Single s-band Half-Metallicity in n-type HgCr$_2$Se$_4$

Tong Guan, Chaojing Lin, Chongli Yang, Yonguo Shi, Cong Ren, Yongqing Li, Hongming Weng, Xi Dai, and Zhong Fang

Beijing National Laboratory for Condensed Matter Physics,
Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

Shishen Yan
School of Physics, Shandong University, Jinan 250100, China

Peng Xiong
Department of Physics, Florida State University, Tallahassee, FL 32306, USA

High quality HgCr$_2$Se$_4$ single crystals have been investigated by magnetization, electron transport and Andreev reflection spectroscopy. In the ferromagnetic ground state, the saturation magnetic moment of each unit cell corresponds to an integer number of electron spins (3µ$_B$/Cr$^{3+}$), and the Hall effect measurements suggest a single band transport with low electron densities. Spin polarizations as high as 97% were obtained from fits of the differential conductance spectra of HgCr$_2$Se$_4$/Pb junctions with the modified Blonder-Tinkham-Klapwijk (BTK) theory. The temperature and bias-voltage dependencies of the sub-gap conductance are consistent with recent theoretical calculations based on spin active scatterings at the superconductor/half metal interface. Our results suggest that n-HgCr$_2$Se$_4$ is a single-band half metal, qualitatively in agreement with theoretical calculations that also predicted undoped HgCr$_2$Se$_4$ is a magnetic Weyl semimetal.

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Half metals are a class of magnetic materials in which the charge current is conducted by carriers of one spin orientation. In a half metal, the Fermi level is located either inside an energy gap or in the localized states for one spin direction, whereas the electronic states are extended for the other. The full electron spin polarization makes half metals very attractive for spintronic applications, such as electrode materials for high performance magnetic tunnel junctions and efficient spin injection into semiconductors. Recent theories also suggest that half metals, and in particular those with single bands and strong spin-orbit interactions, may host p-wave or topological superconductivity when they are in proximity to s-wave superconductors. Nevertheless, the progress in utilizing the half metals has been rather limited, partly because only a handful of materials have been confirmed to be half-metallic in experiments. Moreover, charge carriers in these materials either have d-band characteristics or are holes in p-orbitals. Existence of s-band half-metals still lacks direct experimental evidence.

Chromium chalcogenides of the spinel group, ACr$_2$X$_4$ (A=Hg, Cd, Zn, X=Se, S), have been studied as magnetic semiconductors or insulators for several decades. Ferromagnetic order arises at low temperatures due to the superexchange interactions between the 3d electrons of Cr. The s-d exchange interactions can produce a large spin splitting of the conduction bands (Fig. 1a). Depending on the magnitude of exchange splitting and the strength of spin-orbit coupling, these chalcospins can be either magnetic semiconductors (e.g. CdCr$_2$Se$_4$) or Chern semimetals (i.e. magnetic Weyl metals, HgCr$_2$Se$_4$). Regardless of whether the exchange splitting is strong enough for a band inversion, a common feature of these chromium chalcospins is that there exists a sizable range of chemical potential for fully spin polarized s-band conductivity. This would provide a more complete set of wavefunctions for half metallic materials and hence offer more room for exploring exotic physics and novel functionalities. Despite such an appealing feature, no experimental measurement of the electron spin polarization has been done in the spinel class of materials.

In this work, measurements of magnetization, electron transport and Andreev reflection spectra have been carried out on high quality n-type HgCr$_2$Se$_4$ single crystals. Exchange splitting induced metal-insulator transition is manifested as eight orders of magnitude change in longitudinal resistivity upon ferromagnetic ordering as well as colossal magnetoresistance near $T_C$. In the ferromagnetic state, the saturation magnetization is found to correspond to 3.0 µ$_B$/Cr$^{3+}$, and the Hall effect measurements indicates a single band transport with an electron density on the order of 10$^{18}$ cm$^{-3}$. The zero-bias conductances of Pb/HgCr$_2$Se$_4$ Andreev junctions are nearly completely suppressed. These results coherently point to n-type HgCr$_2$Se$_4$ being a half-metal with carriers in a single s-band.

