Magnetoresistance and Hall effect of the complex metal alloy Mg_2Al_3

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Unusual electronic transport properties have been found in the rhombohedral β'-phase of the complex metallic alloy Mg_2Al_3. The magnetoresistance (MR) is 2 orders of magnitude larger than in the related cubic $\beta-Mg_2Al_3$-phase and Kohler's rule is strongly violated in the β'-phase at higher temperatures. Above about 100 K the Hall coefficient R_H of the β- and β'-phases are similar and free-electron-like, while in the β'-phase, R_H changes sign with decreasing temperature at low fields. We have inquired into the sources of these transport anomalies, but have not been able to clearly identify the grounds. Several conventional mechanisms for a large magnetoresistance are discussed, and found not to be applicable. The different properties in the β- and the β'-phases are puzzling since the magnitudes of the electrical resistivities are similar and $\omega_c\tau$ (cyclotron frequency $\times$ scattering time) is equally small in both phases. The similar temperature range in which anomalies occur in the β'-phase in the resistivity, the Hall effect, and the magnetoresistance indicates an electronic transition or a change of the electron structure in this phase below about 100 K.

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I. INTRODUCTION

In crystalline compounds with large unit cells the electronic mean-free path is at all temperatures typically smaller than a few lattice constants resulting in intense scattering and electronic disorder effects in the transport properties. Usually these alloys display both a large electrical resistivity ρ as well as a strongly negative $d\rho/dT$ in a temperature range from liquid-helium temperatures up to room temperature and higher. Typical examples are several complex metallic alloys. For instance, in the rhombohedral $Al_{63}Cu_{27}Fe_{11}$-phase, which has a lattice parameter of about 30 Å and $\sim$1400 atoms/unit cell, ρ at 4.2 K is about 5000 $\mu\Omega$ cm and $d\rho/dT$ is negative and of large magnitude.[1]

In complex metallic alloys one can also find examples of surprisingly large ρ combined with a positive $d\rho/dT$. In $Al_{68}Cu_7(Fe_{1-x}Ru_x)_{17}Si_8$ for example, ρ(4 K) is in the range 500–700 $\mu\Omega$ cm and $d\rho/dT$ is positive from about 25 K to room temperature.[2] In $Cd_{85.7}Yb_{14.3}$, another complex metallic alloy, ρ is about 300 $\mu\Omega$ cm at room temperature and $d\rho/dT$ is positive in a fairly large temperature range, with ρ (100 K) of about 100 $\mu\Omega$ cm.[3] This situation can arise when the density of states at the Fermi energy is small due, e.g., to a pseudogap, and the electronic mean-free path is not too small, due to a well-ordered structure with small chemical disorder.[2]

$\beta-Mg_2Al_3$ is a metallic alloy of cubic structure with lattice parameter 28.24 Å and approximately 1168 atoms in the unit cell.[4,5] On the length scale of atomic distances the structure is characterized by different clusters such as icosahedra and Friauf polyhedra. However, the transport properties in this case resemble those of simple metallic alloys. ρ for a single crystal at 4 K is only of order 40 $\mu\Omega$ cm and $d\rho/dT$ is positive and small from He temperatures up to room temperature.[6] Neither of the two characteristic features mentioned above for complex metallic alloys is thus obeyed in this case.

Interest in Mg_2Al_3 has recently been revived by the discovery of a first-order phase transformation in the Mg_2Al_3-phase from a cubic β-phase to a rhombohedral $\beta'-Mg_2Al_3$ phase with a smaller unit cell.[5] This structure is based on clusters similar to the β-phase. The symmetry lowering of the β'-phase results from atomic rearrangements within the clusters. The β'-phase is stable at low temperatures and the transition to the β-phase strongly depends on composition. At the composition of our samples, $Mg_{38.5}Al_{61.5}$, the transition temperature is about 200 °C. At higher temperatures $\beta-Mg_2Al_3$ is stable and below $\sim$150 °C it is metastable. In the β'-phase all atomic sites are fully occupied, while in the β-phase there are partly occupied and split positions. The effect on the density is marginal however, only about 0.03%.

The physical properties of $\beta'-Mg_2Al_3$ measured so far are also similar to those found in simple metallic alloys.[7] The resistivity and temperature coefficient are close to those of the β-phase. A superconducting transition was found at 0.87 K to an apparently BSC-like superconducting state with a nodeless gap. However, the $\beta'-Mg_2Al_3$ structure lacks inversion symmetry, which is an unusual property for a superconductor. Thus the complex atomic structures of the β- and β'-phases do not seem to be reflected in these electronic properties.

In the present paper we have investigated electronic transport properties (resistivity, Hall effect, and magnetoresistivity) in the β- and β'-phases of Mg_2Al_3. In spite of the great structural similarities of the β'- and β-phases, striking differences have been found in the electronic transport properties. These differences are apparent in $\rho(T)$, in the Hall effect, and in the magnetoresistance and all develop at temperatures below about 100 K, suggesting that significant differences between the electronic band structure of the β- and the β'-phases occur in this temperature region.

