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Hall sign reversal and electrical transport properties of single crystalline $SrPd_2Ge_2$

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In this study, Hall effects, magnetoresistance, and low temperature resistivity of single crystalline $SrPd_2Ge_2$ were measured. The sign change of the Hall coefficient from negative to positive with increasing temperature in the normal state and the violation of Kohler's rule in magnetoresistance data were observed. These results suggest that electrons and holes would contribute together to the formation of multi bands and that the scattering on the Fermi surface is anisotropic, which should be taken into account for the transport properties of $SrPd_2Ge_2$. In addition, we estimated the upper critical field at $T=0$ K, $H_{c2}(0)=6.1$ kOe, from the temperature-dependent electrical resistivity measurement down to 0.35 K. Including this, we discuss the nature of $SrPd_2Ge_2$ single crystals. © *2011 American Institute of Physics*. [doi:10.1063/1.3549579]

A $ThCr_2Si_2$-type structure has been considered as a mother structure for the discovery of noble superconductors with a high superconducting transition temperature (T_c), because relatively high T_c has been reported in pnictide compounds.[1] Although there has been intensive research recently on superconductivity in pnictide, correlations between superconductivity and the compounds' magnetic and electronic properties are still under investigation. In particular, a significant change in the Hall coefficient with increasing temperature has occurred around the spin density wave (SDW) or the structural transition temperature in several $ThCr_2Si_2$-type pnictide compounds, such as $CaFe_2As_2$,[2] $SrFe_2As_2$,[3] $EuFe_2As_2$,[4] and Cr doped $BaFe_2As_2$.[5] However, interestingly, $EuRh_2As_2$ (Ref. 6) and Ru over-doped $BaFe_2As_2$ (Ref. 7) showed similar behavior even without the significant aspect of SDW or structural transition. Because most of the compounds contain a strong magnetic element, Fe, it would be interesting to investigate the phenomena in a $ThCr_2Si_2$-type structured compound without a magnetic contribution. It is necessary to find compounds that have this type of structure but which contain elements other than Fe or Ni. Recently, superconductivity at 3.04 K in a polycrystalline sample of $SrPd_2Ge_2$ was reported,[8] and this compound would be a suitable candidate for studying the nature of superconductivity and the anomalous properties of the normal state in $ThCr_2Si_2$-type structures without magnetic interference. In this article, we report the transport properties, including the Hall sign reversal, magnetoresistance (MR), and low temperature resistivity, of $SrPd_2Ge_2$ single crystals.

Platelike single crystals of $SrPd_2Ge_2$ were grown by flux method using PdGe as the self-flux; we describe the details elsewhere.[9] We collected well grown, shiny, platelike crystals of $SrPd_2Ge_2$ for this study, with dimensions of $\sim 3.68 \times 2.26 \times 0.14$ mm^3 for the Hall measurement, $\sim 2.73 \times 0.97 \times 0.38$ mm^3 for the MR measurement, and $\sim 2.89 \times 0.50 \times 0.17$ mm^3, which was identical to the measurement of the sample for resistivity down to 2 K in Ref. 9, for the low temperature electrical resistivity measurement. The Hall resistivity of the $SrPd_2Ge_2$ single crystals was measured using an LR700 AC resistance bridge, combined with a temperature control system of a superconducting quantum interference device magnetometer (SQUID, Quantum Design MPMS XL). Epotek H20E silver epoxy was painted on the crystal surface, which itself is perpendicular to the crystallographic *c*-axis, to enable electrical contact with Pt wire. Low temperature resistivity measurements under the various magnetic fields were performed down to 0.35 K with a Heliox ^{3}He refrigerator (Oxford Instruments Ltd.) using a conventional low-frequency AC lock-in method.

The Hall coefficient (R_H) was calculated from the transverse voltage as a function of the magnetic field at a fixed temperature. In order to eliminate the effect of misalignment of the Hall voltage electrodes, the Hall measurements were performed for two opposite directions of applied field, and the value of $\rho_{xy}(H)=[\rho_{xy}(+H)-\rho_{xy}(-H)]/2$ was taken as the Hall resistivity. Figure 1(a) shows the magnetic field dependence of the Hall resistivity (ρ_{xy}) at various temperatures from 5 K to 300 K. The slope of ρ_{xy} in terms of the magnetic field was positive at $T=300$ K, and the absolute value of the slope decreased monotonically down to $T=150$ K. Below $T=100$ K, the slope changed from positive to negative and decreased with decreasing temperature. R_H was calculated from the slope at a specific temperature, i.e., $R_H=\Delta\rho_{xy}(H)/\Delta H$. A positive R_H value indicates that the major carriers are holes at high temperature, and a negative R_H value in a low temperature region indicates that the electrons are major carriers. MR was determined as $\Delta\rho_{xx}(H)/$

