Observation of orbital two-channel Kondo effect in a ferromagnetic L1₀-MnGa film

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The experimental existence and stability of the fixed point of the two-channel Kondo (2CK) effect displaying exotic non-Fermi liquid physics have been buried in persistent confusion despite the intensive theoretical and experimental efforts in past three decades. Here we report an experimental realization of the two-level system resonant scattering-induced orbital 2CK effect in a ferromagnetic L1₀-MnGa film, which is signified by a magnetic field-independent resistivity upturn that has a logarithmic and a square-root temperature dependence beyond and below the Kondo temperature of ~14.5 K, respectively. Our results not only evidence the robust existence of orbital 2CK effect even in the presence of strong magnetic fields and long-range ferromagnetic ordering, but also extend the scope of 2CK host materials from nonmagnetic nanoscale point contacts to diffusive conductors of disordered alloys.

The overscreened Kondo effect displaying non-Fermi-liquid (NFL) physics has been of considerable scientific interest in recent years, especially due to their potential relevance to heavy fermions, topological superconductors, topological Kondo insulators, graphene, and quantum dots. Its simplest manifestation, the two-channel Kondo (2CK) effect, may occur when a spin-1/2 impurity symmetrically couples to conduction electrons in two equal orbital channels via exchange interaction (spin 2CK)⁶–⁸, or when a pseudospin-1/2 of two degenerate macroscopic charge states of a metallic island symmetrically couples to two conduction channels (charge 2CK)⁹, or when a pseudospin-1/2 of structural two-level system (TLS, where an atom or atom group with small effective mass coherently tunnels between two nearby positions at a rate of 10⁸–10¹² s⁻¹) equally couples to two spin channels of conduction electrons via resonant scattering (orbital 2CK)¹⁰–¹³. The 2CK effect is expected to have a unique low temperature (T) resistivity upturn (Δρxx), which scales with ln T beyond the Kondo temperature (TK), followed by an exotic NFL behavior (Δρxx~T⁻¹/₂) as the consequence of two conduction electron spins screening the spin (pseudospin) impurity¹²–¹⁴. The T⁻¹/₂ dependence of Δρxx is a hallmark of the NFL state in the 2CK effect, in striking contrast to the T² scaling of Fermi-liquid (FL) behavior in the case of the fully screened Kondo effect. Recently, the charge 2CK effect and spin 2CK effect were clearly demonstrated and channel asymmetry effect was probed directly and quantitatively⁶,⁷. However, the orbital 2CK physics has been under heated debate despite the intensive studies for almost 30 years. Even the sheer existence of the orbital 2CK effect is still controversial¹⁵–²³, for a TLS model that only considers a particle in a double well potential interacting with a degenerate electron gas, the orbital 2CK behavior can never be observed in the weak coupling limit (JN(EF)~1) because the energy splitting (Δ) between the lowest two eigenstates of the TLSs always dominates the physics, i.e. T_K<Δ²/T_K, even if electron-assisted tunneling and the higher excitation states are taken into account¹⁵. However, as pointed by Zaránd¹⁵, an experimental realization of the orbital 2CK effect with the generic low temperature resistivity upturn is expected for the TLSs with enhanced resonant scattering at E_F and strong Kondo coupling (JN(EF)~1), which is supported by the observation of the NFL behavior in ballistic conductors of Cu and Ti point contacts (PCs) fabricated by electron-beam lithography and diffusive conductors of ThAsSe glasses prepared by chemical vapor transport. Furthermore, the stability of the orbital 2CK fixed point has remained an open question. A breakdown of the orbital 2CK fixed point is predicted at low energies T_D (Δ²/T_K) in the case of a nonzero Δ or asymmetric exchange coupling

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dynamic growth method, e.g. molecular-beam epitaxy 30. Similar to the point 26. Experimentally, it has, however, remained unclear how robust the orbital 2CK fixed point is with respect to the NFL behavior and to produce a crossover to FL behavior at a low temperature.

