

Report on my stay in the SSPC Laboratory in Division of Chemistry, Graduate School of Science, Kyoto University

Rafik BALLOU

Institut Néel, Centre National de la Recherche Scientifique (CNRS) & Université Grenoble Alpes (UGA), 25 Av. des Martyrs, BP 166, Grenoble F-38000, France
E-mail: rafik.ballou@neel.cnrs.fr

Abstract. This report provides an overview of my scientific activities during my stay financed by the International Research Unit of Advanced Future Studies (IRUAFS) as Distinguished Visiting Professor in the Solid State Physics and Chemistry (SSPC) Laboratory in the Division of Chemistry of the Graduate School of Science at Kyoto University. I describe briefly the scientific themes that we discussed in depth, by emphasizing the prospects of future collaboration. A first part pertains naturally to our common field of expertise that includes metallic magnetism, geometric frustration in magnetic networks and topological matter. The second part describes our attempts, embryonic to this stage, to tackle transverse topics in adequacy with the transdisciplinary objectives of the IRUAFS. A first is concerned with quantum entanglement across the time that raises issues of retrocausality. The second is about the physics of the sound of the sanukite stones the magic of which appears to have been poorly investigated. I kept the same spirit, though rather multidisciplinary than transdisciplinary, in my talk at IRUAFS by describing not only my recent works on geometric frustration but also my involvement to an experimental project of search for ultra-light components of dark matter.

Keywords: Itinerant Magnetism, Geometric Frustration, Topological Materials, Weakly Interacting Ultra-Light Particles, Quantum Entanglement and Retro-Causality, Sanukite Stones.

1. Scientific Activities

I worked at the Solid State Physics and Chemistry (SSPC) “Kinso” Laboratory from December 4th, 2017 to January 30th, 2018. I would like to express my deep appreciation to the Division of Chemistry of the Graduate School of Science at Kyoto University for hosting me, to the International Research Unit of Advanced Future Studies (IRUAFS) for providing financial support, and to Professor Kazuyoshi Yoshimura, head of the Kinso Laboratory, for inviting me. During these two months I have had many an opportunity to scientifically interact with Prof. Kazuyoshi Yoshimura, Assoc. Prof. Hiroaki Ueda, Assist. Prof. Chishiro Michioka, and their team of PhD, Master and Undergraduate students. I greatly appreciated in particular to have been invited by the latter to participate to their “Journal Club” that takes place every Friday morning, during which one of them presents works reported in the literature in connection with his topics of research that are then discussed in depth by

the audience. I thank them to have switched from Japanese to English to allow me to follow the presentation and to contribute to the debates. Needless to emphasize that this approach of the literature is extremely enriching for the students as well as for the seniors. Assist. Prof. Chishiro Michioka kindly provided elaborate answers to all my questions on the various physical measurements performed within his team. I thank him for having taken time to show me and to describe in detail their different experimental means, in particular their NMR facilities. I also visited their Chemistry Laboratory and several other experimental installations that they share with other teams, for structural studies, including SEM and TEM, and for magnetization (MPMS) or heat capacity (PPMS) measurements. Assoc. Prof. Hiroaki Ueda was of precious advice concerning all the aspects of the often-complex chemistry inherent to the preparation of poly-crystals or single-crystals of the systems that aroused our interest. I would like to emphasize the relevance of this crucial stage of an experimental project without which it might even fail to start. Prof. Kazuyoshi Yoshimura initiated with me a series of formal and informal discussions on different topics pertaining to our common field of expertise but also well beyond, which have paved the ground of possible wider future collaborations.

