Sr_2IrO_4 magnetic phase diagram, from resistivity

L. Fruchter, G. Collin, D. Colson[+] and V. Brouet
Laboratoire de Physique des Solides, C.N.R.S. UMR 8502, Université Paris-Sud, 91405 Orsay, France and
[+]*Service de Physique de l'Etat Condensé, CEA-Saclay, 91191 Gif-sur-Yvette, France*
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We show that the transition to the antiferromagnetic state in zero magnetic field does show up in the transverse resistivity, for which we point out the possibility for a direct spin orientation effect. In an applied field, we propose that the transition is split into two lines, corresponding to in-plane and out-of-plane magnetic ordering. This picture is corroborated by transverse magnetization measurements. The magnetic phase diagram for Sr_2IrO_4 was investigated, using the angular dependence of the resistivity transverse to the IrO_2 planes.

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I. INTRODUCTION

In the recent years, iridium oxides have become a new playground for the study of electron correlation effects. Indeed, while extended $5d$ orbitals reduce the electron-electron interaction, as compared to the $3d$ transition metal compounds as cuprates, strong spin orbit coupling (SOC) associated to the heavy Ir competes, together with the on-site Coulomb interaction, with electronic bandwidth to restore such correlations[1]. Amongst these compounds, the Ruddlesden-Popper series, $R_{n+1}Ir_nO_{3n+1}$ where R= Sr, Ba and n = 1,2,∞, has attracted much of the attention, in particular due to the structural similarities of these perovskites with the cuprates compounds. Sr_2IrO_4, where one IrO_2 layer alternates with an SrO layer, is structurally similar to the first discovered cuprate superconductor, $(La,Ba)_2CuO_4$. The physics of the latter is the one of an antiferromagnetic Mott insulator, with a magnetic interaction described within the framework of a spin-1/2 Heisenberg model. It was early proposed that the strong SOC in the iridate perovskite actually allows for an effective localized state different from this spin-1/2 state, entangling spin and orbital degrees of freedom, with total angular momentum $J_{eff} = 1/2$. This spin-orbital insulating state was proposed to be the analog of a Mott insulator [1].

The antiferromagnetic order in Sr_2IrO_4 is now well documented[2, 3]. The moments (0.2 μ_B/Ir) lay in-plane and order at $T_N \simeq 240$ K. As for La_2CuO_4, the loss of the inversion symmetry in the non cubic structure, due to a rotation of the oxygen octahedra, allows for a Dzyaloshinsky-Morya interaction, which in turn induces a canting of the spins and a ferromagnetic component in the IrO_2 planes [4–6]. The net moment (0.14 μ_B/Ir), which is coupled antiferromagnetically from plane to plane in zero field, align ferromagnetically with an in-plane field $H \approx 0.2$ T [7]. Recent *ab initio* computations conclude that the dominant magnetic interaction is of Heisenberg type, with little effect of the geometrical factors on the exchange coupling [8]. As shown in ref. 19, the absence of a critical behavior in the in-plane magnetic correlation length at T_N is also in favor of a two-dimensional Heisenberg behavior with large quantum fluctuations. On the basis of such a description, it has been proposed that Sr_2IrO_4 may exhibit electronic properties similar to the ones of the cuprates, including superconductivity [9]. The nature of the insulating state is however the subject of debate. First, the realization of the $J_{eff} = 1/2$ state itself may be questioned, as it requires a perfect orbital degeneracy, which is not obtained in Sr_2IrO_4 where the octahedra are strongly elongated [10]. The location of a metal-insulator transition, either in the paramagnetic state as for a Mott-Hubbard transition [11], or coincident with the magnetic transition as for a Slater-type transition [12] is controversial.