Therefore, in the presence of strong magnetic fields and the spin polarization of the conduction electrons. 

strength in the two channels 25,26. Present theories also expect an imbalance in the channel population to quench the NFL behavior and to produce a crossover to FL behavior at a low T in the neighborhood of T = 0K 2CK fixed point 28. Experimentally, it has, remained unclear how robust the orbital 2CK fixed point is with respect to a channel asymmetry at finite temperatures. A magnetic field (H) of 5 T was reported to result in a breakdown of the NFL behavior at low energies in an early Cu PC experiment 24. Some recent experiments argue for a negligible influence of strong magnetic field of up to 14 T in ThAsSe glasses 22,23 or even a slight spin polarization in Li10-MnAl films 14,27, suggesting a considerable robustness of the orbital 2CK fixed points at finite temperatures. One reason was suggested to be that the electron spins are not directly involved in the Hamiltonian of the TLS coupling to the conduction electrons in the two spin channels 22. It is, therefore, of great importance and interest to develop new Kondo systems with large 2CK and high-density TLSs in order to clarify the controversial physics of the orbital 2CK effect, especially its experimental existence and stability with respect to the population imbalance of two spin channels due to the strong magnetic fields or ferromagnetic exchange splitting.

The fully ordered Li10-MnGa alloy is an itinerant magnet which is predicted to have a spin polarization of ~40% at the Fermi surface, a saturation magnetization (M_s) of ~2.51 μ_B/Mn (i.e. 845 emu cm^-3), a ferromagnetic exchange splitting (E_{exchange}) of ~2.2 eV and a E_F ~11 eV (see Fig. 1a) 28,29. Experimentally, Li10-MnGa films with off-stoichiometry can be achieved in a wide Mn/Ga atomic ratio (x) range (0.76 < x < 1.75) by a non-equilibrium dynamic growth method, e.g. molecular-beam epitaxy 20. Similar to the Li10-ordered MnAl films 31,32, the magnetic and transport properties of Li10-MnGa films are strongly dependent on the structural disorders and may be conveniently tailored by varying the growth parameters 33-35. 

Therefore, Li10-MnGa is an ideal playground for the exploration of disorder-related phenomena, e.g. orbital 2CK effect. In our previous paper 34, we observed in 50 nm thick disordered Li10-Mn1.5Ga films logarithmic low-T resistivity upturns which exhibit a close relevance to growth temperatures (T) and an independence of strong applied magnetic fields. Here we show that the resistivity upturn in Li10-MnGa films most likely arises from the orbital 2CK effect by taking an Li10-MnGa film with enhanced disorder as an example. We observed a low-T resistivity upturn with a clear transition from a ln T dependence to NFL behavior signified by a T^{1/2} dependence. The T dependencies of the resistivity upturn are independent of applied magnetic fields up to 8 T. This result underpins the robustness of orbital 2CK effect even in the presence of strong magnetic fields and the spin polarization of the conduction electrons.

Results

Sample and ferromagnetism. A 30 nm thick Li10-MnGa film was grown on 150 nm GaAs-buffered semi-insulating GaAs (001) substrate at 200°C. The Mn/Ga atom ratio x was determined by high-sensitivity x-ray photoelectron spectroscopy to be 0.94 (Fig. 1b). The chemical composition and the growth temperature were carefully chosen for an enhanced structural disorder. Figure 1c shows a cross-sectional inverse fast Fourier transform (IFFT) transmission electron microscopy (TEM) image of MnGa/GaAs interface, which clearly indicates the existence of the dislocations in the MnGa layer. The dislocations were suggested to be responsible for the TLSs 19,20,36. Figure 2a,b show the well-defined perpendicular magnetization hysteresis loop and hysteretic Hall resistance measured at room temperature, respectively, revealing the ferromagnetism (M_s ~100 emu cm^-3 at room temperature) and perpendicular magnetic anisotropy of this film. Figure 2c displays the T dependence of magnetization (M) along film normal for the Li10-MnGa film under H = 50 Oe. The Curie temperature (T_C) of the film was determined to be 366 K following a three-dimensional (3D) Heisenberg model which expect
$M \propto (T - T_C)^{1/3}$. The quick increase at temperatures below ~25 K is suggestive of nanoscale magnetic clusters embedded in the film due to its high degree structural disorders or due to the two sets of antiferromagnetically coupled Mn atoms which could have different magnetic moments and $T_C$ (similar to a ferrimagnet).