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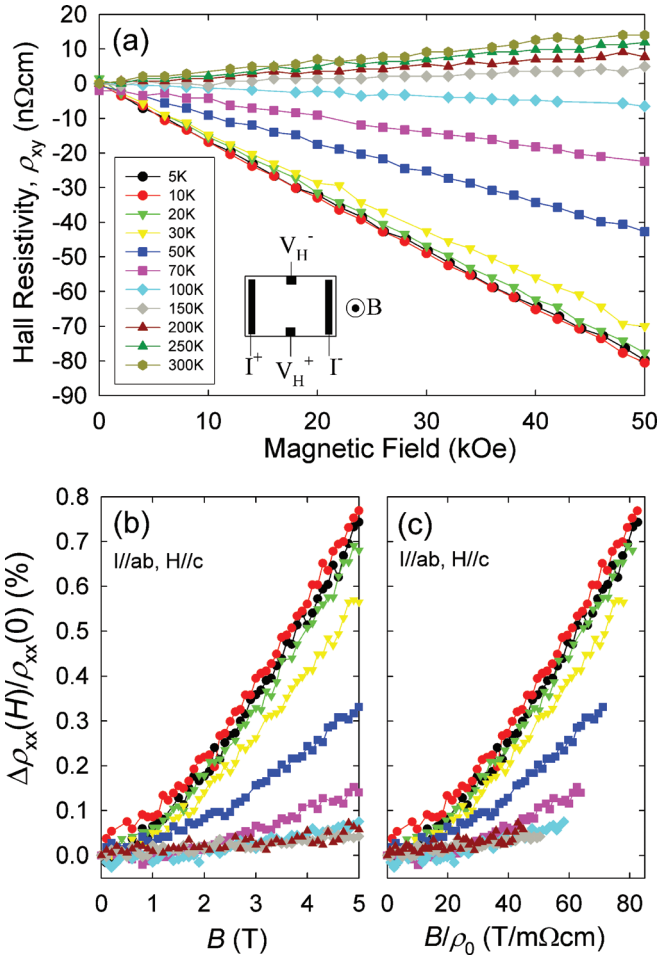

FIG. 1. (Color online) (a) Hall resistivity, ρ_{xy}, in the *ab*-plane with magnetic field along the *c*-axis at various temperatures. The schematic contact configuration for the Hall measurement is shown in the inset. (b) Field-dependent magnetoresistance, $\Delta\rho_{xx}(H)/\rho_{xx}(0)$. (c) Kohler's plot of the MR data in (b).

$\rho_{xx}(0) = [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$, where $\rho_{xx}(H)$ and $\rho_{xx}(0)$ are the longitudinal resistivity at a fixed field and a zero magnetic field, respectively. MR is shown in Fig. 1(b), which depicts positive MR over a whole range of temperatures. However, as is shown in Fig. 1(c), the MR does not follow Kohler's rule, $\Delta\rho_o/\rho_o = f(B/\rho_o)$, where $f(x)$ is a universal function for each material if there is a single band, which contributes to transport. The violation of Kohler's rule means that the scattering on the Fermi surface is anisotropic. Based on both the sign change of the Hall coefficient and the MR in violation of Kohler's rule, it is likely that the formation of multi bands, which is anisotropic, is involved in the transport of $SrPd_2Ge_2$. If we adopt a simple model with two bands, the Hall resistivity can be written as $\rho_{xy} = (1/e)\{[n_h\mu_h^2 - n_e\mu_e^2] + (\mu_h\mu_e)^2 (n_h - n_e) H^2]/[(n_h\mu_h + n_e\mu_e)^2 + (\mu_h\mu_e)^2(n_h - n_e)^2H^2]\}H$, where $\mu_h(\mu_e)$ is a mobility of holes (electrons) and $n_h(n_e)$ is the carrier number of holes (electrons).[7] For $n_h = n_e$, there should be a linear relation between ρ_{xy} and H. Therefore, it was surmised that the numbers of holes and electron carriers are similar in this compound. In fact, the two band model was adopted for other $ThCr_2Si_2$-type pnictide compounds, such as $BaNi_2P_2$,[10] $Ba_{1-x}K_xFe_2As_2$,[11] and $Ba(Fe_{1-x}Co_x)_2As_2$,[12,13] and it was found that the existence of multi bands would be necessary for the analysis of transport data.

