GALVANOMAGNETIC EFFECTS IN THIN FILMS OF SOME TRANSITION METALS

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Electrical conductivity and Hall constants are measured in thin films of the transition metals Cr, W, Pd, and Pt deposited in a high vacuum. The results are interpreted assuming a two-band mechanism of electric current flow. Data on the electron and hole mean free paths in the films are obtained.

It is known that in thin metal films decreased thickness is accompanied by a reduction of their electrical conductivity and temperature coefficient of resistivity as well as by changes in their galvanomagnetic and optical properties. In continuous uniform films these effects are surface-induced and are observed at thicknesses that are commensurable with the mean free path of charge carriers.

The model of Fuchs and Sondheimer[1,2] has been used most successfully for films of the alkali metals, and of the noble metals to a lesser degree. In the case of the transition metals the thickness dependences are complicated by the fact that the different groups of carriers, having different mean free paths as a general rule, participate simultaneously in the electrical conductivity.

In the present work the thickness dependences of electrical resistivity and the Hall constant are investigated experimentally for films of the transition metals Cr, W, Pd, and Pt. The data have been interpreted on the basis of a two-band model, and it has been assumed that the Fuchs-Sondheimer theory can be applied independently to each of the two carrier groups (electrons and holes).

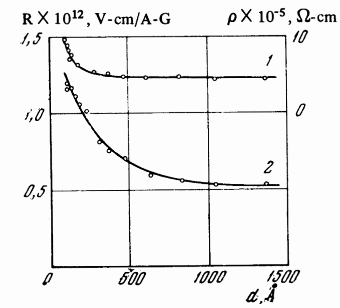

FIG. 1. Curve 1 – electrical resistivity, and curve 2 – Hall constant, as functions of Cr film thickness.

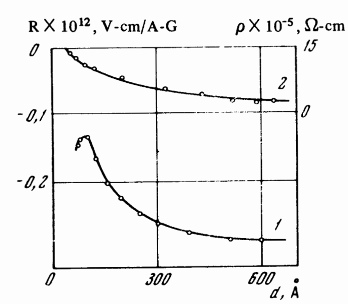

FIG. 2. Curve 1 – Hall constant, and curve 2 – electrical resistivity, as functions of Pt film thickness.

EXPERIMENT

The properties of the films were investigated for thicknesses ranging from about 50 to 1500 Å. Thermal deposition and all measurements were performed in a high vacuum ($\sim 3 \times 10^{-9}$ Torr). The construction of the experimental tube was identical with that used in[3]. Heating of the metals for Cr, W, and Pt evaporation was induced by electron bombardment, while Pd evaporated from a heated tungsten wire. The deposition rate was 2 Å/sec except in the case of Pt, for which the rate was $\sim$ 0.5 Å/min. The substrates were mica slabs that were cooled by boiling nitrogen during the course of film deposition and the measurements. Subsequent to their deposition the films were annealed at 100°C. The film thicknesses were measured by means of a piezoquartz plate[4] and by an optical method. The Hall constants were measured by a balance method in magnetic fields up to 10 kOe.

RESULTS AND DISCUSSION

It was shown by the measured temperature dependences of the resistivity that the films in the given thickness range were continuous, and did not consist of separate yet interconnected granules. The evidence for this consisted in the positive temperature coefficient of resistivity.

Figure 1 shows the basic experimental results for Cr, which exhibits a positive Hall constant R in the bulk metal. In the Cr films R is also positive and increases monotonically, like the resistivity, as the thickness d diminishes.

The results for W do not differ qualitatively from those for Cr, with the exception that for very small d a steep decrease of R is observed.

Figure 2 shows the experimental results for Pt, which has a negative Hall constant in bulk samples. We observe that R is negative for the greater thicknesses, as in a bulk sample, and that it increases as the thickness is diminished, reaching an extremal value near 100 Å.

The Hall constant of thick Pd films (which is negative in the bulk metal) is also negative, but with diminishing film thickness the constant decreases and undergoes a sign reversal at 130 Å. In all cases, as d decreases the electrical resistivity increases monotonically.

