NEGATIVE MAGNETORESISTANCE OF STRONGLY ANISOTROPIC FERROMAGNETIC UAsSe

By A. Wojakowski and Z. Henkie

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

(*Received January 3, 1977*)

The transverse magnetoresistance of single crystal samples of ferromagnetic UAsSe has been measured below and above the magnetic phase transition in the temperature range 77 to 135 K, and in a magnetic field of up to 10 kOe. The magnetoresistance is negative and anisotropic over the entire experimental range, and the curves of $\Delta\varrho(H, T)$ vs T all show a deep and finite minimum at 105.5 K. A comparison of our experimental results with the theoretical predictions of Yamada and Takada is given.

1. *Introduction*

The phenomenon of negative magnetoresistance has been explained for magnetic alloys [1, 2], rare-earth metals [3], Kondo systems [4–6], and magnetic semiconductors [7]. Recently, Yamada and Takada (hereafter YT) [8, 9] have examined the magnetoresistance of ferromagnetic metals due to spin fluctuations. They have obtained the explicit expressions for the temperature and field dependence of the magnetoresistance for both the ordered and paramagnetic regions. Within the YT theory the electron system of a ferromagnetic metal in an external magnetic field consists of the conduction electrons interacting with localized spins through the $s-d$ RKKY-type interaction. A uniform magnetic field is assumed to act upon both the conduction electron and the localized spins. In this model the magnetic field has been assumed to be small enough to neglect the cyclotron motion of the conduction electrons. They have also neglected the anisotropy energy, arguing that anisotropy would not essentially change the magnetoresistance.

YT have calculated the spin correlation function in two ways. In the first treatment [8] the localized spin system was treated in the molecular field approximation (MFA). In the second one [9], the spin Green functions were obtained by a perturbation treatment of the Green functions of the electron and localized spin (the RPA method). Both the

* Address: Instytut Niskich Temperatur i Badań Strukturalnych PAN, 50-950 Wrocław, PO Box 937, Poland.

above treatments lead to qualitatively similar behaviour for the magnetoresistance. The magnetoresistance is negative, because the external magnetic field amplifies the effective field acting on the localized spins and reduces their fluctuations leading to a decrease of the resistivity. For a fixed field the magnetoresistance vs temperature curve shows a deep and finite valley at the transition temperature. In this paper the results of magnetoresistance measurements for UAsSe, as well as their comparison with some theoretical predictions of YT are presented.

The ternary uranium compound, UAsSe, crystallizes in the tetragonal system with the PbFCl-type of structure (space group *P4/nmm*) [15]. It is ferromagnetic below 113 K [10, 11]. A neutron diffraction study [10] has revealed that this compound has the simple uniaxial ferromagnetic structure with an ordered magnetic moment of $1.5 \pm 0.1\ \mu_B$, where the magnetic moment of uranium is aligned along the *c*-axis.

UAsSe exhibits a large anisotropy in the ferro- and paramagnetic states [11, 12, 14]. The anisotropy constant at 78 K was estimated to be equal to $6-10 \times 10^6$ erg/g [14]. The paramagnetic susceptibility (in the range of 140 – 300 K) in the basal plane is about one order of magnitude lower than that measured along the *c*-axis. There is also anomalous behaviour of zero field resistivity in the basal plane $\varrho_{\perp c} = 280 \pm 20\ \mu\Omega$cm at 300 K in contrast to typical simple ferromagnetic metals; the temperature coefficient of resistivity is negative in the ordered state below 45 K, and within the whole examined paramagnetic range (up to 700 K) [13]. The absolute thermoelectric power at the room temperature is about four times larger in the basal plane than that along the *c*-axis. The magnetic moment in this compound is expected to be localized.

2. *Experimental*

Sample preparatiom and zero field d.c. electrical resistivity measurements were described earlier [13]. The samples were cut from the single crystals either in the form of a rectangular slab with dimensions $8 \times 1.5 \times 0.5$ mm³ (sample b), or in the nearly cylindrical form of an 8 mm rod of 0.1 mm² cross section (sample a). The [001] axis was perpendicular to the length (the [100] axis) of both the samples, as well as to the plate surface of the b sample. The crystal quality was good as far as one could determine from a Weissenberg graph examination. Both the current and resistivity were measured along the sample length while the external magnetic field was rotated in the transverse (001) plane. The measurements were carried out by a conventional d.c. method in the temperature range 77 to 135 K and in a magnetic field of up to 10 kOe. The crystallografic orientation of the samples with respect to the magnetic field was controlled with an accuracy of 2 deg or better.

