Linear magnetoresistivity in the ternary $AM_2B_2$ and $A_3RhB_6$ phases ($A =$ Ca, Sr; $M =$ Rh, Ir)

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Abstract

We studied the magnetoresistivity of the $AM_2B_2$ and $A_3RhB_6$ ($A =$ Ca, Sr; $M =$ Rh, Ir) within the ranges $1.8 \leq T \leq 300$ K and $0 \leq H \leq 50$ kOe. The zero-field resistivity, $\rho_0(T)$, is metallic and follows closely the Debye-Grueneisen description. A positive, non-saturating, and dominantly linear-in-$H$ magnetoresistivity was observed in all samples including the ones with a superconducting ground state. Such $\Delta \rho(H)/\rho(0)$, reaching 1200% in favorable cases, was found to be much stronger for the $AM_2B_2$ compounds and to decrease with temperature as well as when Ca is replaced by Sr or Rh is replaced by Ir. Finally, the general features of the observed magnetoresistivity will be discussed in terms of the Abrikosov model for the linear magnetoresistivity in inhomogeneous materials.

I. INTRODUCTION

The recent reports of a positive, extraordinarily high, linear magnetoresistivity (LMR) in nonmagnetic semimetals and semiconductors have attracted much attention. Extensive efforts were directed toward the identification of the involved mechanism(s) as well as toward material optimization for eventual technological applications such as high-density data storage or magnetic sensors and actuators. LMR was observed over wide ranges of temperatures ($\sim $ mK $\leq T \leq 400$ K) and magnetic fields ($\sim $ Oe $\leq H \leq 600$ kOe) and in a variety of materials such as elemental metals, intermetallic compounds ($A_2 \leftrightarrow B_2 \leftrightarrow A_3B_6$, e.g. InSb, Te, Bi), and graphene. Classically, a field dependent normal-ized magnetoresistivity $\Delta \rho(H)/\rho(0) = (\rho(H) - \rho(0))/\rho(0)$ is quadratic in $H$ for $\mu_eH/e < 1$ and saturates for $\mu_eH/e > 1$ (carrier mobility $\mu_e = e\tau/m^*$; symbols have their usual meaning). Various scenarios, with some classical and others quantum mechanical, were proposed for the interpretation of the deviation of LMR from the classical prediction.

The so-called Kapitza’s LMR is expected in metals, such as Bi, with an open Fermi surface and a massless free path which is longer than the electronic Larmor radius. Another scenario discusses the inhomogeneous conducting media, such as InSb semiconductor above 200 K: here disorder causes an intermixing of the off-diagonal components of the magnetoresistance MR tensor and, as such, the associated LMR is due to the distribution in $\mu_e$, rather than $\mu_e$ itself.

Abrikosov identified three classes of materials wherein quantum LMR can be manifested. First are those homogeneous materials at very low $T$ and strong $H$ and a low concentration of charge carriers ($n_c$) and a small $m^*$ such that only the lowest Landau level is populated. Its strength is given by $N_dH/(\pi n_c^2 e^2)$ where $N_d$ is the concentration of the defect centers. Second are those highly inhomogeneous materials, e.g. $A_2X$ ($X =$ Se, Te), wherein metallic inclusions (with higher $n_c$) are dispersed within a matrix having a smaller $n_c$, a linear dispersion relation and a vanishing energy gap. Third are those layered structures such as $LaSb_2$ which – due to a particular configuration of their electronic structure – exhibit a large Fermi surface (with a classical MR contribution) and, in addition, tiny pockets with a small effective mass (thus providing a quantum LMR contribution). In this case, depending on $T$, $H$, and the material properties, LMR may dominate the magnetoresistive feature.