In order to compare the experimental and theoretical thickness dependences quantitatively we used the follow-

	n_1	n_2	σ_{01}/σ_{02}	l_1/l_2	l_1, Å	l_2, Å	l, Å	p_1/p_2
Cr	0.03 [6]	0.03 [6]	0.95	1.1	280	255	260	1.1
W	0.25 [7]	0.25 [7]	0.94	2.0	200	100	200	2.1
Pd	0.36 [8]	0.36 [8]	1 25	1.5	1250	850	1200	1.2
Pt	0.4 [9]	0.4 [9]	1.33	1.3	500	425	510	1.0

ing expressions for the Hall constant and the conductivity:[5]

$$R = \frac{R_1\sigma_1^2 + R_2\sigma_2^2}{(\sigma_1+\sigma_2)^2}, \quad (1)$$

$$\sigma = \sigma_1 + \sigma_2, \quad (2)$$

where σ_1, σ_2, R_1, R_2 are, respectively, the electron and hole conductivities and the Hall constants.

In the present work it has been assumed that the Fuchs-Sondheimer theory[1,2] can be applied to electrons and holes independently. Consequently σ_1, σ_2, R_1, and R_2 are taken to vary with film thickness in accordance with[1,2]: 1)

$$\sigma_1 = \sigma_{01}\Phi(k_1), \quad \sigma_2 = \sigma_{02}\Phi(k_2),$$
$$R_1 = R_{01}F(k_1), \quad R_2 = R_{02}F(k_2);$$

here $\Phi(k_{1,2})$ and $F(k_{1,2})$, which are functions representing the thickness dependences of the conductivity and Hall constant, equal unity in the bulk material (for $k_{1,2} \gg 1$); $k_1 = d/l_1$, $k_2 = d/l_2$, l_1, l_2 are the electron and hole mean free paths.

The electron and hole concentrations n_1 and n_2 used in the calculations were obtained from the literature and are given in the table. Since our test metals were compensated, the initial values $R_{01} = -1/Nn_1ec$ and $R_{02} = 1/Nn_2ec$ are equal in all cases. Here N is the concentration of metal atoms; e is the elementary charge; c is the velocity of light.) The initial values of σ_{01} and σ_{02} were determined experimentally for the thickest samples, using the equations

$$\sigma_{01} = {}^1/_2\,\sigma_0(1 - R_0Nnec), \quad (3)$$
$$\sigma_{02} = {}^1/_2\sigma_0(1 + R_0Nnec). \quad (4)$$

Here σ_0 and R_0 are the conductivity and Hall constant of the thickest films and $n = n_1 = n_2$.

$R(k_1)$ and $\sigma(k_1)$ were calculated from (1) and (2) for all the metals. Figure 3 shows a family of calculated $R(k_1)$ curves for different values of l_1/l_2. In the calculations completely diffuse carrier scattering at surfaces was assumed. The same figure also shows the experimental points of a curve in Fig. 1. The experimental R(d) curve is close to a calculated $R(k_1)$ curve, and we used the two curves to evaluate the mean free paths l_1 and l_2.

The overall appearance of the calculated $R(k_1)$ curves for W does not differ qualitatively from that shown in Fig. 3 for Cr, although we now have no detailed coincidence of the experimental and calculated curves for the thinnest films. The explanation for this probably lies in the damaged structure of very thin tungsten films.

It should be noted that for Cr and W films coincidence

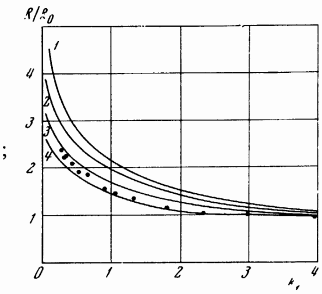

FIG. 3. Calculated $R(k_1)$ curves for Cr. 1 – l_1/l_2 = 1.4; 2 – l_1/l_2 1.3; 3 – l_1/l_2 = 1.2; 4 – l_1/l_2 = 1.1.