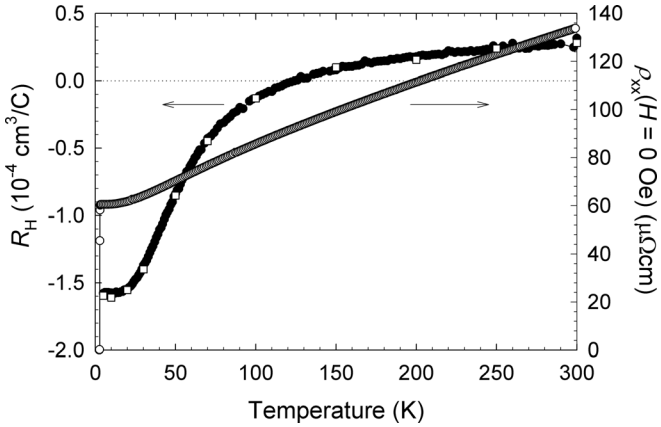

FIG 2. Temperature-dependent Hall coefficient R_H (left axis) and resistivity in the *ab*-plane (right axis) of $SrPd_2Ge_2$ single crystals. R_H values are derived from temperature scan (closed circle) and field scan (open square) of the configuration in Fig. 1(a) for Hall resistivity.

R_H was plotted in terms of temperature in Fig. 2. R_H is positive at 300 K and decreases slowly with decreasing temperature, and then it changes its sign to negative at $T_{rev} = 126$ K and decreases rapidly below T_{rev}. At $T = T_{rev}$, there is no significant change or transition in longitudinal electrical resistivity, which shows normal metallic behavior in a temperature range of $T_c \leq T \leq 300$ K. Therefore, it was supposed that the Hall sign reversal in $SrPd_2Ge_2$ does not result from an abrupt change in electronic structure induced by SDW or a structural transition as with other pnictide compounds, but from a gradual change due to an unbalanced contribution of density of state or scattering rate between the hole and electron pockets with increasing temperature.

The inset of Fig. 3 is a plot of the temperature-dependent resistivity with a field of $H = 50$ kOe, both parallel and perpendicular to the *c*-axis, as well as with zero field. From the data, the MR value, $\Delta\rho_{xx}(H = 5\text{T}, T)/\rho_{xx}(0, T) = [\rho_{xx}(5\text{T}, T) - \rho_{xx}(0, T)]/\rho_{xx}(0, T)$, as a function of temperature was calculated and plotted (Fig. 3). The MR values, with field parallel and perpendicular to the *c*-axis, were found to be similar. This behavior can be understood from the three dimensional

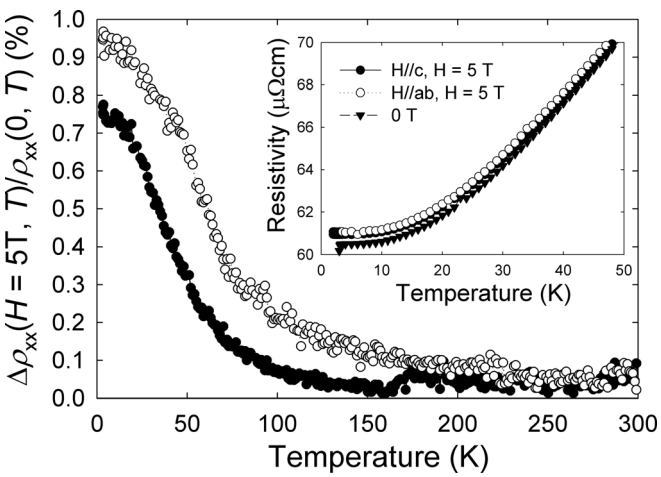

FIG. 3. Temperature-dependent MR with fields along and perpendicular to the *c*-axis. Inset: Temperature-dependent resistivity of $SrPd_2Ge_2$ single crystals with applied fields of 5T, both parallel and perpendicular to the *c*-axis, as well as with zero field.

