DEPENDENCE OF DOMAIN WIDTH ON CRYSTAL THICKNESS IN THE REMANENT HONEYCOMB DOMAIN STRUCTURE IN COBALT

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The dependence of the domain width on the crystal thickness is examined experimentally for the remanent honeycomb domain structure in cobalt single crystals, in the thickness range where no closure domains are formed at the basal crystal surface. The experimental results confirm the half-power law following form theory, the domain width being however nearly five times larger than predicted by theory. Moreover, it is shown that for crystal thicknesses up to 50 μm no closure domains form at the crystal surface, contrary to theory according to which closure domains in cobalt should from if the crystal thickness is larger than 0.2 μm. A critique of similar experiments carried out by other authors on magnetoplumbite is given.

1. Introduction

The remanent honeycomb domain structure, of which a model is shown in Fig. 1, was first observed by Kaczér and Gemperle in magnetoplumbite and studied experimentally and theoretically in [1]. Since then, it has been obtained in various uniaxial ferromagnets by other authors (see, *e.g.*, [2–7]), and the theoretical approach from [1] has been refined in [8]. In either case, [1] and [8], the theoretical considerations are based on the simple model from Fig. 1, with no closure domains at the basel crystal surface. This model seems quite adequate if the crystal is sufficiently thin in the direction of easiest magnetization (hexagonal crystal axis) — the critical thickness depending on the material, since in that case no closure domains are formed at the surface (see [1, 5, 7]; cp. [9–12]). The observations and measure-

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ments in [1] were confined to a thickness range of the magnetoplumbite crystal to which the model from Fig. 1 certainly applies. Nevertheless, there is a significant disagreement between the experimental and theoretical curves for the dependence of the domain width on the crystal thickness, which the authors tried to explain away by declaring the domain structures with domain widths smaller than theoretically predicted as unstable. To bring theory and experiment in agreement, they simply enlarged the domain width (to the desired value) by applying in the magnetically preferred direction a strong magnetic field (up to 2 k0e) which — according to the authors'opinion — is required to make the domain structure stable. In doing so they overlooked the fact that, while trying to improve the agreement between theory and experiment at one point they worsened it by about the same amount at another. This is plainly seen from the following reasoning.

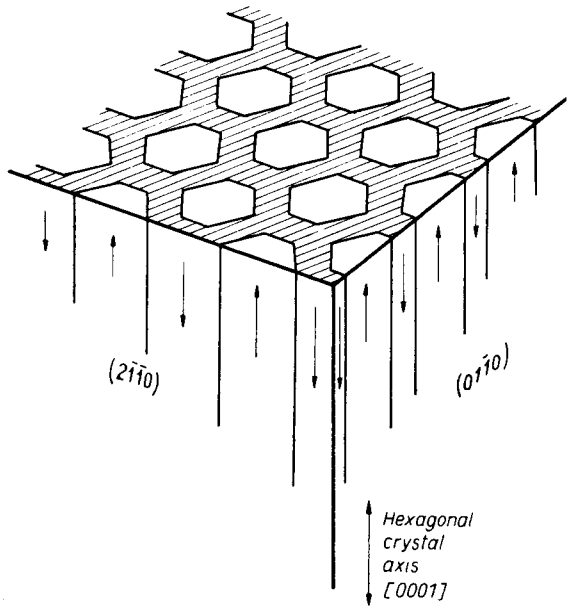

Fig. 1. Model of the simple honeycomb remanent domain structure in uniaxial ferromagnets

According to [1], the theoretical relationship between the domain width D and the crystal thickness T is for the plate-like Landau-Lifshitz domain structure

$$D = 0.767\sqrt{\gamma T}/J \tag{1}$$

where J is the saturation magnetization and γ the Bloch wall energy, and the theoretical dependence of the distance L between the domains in the honeycomb domain structure (see Fig. 2) on the crystal thickness T is

$$L = 2.06\sqrt{\gamma T}/J. \tag{2}$$

Hence, according to theory $D/L = 0.372$, while the experimental curves obtained from the respective "unstable" domain structures (bold curves in Fig. 8 in [1]) lead, according to the

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authors, to $D/L = 0.525$. Upon "stabilizing" the domain structures by applying a strong magnetic field the experimental curves shifted in such a manner (dashed curves in Fig. 8 in [1]) as to bring the experimental value of the ratio D/L pretty close to the theoretical one.