The magnetoresistance $\Delta\varrho(T, H)$ is usually given by the following relation:

$$\Delta\varrho(T, H) = \frac{1}{\varrho(\infty, 0)} [\varrho(T, H) - \varrho(T, 0)], \tag{1}$$

It is not possible, in the case of UAsSe, to determine the temperature independent (in the first Born approximation) values of $\varrho(\infty, 0)$ in the usual way because of the nonlinear decrease of the resisitivity with temperature over the whole paramagnetic region. Hence, the values of $\varrho(77,0)$ instead of $\varrho(\infty, 0)$ were applied in order to define relative magnetoresistance. Furthermore, the UAsSe shows the residual magnetoresistance, and resistivity of sample before apply of magnetic field was usually used as $\varrho(T, 0)$ value.

3. Results and discussion

Below the transition temperature, when a weak external field exists and the following inequalities hold:

$$\left(\frac{T_c - T}{T_c}\right)^3 \gg \left(\frac{\mu H}{T_c}\right)^2; \quad 1 \gg \frac{T_c - T}{T_c} \geqslant 0$$

the following relation in the MFA method is satisfied [8]:

$$\Delta\varrho(T, H) \sim \frac{H}{\sqrt{T_c - T}}. \tag{2}$$

In RPA approximation [9] the formula above is replaced by

$$\Delta\varrho(T, H) \sim \left|\ln \frac{H}{T_c - T}\right| \frac{H}{\sqrt{T_c - T}}, \tag{3}$$

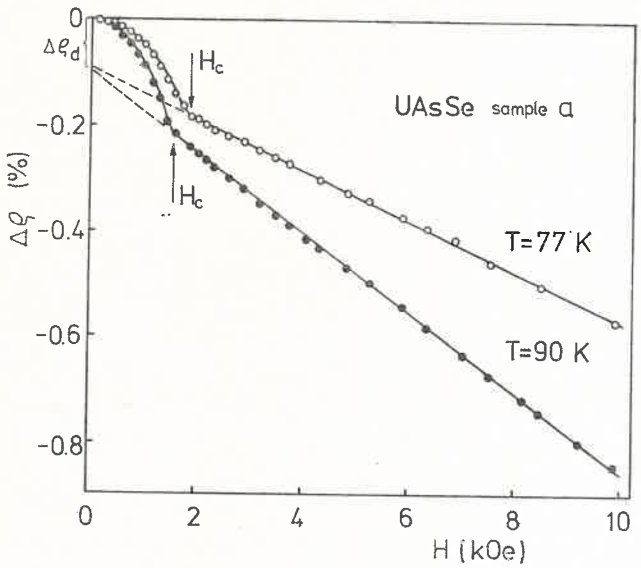

Fig. 1. Magnetic field dependence of the transverse magnetoresistance of UAsSe (sample a) at 77 and 90 K. The experimental points are averaged for two different directions of magnetic field and current

where T_c is the Curie temperature and μ is chemical potential of the conduction electrons. According to the first treatment, the magnetoresistance $\Delta\varrho$ depends linearly on the magnetic field H, whereas in the second treatment $\Delta\varrho/H$ depends linearly on $\ln H$.

The results of our transverse magnetoresistance measurements for sample a at liquid nitrogen and oxygen temperature with the magnetic field vector parallel to the magnetic easy axis [001], are presented in Fig. 1. Note that in all subsequent figures the sign "minus" indicating negative values of $\Delta\varrho$ is omitted. The experimental points are an average of four values — for two different directions of the magnetic field and the electric current. Fairly good agreement with Eq. (2) is obtained. The magnetoresistance is negative and above a certain value of magnetic field H_c, depends linearly on the field intensity.