**Temperature dependence of the longitudinal resistivity.** Figure 2d shows the $T$ dependence of $\rho_{xx}$ for the $L1_0$-MnGa film at zero field ($H = 0$ T) as an example. $\rho_{xx}$ shows a minimum at ~40 K, beyond which $\rho_{xx}$ increases monotonically with $T$ due to increasing thermal phonon and magnon scattering. Below this minimum, $\rho_{xx}$ shows an upturn down to 2 K which is the lowest $T$ that our present setup can reach. The same feature holds for different fixed $H$ of at least up to 8T. In the following we show that the low-$T$ resistivity upturn in our $L1_0$-MnGa film most likely arises from the TLS-induced orbital 2CK effect. In the absence of an external magnetic field, as displayed in Fig. 3a, $\rho_{xx}$ of the $L1_0$-MnGa film first varies linearly with $\ln T$ below a temperature $T_K$ of ~14.5 ± 1.5 K. The dashed lines in (c) are for eye guidance.

Expressed as $\rho_{xx}(T) \propto (T - T_K)^{1/3}$, the temperature dependence of $\rho_{xx}$ below $T_K$ is indicative of the presence of localized magnetic moments. The temperature dependence of $\rho_{xx}$ above $T_K$ is consistent with the model of a single-channel Kondo effect due to static magnetic impurities. In fully screened...
Magnetic field effects. Another characteristic of the TLS-induced orbital 2CK effect is the $H$ independence of the resistivity upturn. Magnetic fields should not have any observable influence on the resonant levels, coupling strength, and thus the effect amplitude via changing the population balance of the two spin channels of the conduction electrons because the Zeeman splitting is negligibly small ($\sim 0.9$ meV at $H = 8$ T) in comparison to the width of energy band and $E_F$ of a host system ($\sim 10$ eV), e.g. ferromagnetic $L_{10}$-MnAl. In order to establish the orbital 2CK physics in $L_{10}$-MnGa film, we further examined the effects of $H$ on both the $\ln T$ and $T^{1/2}$ dependences of $\rho_{xx}$. As shown in Fig. 3a,b, the magnetic fields have no measurable influence on the $T$ dependence: $\rho_{xx}$ scales linearly with $\ln T$ and $T^{1/2}$ at $T_K < T < T_F$ and $2K < T < T_F$, respectively, under different magnetic fields ranging from 0 to 8 T. Note that, under perpendicular $H$, anisotropic MR and spin disorder scattering-induced MR should be negligible in a film with large perpendicular magnetic anisotropy, because of the orthogonal magnetization-current relation and the large energy gap in spin wave excitation spectrum. This is highly amenable to study the intrinsic $H$ dependence of a 2CK effect.