In this work, we report on the observation of a relatively strong LMR effect in the homologous $A_nM_{3n-1}B_{2n}$ series ($A =$Ca, Sr; $M =$Rh, Ir, $n = 1, 3$). Because the evolution of LMR depends on material parameters such as $n_c$, $\mu_e$ and anisotropy, the investigation of MR in different $A_nM_{3n-1}B_{2n}$ members (each with its distinct materials properties) would be helpful in identifying the essential parameters behind the surge of LMR in this series. In fact, the following three reasons highlight our interest in studying the functional dependence of LMR in these intermetallics. First, their structure consists of a combination of alternatively stacked $AM_2B_2$ and $AM_3B_2$ sheets (see Fig. 1). As such, a variation in $n$ (e.g. $1 \leftrightarrow 3$) entails a variation in the number of involved layers and, as a consequence, a variation in the electronic properties. Second, a variation of $A$ (Ca $\leftrightarrow$ Sr) or $M$ (Rh $\leftrightarrow$ Ir) entails also a possible variation in $n_c$, $\mu_e$, or chemical pressure.
Third, because both Sr$_3$Rh$_8$B$_6$ and Ca$_3$Rh$_8$B$_6$ are superconductors while AM$_2$B$_2$ are normal\cite{12} and, furthermore, because all members show LMR effect, it is interesting to investigate the correlation, if there is any, between the electronic ground state (whether superconducting or normal) and their LMR properties.

\begin{align}
\rho_0(T) - \rho_{00} &= 16\pi^2 \omega_D \frac{\lambda}{\omega_p^2} \left( \frac{2T}{\theta_D} \right)^5 \int_0 \frac{x^5}{\sinh(x)^2} dx \quad (1)
\end{align}

where $\lambda$ is the electron-phonon coupling, $\omega_p$ is the Drude plasma frequency and $\omega_D$ is the Debye phonon frequency. Below, only $\theta_D$ and $\lambda/\omega_p^2$ are treated as free parameters.

Low-T \( \rho_T(H)/\rho_T(0) \) isotherms exhibit a strong linear-in-$H$ feature for $H > H_{cr} \approx 10$ kOe ($H_{cr}$ is the crossover field above which the LMR character dominates): thus for $H > H_{cr}$ and $T < 100$ K:

\begin{align}
\Delta \rho_T(H)/\rho_T(0) &= a_0 + a_T H
\end{align}

where $a_T = \left( \frac{\pi \omega_D}{\lambda \omega_p^2} \right)^{\frac{5}{2}}$, depends on $T$ and the material properties. Plots of $\Delta \rho_T(H)/\rho_T(0)$ isotherms against $H/\rho_0$ indicate that the Kohler rule is not satisfied. The thermal evolution of $\Delta \rho_{50K}(T)/\rho_0(T) = (\rho_{50K}(T) - \rho_0(T))/\rho_0(T)$ was found to follow the empirical relation:

\begin{align}
\Delta \rho_{50K}(T)/\rho_0(T) &= b_{1H} \tanh(c_{1H}/T)/(d_{1H} T^2 + 1) \quad (3)
\end{align}

where the phonon contribution was assumed to be $H$-independent\cite{12} and $b_{1H}$, $c_{1H}$ and $d_{1H}$ are sample-dependent parameters that will be used below only for comparative purposes. Above 100 K, this expression (and its LMR character) was found to be extremely small suggesting an energy scale of $\sim 9$ meV. Finally, for $T \to \infty$, $\Delta \rho_{50K}(T)/\rho_0(T) \to T^{-3}$, on the other hand, if both Eqs\cite{22} and \cite{3} hold as $T \to 0$ K, then $b_{1H} \to a_0 + a_T H$: this establishes a link to available theoretical models.