After a brief description of samples and experimental techniques in Sec. II, the experimental results are presented in Sec. III. The anomalous Hall effect of the β'-phase is discussed in Sec. IV. Several commonly occurring causes of a large magnetoresistance are discussed in Sec. V and are found not to be applicable for the case of $\beta' - Mg_{38.5}Als_{61.5}$. Multiband effects are briefly discussed in Sec. VI. Such contributions cannot be completely ruled out in the complex band structure of the β'-phase. The results are summarized in Sec. VII with some concluding remarks.

II. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUES

A large single crystal of nominal composition $Mg_{38.5}Als_{61.5}$ was grown using the flux growth technique.[5] Samples for measurements were cut from the grown crystal with dimensions $\sim 1 \times 2 \times 10$ mm^3 and oriented with a crystallographic cube edge in the long direction parallel to the current.

Samples were prepared in the β- and β'-phases by suitable annealings. Samples referred to below as "as-grown" had been quickly taken out at 300 °C from the furnace retaining the β-phase.[5] Annealings were made in pure Ar gas for four days or more at a series of temperatures in the range 150–200 °C. At 170 °C, a pure β'-phase was obtained with an increasing amount of β-phase expected toward the ends of that temperature interval. The samples retain their single crystalline state during heat treatments.[5]

Electrical contacts were made with silver paint. Standard measurement techniques were used, employing dc current with four contact probes for resistivity measurements and five probes for the Hall effect measurements. Magnetoresistance and Hall effect measurements were made in a cryostat equipped with a 12 T superconducting magnet. The Hall effect at each temperature was determined from the linear slope obtained when sweeping the field to +8 T and back to zero. (Sweeps to −8 T gave the same results.) Sample-size measurements for conversion from resistance to resistivity were performed using an optical microscope. The errors in the values for ρ were estimated to be within $\pm 5\%$. For the Hall measurements the accuracy is limited by the sensitivity of our picovoltmeter. A Hall field of 10 μV/m corresponds to a measured voltage signal of 20 nV, with a scatter of data of $\pm 15\%$. The error in the magnetoresistance measurements is of order 10^{-5}.

III. EXPERIMENTAL RESULTS

Results for ρ(T) of Mg_2Al_3 in different states are shown in Fig. 1 for temperatures up to 300 K. The lower curve corresponds to a sample consisting of the β-phase. The middle curve corresponds to a sample containing a mixture of β- and β'-phases. The topmost curve was obtained on a sample after four days of annealing at 170 °C resulting in a pure β'-phase. The data of the middle curve in Fig. 1 reach down to 1.5 K where the beginning of a superconducting transition can be seen, which is in qualitative agreement with the results for the β'-phase from Ref. 7.

The resistivities depicted in Fig. 1 correspond to values previously obtained for the β'-phase[7] and for single and polygrained samples primarily in the β-phase.[6] In particular, comparison between the results of Refs. 6 and 7 and the

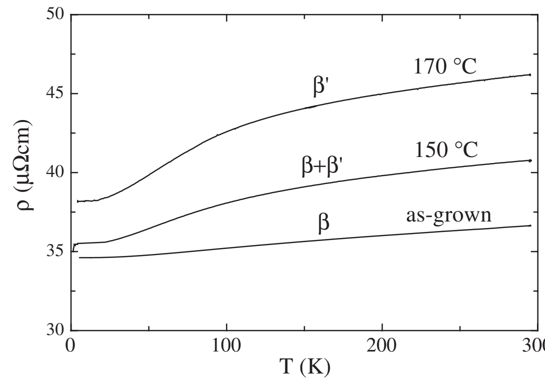

FIG. 1. ρ vs T for one sample in the β'-phase, one sample with a mixture of β and β'-phases, and one sample in a pure β-phase. For the two top curves an as-grown sample was annealed for 72 h at the temperatures indicated.

present results indicate that both ρ and the average $d\rho/dT$ up to room temperature are somewhat larger for the β'-phase than in the β-phase. The values for the resistance ratios $\rho(295\ \mathrm{K})/\rho(4.2\ \mathrm{K})$ from bottom to top in Fig. 1 are 1.06, 1.16, and 1.21, respectively. $\rho(T)$ for the β'-phase sample decreases faster with temperature in a range below about 100 K than for the other samples. The main conclusion from the results shown in Fig. 1 is, however, that considering ρ(T), the behavior of $\beta - Mg_2Al_3$ and $\beta' - Mg_2Al_3$ is similar to that of conventional simple metallic alloys.

Results from the measurements of the Hall effect are shown in Figs. 2 and 3. In Fig. 2 the Hall field E_y vs the magnetic field is shown at 295 K for two samples in the β- and β'-phases, respectively. The Hall constant $R_H = E_y/(j_x B_z)$, where j_x is the current density, is shown in the inset at a few temperatures. It is negative for both samples and the magnitude decreases slowly with increasing temperature.