Figure 3c summarizes the values of the slopes $\alpha$ and $\beta$ as a function of $H$ for the $L_{10}$-MnGa film. It is clear that both $\alpha$ and $\beta$ are independent of $H$, strongly suggesting a nonmagnetic origin of the resistivity upturn observed in $L_{10}$-MnGa. Specifically, there is no measurable change in $T_K$ under different $H$ (Fig. 3a,b), suggesting a negligible effect of $H$ on the Kondo coupling strength, tunneling symmetry, and barrier height of the TLSs. However, our $L_{10}$-MnGa epitaxial film does not show any sign of a breakdown of the NFL behavior due to a magnetic field of up to 8 T in the entire temperature range that is of interest, which suggests both a negligible influence of the applied magnetic fields on the population balance of the two spin channels and the robustness of the 2CK physics to a slight population imbalance. These observations provide strong evidence for the orbital 2CK effect being induced by TLSs originating from nonmagnetic impurities. A negative magnetoresistance (MR) is found to accompany the orbital 2CK effect in several different host systems\cite{10,11,12,13}. Here, the $L_{10}$-MnGa also shows a negative MR at high $H$ in the entire $T$ range (Fig. 4a–c), which monotonically shrinks from $\sim 1.8\%$ at room temperature to $\sim 0.5\%$ at low temperatures and does not saturate even at $H = 8$ T. As shown in Fig. 4b, the negative MR doesn’t scale with $H^2$, which is in contrast the 1CK effect. The Bethe ansatz equations and conformal field theory for the 2CK problem predict an MR that scales with $H^{1/2}$ and $\ln H$ in $T^{1/2}$ and $\ln T$ regimes, respectively. Although the negative MR of the $L_{10}$-MnGa film does scale linearly with $H^{1/2}$ (see Fig. 4c), it should be irrelevant to the orbital 2CK effect as indicated by the energy scale of the $H^{1/2}$ dependence (at least up to 300 K) and the absence of a $\ln H$ scaling.

Coexistence of the 2CK fixed point with ferromagnetism. The evident coexistence of the 2CK physics and ferromagnetism is an intriguing observation. Although the two spin channels are still degenerate in energy because the Kondo coupling with a TLS is nonmagnetic and does not involve any spin variables, the population imbalance of the two spin channels due to the ferromagnetic exchange splitting of the conduction band could be significant in comparison to the magnetic field effects for the TLS model. In fully ordered $L_{10}$-MnGa and $L_{10}$-MnAl, the spin moments of Mn atoms are parallel due to ferromagnetic Ruderman–Kittel–Kasuya–Yoshida interaction and the spin polarization is dominantly determined by the Mn 3d states\cite{14}. In disordered samples, the Mn–Mn antiparallel alignment due to antiferromagnetic superexchange simultaneously reduces $M_s$ and spin polarization as a consequence of the cancelling contributions from the oppositely aligned Mn atoms\cite{15,16,17,18}. For
the disordered $L_{10}$-MnGa film studied here, the value of $M_s$ is only 12.5% of the theoretical value for the fully ordered $L_{10}$-MnGa, indicating a robust antiparallel alignment of Mn-Mn magnetic moments and a very low degree of spin population imbalance. This could be the reason why the ferromagnetism does not quench the 2CK physics here. The robust 2CK effect observed in ferromagnetic systems, e.g. $L_{10}$-MnGa and $L_{10}$-MnAl, also hints that the fixed point of an orbital 2CK effect is more robust to the loss of spin population balance in comparison to that of a spin 2CK effect to the orbital channel asymmetry. However, a dilution of NFL behavior and an enhancement of $T_D$ due to the loss of spin population balance are expected in a ferromagnet with a partially spin-polarized conduction band$^{14}$. It would be very interesting to quantitatively determine how the stability of 2CK fixed point varies with an enhancing population imbalance of the spin channels. More theoretical and experimental efforts are needed to better understand the exotic 2CK physics, especially in ferromagnetic hosts.

The TLSs play a central role in the orbital 2CK model, however, the identification of their microscopic nature is generally challenging. In nanoscale ballistic conductors of Cu and Ti PCs$^{19–21}$, the $1/f$ noises spectrum was introduced to hint the existence of dynamic motion of the atoms. However, it is difficult to separate the component of such $1/f$ noises arising from atomic motion in diffusive conductors$^{40}$, e.g. ThAsSe glasses, $L_{10}$-MnAl and $L_{10}$-MnGa films. A definite identification of the microscopic nature of the TLSs requires future developments of highly sensitive spectroscopic probing techniques. However, we noted that dislocation kinks seem to be responsible for the formation of TLSs as suggested by point contact experiments and theoretical calculations$^{11,19,20,36}$. A logarithmic resistivity increase at low temperatures was also found to be associated with the increase in the dislocation density in different dilute Al alloys where dislocations were introduced by shock loading and extension at different temperatures$^{41}$. Independent resonant scatter centers could also form along one single dislocation with large spatial extent$^{13}$. Taking into account that the average distance of the dislocations (see Fig. 1c) appears to be of the same order with that of the adjacent TLSs (~2 nm), we, therefore, surmise that the nonmagnetic Ga atoms at the high-density dislocations likely play the role of TLS centers in present film. Here, the nonmagnetic nature of orbital 2CK effect and the significant disorder dependence rule out the possibility of the Mn atoms, the electrons nor the embedded magnetic clusters as TLS centers.