For fields below H_c the magnetoresistance obeys approximately an H^2 law (as shown in Fig. 2). The values of H_c are in good agreement with the saturation field of the Hall

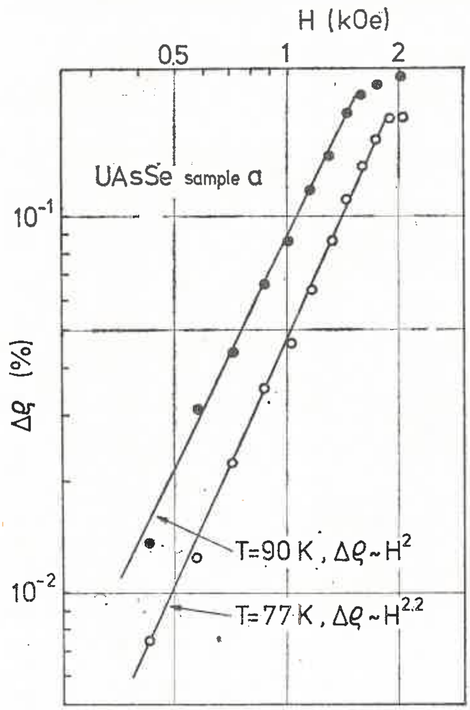

Fig 2. Variation of the magnetoresistance with magnetic field strength below H_c for sample a at 77 and 90 K

constant for UAsSe [17] when the magnetic field is parallel to the [001] direction. The magnetic measurements point out that in the direction [001] UAsSe reaches a saturation magnetization at the same values of the magnetic field H_c.

Fig. 3 shows the angular dependence of the transverse magnetoresistance for two different UAsSe samples of similar shape (a and a′) with the magnetic field vector rotating in the plane (001) perpendicular to the current direction. The transverse magnetoresistance effect is asymmetrical and strongly anisotropic reaching its maximum when the magnetic field is parallel to the magnetic easy axis [001] and minimum, being close to zero, if it is perpendicular to the [001] direction. A residual magnetoresistance is also anisotropic and shows rather a large dispersion connected with the long time need to achieve the equilibrium state (small circles on Fig. 3).

The asymmetry of the magnetoresistance is independent of the manner of cooling the sample. The samples cooled in zero magnetic field, as well as in a magnetic field, display the same asymmetry properties. The asymmetry of the magnetoresistance hysteresis loop is often connected with the magnetic texture as was found for polycrystalline nickel [18]. In the case of UAsSe the hysteresis of $\Delta\varrho(H)$ is small but the magnetoresistance is still

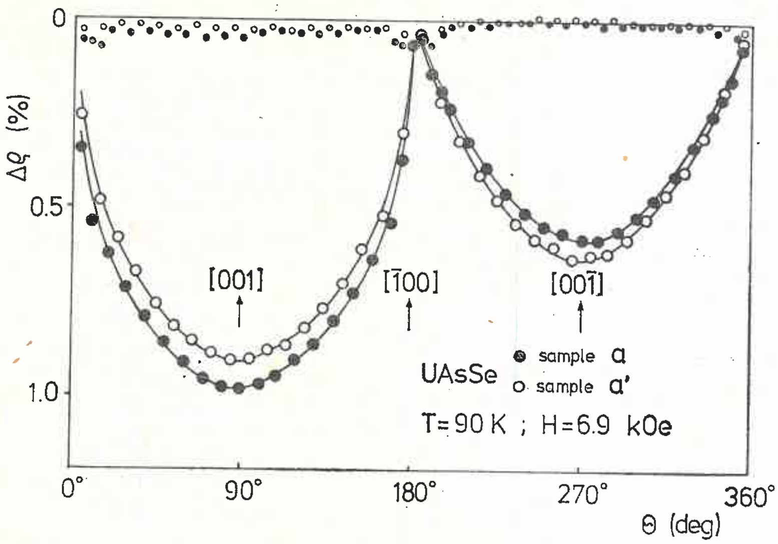

Fig. 3. Angular dependence of magnetoresistance for two different samples (a and a′) of UAsSe at 90 K in the constant field H = 6.9 kOe. θ is an angle between H and [010] direction in the (100) plane. Small circles show the residual magnetoresistance

