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LOW-FIELD HALL COEFFICIENT OF Al–4d DILUTE ALLOYS: THE ROLE OF THE ANISOTROPIC IMPURITY SCATTERING

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The role of the anisotropic impurity scattering in the determination of the low-field Hall coefficient of Al–4d dilute alloys is investigated by means of systematic theoretical calculations, as well as experimental measurements for Al–Zr and Al–Mo. The theoretical results, obtained without using any adjustable parameter, are in excellent agreement with the experimental data and a consistent interpretation of the systematic variation of the low-field Hall coefficient for aluminium-based dilute alloys with transition-metal impurities is given. © 1998 Elsevier Science Ltd. All rights reserved

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The low-field Hall coefficient, R_H^0, of dilute metallic alloys at low temperatures is a valuable tool for the investigation of the topography of the Fermi surface of the host crystal as well as of the details of the scattering mechanisms of the impurities. Although the free-electron model in the case of simple-metal hosts predicts a constant low-field Hall coefficient independent of the nature and scattering properties of the impurity, important deviations of R_H^0 have been observed for different solute atoms [1].

Detailed experimental studies of the low-field Hall coefficient in Al–3d dilute alloys at low temperatures have been reported by Papastaikoudis and co-workers [2, 3]. Their results show a strong dependence on the species of the impurity and exhibit a parabolic behaviour within the 3d series. Since many of the scattering properties of transition-metal impurities in simple metals can be explained using the virtual-bound-state model, it was speculated that the observed parabolic behaviour could be attributed to the crossing of the Fermi level by the impurity 3d virtual bound state as we move along within the 3d series. Recent theoretical calculations in Al–3d alloys [4], however, have shown that this is not quite correct. In particular, these calculations showed that there is a monotonic increase of R_H^0 versus the impurity atomic number, with an upward bending at the beginning of the 3d series. A careful analysis revealed that the bending results from a sensitive balance between the s and p scattering rather than the d resonant scattering. Therefore, one would expect the 4d impurities to behave quite differently, since the s versus p scattering strength is reduced for the early 4d impurities as compared to their 3d counterparts [5]. Unfortunately, no investigation of the galvanomagnetic properties of Al–4d alloys has been reported so far, due partly to the difficulty of the sample preparation arising from the very low solubility of the 4d atoms in aluminium which in most cases lies below 100 ppm [6].

In this work we report an experimental and theoretical investigation of the low-field Hall coefficient of aluminium containing isolated 4d impurities. The analysis of the results, in conjunction with the previous for the Al–3d alloys, elucidates the physical mechanisms that determine the behaviour of R_H^0 in these systems.

From a series of Al–4d alloys that were prepared, only Al–Zr and Al–Mo gave a residual resistivity vary-

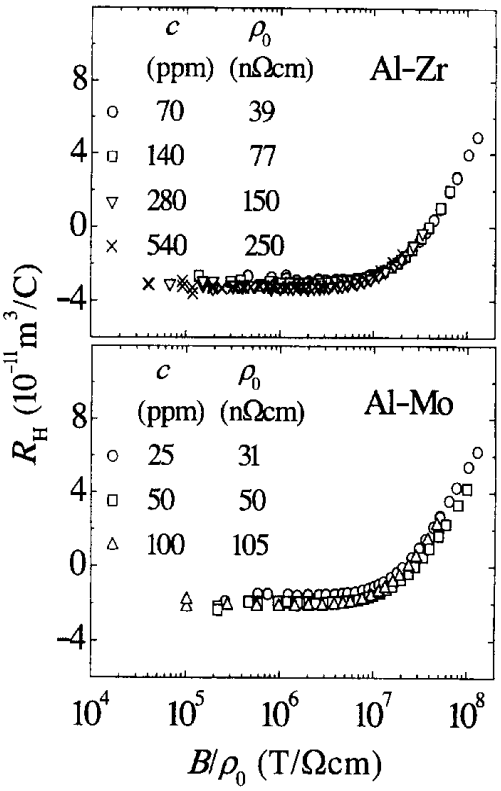

Fig. 1. Kohler diagram of the measured Hall coefficient for Al–Zr and Al–Mo dilute alloys at 4.2K.