Single crystals of HgCr$_2$Se$_4$ were grown with chemical vapor transport in a sealed tube. The single crystalline samples usually are octahedral shaped and
Temperature dependence of longitudinal resistivity near the zero-field is due to the anomalous Hall effect (AHE). The temperature dependence of resistivity $\rho_{xx}$ can be attributed to a metal-insulator transition driven by the ferromagnet-paramagnet transition. In the paramagnetic phase, the chemical potential is located inside the bulk band gap. Below $T_C$, the s-d exchange interaction spin-splits the conduction band, and gradually raises the chemical potential above the conduction band minimum. The $\rho_{xx}$ data shown in Fig. 1d span from $\sim 10^{-2}\, \Omega\cdot cm$ (metallic) to $\sim 10^4\, \Omega\cdot cm$ (very insulating). Such a drastic variation in resistivity is comparable to the best values of magnetic semiconductors or other materials exhibiting colossal magnetoresistance (CMR) effect. The data in Fig. 1d also indicate that the HgCr$_2$Se$_4$ sample has very large magnetoresistance (MR) around the phase transition region. The MR, defined here as $\Delta M/M = (\rho(B) - \rho(0))/\rho(0)$, reaches $7 \times 10^4$ at $B = 8\, T$ and $T = 110\, K$.

We used the Andreev reflection spectroscopy to directly probe the electron spin polarization in HgCr$_2$Se$_4$. It has been used to measure the spin polarization in many ferromagnetic materials including several candidates for half metals, such as (La, Sr)MnO$_3$, CrO$_2$, EuS. This method is based on the fact that the Andreev reflection probability at the interface between a superconductor and a ferromagnet is partially or completely suppressed by spin-imbalanced density of states at the Fermi level in the ferromagnet. The spin polarization can be extracted by fitting the conductance spectra to the modified BTK theory. The measurement can be implemented either with point contact geometry or with planar junction geometry. Both geometries have been proven effective in revealing the half metallicity in CrO$_2$, a classic half metal that has also been confirmed with other methods, such as Meservey-Tedrow experiment.

Fig. 2a shows a sketch of the HgCr$_2$Se$_4$/Pb planar junctions used in this work. A typical device was fabricated on a lustrous, triangle-shaped surface of a HgCr$_2$Se$_4$ single crystal (Fig. 1b, inset). The differential conductance, defined as $dI(V)/dV$, was measured as a function of the dc bias voltage $V$ for HgCr$_2$Se$_4$/Pb junctions using phase-sensitive lock-in detection. The amplitude of the ac modulation was kept sufficiently small in order to avoid heating and other spurious effects. Figs. 2b shows the conductance curves of sample J1 for temperatures from 0.34 K to 8.5 K. The Pb films used in this work has a superconducting transition temperature $T_c = 7.2\, K$, nearly identical to the bulk $T_c$ value in literature. The conductance data at 8.5 K are close to the normal state.

\[ \rho_{xx}(B) = \rho_{xx}(0) + \Delta \rho_{xx}(B) \]

kg, this is consistent with the results of the first principles calculations. As illustrated in the band diagram in Fig. 1a, the measured magnetization corresponds to the fully occupied spin up $t_{2g}$ orbital of Cr. At low temperatures, Hall resistivity $\rho_{xy}$ has a linear dependence on magnetic field, except at very low fields where magnetization is not saturated and the small curvature is cause by the anomalous Hall effect (Fig. 1c). The linearity in the Hall resistance for a large range of magnetic fields suggests that the carriers come from a single band. From the magnitude and the sign of the Hall coefficient, an electron density of about $2 \times 10^{18} \, cm^{-3}$ is obtained at $T < 70\, K$. Measurements of many other n-type samples consistently reproduce carrier densities on the same order of magnitude and electron mobilities of the order of $10^2 \, cm^2/V\cdot s$ at $T = 2\, K$.

In addition to the ideal magnetization value, the high quality of our n-HgCr$_2$Se$_4$ samples is also shown by the strong temperature dependence of the longitudinal resistivity $\rho_{xx}$. As depicted in Fig. 1d, $\rho_{xx}$ increases by more than eight orders of magnitude as $T$ is increased from 10 K to temperatures above the Curie temperature. This is in contrast to only up to 3-4 orders of magnitude change in $\rho_{xx}$ obtained in pervious works.

\[ MR = \rho_{xx}(0)/\rho_{xx}(B) - 1, \quad MR < 7 \times 10^4 \, \Omega\cdot cm \]