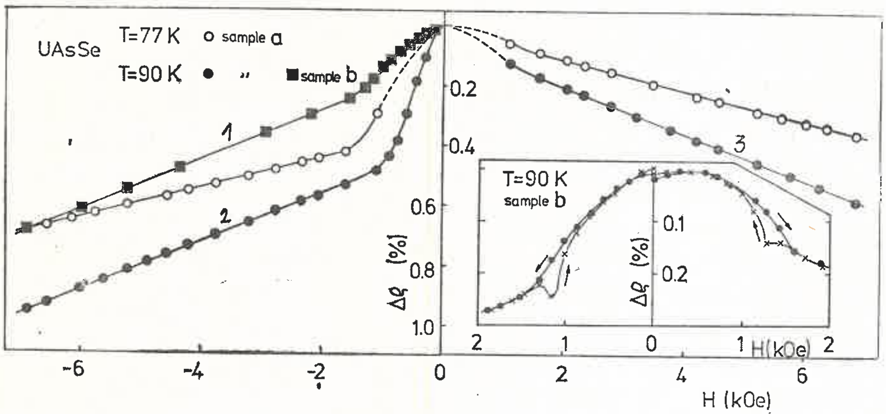

Fig. 4. Magnetoresistance isotherms at 77 and 90 K for two different samples (a and b) with increasing magnetic field for two opposite directions. Inset shows the details of the hysteresis observed for sample b.

asymmetric over the whole magnetic field range (the inset on Fig. 4 shows the hysteresis for sample b). However, the asymmetry comes from the phenomena occuring below the H_c. This can easily be seen from Fig. 4, in which the magnetoresistance isotherms for two different samples and for two opposite direction of the magnetic field are presented. The slope of the linear parts of the magnetoresistance vs magnetic field curve at a fixed temperature is identical for both samples (curves *1* and *2*) and for opposite directions of the magnetic field (curves *2* and *3*). Therefore, the magnetoresistance of UAsSe above the H_c can be expressed as

$$\Delta\varrho_c(T, H) = \chi_1 H + \Delta\varrho_d, \tag{4}$$

where $\chi_1 H = \Delta\varrho_{sf}$ is a term symmetric with respect to the direction of the magnetic field (the parameter χ_1 depends on the temperature according to the Eq. (2)). Hence, it may be concluded that above H_c, where the magnetic field increases, the observed linear decrease of the resistivity is connected with the suppresion of the spin fluctuations as shown by YT [8].

$\Delta\varrho_d$ is a term asymmetric in relation to the direction of the magnetic field, nearly proportional to H^2 for $H < H_c$, and independent of the magnetic field for $H > H_c$. It seems that appearance of the additional term $\Delta\varrho_d$ could arise from domain processes in the presence of a potential barrier which depends on structure defects. This kind of processe, e.g., the moving and rotation of the domains, was not taken into account by YT. A more detailed investigation of the magnetic properties of UAsSe at low temperatures [11] has revealed the existence of 180° compensated domains with the spin alignment along the [001] axis. Since the magnetoresistance is an even galvanomagnetic effect, the movement of 180° domains should not generate any change of the electrical conductivity [19]. Furthermore, we have found that the magnetoresistance anisotropy of UAsSe occurs in the paramagnetic region as well.

For the antiferromagnetic case the external magnetic field, applied parallel to the staggered magnetization, increases the spin fluctuations in one sublattice and simultaneously suppresses it in the second one. The behaviour of the magnetoresistance is determined by the change of the sum of the spin fluctuations of both the sublattices, and for H small, leads to a positive magnetoresistance proportional to H^2 [8].

If one assumes that the ferromagnetic domains in strongly anisotropic UAsSe behave like the sublattices in antiferromagnets, the resistivity should be conditioned by spin fluctuations in 180° domains. When the external magnetic field raises, the volume of the domains in which the magnetic field generates larger spin fluctuations, decreases and some crystal defects could be responsible for asymmetry, because for two opposite directions of the field, the volume of the domains with enlarged and reduced spin fluctuations would be different. Unfortunately, our X-ray analysis was not sufficiently accurate to detect these kinds of "small defects" in the UAsSe single crystals.