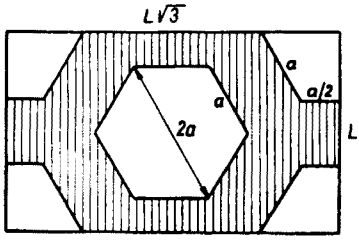

Fig. 2. Elementary rectangular cell of the regular honeycomb pattern as used for the Fourier-series expansion in [1, 8]

Now, from the experimental curves in Fig. 8 in [1] we have for the "unstable" honeycomb domain structure

$$L_u = 1.45\times10^{-2}\sqrt{T}\ [\mathrm{cm}], \tag{3}$$

and for the "stable" one

$$L_s = 1.85\times10^{-2}\sqrt{T}\ [\mathrm{cm}]. \tag{4}$$

On the other hand, upon inserting in Eq. (2) the values for magnetoplumbite ($\gamma = 4.82$ erg/cm^2, $J^2 = 1.02\times10^5$ erg/cm^3) one has

$$L = 1.42\times10^{-2}\sqrt{T}\ [\mathrm{cm}] \tag{5}$$

which is evidently in much better agreement with L_u than L_s. From this point of view one can as well reverse the conclusions drawn in [1] about the stability of the respective domain structures.

In our opinion, this contradiction has nothing to do with the problem of stability, for reasons repeatedly pointed out elsewhere [6–8] (see also Section 4 of the present paper). We believe that, instead of jumping to premature conclusions and offering *ad hoc* explanations, some more research is to be done both, theoretically and experimentally, before any satisfactory answer to this problem can be given. The more so as the plain disagreement between theory and experiment is by no means confined to the honeycomb structure in magnetoplumbite or, for that matter, to any particular remanent domain structure in any particular uniaxial ferromagnet, as shown here and in [12]. On the theoretical part, further refinements of the existing theory are certainly needed, regarding both the simplicity of the models commonly in use and the numerous simplifications usually made in formulating the problem and carrying out the calculations. On the other hand, of the enormous number of papers devoted to experimental investigations of domain structures only a small part provides results of systematic observations or measurements, to that extent that reliable experimental curves for the thickness dependence of the domain width are still a rarity.

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The purpose of the present paper is confined to examining experimentally the thickness dependence of L, as defined in Fig. 2, for the honeycomb domain structure in cobalt, and to comparing the experimental results with the theoretical curves following for cobalt from [1] and [8]. Our results show that the disagreement between theory and experiment is for cobalt even largerthan for magnetoplumbite, despite the fact that the curve obtained from the refined theoretical approach proposed in [8] comes closer to the experimental curve than that following from the simpler theory given in [1]. At present, no reasonable explanation of this disagreement can be offered.

2. *Theory*

The formula (2) which has been derived in [1] leads for cobalt to the relation

$$L = 0.42 \times 10^{-2} \sqrt{T} \text{ [cm]} \tag{6}$$

as $\gamma = 8.2$ erg/cm^2, $J^2 = 20.2 \times 10^5$ erg/cm^3. This dependence is shown by curve *3* in Fig. 3.

The simple theoretical approach in [1] was based on the assumption that in the demagnetized state the total areas of opposite magnetic poles on the basal crystal surface (shaded and white areas in Figs 1, 2) should be equal, which leads to the condition

$$a/L = \sqrt{6}/6 \approx 0.4082 \tag{7}$$

as easily concluded from Fig. 2. This assumption is fairly justified for large crystals (large T). For small T, however, one may well expect a/L to depart from its asymptotic value (7), as with decreasing crystal size the relative contribution of the anisotropy energy and the magnetostatic self-energy to the total energy of the crystal changes (*e.g.*, single-domain