As first noticed by Kini et al [13], no anomalies can be detected in resistivity at T_N. The authors proposed that this could result from the fact that localized states shifts the Fermi level away from the band edges affected by spin polarization. A time-resolved optical study found that the metal-insulator transition takes place over a wide temperature range $0.7 \lesssim T/T_N \lesssim 1.4$, thus accounting for the absence of any sharp anomaly in transport and thermodynamic quantities[14]. Well below T_N, for $T \lesssim 100$ K, large anisotropic magnetoresistance as well as magnetodielectric effects were observed (Refs. 15, 16). It was proposed that the magnetoelectric effects result from the competition of antiferromagnetic and ferromagnetic coupling, at low and high temperature respectively[16]. Here, we show that there is actually a small but clear signature of the magnetic transition in zero field in the transverse resistivity. This allows to use magnetotransport as a probe to establish a magnetic phase diagram up to T_N for this compound. We suggest that the magnetic transition under magnetic field is split into two lines, corresponding to in-plane and out-of-plane magnetic ordering. In the ordered region, we propose that spin orientation has a direct impact on resistivity.

II. RESISTIVITY AND TRANSVERSE MAGNETOMETRY

The results below where obtained with a Sr_2IrO_4 single crystal, with dimensions 300 x 200 x 30 μm^3. It was grown using a self-flux technique in platinum crucibles,

similar to the one in Ref. 2. We denote a and b the crystal lattice vectors of the superstructure in the IrO_2 planes[4]. Low-resistance contacts were achieved using silver epoxy annealed at 500 C in oxygen atmosphere. Although we present only the data obtained with one crystal, we have checked that the reported behavior is typical of what can be observed in several other similar undoped samples.

Careful investigation of the c-axis resistivity reveals the existence of an anomaly at a temperature close to the reported T_N for the undoped material. The anomaly is actually very small (Fig. 1, inset), but the sharp jump in the resistivity temperature derivative in zero magnetic field unambiguously points towards a well defined phase transition at $T_N = 217 \pm 0.5$ K (Fig. 1). Quantitatively, the evaluation of the resistance change below T_N requires subtracting some arbitrary background, as obtained from the high temperature resistivity, and we have used a linear fit of the resistance logarithm to reveal an *increase* of the resistivity in zero field below the transition (Fig. 1, lower inset). At least close to the transition temperature, little error is likely made due to the sharp transition, and a scaling of the resistivity change may be attempted. This yields, within a ≈ 20 K interval, a scaling exponent 0.55 ± 0.05. We did not observe such an anomaly for the in-plane resistivity. Previous studies did not uncover such a feature, although a tiny specific heat jump could be evidenced at T_N (Ref. 16).

Applying a magnetic field along the c-axis and using the same procedure to uncover the resistivity anomaly shows that there is a "splitting" of the transition with the field (Fig. 1). The *positive* magnetoresistance anomaly shifts to *lower* temperature (as an example, data in Fig. 1 allows to assign a transition temperature, noted T_{3D}, 215.5 ± 0.5 K with $H = 0.2$ T), while a *negative* contribution to the magnetoresistance shows up *above* T_N (as an example, data in Fig. 1 allows to assign a transition temperature, noted T_{2D}, 236 ± 2 K with $H = 9$ T). With the applied field the transition at T_{3D} retains its sharp character at small field, while the transition at T_{2D} appears to be smoother. The existence of the upper branch implies that the transition temperature obtained from high field studies (as in magnetometry) must be overestimated. Data in Fig. 1 suggests, in the present case, that a maximum shift $\Delta T_{2D} \approx 20$ K is obtained for $H \approx 7$ T. With a field applied parallel to the conducting planes, the behavior is found similar, with a transition temperature increase somewhat larger (Fig. 2, open symbols). At T_{3D}, both the sign of the resistance anomaly and the sharpness of the transition point towards similarity with the zero field low temperature phase, but the anomaly is quickly washed out in a small magnetic field (Fig. 1). However, we find that a kink in the magnetoresistance substitutes to this anomaly (Fig. 3), allowing to extend the definition of T_{3D} to low temperature in Fig. 3 (circles). This defines two branches emerging from the (T_N, $H = 0$) point (which we denote $T_{3D}(H)$ and $T_{2D}(H)$, respectively in Fig. 2).