Independent of the reason of the appearance of the $\Delta\varrho_d$ term it seems that we are able to separate it by simple extrapolation of the linear part of $\Delta\varrho(H)_T$ to $H = 0$, i.e., in a similar way to that adopted for transition metals and their alloys [19]. This procedure has been employed by us for the strongly anisotropic sample a with the magnetic field direction generating the larger $\Delta\varrho_d$. We have used only two value of $\Delta\varrho(H)_T$, one at 2.95 kOe (curve *3* in Fig. 5) and a second at 9.23 kOe (curve *1* in Fig. 5), assuming that below T_c the isotherms of $\Delta\varrho(H)$ have the same shape as at 77 and 90 K. The temperature dependence of $\Delta\varrho_d$ (curve *4* in Fig. 5) and $\Delta\varrho_{sf}(H, T)$ have been calculated in this manner. $\Delta\varrho_d$ depends weakly on temperature and displays a flat minimum at 90 K. The presence of a $\Delta\varrho_d$ term, independent of the magnetic field above H_c, is responsible for the anomaly in $\Delta\varrho(T)_{H=\text{const}}$ occurring in the form of plateau (curve *3* in Fig. 5) which disappears at higher magnetic field (curve *1* in Fig. 5).

The linear character of the dependence of $\Delta\varrho_{sf}^{-2}$ vs T below T_c shown in Fig. 6 (curve *1*) indicates that the Eq. (2) is satisfied. Curve *2* in Fig. 6 shows the $\Delta\varrho_c^{-2}$ vs T dependence for the same sample but for an opposite direction of the magnetic field for which $\Delta\varrho_d$ is small and does not exceed ~3% of $\Delta\varrho_c$ at 90 K, and in a field $H = 9.23$ kOe. Thus,

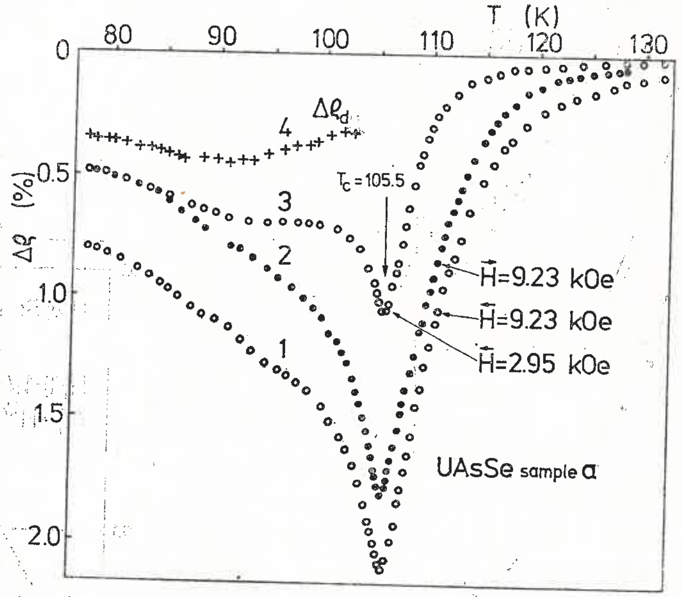

Fig. 5. Temperature variation of the transverse magnetoresistance of UAsSe (sample a) in a constant field; curve *1* — $H = 9.23$ kOe, curve *2* — $H = 9.23$ kOe (opposite direction of the field for which $\Delta\varrho_c \approx \Delta\varrho_{sf}$), curve *3* — $H = 2.95$ kOe, curve *4* — represents the variation of the asymmetric term $\Delta\varrho_d$

we can roughly assume that $(\Delta\varrho_c)_{\vec{H}} \approx (\Delta\varrho_{sf})_{\vec{H}}$. In both cases the experimental points lie on two straight lines which bend near 90 K. Curves *3* and *4* in Fig. 6 show, according to Eq. (3), the dependence of $\Delta\varrho\sqrt{T_c-T}$ vs ln $\sqrt{T_c-T}$ for the same sample. It is clearly seen that in this case a linear dependence is also approximately satisfied.

Above the Curie point, in the region where $(T_c-T)/T_c \gg 1$ and $1 \gg \mu H/T_c$ inequalities hold, in both i.e., MFA and RPA approximations we have the following relation [8, 9]:

$$\Delta\varrho_{sf}(H, T) \sim H^2/(T-T_c)^2, \qquad (5)$$

whereas in the region close to T_c, when $[(T-T_c/T_c]^3 \gg (\mu H^2/T_c)^2$ and $1 \gg (T-T_c)/T_c \geqslant 0$, the RPA treatment gives [9]:

$$\Delta\varrho_{sf}(H, T) \sim -|\ln(T-T_c)|H^2/(T-T_c)^a, \qquad (6)$$

where a is a linear combination of the critical indices.