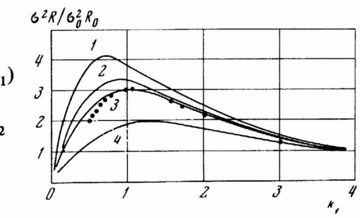

FIG. 4. Calculated $R(k_1)\sigma^2(k_1)$ curves for W. 1 – l_1/l_2 = 3.0; 2 – l_1/l_2 = 2.2; 3 – l_1/l_2 = 2.0; 4 – l_1/l_2 = 1.5

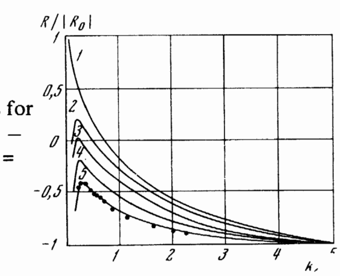

FIG. 5. Calculated $R(k_1)$ curves for Pt. 1 – l_1/l_2 = 1.7; 2 – l_1/l_2 = 1.6; 3 – l_1/l_2 = 1.5; 4 – l_1/l_2 = 1.4; 5 – l_1/l_2 = 1.3.

of the experimental and calculated curves is achieved more conveniently not directly for R(d) and $R(k_1)$ or for σ(d) and $\sigma(k_1)$, but for their products $R(d)\sigma^2(d)$ and $R(k_1)\sigma^2(k_1)$ [representing the numerator in the right-hand side of Eq. (1)]. The $R(k_1)\sigma^2(k_1)$ curve exhibits a pronounced maximum; the correspondence between the units of d and k_1 can therefore be determined more reliably. Figure 4 shows the calculated and experimental curves $R(k_1)\sigma^2(k_1)$ and $R(d)\sigma^2(d)$ for W.

Figure 5 shows a family of calculated $R(k_1)$ curves and the experimental R(d) curve for Pt. Some of the $R(k_1)$ curves exhibit a maximum that also appears on the experimental R(d) curves. A similar effect has previously been observed for Ni.[10]

Figure 5 shows that as l_1/l_2 increases the maximum of $R(k_1)$ is elevated, and that for sufficiently large values of l_1/l_2 the sign of the Hall constant may be reversed for thin films, as appears to be the case for Pd.

The table gives the electron and hole mean free paths that were determined by comparing the calculated and experimental curves; in all cases $l_1 > l_2$. The table also gives, for comparison, the values of l averaged over electrons and holes, based only on σ(d) (or, more precisely, from $d\rho(d)$, where $\rho = 1/\sigma$).

1)V. V. Vladimir brought to our attention the thickness dependences of R_1 and R_2 in accordance with [2].

As already mentioned, some of the $R(k_1)$ curves in Fig. 5 have a maximum at a small value of k_1. Figure 5 shows that an increase of the parameter l_1/l_2 within a certain narrow range leads to a qualitative change of the $R(k_1)$ curve; the maxima disappear and the curves become monotonic. It may be inferred that in this transition region of l_1/l_2 (for small k_1) the Hall constant is more sensitive to changes of l_1/l_2. This situation can exist only for particular relations among σ_{01}, σ_{02}, R_{01}, R_{02}, and l_1/l_2 and may occur by chance; therefore this is not a general characteristic of all metal films. Nevertheless, the given quantities probably exist with the required relationships for Ni films, where the Hall constant is consequently highly sensitive to external influences. Thus, it has been shown experimentally in[11] that for Ni films R can change by several tens per cent (while the electrical conductivity changes by only $\sim 0.01\%$) when their surfaces are acted upon by a high electric field or by adsorption. Under these conditions the interaction between charge carriers and the metal surface is modified, and the electron and hole mean free paths are changed.

Preliminary experiments on Pd and Pt films indicated that R is here not appreciably affected by electric fields. This result was to be expected on the basis of the foregoing discussion, because the thickness dependences of the Hall constant (shown for Pt in Fig. 5) are represented by curves that are distant from the aforementioned transition region.

We note in conclusion certain considerations that support the foregoing hypothesis of predominantly diffuse carrier reflection from the film surfaces. The data on l_1/l_2 and σ_{01}/σ_{02} can be used to determine the ratio p_1/p_2 of electron and hole average quasimomenta. The results, obtained by means of the Drude-Lorentz formula $\sigma = Nne^2 l/p$, are given in the table, where we observe that p_1/p_2 equals unity, or is somewhat larger, for Cr, Pd, and Pt, but is ~ 2 for W. These results are consistent qualitatively with conclusions reported in[6-9,12-14] concerning the Fermi surface parameters of the test metals, and furnish some degree of confirmation for the aforesaid hypothesis.

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