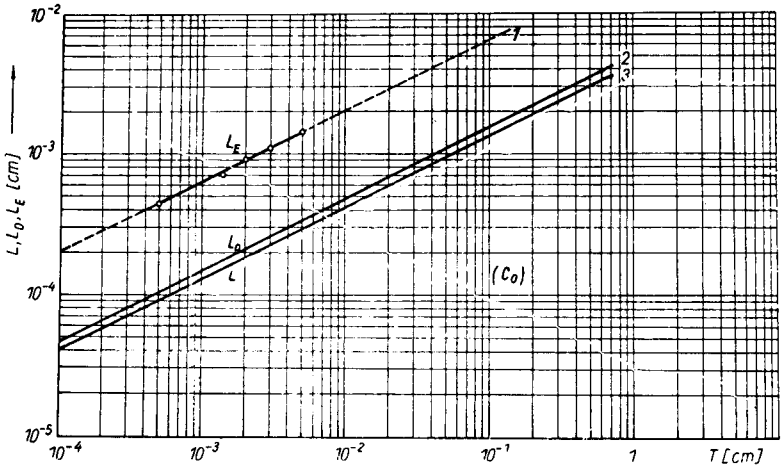

Fig. 3. Dependence of the distance L (for cobalt) as defined in Fig. 2 on the crystal thickness T in the direction of easiest magnetization [0001]. Experimental curve *1* leading to Eq. (16) is compared with theoretical curves *2* and *3* given respectively by Table I and Eq. (6). Curve *2* leads to the relation (15)

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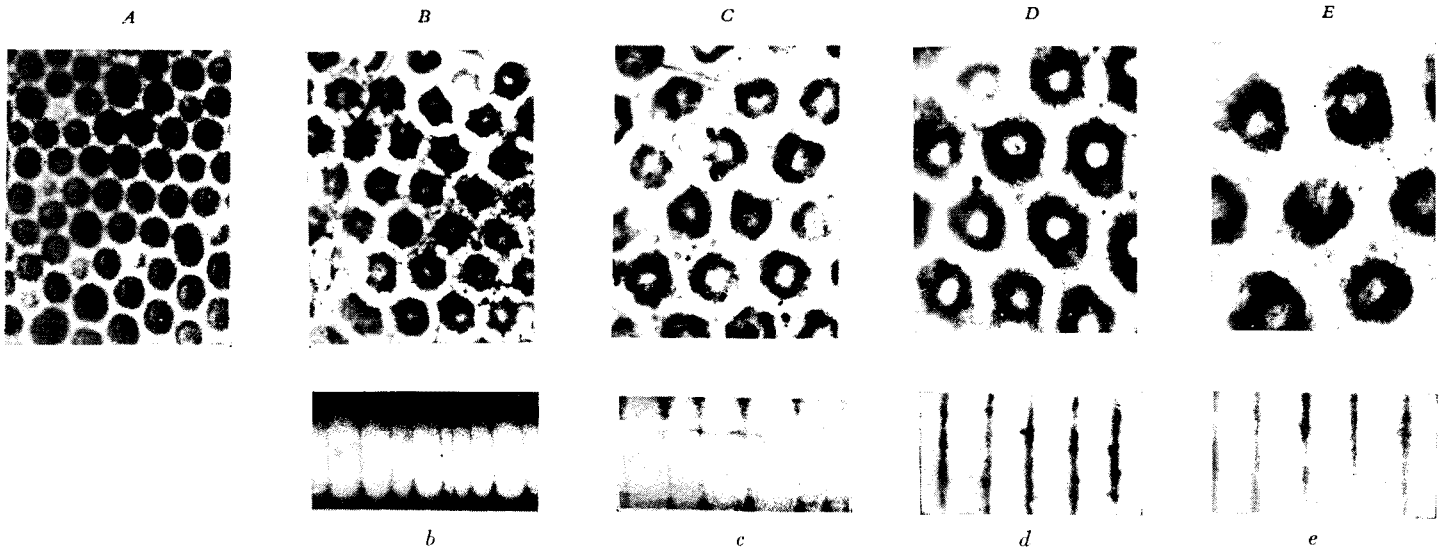

Fig. 5. Powder patterns of the simple honeycomb remanent domain structure in cobalt single crystals of rectangular shape and crystallographic orientation as shown schematically in Fig. 1. Patterns A—E from the basal crystal surface (0001), and patterns b—e from the axial surface $(2\bar{1}\bar{1}0)$. Crystal thicknesses in the direction [0001]: A — 5 μm; B, b — 15 μm; C, c — 20 μm; D, d — 30 μM, E, e — 50 μm. Thin plastic coating applied on the basal crystal surface in C(4 μm), D(6 μm) and E(10 μm), to enhance the legibility of the powder pattern (see [7, 17])