Finally, investigating lower temperatures in the same way shows that a third characteristic temperature may be evidenced, which coexists with the former in the temperature range 80 K $\lesssim T \lesssim$ 175 K (Fig. 3, squares). We denote this temperature $T_{spinflip}(H)$ and also report it in Fig. 2. For all three branches of the diagram, the characteristic magnetic field allowing to cross a branch is found smaller when applied parallel to the conducting plane (Fig. 3). Then, the crossovers observed in the parallel configuration are not a consequence of the sample misalignment that unavoidably occurs (which we estimate a few degrees), but are genuinely induced by the parallel field component. The large negative magnetoresistance that we observe at low temperature is similar to the one reported earlier by Ge *et al* below $T = 100$ K (Ref. 15), although $T_{3D}(H)$ appears to occur at somewhat lower field in our case (for instance, $H \approx 1$ T at $T = 50$ K in the present case, and $H \approx 3$ T in ref. 15). Our data allow to draw the $T_{3D,spinflip}(H)$ lines up to temperatures close to T_N and indicate that the resistance change at small field at the $T_{3D}(H)$ line is similar to the one observed at $T = T_N$ and $H = 0$.

As noticed above, the procedure to evaluate the resistance change at the transition is quite arbitrary, which may be a problem in the case of a smooth variation. Angular dependent magnetoresistance, however, confirm the general trend for $T_{2D}(H)$. Indeed, rotating a large magnetic field around the c-axis reveals the existence of a four-fold contribution to the (negative) magnetoresistance (Figs. 5). The angular-dependent contribution is typically only a few percent of the total magnetoresistance in Fig. 3, and is found maximal along the a and b axis of the crystal. The temperature dependence of the four-fold component extracted in Fig. 5 confirms, with no need for a high-temperature background fit, the existence of an onset at T_{2D}. A two-fold angular component is also present, which vanishes with increasing temperature simultaneously with the four-fold component. However, while the four-fold component temperature dependence appears similar to the one of some order parameter (as for the zero field anomaly), the two-fold component saturates with decreasing temperature.

Finally, we have performed torque measurements on this crystal, $\mathbf{\Gamma} = \mu_0 \boldsymbol{m} \times \boldsymbol{H}$, which provides the magnetization component transverse to the applied magnetic field. Typical results are shown in Fig. 4 for three points in the H-T plane, representative of three distinct regimes in Fig. 2. In the paramagnetic domain, torque is a pure four-fold sine, suggesting a simple anisotropic magnetic susceptibility contribution, as for a two-dimensional system. In the high field regime, a distorted sine indicates that the magnetic moment may no longer be decomposed into two independent linear contributions, which could sign a smooth rotation of the magnetization. Finally, in the intermediate field regime, the torque signal develops a discontinuity (for the data shown, at ≈ 12 deg. from the c-axis), which possibly originates from a spin flip.

III. DISCUSSION

We first comment on the lower line, $T_{spinflip}(H)$. The problem of the observation of a very large magnetoresistance associated to the presence of a weak ferromagnetic state induced by the magnetic field was already encountered in the case of cuprates[6, 17], and received a quantitative interpretation in the case of La_2CuO_4[18]. Two contributions should actually be distinguished: one due to the change in the transfer integrals and linked to the orientation of the IrO_6 octahedra, as proposed in Ref. 15, the other one as a pure spin configuration effect. Could the latter contribute in the present case ? In La_2CuO_4, in the orthorhombic phase, the CuO_6 octahedra are tilted from the CuO planes by $\alpha \approx 3$ deg. (inducing a ferromagnetic component perpendicular to the planes). This allows an antisymmetric superexchange term in the spin Hamiltonian, which would otherwise be zero due to symmetry in the tetragonal phase[6]. In Sr_2IrO_4, IrO_6 octaheadra are tilted in the IrO planes by $\alpha \approx 11$ deg. (inducing a ferromagnetic component in the planes) and the compound is tetragonal. This also destroys the inversion center which exists midway between the Ir atoms, and allows for a non zero antisymmetric superexchange term[4] (Fig. 6). Equivalently, due to the tilt of the IrO_6 octahedra, the transfer integral between one Ir atom and its four nearest neighbors in the next adjacent plane are unequal. As a consequence, interchanging magnetic sublattices, as could be induced by a spin flip, may strongly influence the transverse conductivity in this case also.