Evaluating an apparent charge density, n, from $R_H = -1/ne$ with $R_H = -10^{-10}$ m$^3/C \pm 10\%$ as in Fig. 2 gives $n \approx (5-7) \cdot 10^{28}$ m^{-3}. This corresponds to free-electron estimates assuming 1 conduction electron/Al atom and 1 or 2

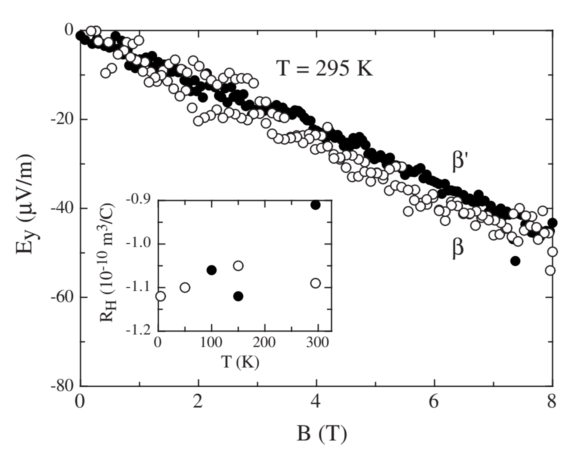

FIG. 2. Hall field E_y vs B at 295 K. •; the β'-phase sample from Fig. 1. ∘; the β-phase sample from Fig. 1. Inset: Hall constant R_H vs T for the same samples at a few temperatures. The results for the β'-phase below 100 K are shown in Fig. 3.

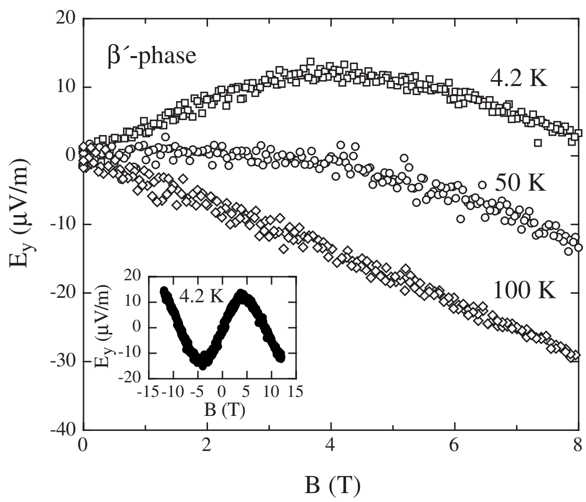

FIG. 3. Hall field E_y for the β'-phase vs B at low temperatures. Inset: Results at 4.2 K from −12 to +12 T.

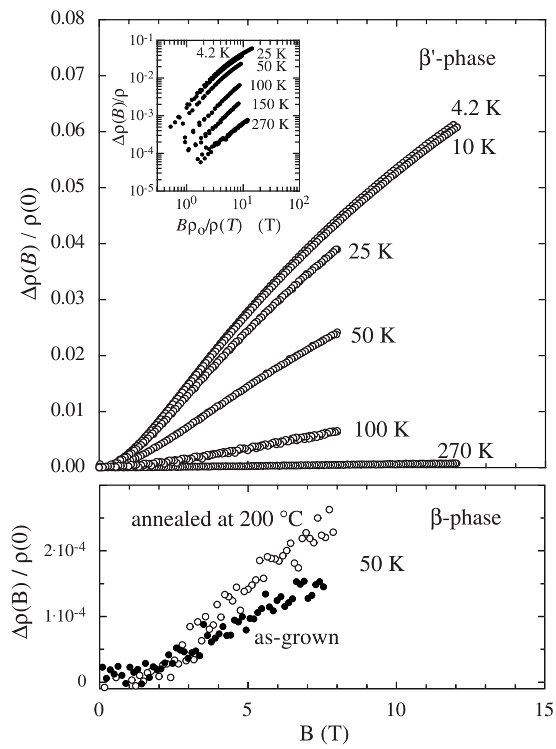

FIG. 4. Transverse magnetoresistance $\Delta\rho(B)/\rho(0)$ vs B for β'-and β-phase samples. Upper panel: Results for a β'-phase sample in the temperature range 4.2–270 K. Inset: Kohler plot; $\rho_o = \rho(295\ \mathrm{K})$. Deviations from Kohler's rule can be seen for $T \geq 25$ K. Lower panel: Two β-phase samples at 50 K: •; as-grown, ◦; annealed for ten days at 200 °C.

electrons/Mg atom giving $n=5.2$ and $7.2\times10^{28}\ \mathrm{m}^{-3}$, respectively. The reason for a reduced number of conduction electrons in Al is that in metallic Al the first Brillouin zone is filled so that there is no Fermi surface for two of the three conduction electrons.[8] We thus find that the Hall effect is similar at high temperatures in the β- and β'-phases and of a magnitude which is consistent with a free-electron model.

In the β-phase E_y is linear in B at all temperatures and the Hall coefficient remains almost constant down to the lowest temperatures. In contrast, in the β'-phase at $T<100$ K, the slope of $E_y(B)$ vs B starts to decrease and changes sign below 50 K at low fields ($B<4$ T). At 4.2 K, there is a crossover at $\sim$4 T from a holelike to an electronlike Hall effect with increasing magnetic field. This behavior is fully symmetric when sweeping the field from −12 to +12 T as shown in the inset of Fig. 3. The slopes of $E_y(B)$ vs B are almost the same in the holelike and electronlike parts of this relation, suggesting a transition with increasing magnetic field from holelike to electronlike orbits with conserved density of charge carriers.