\[ \rho_{xx} \sim 1 \, mm \text{ in size (Fig. 1b inset). As shown in Fig. 1b, HgCr$_2$Se$_4$ is a soft ferromagnet below the Curie temperature ($T_C = 105.5\, K$), and no hysteresis was clearly resolved. The saturation magnetization $M_s$ corresponds to 3.00 \pm 0.05 \, \mu_B$ per Cr$^{3+}$ ion. In contrast, all previous works non-integer values of the magnetic moment $2.7-2.9\, \mu_B/\text{Cr}^{3+}$ obtained in pervious works. This is in consistent with the results of the first principles calculations. As illustrated in the band diagram in Fig. 1a, the measured magnetization corresponds to the fully occupied spin up $t_{2g}$ orbital of Cr. At low temperatures, Hall resistivity $\rho_{xy}$ has a linear dependence on magnetic field, except at very low fields where magnetization is not saturated and the small curvature is cause by the anomalous Hall effect (Fig. 1c). The linearity in the Hall resistance for a large range of magnetic fields suggests that the carriers come from a single band. From the magnitude and the sign of the Hall coefficient, an electron density of about $2 \times 10^{18} \, cm^{-3}$ is obtained at $T < 70\, K$. Measurements of many other n-type samples consistently reproduce carrier densities on the same order of magnitude and electron mobilities of the order of $10^2 \, cm^2/V\cdot s$ at $T = 2\, K$.

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small, but noticeable superconducting coherence peaks 

expected for a half metal. (2) There exist a pair of 

The subgap conductance is significantly suppressed, as 

junctions have the following common characteristics: (1) 

FIG. 2: (color online). (a) Sketch of the HgCr$_2$Se$_4$/Pb planar 

junctions used for Andreev reflection spectroscopy. (b) Differential 

conductance $G(V) = dI/dV$ of the sample J1 plotted 

as function of dc bias. (c) Normalized differential conduc-

tance $G(V)/G_N(V)$ spectra for sample J2. Here, the bias 

V is defined as the dc voltage of the Pb electrode relative to the 

crystal, and 

Se$_4$/Pb interfaces. (2) The inelastic broadening 

features (see for instance, the small hump-and-dip struc-

tures at about 7 meV in the conductance spectra shown 

in Fig. 2b) imply high quality of the Pb films and the 

HgCr$_2$Se$_4$ crystal, and $G_N(V)$ is normal state differential conduc-
tance recorded with a small field ($\mu_0H = 0.2$ T) applied.

values, $G_N(V)$, which were recorded when a perpen-
dicular magnetic field $\mu_0H = 0.2$T was applied at corre-
sponding temperatures. For sample J1, $G_N$ is about 

34Ω, and it has a weak bias dependence.

Many other samples with low junction resistances have 

also been studied, in which $R_N$ is mostly in the range of 

14-75Ω. Fig. 2c shows a set of normalized conductance 

$(G(V)/G_N(V))$ curves for a HgCr$_2$Se$_4$/Pb junction (sam-

ple J2) with $R_N \approx 14$Ω. The low junction resistance in-
dicates a weak barrier strength at the HgCr$_2$Se$_4$/Pb in-
terface, thus allowing for reliable extraction of the elec-
tron spin polarization with the modified BTK theory. 

This is in stark contrast to the planar Andreev junctions 

of EuS, a magnetic semiconductor with much higher elec-
tron densities. In the latter, $R_N$ is on the order of 10kΩ 

and the I-V trace is highly nonlinear due to the forma-
tion of a Schottky barrier [34]. As shown in Ref. [35], 

very strong barrier strength could lead to the conduc-
tance spectra hardly discernible from the tunneling spec-

tra between an unpolarized metal and a superconductor. 

The conductance spectra of our low-$R_N$ HgCr$_2$Se$_4$/Pb 

junctions have the following common characteristics: (1) 

The subgap conductance is significantly suppressed, as 

expected for a half metal. (2) There exist a pair of small, 

but noticeable superconducting coherence peaks at the superconducting gap edges, i.e. $eV = \pm \Delta$. The 

energy gap $\Delta$ is 1.4-1.5 meV, close to the bulk value of 

Pb. (3) The heights of both coherence peaks are signifi-
cantly lower than what is expected of a tunnel junction, 

and the height of the peak at the positive bias (see Fig. 2) 

is greater than that at the negative bias. Such an asym-

metry has not been observed in the Andreev junctions 

of CrO$_2$ and EuS, despite that the barrier strength can 

be varied systematically from very weak to very strong. 

However, it has been observed in the Andreev reflect-

ion spectra of several strongly correlated electron sys-

tems, such as heavy fermion superconductor CeCoIn$_5$ [36] 

and ferromagnetic semimetal EuB$_6$ [37]. Further work is 

needed to confirm whether there is a link between the asymmetry and electron correlation.