In Ref. 18, it was proposed that the transverse resistivity is controlled by the localization length in the 3D variable-range hopping regime (VRH). In the present case, resistivity can be fitted with the conventional expression for three-dimensional VRH, $\rho \propto \exp{(T_0/T)^{1/4}}$, using $T_0 = 6\,10^5$ K (where $T_0 \propto \lambda^{-3}$, and λ is the transverse localization length – see Ref. 18 and Refs therein). This yields for the ratio of the hopping length to the localization radius $l/\lambda \approx (T_0/T)^{1/4} \approx 7$, indicating that charge hopping from impurity centers is controlled by the pure material transfer integrals between sites[18]. Using $\lambda \propto t^{1/2}$, where t is some effective transfer integral between planes, we expect in this case a relative change $\delta\rho/\rho \approx -\frac{3}{8}(T_0/T)^{1/4}\delta t/t$. According to Ref. 18, a flip of the spins required to align ferromagnetic moments in the plane is associated to a change $\delta t/t \approx \frac{1}{2}\,(J_\perp/J_\parallel)(\kappa_\parallel/\kappa_\perp)^4(m_\perp/m_\parallel)^2$, where $m_{\parallel(\perp)}$ is the in-plane (transverse) effective mass, $J_{\parallel(\perp)}$ is the in-plane (transverse) exchange coupling, and $\kappa_{\parallel(\perp)}$ is the corresponding reciprocal lattice constant. We have $\kappa_\parallel/\kappa_\perp = 1.8$, $J_\perp/J_\parallel \approx 10^{-5}$ (a value comparable to that for La_2CuO_4, Ref. 19) and $m_\perp/m_\parallel > 20$ (this is evaluated by the ratio of the bandwidth for J=1/2 along ΓX and NC[12]). This yields $\delta\rho/\rho = \frac{1}{2}\,\delta t/t > 6\,10^{-2}$. Though this is only a rough estimate and magnetic configurations for both cases are different, this illustrates that an effect comparable to the one that we observe may be expected from the spin contribution alone. We expect this contribution to be significant at the $T_{spinflip}(H)$ line, where there is a field-induced ferromagnetic moment[15]. Then, within this hypothesis that the resistivity change results from the larger transverse transfer integral associated to magnetic ordering, we may tentatively relate the observed scaling of the resistivity to a critical exponent. We expect the resistance change to be proportional to the phase transition order parameter associated to interplane spin ordering, M (this may be assumed in the framework of a two-fluid model, for which there is an amount $n \propto M$ of ordered moments associated to a larger transfer integral). As a result, the scaling exponent for the resistivity is identical to the conventional exponent β for the order parameter. The value obtained, $\beta \simeq 0.55$, is close to the one for a mean field type transition ($\beta = 0.5$). In Ref. 19, the transverse fluctuation correlation length above T_N yielded a critical exponent $\nu = 0.75 \pm 0.05$. This value is far off the mean field value ($\nu = 0.5$). This discrepancy could sign the limit of the present analysis for the resistivity scaling, made within a simple static picture. Also, the scaling in Ref. 19 relates to long range correlations ($\simeq 3-20\ c$), while we expect resistivity to be essentially driven by magnetic correlations at the scale of the interplane distance.