The magnetoresistance, $\Delta\rho(B,T)/\rho(0,T)$, is shown in Fig. 4 for β'- and β-phase samples with $B\perp j$ and the sample plane. The β'-phase sample was prepared as described in Fig. 1. The β-phase was studied in one as-grown sample and in the same sample after a series of annealing steps (described in Fig. 7 below), which ended with an anneal at 200 °C for ten days, giving a similarly pure β-phase.[5]

In Fig. 4, $\Delta\rho(B,T)/\rho(0,T)$ of the β'-phase sample reaches more than 6% at 12 T and 4.2 K. Beyond an initial B^2 region, the magnetoresistance is almost linear with a tendency of beginning saturation at the highest fields. Above 50 K, $\Delta\rho(B,T)/\rho(0,T)$ decreases strongly as a function of T toward $\sim 7\times10^{-4}$ at 270 K and 12 T. In the $\beta'+\beta$-phase sample, $\Delta\rho(B,T)/\rho(0,T)$ (not shown) reaches almost 3% at 12 T and 4 K with a similar strong decrease in amplitude for increasing temperatures above 50 K.

In contrast, for the β-phase at 50 K, $\Delta\rho(B)/\rho(0)$ is about 2 orders of magnitude smaller than in the β'-phase. This difference is surprising recalling the similar results, characteristic for simple alloys, which have been observed in the electrical resistivity and resistance ratios of both phases.

Kohler's rule for the magnetoresistance states that $\Delta\rho(B,T)/\rho(0,T)$ should scale as $B/\rho(0,T)$;

$$\frac{\Delta\rho(B,T)}{\rho(0,T)} = f\left[\frac{B\rho_o}{\rho(0,T)}\right], \tag{1}$$

where ρ_o is a normalization constant, here taken as ρ at 295 K, and f is an arbitrary function. In the low-field limit one usually finds $f(x)\sim x^2$. This rule is frequently obeyed for conventional metals since in semiclassical transport theory, the magnetoresistance depends only on the product τB, where the relaxation time $\tau\sim 1/\rho$. In the β'-phase, the strong temperature dependence of $\Delta\rho(B,T)/\rho(0,T)$ above 25 K in Fig. 4 in combination with the small variation of $\rho(T)$ from Fig. 1 immediately suggests that Kohler's rule is violated. The inset of Fig. 4 shows a Kohler plot of the magnetoresistance. At low temperatures, data coalesce and Kohler's rule is obeyed. Deviations are observable at 25 K and become more pronounced with increasing temperature.

IV. ANOMALOUS HALL EFFECT

The phenomenology of the Hall effect can often be quite complex also in simple alloys and pure elements with sign changes of R_H as a function of temperature, magnetic field, or both.[9] For example, in Cd, the Hall resistivity, ρ_H [$=E_y/j_x$] at 2 K is negative and displays a minimum at about

1.2 T, with successively increased importance of hole contributions for further increasing fields. At 4 K, however, ρ_H(4 K) was positive at all measuring fields.[10]

One explanation of this phenomenon is tunneling between different sheets of the Fermi surface separated by a small gap, E_g. Spin-orbit splitting or accidental degeneracies at points close to the Fermi surface can induce small gaps, so that the cyclotron frequency ω_c [$=eB/m*$] exceeds $E_g/\hbar$ at moderate magnetic fields.[11] In Mg, gap energies were estimated to be in the range 10^{-5} to 10^{-3} eV.[12] This effect has been termed magnetic breakdown. In this process, the nature of an orbit changes under the influence of a magnetic field, e.g, from a holelike to an electronlike contribution. The probability for such processes increases with increasing magnetic field. Alternatively, intersheet scattering has also been considered.[13] Such processes also involve tunneling between different sheets of the Fermi surface. For intersheet scattering the probability for switching bands decreases with increasing magnetic field.[14]

For $\beta'-Mg_2Al_3$, the change of charge carrier type in the Hall effect as a function of increasing magnetic field at low temperatures and the disappearance of this effect at a fairly low temperature of ~50 K, qualitatively support that such band transitions may account for the observations. However, conditions for magnetic breakdown processes are that $\hbar\omega_c > E_g > k_BT$, where the first inequality assures that a band transition can occur and the second one that the gap is not swamped by the thermal energy. For our data in Fig. 3 these estimates imply that a gap of a few tenths of a meV, consistent with the order of magnitude for a gap arising from spin-orbit splitting, could still be operative at 4 K and 4 T, the position of the maximum of E_y in Fig. 3. The thermal energy is then ≈0.35 meV, and a free-electron estimate of $\hbar\omega_c \approx 0.46$ meV. These conditions for magnetic breakdown are fragile. A conjecture of magnetic breakdown is therefore somewhat questionable.