Fig. 7 shows the magnetic field dependence of $\Delta\varrho_c$ above the transition tmperature T_c, for two opposite directions of the magnetic field vector $\vec{H}$. For both directions an exponential dependence of the type $\Delta\varrho_c \sim H^\alpha$ is observed, with α equal to 2.1 ± 0.1 when $\Delta\varrho_c \approx \Delta\varrho_{sf}$, and α equal to 1.7 ± 0.1 for the opposite magnetic field vector. The latter is distinctly lower than that appearing from Eqs (5) and (6). This points out that $\Delta\varrho_d$ is

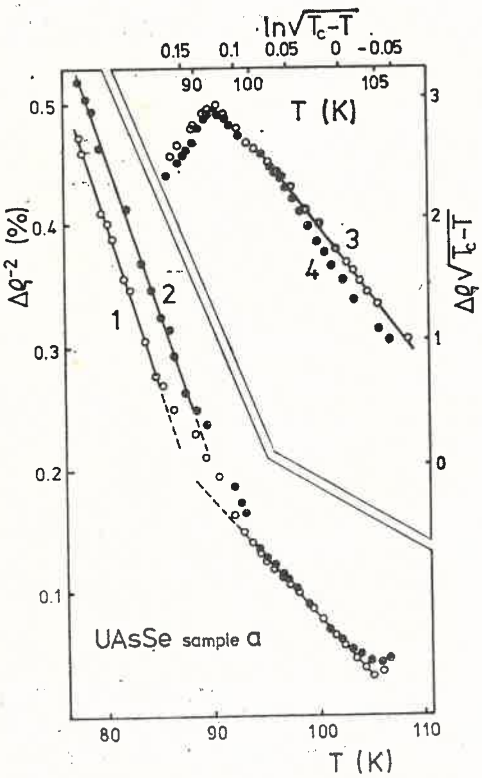

Fig. 6

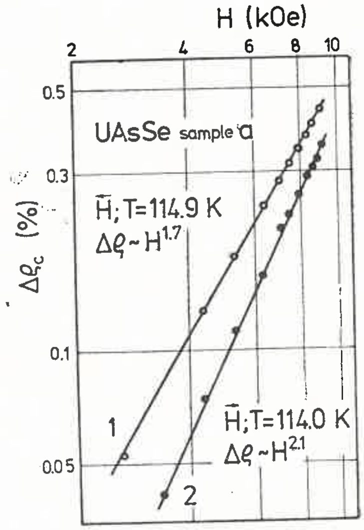

Fig. 7

Fig. 6. Plots of $\Delta\varrho_c^{-2}$ (curve *2*) and $\Delta\varrho_{sf}^{-2}$ (curve *1*) vs temperature below T_c; comparison with Eq. (2), (see text). Plots of $\Delta\varrho_c\sqrt{T_c-T}$ (curve *4*) and $\Delta\varrho_{sf}\sqrt{T_c-T}$ (curve *3*) vs ln $\sqrt{T_c-T}$ below T_c; comparison with Eq. (3), (see text)

Fig. 7. Log-log plots of $\Delta\varrho_c$ vs magnetic field strength for sample a of UAsSe above T_c for two opposite directions of the magnetic field. The slope is equal to 2.1 ± 0.1 (curve *2*) for the direction of the field for which $\Delta\varrho_c \approx \Delta\varrho_{sf}$, and is equal to 1.7 ± 0.1 (curve *1*) for the opposite direction

an odd effect even for $T > T_c$. However, in this region we can not separate the $\Delta\varrho_d$ term so unambiguously as was possible for $T < T_c$. As a consequence, the results of the temperature dependence of $\Delta\varrho(H, T)$ represented by curve *2* in Fig. 5 (i.e., when $\Delta\varrho_c \approx \approx \Delta\varrho_{sf}$) are compared with theoretical predictions.