We now comment on the upper line, $T_{3D}(H)$. In Ref. 15, it was proposed that the occurrence of a large magnetoresistance with no relevance to the magnetization could be interpreted as a the result of the concomitant rotation of the spins in a spiral configuration under applied field, and of the Ir-O bond (due to spin-orbit coupling[5]), charge hopping being in turn controlled by the orbitals configuration. While our data do not allow to conclude on the validity of such a scenario, our data brings some new insight on the nature of the $T_{3D}(H)$ line. Results in Fig. 1 suggest that $T_{3D}(H)$ retain the second order phase transition character of the zero field transition. In particular, crossing this branch, the resistance change is observed *positive* (although the overall magnetoresistance is negative, see Fig. 3). Measurements of the angular dependence of the magnetoresistance along constant T and H intervals crossing $T_{3D}(H)$ bring additional information (Fig. 7). The striking features of such results are: i) a large angular susceptibility of the transverse magnetoresistance develops at the crossing of the line along the a and b directions; ii) just below this line (at 200 K in Fig. 7), there is a two-fold periodicity (we have checked, deliberately tilting the crystal and observing no qualitative change in the $R_c(\theta)$ behavior, that this cannot be due to the sample misalignment), and the peaks in angular susceptibility are hysteretic.

We propose that a magnetic ordering transition along c-axis may account for these observations. Indeed, such a transition would have a large effect on the transverse resistivity by the virtue of the mechanism described above. Then, ferromagnetic domains being linked to the possibility to order magnetism from plane to plane, it is natural to expect their signature to show up below this transition, as well as some potential a/b unbalance due to inequiva-

lent domains. Finally, the susceptibility to the magnetic field at the transition should be the larger when the field is applied along one of the two possible directions for the ferromagnetic moment. The scenario in Ref. 15 proposed that a c-axis field induces an in-plane rotation of the spins, while at a constant angle from the c-axis. If so, we expect a reduction of the ferromagnetic component with the applied field. It is then possible that this reduction favors magnetic decoupling along the c-axis. To the credit of our picture, preliminary measurements on $Sr_{2-x}La_xIrO_4$ indicate – beside the known T_N decrease – that the $T_{3D}(H)$ line is suppressed in zero field (*i.e.* the positive transverse resistivity anomaly is replaced by the negative one, as in an applied field). This suggests that disorder may also contribute to weaken the c-axis magnetic ordering, just as for $Sr_2Ir_{1-x}Rh_{x>0.03}O_4$ exhibits a modified c-axis magnetic ordering to the so-called 'AF-II' phase with ferromagnetic inter-plane order[20]. The observation of a smooth crossover distinct from the sharp transition to the 3D ordered state seems also to corroborate the one that the in-plane magnetic fluctuation correlation length decreases above T_N much less rapidly than the out-of-plane one, a feature interpreted as the consequence of a two-dimensional S= 1/2 quantum Heisenberg model[19].

The overall picture would then be the following: in zero field, we observe a single transition temperature at T_N were 3D magnetic order establishes. Applying a magnetic field splits the transition into two branches: a near vertical branch at $T > T_N$ at which AF order is established in the plane, and a low temperature one, where a long range c-axis magnetic order is established. A second line in the ordered phase delimits field-induced ferromagnetic interplane ordering. We propose that magnetotransport is influenced strongly by direct spin reorientation effects, in addition to bond reorientation ones. Our data clearly contradicts the general belief that there would be no influence of the magnetic state on the transport properties. We have proposed that this influence should be evaluated here considering hopping of the localized charge.