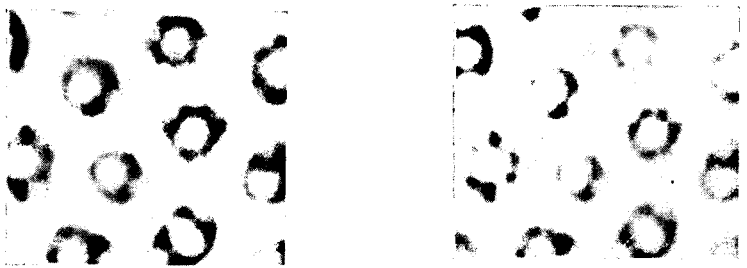

Fig. 6. Powder patterns from the same site on the basal surface of the cobalt single crystal used in [18] (crystal thickness in the direction [0001] — 12 mm), showing the stability of the remanent honeycomb domain structure. Pattern (a) observed in 1964 just upon formation of the domain structure, pattern (b) obtained in 1968

state in small particles). For this reason, the approach from [1] has been refined in [8], by making $a/L = \beta$ a variable (beside L) confined to the interval

$$0 \leqslant \beta \leqslant \sqrt{3}/3, \quad \beta = a/L. \tag{8}$$

In that case, however, the minimum condition for the energy E with respect to L and β leads to a transcendental equation for β which must be solved numerically. Unfortunately, due to an oversight this equation has been solved in [8] for $\gamma = 8.2$ erg/cm^2, $J^2 = 1.02 \times$ $\times 10^5$ erg/cm^3, the value of γ corresponding to cobalt, and that of J^2 to magnetoplumbite. Therefore, for the present purpose we had to solve this equation once again, with the values of γ and J^2 for cobalt as used in Eq. (6).

From Eq. (16) in [8] we have

$$E = 4\gamma\beta\, T\sqrt{3}/L + 2\pi T J^2 (6\beta^2 - 1)^2 +$$
$$+ (27 J^2 L \sqrt{3}/\pi^4) [\cos(2\pi\beta/\sqrt{3}) - \cos(4\pi\beta/\sqrt{3})]^2, \tag{9}$$

the necessary conditions for E to be minimum being

$$\partial E/\partial L = 0, \tag{10}$$

$$\partial E/\partial \beta = 0, \tag{11}$$

and the sufficient ones

$$\Delta = (\partial^2 E/\partial\beta^2)(\partial^2 E/\partial L^2) - (\partial^2 E/\partial L \partial\beta)^2 > 0,$$

$$\partial^2 E/\partial L^2 > 0. \tag{12}$$

From (9)–(11) one has for β the transcendental equation

$$8\pi^3 J\beta(6\beta^2 - 1)\sqrt{\beta T/9\gamma} = (4\pi\beta/\sqrt{3}) \sin(2\pi\beta/\sqrt{3}) - \tag{13}$$
$$-\cos(2\pi\beta/\sqrt{3}) + \cos(4\pi\beta/\sqrt{3}) - (8\pi\beta/\sqrt{3}) \sin(4\pi\beta/\sqrt{3})$$

to be solved for different values of the parameter T. Denoting by β_0 the solution of the above equation and by L_0, a_0, E_0 the corresponding values of L, a, E determined from Eqs (8)–(10) we have for $\beta_0(T)$ and $E_0(T)$ the curves in Fig. 4, and for $L_0(T)$ the curve 2 in Fig. 3. A representative set of numerical data is given in Table I. As seen from the signs of Δ and $\partial^2 E/\partial L^2$ in Table I, the examined domain structure is stable.

One easily proves from Fig. 3 that the curve 2 is quite accurately described by the relation

$$L_0 = 0.46 \times 10^{-2} \sqrt{T}\ [\text{cm}] \tag{14}$$

which, when converted to the form (2) would read

$$L_0 = 2.27 \sqrt{\gamma T}/J. \tag{15}$$

By comparing (2) and (15) or (6) and (14) one has $L_0/L \approx 1.1$. The formula (15) when applied to magnetoplumbite leads to $L_u < L_0 < L_s$, with L_u and L_s from Eq. (3) and (4).