\section*{III. RESULTS}

\subsection*{A. Ca$_n$Rh$_{3n-1}$B$_{2n}$ (n =1,3) and CaIr$_2$B$_2$}

Figure 2(a) emphasizes the metallic character of the zero-field $\rho_0(T)$ curve of Ca$_2$Rh$_2$B$_2$: it follows the BG description (see Eq. 1 and Table 1) emphasizing the dominant strength of the phonon-electron interaction. In addition, Fig. 2 in particular the insets, manifests a relatively strong $\Delta \rho/\rho_0$, in which $\Delta \rho_{50K}(T)/\rho_0(T)$ is relatively strong (>100%) at low-T but decreases sharply with temperature (below 4% for temperatures above 100 K). The residual resistivity ratio, $\text{RRR} = \rho(300K)/\rho(1.8K)$, was found to be $\sim 6 - 32$. $\rho(T, H = 0)$ was considered to be a sum of a residual contribution $\rho_{00}$ and a Bloch–Grüneisen (BG) expression\cite{32}.
FIG. 2: (Color online) $\rho(H,T)$ curves of CaRh$_2$B$_2$. (a) Isofield $\rho_0(T)$ curves at $H=0$ and 50 kOe (solid line, $H=0$), is a fit to Eq. 1. Inset (Upper-left): Isofield $\rho_0(T)$ curves at $H=0$, 5, 10, 20, 30, 40, 50 kOe. Inset (Lower-right): Thermal evolution of $\Delta\rho_{50kOe}(T)/\rho_0(T)$ (solid line represents Eq. 3). (b) Longitudinal $\Delta\rho(T)/\rho_0(0)$ isotherms (solid lines are fits to Eq. 2).

FIG. 3: (Color online) Thermal evolution of (a) longitudinal $\Delta\rho_{50kOe}(T)/\rho_0(T)$ (solid lines are fits to Eq. 3), (b) $\alpha_T$ (based on fit to Eq. 2), and (c) the measured zero-field $\rho_0(T)$. For the three panels, the anomalous features of $A_3$Rh$_3$B$_6$ ($A=Ca$, Sr) at the lowest temperatures are related to the onset of superconductivity.

FIG. 4: (Color online) $\rho(H,T)$ curves of Ca$_4$Rh$_8$B$_6$. (a) $\rho_0(T)$ curves at $H=0$ and 50 kOe (solid line, $H=0$), is a fit to Eq. 1. Inset (upper-left): thermal evolution of isofield $\rho_0(T)$ curves at $H=0$, .., 50 kOe. Inset (lower-right): thermal evolution of $\Delta\rho_{50kOe}(T)/\rho_0(T)$ curves (triangles are transversal; stars are longitudinal; solid lines are fits to Eq. 3). (b) Longitudinal $\Delta\rho(T)/\rho(0)$ isotherms (solid lines are fits to Eq. 2). Inset: A semilog plot of the $\Delta\rho(T)/\rho(0)$ isotherms at 2 and 10 K; filled (open) symbols represent transversal (longitudinal) arrangement.

K) following approximately Eq. 3 (see inset of Fig. 2(a), Fig. 3(a) and Table I).

Similar conclusions were drawn from the analysis of various $\rho_T(H)$ isotherms, where all $\Delta\rho_T(H)/\rho_T(0)$ isotherms of Fig. 2(b) manifest a positive MR with a positive curvature and a predominant high-$H$ LMR character. Fitting $\Delta\rho_T(H > 10kOe)/\rho_T(0)$ to Eq. 2 gave the parameter plotted in Fig. 3(b) which, once more, emphasizes the strong $T$-dependence of $\Delta\rho_T(H)/\rho_T(0)$.

In contrast to CaRh$_2$B$_2$, low-$T$ $\rho(H,T)$ of Ca$_4$Rh$_8$B$_6$ (Fig. 3) show a superconducting state below $T_c \approx 4$ K and, surprisingly, the resistivity within the superconducting phase does not completely vanish indicating an absence of percolation. Above $T_c$, the normal metallic state follows a BG description (Fig. 3(a) and Table I) however, for temperatures above 250K, there is a weak deviation, away from Eq. 1. A sizable $\Delta\rho_T(H)/\rho_T(0)$ is evident in most curves of Fig. 4 in particular, Fig. 4(b)
shows that $\rho_{T<T_c}(H > H_{c2})$ manifests a negative curve while $\rho_{T>T_c}(H)$ manifests a positive and almost linear evolution. Fitting these curves to Eq. 2 yielded $\alpha_T$, the thermal evolution of which is plotted in Fig. 4. The normalized $\Delta\rho_{1.8K}(H)/\rho_{1.8K}(0)$ reaches, at 50 kOe, an impressive value of 1200% (see inset of Fig. 4(b)); this is attributed to the presence of the superconducting state (much higher value would be attained if $\rho_T(0)$ is decreased further).