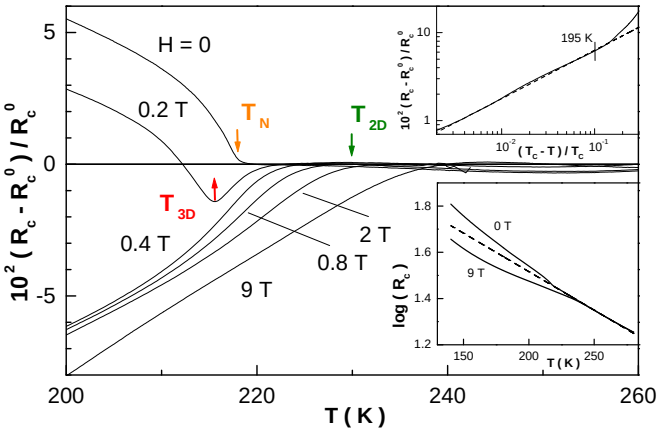

FIG. 1: c-axis resistance anomaly, as obtained subtracting a high temperature linear logarithmic resistance (dashed line in lower inset). Upper inset: scaling of the resistive anomaly. The magnetic field is applied along the c-axis.

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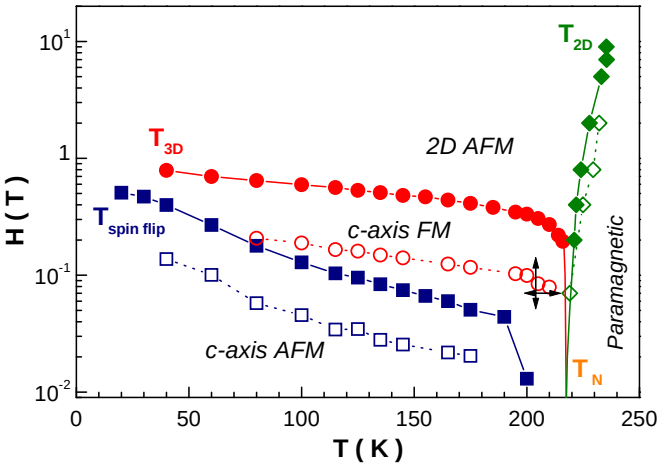

FIG. 2: Phase diagram for the transverse resistivity, as obtained from the data in Fig. 3 (squares and circles) and the onset in Fig. 1 (diamonds). Filled symbols are for field applied along the c-axis; open ones, in the a-b plane. T_N is the zero field transition temperature, as given by the data in Fig. 1.