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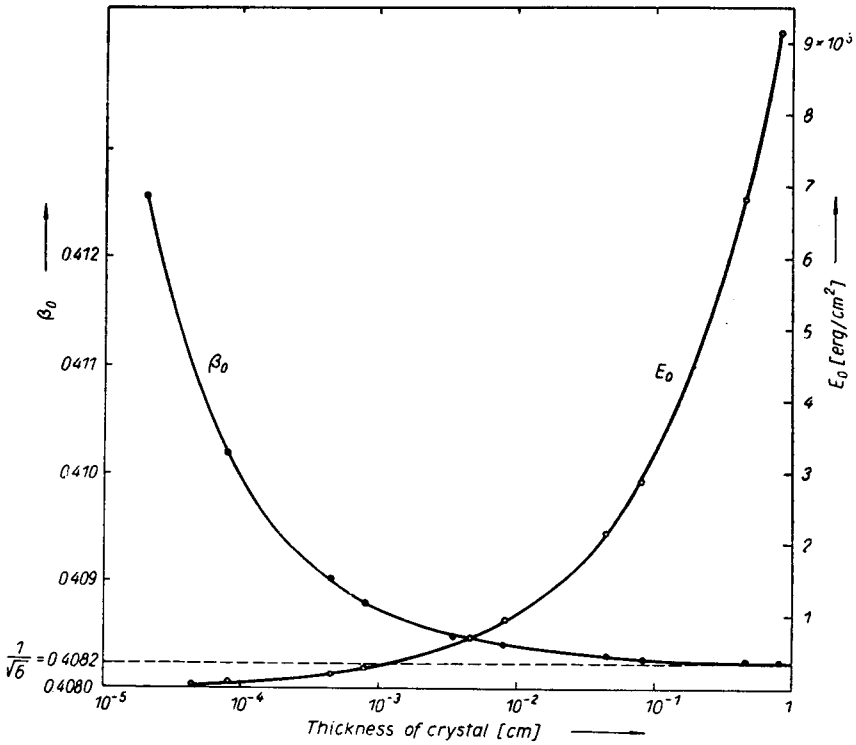

Fig. 4. Minimum values β_0, E_0 for cobalt of the coefficient β defined by Eq. (8) and the energy E given by Eq. (9), plotted as functions of the crystal thickness T, according to Table I

TABLE I

Representative numerical data for cobalt of the minimum values β_0, L_0, a_0, E_0 of β, L, a and E, for several values of the crystal thickness T

T [cm]	$10^4\beta_0$	L_0 [μm]	a_0 [μm]	E_0 [erg/cm²]	Sign of	
					Δ	$\partial^2 E/\partial L^2$
8.06×10^{-1}	4082.6	40.90	16.70	9142.40	+	+
4.5×10^{-1}	4082.7	30.56	12.48	6831.21	+	+
8.06×10^{-2}	4083.0	12.93	5.28	2891.03	+	+
4.05×10^{-2}	4083.2	9.67	3.95	2160.16	+	+
8.06×10^{-3}	4084.2	4.09	1.67	914.17	+	+
4.5×10^{-3}	4084.8	3.06	1.25	685.05	+	+
8.06×10^{-4}	4088.1	1.30	0.53	289.03	+	+
4.5×10^{-4}	4090.2	0.97	0.40	215.95	+	+
8.06×10^{-5}	4101.7	0.41	0.17	91.34	+	+
2.0×10^{-5}	4125.8	0.21	0.09	45.45	+	+

3. Experimental results

The measurements of L for the honeycomb domain structure in cobalt were performed on five samples cut from a cobalt single crystal (purity 99.46% Co) having the shape of a rectangular block with faces perpendicular to the crystallographic directions [0001], $[01\bar{1}0]$ and $[2\bar{1}\bar{1}0]$ (see Fig. 1), the dimensions in the last two directions being respectively 8 mm and 7 mm, and the thickness T of the samples in the hexagonal direction [0001] being 50, 30, 20, 15 and 5 μm. The preparation of the samples (polishing, determination of crystallographic orientation, *etc.*) and the colloid used for the powder-pattern observations were as usual (see, *e.g.*, [6, 7, 10, 13–16]). To improve the legibility of the powder patterns on the basal surface of the thicker samples, a thin plastic coating (see [17]; cp. [6, 7, 12]) has been used on this surface, its thickness being about 10, 6 and 4 μm for the samples $T = 50$, 30 and 20 μm respectively.