The band structure of $\beta'-Mg_2Al_3$ or $\beta-Mg_2Al_3$ is not known and a firm confirmation of magnetic breakdown or intersheet scattering in the β'-phase is presently out of reach. In order to make quantitative predictions, models of the scattering processes may also be necessary.[13]

V. SOURCES OF LARGE $\Delta\rho(B,T)/\rho(0,T)$

Several frequently occurring sources of contributions to the magnetoresistance are superconducting fluctuations, electronic disorder effects, Fermi surface shape effects, and extrinsic effects. Superconducting fluctuations can be straightforwardly ruled out. Although this effect can give significant and usually positive contributions to the magnetoresistance in a temperature range extending up to a few times the transition temperature T_c, they fall off rapidly for increasing temperature. To safely rule out such contributions, discussion of subsequent results is mainly limited to $T \geq 10$ K, corresponding to T at least $>6 \cdot T_c$. The electron-phonon interaction can affect transport properties at higher temperatures. However, the differences in ρ and $d\rho dT$ (above 200 K) between the samples in Fig. 1 are insignificant on the scale of the differences in the magnetoresistance (Fig. 4), and cannot account for the differences between β- and β'-phase samples.

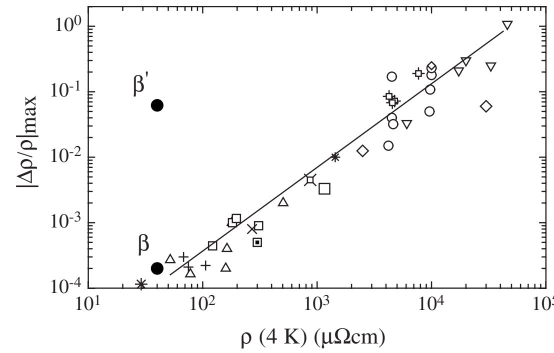

FIG. 5. Maximum magnetoresistance, $|\Delta\rho/\rho|^{max}$ vs ρ(4 K) for several metallic alloys including crystalline CuGe and complex metallic alloys, such as amorphous metals, decagonal and icosahedral quasicrystals and crystalline approximants (Ref. 15). This $\Delta\rho/\rho$ arises in disordered electron systems from weak localization and enhanced electron-electron interactions. The present alloys are depicted by solid circles. For the β-phase, $|\Delta\rho/\rho|^{max}$ is consistent with electronic disorder effects. For the β'-phase, it is more than 2 orders of magnitude larger.

A. Electronic disorder effects

In electronically disordered metals, quantum corrections to the Boltzmann conductivity occur when electron scattering is intense enough that scattered electron waves can interfere during the time of preserved phase coherence. A large resistivity with frequent elastic scattering events leads to large quantum corrections. Usually these contributions also have a significant temperature and magnetic field dependence with magnitudes which increase with magnetic field at most laboratory fields and increase with decreasing temperature.

An empirical relation has been found between the maximum of the observed $|\Delta\rho(B,T)/\rho(0,T)|$ and ρ(4.2 K) which extends over 4 orders of magnitude in $|\Delta\rho/\rho|$.[15] The maximum $|\Delta\rho/\rho|$ for each alloy system was estimated from the largest value reported for that alloy system. This relation is shown in Fig. 5. The data include results from a crystalline low resistivity alloy (fcc CuGe), and different complex metallic alloys.

The straight line corresponds to a universal curve approximately summarizing the data. This relation provides a convenient tool to find the order of magnitude of the magnetoresistance for a given level of electronic disorder, roughly estimated by ρ(4 K). The present data are given by the full black circles at ~40 $\mu\Omega$ cm. For the β phase, $\Delta\rho(B)/\rho(0)$ corresponds to the electronic disorder contribution for the resistivity of this phase. For the β'-phase electronic disorder is clearly an inadequate explanation.

B. Fermi surface shape effects and anisotropy

With the cyclotron frequency in a magnetic field $\omega_c = eB/m^*$ and scattering time τ, the number of revolutions without scattering is $\approx\omega_c\tau$. At high magnetic fields, $\omega_c\tau \geq 1$,

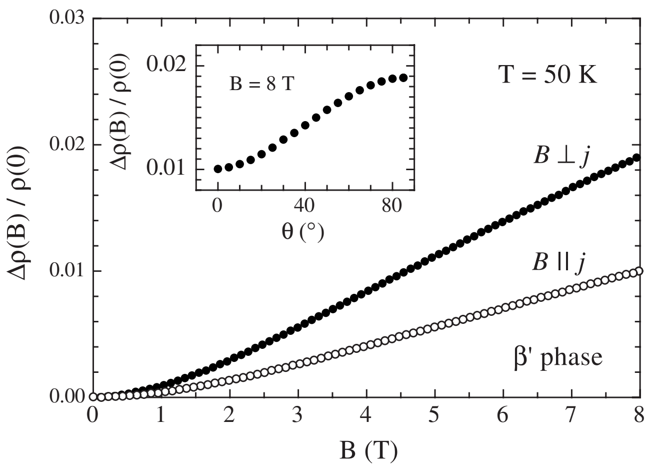

FIG. 6. $\Delta\rho(B)/\rho(0)$ of a β' sample at 50 K as a function of B. Transverse and longitudinal magnetoresistances are shown. Inset: Magnetoresistance for the same sample at 8 T and 50 K as a function of angle between **j** and **B**. At $\theta=0^o$, $\mathbf{B}\|\mathbf{j}$.