Figure 5 of CaIr$_2$B$_2$ reflects the same features that were observed in CaRh$_2$B$_2$: a metallic $\rho_T(0)$ with a BG character (Table I), a predominant LMR feature and a relatively strong $\Delta\rho_{50kOe}(T)/\rho(0)$ effect at low $T$ but decays rapidly at higher $T$, dropping to below 3% above 100 K. The $\alpha_T$ parameter (the fit of $\rho_T(T)$ to Eq. 2, $\alpha_T=0$, .., 50 kOe. Inset (upper-left): Thermal evolution of isofield $\rho_T(T)$ curves at $H=0$, .., 50 kOe. Inset (lower-right): The $\rho(0)$ isotherms (solid lines are fits to Eq. 2). (b) The $\Delta\rho_T(H)/\rho_T(0)$ isotherms (solid lines are fits to Eq. 2).

**B. Sr$_x$Rh$_{3n-1}$B$_{2n}$ (n=1,3) and SrIr$_2$B$_2$**

$\rho(T,H)$ curves of SrRh$_2$B$_2$ (Fig. 6) manifest magnetoresistive features that are very similar to those found in CaRh$_2$B$_2$ except that the strength of the effect is smaller and there is a weak superconducting secondary phase (namely Sr$_3$Rh$_5$B$_6$) which is believed to be behind the drop in the magnetoresistivity of SrRh$_2$B$_2$ below that of SrIr$_2$B$_2$ (compare Figs. 3 and 6). On the other hand, Fig. 7 shows that Sr$_3$Rh$_5$B$_6$ superconducts below $T_c \approx 3.5$ K, exhibits a BG-type resistivity above $T_c$ and has MR features that are very similar to, but almost two orders of magnitude weaker than, those of Ca$_3$Rh$_5$B$_6$.

Similar to the cases found in CaRh$_2$B$_2$ isomorphs, $\rho(T,H)$ of SrIr$_2$B$_2$ (Fig. 6) show all the features that we mentioned above: the metallic resistivity obeying a BG description, the predominantly LMR character and the strong $T$-dependence of $\Delta\rho_{50kOe}(T)/\rho(0)$ up to 100 K. From a fit of $\Delta\rho_T(H)/\rho_T(0)$ to Eq. 2, we obtained $\alpha_T$ (given in Fig. 6).

**IV. DISCUSSION AND CONCLUSIONS**

Our experiments indicated that $\Delta\rho_T(H)/\rho_T(0)$ of $A_nM_{3n-1}B_{2n}$ is positive, non-saturating and dominantly
linear above ~10 kOe, and that a relatively strong
\( \Delta \rho(T)/\rho_0(T) \) was observed in both superconducting
and normal members, which decreases sharply with tem-
perature and whenever \( n_c \) is increased, Rh is replaced by
Ir, or Ca is replaced by Sr. These features as well as other
magneto-resistive properties of the studied members were
compared in Fig. 3 and Table 1. Evidently the Ca-based
isomorphs exhibit a higher RRR, a higher \( \lambda/\omega_p \) and a
higher \( \Delta \rho(T)/\rho_0(T) \).

It is recalled that a reduction in the LMR is usually
related to an increase in \( n_c, \mu_c \), or a decrease in \( N_d \) (for
a quantum LMR, a smearing of the Landau levels). In turn,
the variation in any of \( n_c, \mu_c \), or \( N_d (\rho_0) \) can be
straightforwardly associated with a related variation
in \( T, H \) or the material properties. Along this line of
arguments, we discuss the above-mentioned MR features
of \( A_nM_{3n-1}B_{2n} \) series.