As already pointed out in [6, 7], the creating (saturation) field H_0 used in producing the remanent honeycomb domain structure in large crystals (*i.e.*, large T) must be parallel to the hexagonal axis [0001], as opposite to the case of thin samples where H_0 must be perpendicular to this direction [1, 5]. In our case, we had to apply a field $H_0 \perp [0001]$ for the thinnest sample, and $H_0 \parallel [0001]$ for the remaining four samples. It thus appears that the critical thickness below and above which H_0 must be respectively perpendicular or parallel to the direction [0001] lies for cobalt somewhere between 5 and 15 μm. This effect is examined in more detail in [12] where the critical thickness is found to be about 12 μm.

As shown in [6], the pattern of a remanent domain structure also depends on the intensity of H_0, as well as on the switching-off velocity of H_0. According to our experience, the highest regularity in the domain pattern is obtained for $H_0 = 1.5$ k0e in the case $H_0 \parallel [0001]$, and $H_0 = 18$ k0e for $H_0 \perp [0001]$, the switching-offvelocity being in both cases 300 0e/sec.

Examples of powder patterns observed on the basal (0001) and axial $(2\bar{1}\bar{1}0)$ crystal surface of each sample are shown in Fig. 5. In this way, the distance L between the centres of two neighbouring domains could in each case be measured on both the crystal surfaces. For each sample, the honeycomb domain structure was destroyed and reconstructed several times, and the average experimentally determined value of L which we shall denote by L_E was in each case the same, within the accuracy of measurements. Our experimental results lead to curve *3* in Fig. 3 (bold part of the curve) which is very accurately described by the relation

$$L_E = 2.02 \times 10^{-2} \sqrt{T}\ [\mathrm{cm}]. \tag{16}$$

Eq. (16) when converted to the form (2) yields

$$L_E = 9.96 \sqrt{\gamma T}/J. \tag{17}$$

By comparing Eqs (2), (15) and (17) or (6), (14) and (16) we obtain

$$L_E/L_0 \approx 4.4, \quad L_E/L \approx 4.8, \tag{18}$$

i.e., the experimental values for cobalt are nearly five times larger than the theoretical ones.

4. *Concluding remarks*

As seen from the curves in Fig. 3, the disagreement between theory and experiment is for the remanent honeycomb domain structure in cobalt even larger than in magnetoplumbite, particularly when taking into account the fact that the domain structures examined here correspond to those which in [1] were classified as "unstable". From this point of view, namely, the relation (16) for cobalt corresponds to the relation (3) for magnetoplumbite and, while (3) agrees fairly enough with the theoretical formula (5) obtained from (2) and differs not so badly from formula (15) when specified for magnetoplumbite, the respective differences between theory and experiment are far more pronounced for cobalt, as seen from Eq. (18). Moreover, should we apply the "stabilizing procedure" recommended in [1, 5] to our domain structures which — if anything — cannot but widen the domains and enlarge the distance between domain centres, the disagreement between theory and experiment would be all the more severe.

Although it is not the purpose of the present paper to study the problem of stability of domain structures, some remarks are due in this respect in connection with the papers [1, 5]. First of all, for a honeycomb domain structure obtained with a creating field $H_0 \| [0001]$ (thicker crystals) the "stabilizing procedure" from [1, 5] would simply be useless, as the "stabilizing field" being in that case parallel to the creating field H_0 does not change the domain structure at all up to 1.5 k0e, and gradually destroys it if further increased, as can be concluded from [6]. What more, the remanent domain structure appears (at least in that case) to be pretty stable without any "stabilizing" manipulations, as seen from Fig. 6 where two powder patterns are reproduced that reveal the same remanent honeycomb domain structure at the same site on the basal surface of the same cobalt single crystal (used in the experiments reported in [18]; crystal thickness in the direction [0001] — 12 mm), except that the pattern (a) was obtained in 1964 just upon producing the domain structure while the pattern (b) was obtained a few days ago. During those four years between the observations the crystal was kept at room temperature and away from magnetic fields, to preserve the domain structure intact. As expected, there is hardly any difference at all between the two patterns.