Curve *1* in Fig. 8 shows the dependence of $\ln\Delta\varrho_c$ vs $\ln(T-T_c)$. Above $\sim$113 K the linear dependence of $\Delta\varrho_c \sim (T-T_c)^a$ is observed with a equal to 2.2 ± 0.1, somewhat

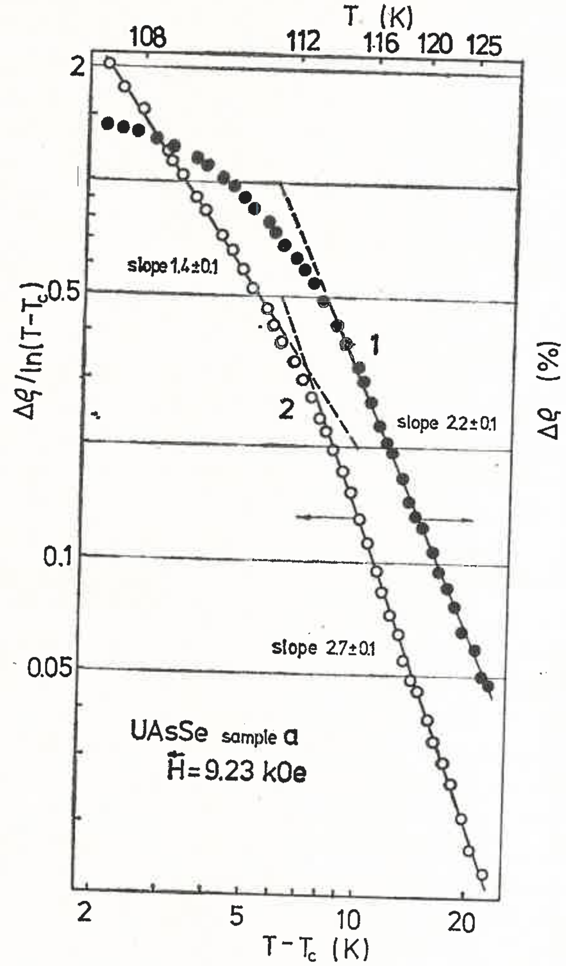

Fig. 8. Log-log plots of $\Delta\varrho_c$ and $\Delta\varrho_c/\ln(T-T_c)$ vs $(T-T_c)$ for sample a of UAsSe at a constant magnetic field of $H = 9.23$ kOe (direction for which $\Delta\varrho_c \approx \Delta\varrho_{sf}$)

higher than that resulting from the MFA approximation (Eq. (5)). The dependence of $\Delta\varrho/\ln(T-T_c)$ vs $(T-T_c)$ for the same sample is represented by curve *2* in Fig. 8, and demonstrates that the Eq. (6) is also obeyed, but with a equal to 2.7 ± 0.1. At temperatures closer to T_c the slope is almost two times lower than the latter and equal to 1.4 ± 0.1.

4. *Final remarks*

Results of the theoretical calculations by Yamada and Takada [8, 9] in the MFA and RPA approximations were obtained for an isotropic ferromagnet neglecting its domain structure. We have examined the transverse magnetoresistance for a strongly anisotropic uniaxial ferromagnetic compound, UAsSe, in which the domain structure plays an important role. If a weak magnetic field (up to 10 kOe) is perpendicular to the staggered magnetization, the resulting magnetoresistance of UAsSe is not higher than the residual magnetoresistance, amounting to about — 0.07% at 90 K, and if the field is parallel to the c axis it is of order of — 1,0% at 90 K. The temperature dependence of the total magnetoresistance manifests a qualitative agreement with the theoretical calculation by YT — in that a negative magnetoresistance displays a minimum near Tc. However, a more detailed analysis of the experimental results shows that the measured values of the magnetoresistance contain, apart from the even $\Delta\varrho_{sf}$ term (the same for different samples), a second $\Delta\varrho_d$ term appearing in the region of the magnetic field in which domain processes are observed. This latter term is asymmetric in the direction of the magnetic field and its magnitude varies from sample to sample. The anomaly in the temperature dependence of $\Delta\varrho$ observed at about 90 K (see curve *3* in Fig. 5) has the same origin as $\Delta\varrho_d$, indicating that it is connected with the domain processes. Field and temperature dependence of the $\Delta\varrho_{sf}$ term yield a general quantitative agreement with the MFA theoretical calculations. Based on the above observations we suppose that in the case of UAsSe, the $\Delta\varrho_{sf}$ term results from the suppresion of the fluctuation of spins by the magnetic field. The experimental results for the temperature dependence of UAsSe magnetoresistance agree with the RPA calculation too, whereas the predictions for magnetoresistance dependence on the external magnetic field is not observed.