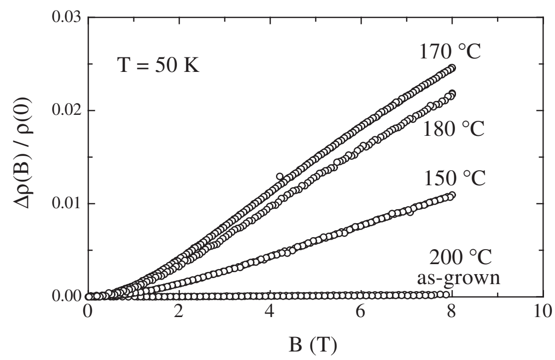

FIG. 7. $\Delta\rho(B)/\rho$ (0) vs B at 50 K for one sample, starting from the β-phase (as-grown) and after annealing for several days at the temperatures indicated and in the sequence of increasing temperatures. The two β-phase samples in Fig. 4 have here collapsed onto one curve at the bottom of the figure.

there can then be a significant effect on the magnetoresistance. For low fields, $\omega_c\tau<1$, the effect on electron motion is small.

Assuming a free-electron-like formalism we can estimate $\omega_c\tau$ from $\rho=m^*/ne^2\tau$ and $R_H=-1/ne$. Substituting for $\tau/m*$ in $\omega_c\tau$ gives

$$\omega_c\tau=\frac{B|R_H|}{\rho}. \tag{2}$$

$R_H\approx-10^{-10}$ m^3/C for the β-phase at all measuring temperatures and for the β'-phase above 100 K. At 4.2 K we have $\rho(4\ \mathrm{K})\approx40$ $\mu\Omega$ cm from Fig. 1. One then finds

$$\omega_c\tau(4\ \mathrm{K})\approx2.5\times10^{-4}B, \tag{3}$$

with B in Tesla. This result can be taken as an upper limit for the β'-phase below 100 K. The local slopes of the curves in Fig. 3 are always smaller or at most equal to the slope of the straight line through the data at $T=100$ K. Both the β'- and β-phase samples are thus in the extreme low-field limit at all laboratory magnetic fields. Furthermore, the temperature dependence of $\omega_c\tau$ is small. Although the increase with temperature of ρ and the decrease of $|R_H|$ add to a correction in Eq. (2), the total change of the coefficient in Eq. (3) can be estimated to be within a 40% decrease up to room temperature. The samples therefore remain in the low-field limit at all fields and temperatures studied.

The Hall angle arctan $(\omega_c\tau)$ between **j** and resulting electric field **E** consequently also has a small temperature dependence. At 10 T and 4 K one has arctan $0.003\approx0.7°$, illustrating that electron trajectories are almost parallel to **E**. This suggests that Fermi surface shape effects are absent.

To further study anisotropy effects, the β'-phase sample was rotated in magnetic field. Rotation was performed around an axis perpendicular to both **j** and **B**. Some electron paths on the Fermi surface can then be rotated into a position $\perp$ **B** when the angle θ between **j** and **B** is varied. The result is shown in Fig. 6.

If Fermi surface shape effects are operative, one may observe an oscillatory behavior of $\Delta\rho(B)/\rho(0)$ as a function of B. In the absence of other effects, one would expect the same values of $\Delta\rho(B)/\rho(0)$ at $\theta=0$ and 90° since these states are equivalent. However, in this mode of rotation there is an additional contribution to the magnetoresistance, which increases with increasing θ as illustrated in the inset of Fig. 7. The curve form is sinusoidal, suggesting that the origin of this anisotropy is a Lorentz force $-e\mathbf{v}\mathbf{x}\mathbf{B}$, successively turned on when θ increases from 0. The result thus suggests that in this case almost 50% of the magnetoresistance arises due to a bending effect on electron trajectories.

The importance of a Lorentz force was confirmed by rotating the sample around an axis approximately parallel to **j** and the sample length. In this geometry the Lorentz force does not vary significantly with rotation angle. $\Delta\rho(B)/\rho(0)$ was now found to be almost constant.

In conventional studies of the magnetoresistance for pure metals and alloys,[16] much larger values of $\Delta\rho/\rho$ than the present ones can be observed. However, present values of $\omega_c\tau$ are limited to small values making studies inaccessible in the region $\omega_c\tau>$ or $\gg0.1$, where the magnetoresistance of pure metals is usually studied. Results for pure Al and dilute Al alloys by Snodgrass and coworkers[8] illustrate this point. For decreasing $\omega_c\tau$ in Al, $\Delta\rho/\rho$ decreased to 3% at $\omega_c\tau\approx0.1$, while from Fig. 4 and Eq. (3) a corresponding value of $\Delta\rho/\rho$ is attained at $\omega_c\tau\approx0.0015$ at 25 K and at somewhat smaller values at lower temperatures. This emphasizes that the most challenging problem with the present magnetoresistance is not its magnitude but the fact that it is observed at such a small $\omega_c\tau$.