**Discussions**

We have presented the experimental evidence which strongly suggests the occurrence of a robust orbital 2CK effect due to the electron scattering by high-density TLSs in ferromagnetic $L_{10}$-MnGa film. The $H$-independent resistivity upturn scaling with $\ln T$ and $T^{1/2}$ in the two $T$ regimes below the resistance minimum are well consistent with the TLS model. The large $T_K$ of ~14.5 K suggests a strong Kondo coupling between the TLS centers and the surrounding conduction electrons via resonant scattering. The orbital 2CK effect in a ferromagnetic material points to a more robust fixed point for orbital 2CK with respect to the slight spin population imbalance in comparison to that for spin 2CK to the orbital channel asymmetry. Our observation also suggests that diffusive films of disordered ferromagnetic alloys, which were overlooked in the past 2CK physics studies, can be better TLS host materials than conventional nanoscale point contact devices fabricated by electron-beam lithography due to their high TLS densities ($\sim 10^{20}$ cm$^{-3}$) and high Kondo temperatures. Our findings also imply that the nonmagnetic

**Figure 4. Magnetoresistance.** (a) MR versus $H$, (b) MR versus $H^2$, and (c) MR versus $H^{1/2}$, respectively. The dashed lines in (c) represent the best linear fits of MR-$H^{1/2}$ for each temperature.
disordered alloys may also be potential host materials for realizing orbital 2CK physics because of the absence of the spin polarization that actually hurts the orbital 2CK effect. This greatly extends the scope of TLS host systems for future studies of the orbital 2CK physics, which should be inspiring for future 2CK physics studies. More experimental and theoretical efforts are needed in the future in order to better understand the intriguing robustness of the 2CK physics even in the presence of ferromagnetism.

Methods
Sample preparation and characterizations. The sample was prepared by a VG-80 molecular-beam epitaxy system with two growth chambers (one for growing III–V group semiconductors, the other for growing magnetic alloys). A semi-insulating GaAs (001) substrate was first loaded into the semiconductor chamber to remove the oxidized surface by heating up to 580 °C in arsenic atmosphere (~1 × 10⁻² mbar) and to get a smooth fresh surface by growing a 150 nm GaAs buffer layer. Afterwards, the sample was transferred to second growth chamber to grow a 30 nm thick L₁₂-MnGa film at 200 °C and a 5 nm thick MgO capping layer for protection from oxidation. The composition, structure, and magnetism were measured by an x-ray photoelectron spectroscopy (Thermo Scientific ESCALAB 250Xi), a transmission electron microscopy (JEOL 2010), and a Quantum Design superconducting quantum interference device (SQUID-5) magnetometer, respectively.

Device fabrication and transport measurement. The film was patterned into 60μm wide Hall bars with an adjacent electrode distance of 200μm using standard photolithography and ion-beam etching for transport measurements. The Hall resistivity (ρ_H) and longitudinal resistivity (ρ_L) were measured in a Quantum Design physical property measurement system (PPMS-9) as a function of temperature and magnetic field with a superconducting quantum interference device (SQUID-5) magnetometer, respectively.

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**Author Contributions**

L.Z. designed and performed the experiments, L.Z., G.W. and J.Z. analyzed the data and wrote the manuscript.

**Additional Information**

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