First, the drop in \( \Delta \rho/\rho_0 \) with increasing \( n \) is attributed
to an increase in \( n_c \). Because the structure of \( A_3M_3B_6 \),
in contrast to \( AM_2B_2 \), includes additional, sandwiched
\( AM_2B_2 \) layers (Fig. 1), it is inferred that the intro-
duction of \( AM_2B_2 \) enhances \( n_c \). In fact, rewriting the
\( n=3 \) member as \( A_1M_8/3B_2 \) already suggests that this
enhancement is due to a contribution from the \( 4d^85s^1 \)-subbands
of the extra Rh. Such a higher \( n_c \) is consistent with
the surge of superconductivity in the \( n=3 \) members.
Second, the fact that the resistivity within the supercon-
ducting state of \( A_3RhB_6 \) does not vanish is an indication

FIG. 8: (Color online) \( \rho(H, T) \) curves of \( A nIr_2B_2 \), (a) Isofield
\( \rho_\| (T) \) curves at \( H=0 \) and 50 kOe (solid line, \( H=0 \), is a fit to
Eq. 1). Inset (upper-left): Thermal evolution of isofield
\( \rho_\| (T) \) curves at \( H=0 \), .., 50 kOe. Inset (lower-right):
\( \Delta \rho(T)/\rho_0(T) \) curve (solid line is a fit to Eq. 2). (b) \( \Delta \rho(H)/\rho_0(0) \) isotherms (solid lines are fits to Eq. 3).

that these \( n=3 \) samples contain superconducting regions
dispersed within a nonsuperconducting matrix. This feature
excludes the applicability of the classical LMR models;
rather it supports the Abrikosov LMR scenario for
inhomogeneous media.

By generalizing this inhomogeneous configuration to
the \( n=1 \) members and assuming the variation in the
LMR effect to be related to a corresponding variation
in either \( n_c \) or carrier dynamics (influenced by pressure,
charge doping, \( T \), or \( H \)), the above-mentioned experi-
mental results can be satisfactorily explained. As an ex-
ample, the fact that \( \Delta \rho/\rho_0 \) of Sr-based compounds
are lower than their Ca-based isomorphs is attributed to
a negative chemical pressure which is induced by the sub-
stitution of isovalent, relatively large-sized Sr into the
Ca-site. Similarly, the reduction of \( \Delta \rho/\rho_0 \) caused by the
replacement of Rh by Ir \((5d^55s^2)\) is attributed to
an increase in \( n_c \) that overwhelms the influence of an
increased antisymmetric spin-orbit interaction. It is re-
called that the space group of \( AM_2B_2 \) is \( Fdd2 \) (having
no inversion symmetry operator) while the space group
of \( A_3M_3B_6 \) is \( Fmmn \) (with an inversion symmetry op-
erator). Accordingly, the antisymmetric spin-orbit in-
teraction in the former series would exercise a consid-
erable influence (via a spin splitting of the quasi-particle states)
on the electronic properties. According to Abrikosov, a
linear spectrum may arise due to an absence of a sym-
metry inversion centre. Because a linear spectrum im-
plies a smaller effective mass, the absence of a symme-
try inversion would enhance the quantum LMR of the
\( n=1 \) members. Finally, the thermal rate of decrease of
\( \Delta \rho(T)/\rho_0(0) \) in \( A_3M_{3n-1}B_{2n} \) is much faster than that
of, say, \( A_{2/3}X \) \((X=Se, Te)\) (Ref. 8) but similar to that
of \( LaSb_2 \) (Ref. 25): as \( n_c \) hardly varies below 300 K,
this thermal decrease is attributed to the phonon-driven
decline in \( \mu_c \) and a smearing of the Landau levels.

In summary, a positive, nonsaturating and domi-
nantly linear MR was observed in the \( A_nM_{3n-1}B_{2n} \) series
\((A=Ca, Sr; M=Rh, Ir, n=1, 3)\). This effect was found
to decrease whenever \( n \) is increased, Ca is replaced by
Sr, Rh is replaced by Ir, or the temperature is raised.
Comparative MR studies among the different members
suggest that LMR can be described by the Abrikosov
model for inhomogeneous media.

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