C. Extrinsic effects

Impurities at a level escaping detection also in careful standard analyses can cause large contributions to the magnetoresistance. In fact, the long-standing problem why the magnetoresistance in amorphous alloys is often larger than expected from quantum corrections[17] could be due to precipitation of minute quantities of an impurity phase.[18] An example is amorphous $Al_{90}La_{10}$ of resistivity comparable to the present samples. When a sample with anomalously large magnetoresistance was investigated by high-resolution x-ray diffraction and in an analytical transmission electron microscope, only a homogeneous amorphous phase was observed. However, in high-resolution electron microscopy, the presence of small amounts of crystalline $Al_{11}La_3$ was detected, which could be associated with the anomalous magnetoresistance.[18]

The possibility of an impurity dominated magnetoresistance in $\beta'-Mg_2Al_3$ was investigated in the following way: A sample in as-grown state, i.e., consisting of pure β-phase, was annealed for several days at the subsequent temperatures of 150, 170, 180, and 200 °C, and the magnetoresistance at 50 K was measured up to 8 T after each annealing step. We found that $\Delta\rho/\rho$ has a maximum at $\approx 170°$, in the β'-phase, and decreased with further increase in the annealing temperature. The two curves for the β-phase in Fig. 4 coincide on the scale of Fig. 7.

These results show that in a single sample one can monitor the magnetoresistance from its large value in the β'-phase to the low value of electronic disorder magnetoresistance of the β-phase by a series of low-temperature annealings. Dissolution or precipitation of impurity phases do not occur at these temperatures.[5] Hence impurities can be ruled out as a cause for the large magnetoresistance.

The same argument is applicable also to other extrinsic effects. An anomalous linear magnetoresistance extending up to high fields has frequently been observed, e.g., in some pure metals,[16] and the origin has been a subject of controversies. Suggested explanations for this phenomenon include voids[19] and thickness variations over the sample length.[20] Neither of these models is consistent with the results in Fig. 7. We conclude that the magnetoresistance observed in β'-phase samples is an intrinsic effect of this phase.

VI. MULTIBAND MODELS

Multiband effects may considerably change simple model results for the Hall effect and the magnetoresistance. An extension of a free-electron model to two electron bands qualitatively changes the nature of the magnetoresistance, and a nonzero $\Delta\rho(B)/\rho(0)$ is now obtained in free-electron-like models. Such models can apply also to metals with partly filled bands in different Brillouin zones, occurring, as mentioned, in pure Al where the first Brillouin zone is filled, leaving the last electron to be shared between a holelike Fermi surface in the second zone and an electronlike Fermi surface in the third zone. Already in free-electron-based derivations of multiband expressions, the Hall effect and the magnetoresistance depend on a number of unknown parameters, such as electron densities, effective masses, and conductivities in each subband. A range of different behaviors can be obtained by fitting to such models. However, in the present case, a significant simplification can be obtained at $T > 100$ K, where $\Delta\rho(B)/\rho(0)$ of the β'-phase is still anomalously large (Fig. 4) and the Hall effect is free-electron-like and close to the value $-1/ne$ (Fig. 2).

Using $R_H = -1/ne$ in a standard expression for the two-band Hall effect[21] one has

$$R_H = \frac{R_1\sigma_1^2 + R_2\sigma_2^2}{(\sigma_1+\sigma_2)^2} = -\frac{1}{ne}. \tag{4}$$

R_ν and σ_ν, $\nu = 1,2$, are the Hall constants and the conductivities of the subbands, respectively, and the charge densities obey $n = n_1 + n_2$. The last equality of Eq. (4) is the present result. It requires that $\tau/m*$, and hence also $\omega_c\tau$, are equal in the two subbands. This simplifies the magnetoresistance for a two-band free-electron model to

$$\frac{\Delta\rho(B)}{\rho(0)} = \frac{1+(\omega_c\tau)^2}{1+(\omega_c\tau)^2(1-c)^2/(1+c)^2} - 1, \tag{5}$$

where $c = n_1/n_2$ is the charge compensation. Equation (5) was calculated as a function of $\omega_c\tau$ for a range of c values[16] showing that the magnetoresistance grows initially as B^2, which for small and nonzero $1-c$ is followed by an extended region where $\Delta\rho(B)/\rho(0)$ is linear in B (before saturation). Although this curve form resembles the observations for the β'-phase of Mg_2Al_3 (Fig. 4), $\omega_c\tau$ of the present samples is small enough that Eq. (5) remains in the B^2 region for all accessible values of B. From Eq. (3) the present samples reach $\omega_c\tau = 10^{-3}$ for $B \approx 4$ T and, hence, $\Delta\rho(B)/\rho(0)$ from Eq. (5) is at most 10^{-6}. This is more than an order of magnitude smaller than the observations at 4 T for the β-phase (Fig. 4) and more than 2 orders of magnitude smaller than $\Delta\rho(B)/\rho(0)$ for a β'-phase sample at 100 K and 4 T (Fig. 4).

A simple two-band model is thus inapplicable. However, the complications of the real structure are formidable. The β'-phase has about 352 Mg atoms and 528 Al atoms in the rhombohedral cell and an irreducible cell is 1/18 of the crystallographic cell.[5] This implies about 130 electrons in the irreducible cell and ~65 electron bands to consider. The large number of parameters of such expressions for the Hall effect and the magnetoresistance could be expected to describe a wide range of different behaviors. Lacking independent information on band parameters, fitting to such models usually gives limited physical insight.