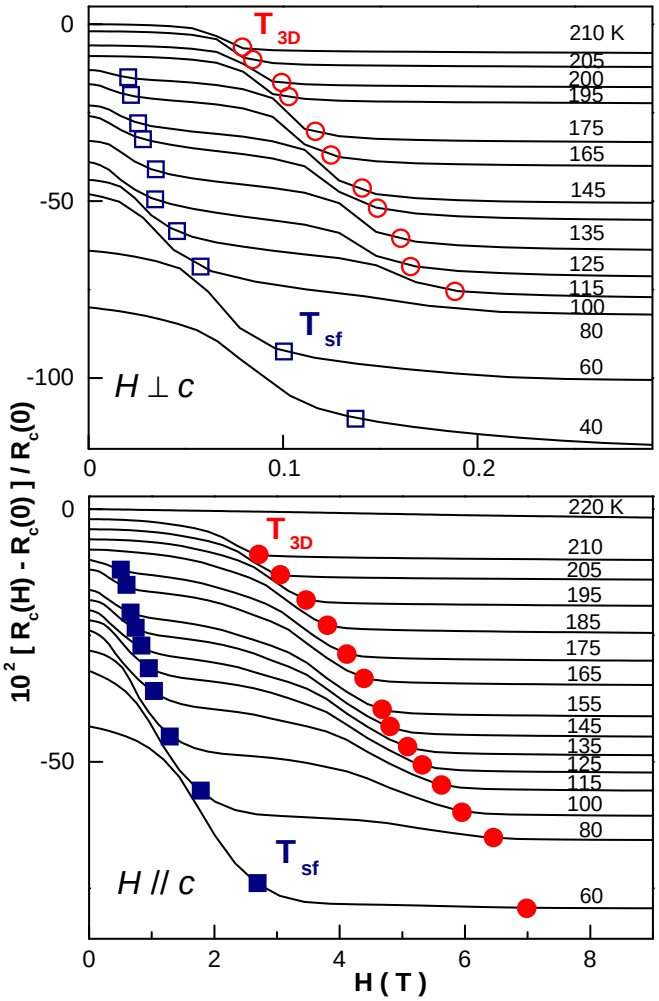

FIG. 3: c-axis magnetoresistance, for field along (lower panel) and perpendicular to c-axis (upper). Symbols mark the characteristic fields in Fig. 2. (curves have been shifted from their $H = 0$ zero value, for clarity)

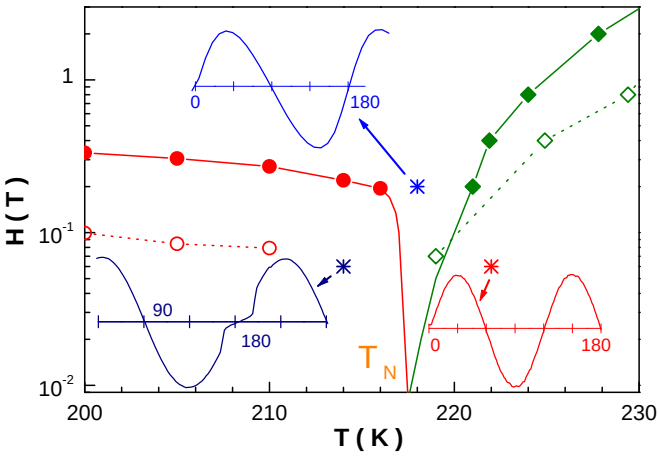

FIG. 4: Torque (Γ) angular dependence at typical locations in the H-T plane. Magnetic field is rotated in the $a(b)$-c plane. The angle is the one from the c-axis. Symbols are from Fig. 2.

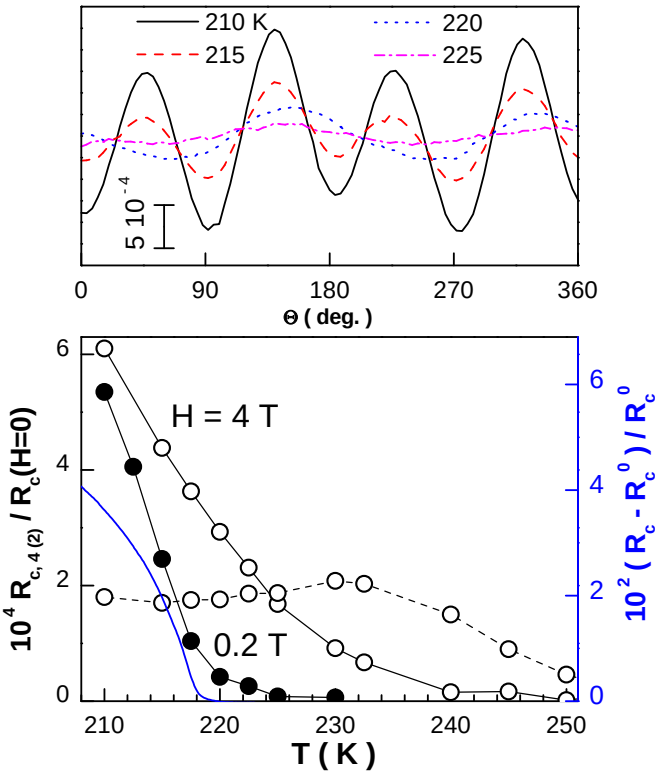

FIG. 5: Upper panel: angular dependence of the magnetoresistance (in-plane field $H = 0.2$ T); θ is the angle between $a(b)$ and the magnetic field. Lower panel: Symbols: four-fold and two-fold (dotted line) components of the angular c-axis magnetoresistance (left scale). Full line: zero field c-axis resistance anomaly, as in Fig. 1