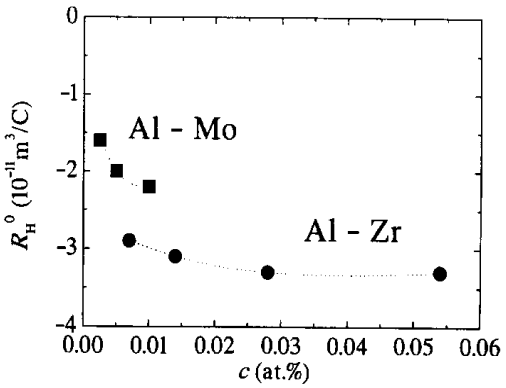

Fig. 2. The measured Hall coefficient for Al–Zr and Al–Mo dilute alloys at 4.2K as a function of the impurity concentration at a constant value of the effective magnetic field: $B/\rho_0 = 3 \times 10^6$T/Ωcm.

ing linearly with the solute concentration at 4.2K. Consequently, only these two alloys were further studied experimentally. The dilute Al–Zr and Al–Mo alloys were prepared from 99.9997% pure aluminium (VAW, Bonn, Germany), 99.99% Zr (MRC) and 99.99% Mo (MRC) by induction melting under a pure argon gas atmosphere, and then rolled into foils of about 80μm thickness. The samples were stamped out from these foils in rectangular shapes (20×2mm^2) with extensions for Hall and magnetoresistance contacts. The samples were homogenized at 560°C for 30h in a vacuum of about 10^{-5}mbar.

The measurements were carried out in a conventional stainless steel helium cryostat that contained a superconducting solenoid. The resistivity and Hall voltage were measured using a standard four-terminal technique. The dc signals were amplified by a galvanometric photocell amplifier (Sefram "Amplispot"). The final resolution was of the order of 2×10^{-9}V.

The results for the Hall coefficient, R_H, of the Al–Zr and Al–Mo alloys, at 4.2K, are shown in Fig. 1. R_H is represented in a Kohler diagram, i.e. it is plotted as a function of the effective magnetic field, B/ρ_0, where ρ_0 is the residual resistivity of the samples in a zero magnetic field. The inset tables show the values of ρ_0 of the samples for both systems. The residual resistivity at 4.2K varied linearly with the solute concentration. The values of the residual resistivity per atomic percent in the present work were 5.3$\mu\Omega$cm/at% and 10.9$\mu\Omega$cm/at% for Al–Zr and Al–Mo, respectively. These values are in good agreement with those reported by Toyoda *et al.* [6]: 5.9$\mu\Omega$cm/at% for Al–Zr and 9.5$\mu\Omega$cm/at% for Al–Mo. The experimental results reported in Fig. 1 show that the low-field Hall coefficient is practically independent of the impurity concentration.

In Fig. 2 the measured low-field Hall coefficient of both alloys at 4.2K, taken at a constant value of the effective magnetic field: $B/\rho_0 = 3\times10^6$T/Ωcm, is plotted as a function of the impurity concentration, c. It can be seen that R_H^0 tends to constant values. The initial decrease of R_H^0 for both systems may be attributed to the presence of other impurities, grain boundaries or dislocations, which are produced during the mounting and cooling of the samples.

In addition to the experimental measurements, we carried out systematic calculations of the low-field Hall coefficient for all the 4d impurities diluted in aluminium. Our theoretical method relies on the so-called on-Fermi-sphere approximation. This allows us to combine the exact topography of the Fermi surface described by the 4-OPW model with the scattering phase shifts which are obtained by self-consistent impurity-in-jellium calculations. The approach outlined above gives an all-electron description of the impurity that enables us to reliably represent the case of the deep d potentials of the 4d impurity series for which an OPW method is not suitable. Thus, weak sp as well as strong d resonant scattering are treated on the same footing. The exchange and correlation contributions to the impurity potential are dealt with in the

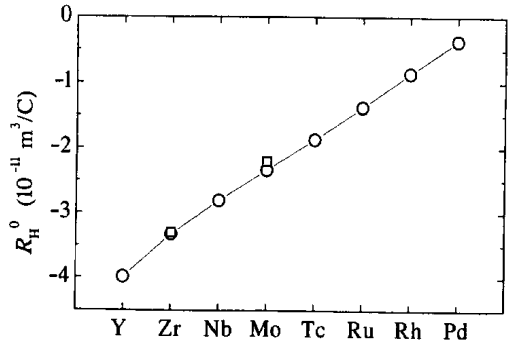

Fig. 3. The low-field Hall coefficient for 4d substitutional impurities in Al. The circles show the theoretical results and the squares the experimental data.