VII. DISCUSSION AND CONCLUSIONS

We summarize and discuss the main findings. $\rho(T)$ of the β- and β'-phases of Mg_2Al_3 is similar to the behavior found for simple metallic alloys. $\rho(T)$ is of order 40 $\mu\Omega$ cm and resistance ratios $\rho(296\text{ K})/\rho(4.2\text{ K})$ are 1.2 or smaller, with slightly smaller values for both quantities in the β-phase than in the β'-phase. The Hall coefficient $R_H(T)$ shows a free-electron-like behavior above 100 K in both phases. $R_H(T)$ has a similarly weak temperature dependence in both phases and its magnitude corresponds to a free-electron result with 1 conduction electron/Al atom and 1–2 electrons/Mg atom. For $T < 100$ K, the Hall effect of the β'-phase changes sign and displays a maximum as a function of magnetic field at 4 K.

The magnetoresistance is strikingly different in the two phases with values of $\Delta\rho(B)/\rho(0)$ of order 10^{-4} in the β-phase and several percent in the β'-phase. Only the result for the β-phase is consistent with quantum corrections in a weakly disordered metal. Evidence has been presented that the large magnetoresistance of the β'-phase is an intrinsic property. The strong temperature dependence of $\Delta\rho(B)/\rho(0)$

and the small temperature dependence of $\rho(T)$ lead to strong violations of Kohler's rule at $T \geq 50$ K. Breakdown of Kohler's rule has been observed previously in alloys of roughly comparable resistivities, and at temperatures above 100 K, e.g., in the normal state magnetoresistance of some high-temperature superconductors.[22] In this case, the violation of Kohler's rule was traced to the unusual temperature dependence of the Hall angle. This explanation does not apply to $\beta'-Mg_2Al_3$ with its small temperature dependence of the Hall angle.

$\omega_c\tau$ is small in these alloys and our measurements in the temperature range 4–300 K in magnetic fields up to 12 T are well within the low-field region. Yet, $\Delta\rho(B)/\rho(0)$ in the β'-phase increases as B^2 only up to a few Tesla at low T and then follows an almost linear relation up to high fields. An extended linear magnetoresistance is, as mentioned, a subject of dispute. Two suggestions for this effect were briefly mentioned in Sec. V C and were found not to be applicable in the present case. The linear magnetoresistance can be quantified by the Kohler slope, which is a dimensionless number obtained from the linear slope of the magnetoresistance when plotted vs $\omega_c\tau$. This number is usually in the range 0.001–0.1.[23] The largest value which we are aware of is a result quoted by Pippard[16] for polycrystalline Cu of $\sim$0.5. For the present results we can use Eq. (3) to rescale the abscissa in Fig. 4 to $\omega_c\tau$. In the region of a linear $\Delta\rho(B)/\rho(0)$ the Kohler slope is then found to decrease from about 25 to 3 when temperature increases from 10 to 100 K. On the other hand if data for the β-phase in Fig. 5 are approximated by straight lines above 2 T the Kohler slopes are roughly 0.10 and 0.07 and thus fall in the range of conventional values. These observations reinforce the remarkable differences between the β- and β'-phases.

It is interesting to note that transport anomalies of $\beta'-Mg_2Al_3$ have been observed at similar temperatures in all studied transport properties; At about 50–100 K, $\rho(T)$ in Fig. 1 displays a knee, E_y starts to deviate from the free-electron behavior at higher temperatures (Fig. 3), and $\Delta\rho(B)/\rho(0)$ grows rapidly with decreasing temperature (Fig. 4). This suggests a common origin, such as a transition or modification of the electron band structure in the β'-phase, giving the experimental conditions which allow these anomalies to be observed.

Can band splittings leading to magnetic breakdown and intersheet scattering explain these observations in $\beta'-Mg_2Al_3$? Models have been developed, which treat anomalies in the Hall effect and the magnetoresistance, and the breakdown of Kohler's rule on a common ground of intersheet scattering or magnetic breakdown.[9,14,24] Comparing such results with $\beta'-Mg_2Al_3$, one finds on the one hand that model calculations, based on intersheet scattering in dilute CdZn alloys,[14] have given results for both $\Delta\rho(B)/\rho(0)$ and R_H vs B of curve forms which may resemble the results for $\beta'-Mg_2Al_3$, although at much larger $\omega_c\tau$ and $\Delta\rho(B)/\rho(0)$ in the magnetoresistance. On the other hand, in the present case Kohler's rule is obeyed at low temperatures where $\Delta\rho(B)/\rho(0)$ is the largest and the magnetoresistance does not saturate, as often expected in magnetic breakdown.[24] Lack of knowledge of the band structure does not allow us to select a particular mechanism for the observed results. We therefore conclude, more generally, that the similar temperature range for the anomalies in $\rho(T)$, R_H, and $\Delta\rho(B)/\rho(0)$ suggests common or related mechanisms, such as a transition or modification of the electron band structure of the β'-phase below about 100 K.

An interesting and unresolved problem is the striking difference between the two phases in the magnetoresistance and the Hall effect, and how this occurs in the β'-phase which is deformed only by a 0.8% shrinking of the cube cell diagonal of the β-phase. Studies of the Hall effect and the magnetoresistance of the β- and β'-phases of Mg_2Al_3 raise a number of challenging problems about the relation between structure and properties of these compounds.

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