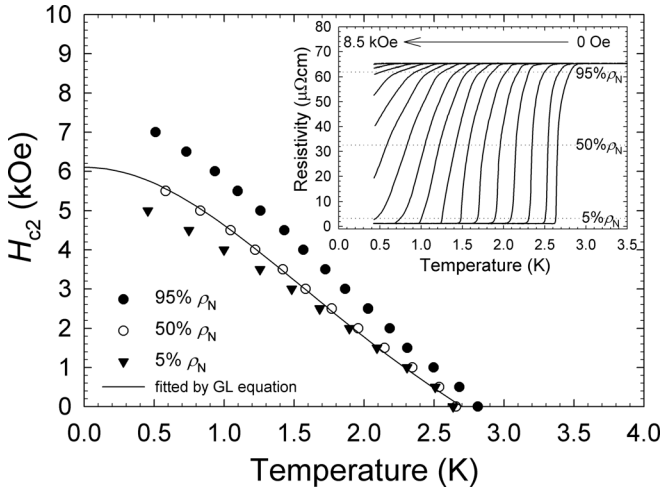

FIG. 4. Upper critical field (H_{c2}) of $SrPd_2Ge_2$ single crystals as a function of temperature from the resistivity data presented in the inset. The H_{c2} values were determined from 95% (closed circles), 50% (open circles), and 5% (triangles) of the normal-state value, ρ_N. The solid line is a fit of H_{c2} of 50% ρ_N to the Ginzburg–Landau equation (see text). Inset: Temperature-dependence of resistivity down to 0.35 K with increasing magnetic fields from right to left ($H = 0$, 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5, 5, 5.5, 6, 6.5, 7, 7.5, 8, and 8.5 kOe).

electronic properties of this compound, which was verified in both theoretical calculations[14] and experimental observations.[9] The MR values start to increase rapidly with decreasing temperature around 125 K, which roughly corresponds to T_{rev}. Similar behavior was observed in $EuRh_2As_2$ single crystals.[6] In $EuRh_2As_2$, the MR value started to increase at the same temperature, at which the Hall sign reversal occurred without any noticeable peak, due to SDW or structural transition, in temperature-dependent resistivity. In addition, the decrease in and sign change of R_H were understood in terms of the redistribution of carriers between electron- and holelike Fermi surfaces with increasing temperature, and was not supposed to be related to magnetic ordering, because the temperature was well below the antiferromagnetic transition temperature in $EuRh_2As_2$. Therefore, it was believed that the Hall sign reversal in $SrPd_2Ge_2$ is likely to have the same mechanism as in $EuRh_2As_2$.

Temperature-dependent resistivity near superconducting transition temperature was plotted under various magnetic fields (see the inset of Fig. 4). The current was applied perpendicular to the c-axis, and the field was parallel to the c-axis. The transition temperature was systematically suppressed with increasing magnetic field. Although the transition width increased slightly under the field, it was smaller than 1 K even at $H = 5$ kOe. From the definitions of 5% ρ_N, 50% ρ_N, and 95% ρ_N as indicated in the inset (Fig. 4), the upper critical field (H_{c2}) was estimated and plotted in terms of temperature. The upper critical field, determined from 50% ρ_N data, was fitted to the Ginzburg–Landau (GL) equation, $H_{c2}(T) = H_{c2}(0)(1 - t^2)/(1 + t^2)$, where $t = T/T_c$, and the result was plotted as a line (Fig. 4). The estimated value of $H_{c2}(0)^{GL} = 6.1$ kOe is larger than $H_{c2}(0)^{WHH} = 4.9$ kOe, which was extrapolated from the Werthamer–Helfand–Hohenberg (WHH) theory, and lower than the value from a linear fit, $H_{c2}(0) = 7.1$ kOe (which was found in Ref. 9), and the values from the extrapolation of H_{c2} data near T_c. The value of $H_{c2}(0)^{WHH} = 4.9$ kOe in Ref. 9 is not acceptable as an upper critical field at $T = 0$ K because the value is already achieved at $T = 0.83$ K in this study. In general, the origin of a higher upper critical field than in the WHH theory was explained by the anisotropy of the Fermi surface, highly disordered superconductors, and the strong electron–phonon coupling.[15] Because the heat capacity data suggested a strong electron–phonon coupling in $SrPd_2Ge_2$,[9] the strong coupling scenario would be the origin of a larger H_{c2} than in the WHH theory in $SrPd_2Ge_2$.

In conclusion, we measured the Hall effects, MR, and low temperature resistivity of $SrPd_2Ge_2$ single crystals. According to our observations of Hall sign reversal and violation of Kohler's rule, we could confirm a formation of multi band structure and probably the anisotropic scattering on the Fermi surface in $SrPd_2Ge_2$. In particular, the Hall sign reversal was observed at $T_{rev} = 126$ K while there was no noticeable anomaly in the temperature-dependent resistivity measurement. Therefore, the effect in $SrPd_2Ge_2$ is not likely to be related to the abrupt change of electronic structure induced by SDW or structural transition as in several pnictide compounds. Instead, we assume that the effect originated from the redistribution of carriers between electron- and holelike Fermi surfaces as a result of temperature changes. In addition, a larger upper critical field (H_{c2}) than the $H_{c2}(0)$ from WHH theory was observed in low temperature resistivity measurements, and the strong electron–phonon coupling was suggested as the origin.

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