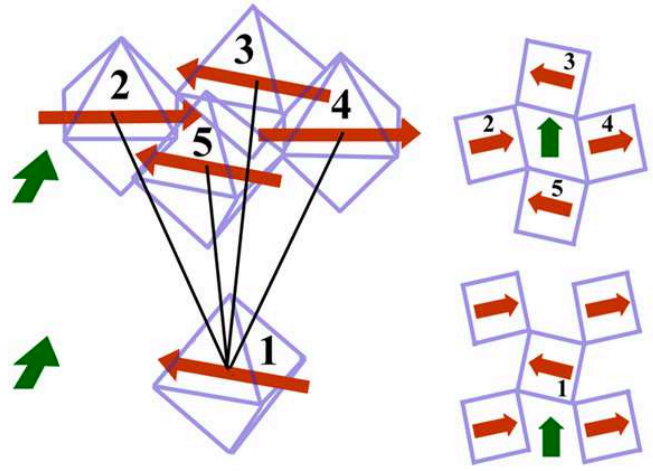

FIG. 6: With ferromagnetic coupling of the in-plane magnetic moment, as shown (green arrow), transverse hopping occurs within a magnetic sublattice of equivalent Ir, 1-5-3. Antiferromagnetic coupling is obtained reversing the spins in one layer, and hopping within a sublattice occurs between inequivalent 1-2-4.

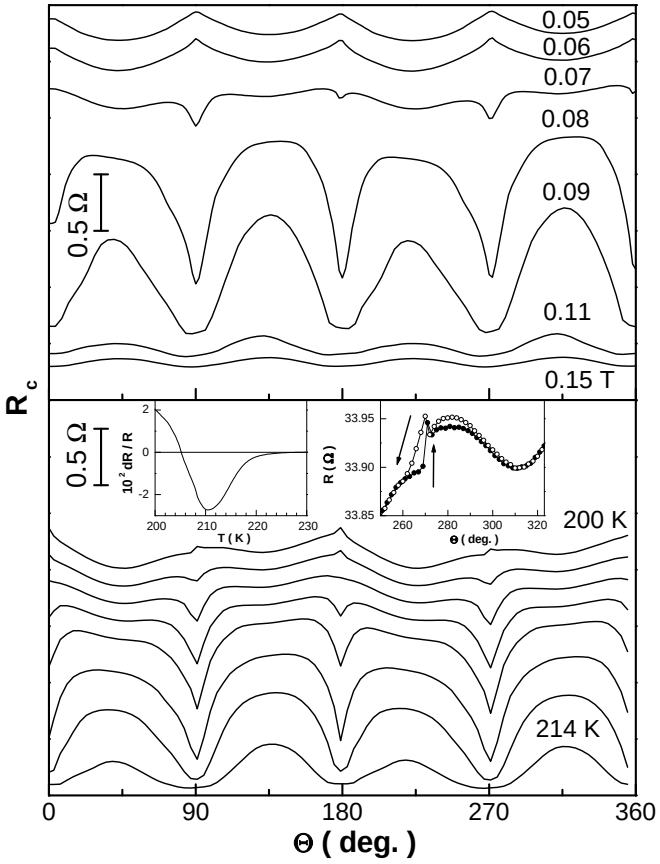

FIG. 7: Angular dependence of the magnetoresistance along the two segments in Fig. 2 (field in the a-b plane). Upper panel: $T = 204$ K. Lower panel: $H = 0.07$ T (T varies in steps of 2 K); left inset: resistance anomaly in zero field, obtained as in Fig. 1; right inset: occurrence of a uniaxial hysteretic feature ($T = 200$ K).