and have a more serious influence on the forced magnetostriction.

(d) The anisotropic forced magnetostriction can be separated experimentally by making a measurement with a single crystal. By subtracting these contributions from the total effect, we can expect a more satisfactory agreement between theory and experiment.

The limitations of the present treatment concerning the origin of the anisotropic magnetostriction are discussed. In addition to this, the limitations of deducing the relationships between the isotropic forced volume magnetostriction and the pressure dependence of the Curie point are also discussed, since these effects are important to understanding the fundamental interaction between elementary magnetic moments.

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<sup>15</sup> H. Sato, unpublished work presented before the meeting of the Physical Society of Japan, April, 1949.

<sup>16</sup> J. J. Went, *Physica* **17**, 98 (1951).