In order to compare the experimental data with the 

modified BTK theory [29], a symmetrization procedure, i.e. $G(V) \rightarrow [G(V) + G(-V)]/2$, was taken for the con-
ductance spectra. In this theory, there are four fitting 

parameters: spin polarization $P$, barrier strength param-

eter $Z$, superconducting gap $\Delta$ and inelastic broadening 

parameter $\Gamma$. In this work, we fixed $\Gamma$ to zero for the 

fits due to the following two considerations: (1) Both the large gap energies of our Pb films and the phonon 

features (see for instance, the small hump-and-dip structures 

at about $\pm 7$ meV in the conductance spectra shown 

in Fig. 2b) imply high quality of the Pb films and the 

HgCr$_2$Se$_4$/Pb interfaces. (2) The inelastic broadening 

energy extracted from tunnel junctions of the Pb films 

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with similar quality is on the order of tens of $\mu$eV, two orders magnitude smaller than the gap energy. Fig. 3 shows the normalized conductance curves after the symmetrization and their BTK fits with three adjustable parameters ($P$, $Z$ and $\Delta$) for four junctions. The fits yield $P = 95\%$, 97%, 92% and 92%, and $Z=0.66$, 1.04, 0.75 and 1.00, for samples J1-J4, respectively. There is no apparent trend of decreasing in $P$ with increasing $Z$, consistent with previous work on planar CrO$_2$/Pb junctions. Similar fits were also carried out for the positive (or negative) halves of the conductance spectra, namely $G(V)$ at $V > 0$ (or $V < 0$), the extracted $P$ only differs up to a few percent for different polarities. This can be regarded as a posteriori justification for the symmetrization procedure described above.

The conductance spectra of the HgCr$_2$Se$_4$/Pb junctions and the extracted high degree of spin polarization from the modified BTK fits have some resemblance to those of CrO$_2$ planar junctions, in which spin polarizations of 90% to 97% were obtained for a wide range of barrier strengths ($Z = 0$ to 2.7). Nevertheless, it should be pointed out that a 100% spin polarization (or correspondingly, completely suppressed subgap conductance) has never been observed in any Andreev reflection experiment on CrO$_2$ or other half metals despite an enormous amount of experimental efforts. Recent theoretical advances have rendered several mechanisms that can account for the small deviation from full spin polarization [39]. These include various spin active scatterings at the superconductor/half metal interface [40], the spin-orbit coupling in the superconductor [41] and inelastic scatterings of quasiparticles in the half metal [42]. These scatterings can relax the rule of spin conservation and lead to finite conductance at zero bias. Furthermore, the lifting of the particle-hole symmetry away from zero-bias brings a conductance correction of the form $\delta G \propto V^2$, if $V$ is not too large [40, 43]. Following these models, one would expect a quadratic dependence of the junction conductance on the bias voltage. Indeed, this is borne out in the conductance spectra of HgCr$_2$Se$_4$/Pb junctions (Fig. 3a-d): As shown clearly in Fig. 4a, the low bias $G(V)$ of sample J2 deviates substantially from the BTK fit, but follows very well a quadratic function.

Further evidence for the deviation from conventional BTK-type Andreev reflection comes from the temperature dependence of zero-bias conductance $G_0$. As shown in Fig. 4b, $G_0$ of sample J2 roughly follows a $T^3$ dependence: $G_0/G_{0,N} \simeq (T/T_C)^3$. In contrast, the modified BTK model predicts substantially smaller $G_0$ values at intermediate temperatures. The excess of conductance at finite temperatures is consistent with the calculation of Löfwander et al. with a model of spin active scatterings at a clean superconductor/half-metal interface [39], for the case of a typical spin mixing angle. An interesting outcome of their work is that $G(V)$ vanishes at the $T = 0$ limit. As shown in Figs. 2a and 2b, the zero-bias conductance $G_0(T)$ saturates at very small values as $T$ approaches zero. The low temperature $G_0$ appears to vary from sample to sample (Fig. 4a), suggesting the interface quality is important. This may lend support to the disorder assisted spin active scatterings proposed in Ref. [44] for finite $G_0$ at $T = 0$. The additional conductance brought by the spin active scatterings may be responsible for the underestimation of the spin polarizations in half-metals with the fits to the modified BTK theory reported in the literature [32, 39]. The values obtained from the modified BTK fits may only provide lower bounds for the spin polarization.

In summary, the high degree of spin polarization derived from fits to the modified BTK theory as well as the integer moment magnetization provide strong evidence for half-metallicity in n-HgCr$_2$Se$_4$. This is consistent with the first principle calculations that predicted the half-metallic and Chern semimetallic phases for the n-doped and undoped HgCr$_2$Se$_4$, respectively [22]. Furthermore, the small but finite zero-bias conductance and its temperature dependence are compatible with recent theoretical models developed for spin-active scatterings, which have been proposed as a venue for realizing the exotic triplet paring with the superconducting proximity effects in half metals [6, 7, 9, 40].

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