local-density approximation of the density-functional theory with the parametrization of Vosko *et al.* [7]. In the case of 4d impurities, where relativistic effects are non negligible, the scalar-relativistic approximation is employed for the description of the electron scattering [8]. The transport coefficients are calculated in the framework of Boltzmann transport theory. Our approach relies on the expansion of the anisotropic vector mean free path in powers of the applied magnetic field. Thus, departing from the linearized Boltzmann integro-differential equation, we end up with a hierarchy of integral equations of the Jones–Zener type, one for each power of the magnetic field. These are solved self-consistently with an iterative procedure. The coefficient of the zeroth power of the mean free path gives the residual resistivity, that of the first power the low-field Hall coefficient, that of the second power gives the magnetoresistance coefficients, etc.. In short, the solution of each integral equation is used to compute the conductivity tensor in the respective order of the magnetic field. Details about our method of calculation can be found elsewhere [9].

For a Zr impurity we calculate a residual resistivity per atomic percent equal to $8.4\mu\Omega$cm/at%, which is higher than the experimental results cited above. On the other hand, for a Mo impurity we find $10.6\mu\Omega$cm/at%, which is in very good agreement with the experimental data. Our theoretical results for the low-field Hall coefficient turn out to be independent of the direction of the magnetic field as expected for cubic crystals and, therefore, they can be directly compared with the experimental data obtained for polycrystalline samples.

In Fig. 3 we show our results for the variation of R_H^0 values within the 4d impurity series. It can be seen that the low-field Hall coefficient increases almost linearly with the impurity atomic number. There is a surprisingly excellent agreement between the theoretical results and the measured R_H^0 for Zr and Mo impurities.

A detailed analysis of the numerical results shows that R_H^0 is dependent on the fine balance between s and p scattering, especially in cases where d scattering is weak. The reason is that the aluminium Fermi surface consists of an extended free-electron-like part, S_-, of negative curvature, a small hole-like region, S_{++}, of high positive curvature, and a small region, S_{--}, of strong negative curvature. The S_{--} region has an sd character, while the S_{++} part is rather pd-like [10]. Thus, p scattering translates to hole scattering, diminishing the positive contribution of S_{++} to R_H^0, while s scattering weakens the negative contribution of S_{--}. We conclude that, as a rule of thumb, s scattering contributes positively to R_H^0 and p scattering negatively; interference effects in the scattering of the various spherical-waves components will, however, blur this simple picture.

Let us for instance consider the case of an Y impurity where the calculated phase shifts at the Fermi level: $\delta_s(E_F) = -1.28$, $\delta_p(E_F) = -0.67$, $\delta_d(E_F) = 0.67$, $\delta_f(E_F) = 0.01$, indicate that we have s dominant scattering. If we strengthen further the s scattering by hand, putting $\delta_s(E_F) = -1.40$ and -1.55 (all other phase shifts are kept fixed), we find that R_H^0 increases from its actual value ($= -3.99 \times 10^{-11}$m^3/C) to -3.73×10^{-11}m^3/C and -3.45×10^{-11}m^3/C, respectively. This explains the upward bending of the R_H^0-curve in the beginning of the 3d series [4, 9], because the s over p scattering-strength ratio is higher for the early 3d impurities as compared to their 4d counterparts [5].

Assuming scattering of only a specific symmetry, ℓ, we obtain a constant low-field Hall coefficient, independent of the value of $\delta_\ell(E_F)$. In this respect, we find that s scattering alone leads to a high positive R_H^0, while p scattering gives a strongly negative low-field Hall coefficient. The d electrons, on the other hand, are quite isotropically distributed on the Fermi surface, their scattering thus giving a total of a negative contribution, roughly equal to the free-electron value of R_H^0 for aluminium ($= -3.47 \times 10^{-11}$m^3/C). Thus, a simple superposition of the different partial-wave contributions cannot account for the variation of the low-field Hall coefficient within a given series of transition-metal impurities. Interference effects in the scattering of the various spherical waves are important, and a reliable determination of R_H^0 for dilute Al-transition metal alloys can be obtained only by taking into account the anisotropic scattering of all the electrons of the Fermi surface in a detailed transport calculation.

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