Research in condensed matter magnetism has mainly focused on materials incorporating elements of the 3d transition-metal series (Fe, Co.) and elements of the 4f lanthanide rare earth series (Nd, Tb, Er...). The 3d and the 4f electrons have very different magnetic properties. Both properties can be combined to obtain a great variety of magnetic phases and to make outstanding permanent magnets, like Nd$_2$Fe$_{14}$B, used industrially in numerous devices. Now, new and fascinating territories are being explored using the 5d transition-metal elements – such as iridium – two rows below the 3d elements in the Mendeleev table.

The 5d electron shell has much more spatial extension than the 3d shell, which at first sight would imply more-delocalized electrons with weaker electronic correlations leading only to trivial paramagnetism. However, the 5d electrons are characterized by much larger spin-orbit interactions that strongly increase with the atomic number Z. It follows that the spin and orbital degrees of freedom are strongly entangled (mixed quantum mechanically), as pictured in Fig. 1. As a matter of fact, the spin-orbit interactions become comparable to the electron-electron correlations and to the crystal-field interactions. Consequently, one can expect unusual metal-insulator transitions, electronic phases with novel topological properties (meaning that they cannot deform continuously into standard electronic phases), or spin-orbit entangled magnetism producing exotic magnetic states and excitations.

We are currently studying a family of oxides with formula R$_2$Ir$_2$O$_7$. The iridium ions (Ir) and the rare earth ions (R) form two interpenetrating lattices of the “pyrochlore” type within the crystal. Each of these lattices consists of tetrahedra joined by their corners (e.g. Fig. 2a shows the Ir sublattice).

It is known that the pyrochlore iridates R$_2$Ir$_2$O$_7$ can undergo an electronic metal-insulator transition, driven by the iridium spin-orbit interaction. This occurs at the same time as the magnetic ordering of the iridium sublattice. Besides, these compounds have been predicted to host a novel type of electronic phase below this transition, called a Weyl semi-metal. An experimentally observable signature of such a phase would be the occurrence of a peculiar “all-in all-out” magnetic order of the iridium sublattice at the metal-insulator transition. That is, all the iridium magnetic moments would be pointing either toward or away from the centre of each tetrahedron (Fig. 2a).

Unfortunately, the magnetic order in the iridates is very difficult to probe since the iridium magnetic moment is tiny. To overcome this problem, we have chosen to probe the iridium magnetism via its influence on the magnetic rare-earth. We have thus decided to compare two different rare-earths, Terbium (Tb) and Erbium (Er), which have very different magnetic anisotropy properties: In terbium iridate Tb$_2$Ir$_2$O$_7$, the terbium ions’ magnetic moments want to align along their local <111> directions (i.e. along the diagonal directions joining the corners and the centres of each tetrahedron (Fig. 2)). In Er$_2$Ir$_2$O$_7$, the erbium ions moments want to lie in a plane perpendicular to the <111> directions.

We have done magnetization measurements and neutron scattering to investigate these two compounds. We find that the Tb sublattice in the terbium iridate develops an all-in all-out magnetic order below 40 K. On the other hand, in the erbium compound, the Er sublattice remains disordered down to our lowest measurement temperature of 80 mK.

This contrasting behaviour is explained by the interaction between the iridium and the rare earth magnetic moments. The all-in all-out order of the Iridium moments produces a local magnetic field along the <111> directions on each rare earth ion site (Fig. 2). This magnetic field is compatible with the magnetic anisotropy of terbium, the Tb magnetic moment getting aligned along the field in the induced all-in all-out configuration (Fig. 2). This indirectly proves the all-in all-out order of the iridium in this pyrochlore. On the contrary, the magnetic field produced by the iridiums is competing with the erbium’s magnetic anisotropy, thus preventing magnetic ordering of the erbiums. This is again compatible with the all-in all-out order of the iridiums.

From this novel approach to the iridium’s magnetism, through the induced magnetic behaviour of the rare-earth, our experiments have firmly established the all-in all-out order of the iridium sublattice. This special type of order is a necessary condition for the occurrence of exotic electronic transport properties, to be searched for in future work.

Fig. 2 (a): The “all-in all-out” ordering in the iridium sublattice of R$_2$Ir$_2$O$_7$.
For any given tetrahedron of iridium ions, all four iridium magnetic moments either point in or point out together along the local <111> directions.
(b) The six Ir nearest-neighbours are situated on a hexagon, surrounding a central Terbium ion. Their magnetic moments, in the all-in all-out configuration, all have a component pointing out of plane (+ direction), thus producing a magnetic field at the Tb site along the <111> direction.