On the other hand, in thin crystals where $H_0 \perp [0001]$ the "stabilizing field" is perpendicular to the creating field H_0. Hence, it obviously changes the remanent domain structure. As shown in [6] on a large cobalt single crystal, in that case irreversible changes may occur for field intensities as low as 500 0e. Therefore, in our opinion the so-called "stable" structures in [1, 5] are simply forcibly enlarged and irreversibly changed (thus different) honeycomb structures which, for better or worse, may or may not be stable much like the supposedly "unstable" ones. As a matter of fact, it is somewhat startling to call a domain structure unstable if it requires a field of up to 2 k0e to be converted into a stable one. It seems to be a very stable instability, indeed.

For the above stated reasons, we do not ascribe the disagreement between theory and experiment in [1] and in the present paper to any instability of the examined domain structures, the more so as a similar discrepancy is obtained for the plate-like remanent domain structure in cobalt [12]. However, it seems also unlikely that the disagreement is due to simpli-

fications in the theory, as for these particular structures the models as well as the approximations made in the calculations seem fairly justified, and in any case the disagreement is far too large to be attributed to subtle theoretical simplifications, unless there is something wrong in the very formulation of the whole problem. At any rate, at present we see no reasonable explanation of the disagreement, and we believe that more experimental evidence is needed — concerning remanent as well as non-remanent domain structures — before a satisfactory answer to this problem can be given.

Finally, we would like to draw attention to another theoretical defficiency. In [11], a formula was given for the critical crystal thickness above which dagger-like closure domains should form at the crystal surface, and this thickness was found for cobalt to be approximately 0.1 μm. Now, as seen from Fig. 5 no closure domains are formed in the honeycomb domain structure in cobalt for crystal thicknesses up to 50 μm, and as shown in [12] the same is true for the plate-like domain structure in cobalt. In this case the disagreement between theory and experiment is enormous, notwithstanding the fact that the same formula yields for magnetoplumbite a value which is (apparently incidentally) in reasonable agreement with experiment. Unfortunately, no derivation of the formula is given in [11], so that the author's reasoning in deriving it cannot be checked for possible inconsistencies or crude simplifications, which are not unlikely if only because the adaptation of the method given in [19] to domain structures with an open magnetic flux is by no means straightforward.

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REFERENCES

[1] J. Kaczér, R. Gemperle, *Czech. J. Phys.* B, **11,** 510 (1961).
[2] H. Kojima, K. Goto, *J. Phys. Soc. Japan*, **17,** 584 (1962).
[3] A. Wrzeciono, R. Gemperle, *Phys. Status Solidi*, **2,** 1384 (1962).
[4] G. S. Kandaurova, J. S. Shur, *Fiz. Metallov Metalovedenie*, **15,** 839 (1963).
[5] R. Gemperle, *Phys. Status Solidi*, **6,** 89 (1964).
[6] B. Wysłocki, *Acta Phys. Polon.*, **27,** 783, 955, 969 (1965).
[7] B. Wysłocki, W. J. Ziętek, *Acta Phys. Polon.*, **29,** 223 (1966).
[8] G. Kozłowski, W. J. Ziętek, *J. Appl. Phys.*, **36,** 2162 (1965).
[9] J. Kaczér, R. Gemperle, *Czech. J. Phys.* B, **10,** 505 (1960).
[10] B. Wysłocki, *Phys. Status Solidi*, **3,** 1333 (1963).
[11] J. Kaczér, *Zh. Eksper. Teor. Fiz.* (USSR), **46,** 1787 (1964).
[12] B. Wysłocki, *Acta Phys. Polon.*, **34,** 327 (1968); *ibid.*, in the press.
[13] W. C. Elmore, *Phys. Rev.*, **51,** 982 (1937); **53,** 757 (1938).
[14] H. J. Williams, R. M. Bozorth, W. Shockley, *Phys. Rev.*, **75,** 155 (1949).
[15] J. R. Garrood, *Proc. Phys. Soc. A*, **79,** 1252 (1962).
[16] B. Wysłocki, W. J. Ziętek, *Acta Phys. Polon.*, **21,** 433 (1962).
[17] B. Wysłocki, *Ann. Phys.* (Germany), **13,** 109 (1964).
[18] B. Wysłocki, W. J. Ziętek, *Proceedings of the International Conference on Magnetism* (7—11 September 1964, Nottingham, England), *Magnetization Processes* II, pp. 716—719.
[19] E. M. Lifshitz, *Zh. Eksper. Teor. Fiz.* (USSR), **15,** 97 (1945).