As observed for rare-earth metals [20], particularly for Gd, the magnetoresistance depends on magnetic structure more distinctly than the resistivity. On the curve of $\Delta\varrho$ vs T for Gd, besides a maximum of magnetoresistance near the Curie temperature (~290 K), an additional, well pronounced maximum occurs at lower temperatures (240–250 K) corresponding to the rapid deviation of the magnetic moment from the hexagonal axis [21]. The latter effect manifests itself in the temperature dependence of the electrical resistivity as a small change of the temperature coefficient of the resistivity [22]. In the case of UAsSe the additional maximum below T_c, though considerably weaker than that for gadolinium, becomes, like for gadolinium, less and less pronounced with increasing magnetic field.

Lastly, it seems worth mentioning that a neutron diffraction study of the $UAs_{2-x}Se_x$ solid solution has shown, within a fairly broad composition range (from $x = 0.6$ to $x = 0.65$), the simultaneous existance of antiferromagnetic and ferromagnetic phases. This fact has been explained as a result of the poor homogeneity of samples [16]. A local inhomogeneity of the composition of our samples is not unlikely and may be considered as some small defect in the single crystal. This could be responsible for the effects observed by us.

It may be well to add that the minimum of the magnetoresistance at 105.5 K lies slightly below the discontinuity point of the derivative of the zero magnetic field resistivity with respect to the temperature (109 K [13]), as well as below the Curie temperature deduced from the magnetization data (113 K [11, 12]). A similar displacement was observed in the case of $FeCr_2S_4$ [23].

The authors would like to thank Professor W. Trzebiatowski for his stimulating interest throughout this study, as well as Professor Yu. P. Irkhin and Dr. A. Zygmunt for helpful discussions.

REFERENCES

[1] K. Yosida, *Phys. Rev.* **107**, 397 (1957).
[2] J. Kondo, *Progr. Theor. Phys.* **27**, 772 (1962).
[3] G. Williams, *Solid State Commun.* **8**, 1451 (1970).
[4] R. B. Stinchcombe, G. Horwitz, F. Englert, R. Brout, *Phys. Rev.* **130**, 155 (1963).
[5] M. T. Beal-Monod, R. A. Weiner, *Phys. Rev.* **170**, 552 (1968).
[6] R. Moro, H. Suhl, *Phys. Rev. Lett.* **20**, 500 (1968).
[7] C. Haas, *Phys. Rev.* **168**. 531 (1968).
[8] H. Yamada, S. Takada, *J. Phys. Soc. Jap.* **34**, 51 (1973).
[9] H. Yamada, S. Takada, *Progr. Theor. Phys.* **48**, 1828 (1972).
[10] J. Leciejewicz, A. Zygmunt, *Phys. Status Solidi* (a) **13**, 657 (1972).
[11] C. Bazan, A. Zygmunt, *Phys. Status Solidi* (a) **12**, 649 (1972).
[12] A. Zygmunt, M. Duczmal, *Phys. Status Solidi* (a) **9**, 659 (1972).
[13] A. Wojakowski. Z. Henkie, Z. Kletowski, *Phys. Status Solidi* (a) **14**, 517 (1972).
[14] K. P. Belov, A. S. Dmitrijevskij, A. Zygmunt, R. Z. Levitin, W. Trzebiatowski, *Zh. Eksp. Teor. Fiz.* **64**, 582 (1973).
[15] F. Hulliger, *J. Less-Common Met.* **16**, 113 (1968).
[16] S. Ligenza, A. Murasik, A. Zygmunt, J. Leciejewicz, *Phys. Status Solidi* (a) **16**, K171 (1973).
[17] A. Wojakowski, to be published.
[18] W. J. Zajcev, *Zh. Eksp. Teor. Fiz.* **19**, 95 (1949).
[19] K. P. Biełow, *Zjawiska w materiałach magnetycznych*, PWN, Warszawa 1962, pp. 256–314 (in Polish).
[20] K. P. Belov, R. Z. Levitin, S. A. Nikitin, *Usp. Fiz. Nauk* **82**, 449 (1964).
[21] G. Will, R. Nathans, H. A. Alperin, *J. Appl. Phys.* **35**, 1045 (1964).
[22] W. J. Nellis, S. Legvold, *J. Appl. Phys.* **40**, 2267 (1969).
[23] T. Watanabe, *J. Phys. Soc. Jap.* **34**, 1695 (1973).