I share with the team of Prof. Yoshimura a strong interest for the experimental investigation of the novel topological electronic states that might be awaited from the strong spin-orbital entanglement of the metallic electrons and of the magnetic phases always so fascinating that might arise from the geometrical frustration in a variety of magnets. It was, in these domains, almost immediate to find topics of common interest. I initially focussed my attention at iridium-based oxides following my recent investigations of the pyrochlore oxides with formula $R_2Ir_2O_7$, where the trivalent Rare earth (R) ions and the tetravalent Iridium ions form two interpenetrating networks of corner sharing tetrahedra, each potentially a generator of geometric frustration. A motivation of the study besides surprises awaited from the geometric frustration was that the Iridium 5d electrons are showing an electronic transition that could be associated with the stabilisation of a Weyl semimetal. A necessary condition is that the electronic transition is accompanied with a magnetic order that breaks solely the time reversal symmetry. A main experimental obstacle of the study was the difficulty to grow single crystals of sufficient size in which to directly explore the electronic state and magnetic order on synchrotron sources. We however succeeded to demonstrate indirectly that Iridium do order in the “all-in/all-out” magnetic structure, which is the only one preserving the whole crystal symmetry, through the influence this order exerts through the R-Ir exchange interactions on the R magnetism, which gave well interpretable signals in neutron scattering experiments on polycrystals. Novel magnetic behaviours occurred at very low temperature when the exchange interactions between the R ions become relevant and are in competition against the exchange field from the Iridium network. We found out for instance that the outcome of this conflict in $Ho_2Ir_2O_7$ is a fragmentation of the Ho magnetic moments that generates a monopole crystal coexisting with a spin ice, a state of fluctuating moments correlated by the constraint “2-in/2-out”. This is a quite unusual phase, where a same degree of freedom contributes thermodynamically to a solid and a fluid, which might be dubbed a “thermodynamic Schrödinger Cat” [E. Lefrançois *et al.* Nature Comm. 8 (1), 209 (2017)]. I discussed these results in the first part of the talk I gave at the IRUAFS. I hoped that my hosts could propose alternative methods to grow single crystals so as to examine the fascinating behaviors of the series in more details, but it seemed during our discussions that we already had exhausted all the possible attempts. The recent works of the team of Prof. Yoshimura on the iridates $ZnIrO_3$ and $MgIrO_3$ also strongly aroused my interest. In these compounds the Iridium ions form a honeycomb lattice and the O-Ir-O bonds form an angle close to 90° from which directional exchange interactions are awaited. Notice that in the $R_2Ir_2O_7$ oxides the O-Ir-O bonds form an angle close to 180° giving rather rise to a pseudo-dipolar contribution to the exchange interactions. The ideal model of directional exchange on the honeycomb lattice was exactly solved [A. Kitaev, Ann. Phys. 321, 2 (2006)]. According to the strength distribution of the exchange interactions it leads to either a gapped quantum spin liquid carrying abelian anyon excitations or a gapless phase that becomes gapped under magnetic field and carries then non-abelian anyon excitations. None of these fascinating phases was observed in $ZnIrO_3$

and MgIrO_3 . An antiferromagnetism in the first compound and a canted antiferromagnetism giving rise to a weak ferromagnetism in the second compound are instead inferred from the magnetic measurements on magnetic field oriented polycrystals, most probably because of additional first neighbor isotropic exchange interaction, next neighbor frustrating isotropic and antisymmetric interactions, as in many other similar systems. These magnetic phases, especially the canted antiferromagnetism, are however intriguing. It would be of utmost interest to probe to what extent they would show reminiscences of the spin liquid phases by carrying excitations with anyonic features, what of course would be best investigated on single crystals together with the exact magnetic orders. Unfortunately again the chemistry is again complex and delicate, proceeding through a metathesis reaction that leaves few chances to grow a single crystal with sufficient size. We can however anticipate performing powder elastic and inelastic neutron scattering and X-ray scattering at the Ir L_{III} edge on a synchrotron source. Besides iridium-based oxides I got also strongly interested by their experimental investigations of fluorides of formula $A_2BM_3F_{12}$ ($A = \text{Cs, Rb}$; $B = \text{K, Na}$; $M = \text{Ti, V, Cr}$) where the 3d-metal ions M^{3+} form distorted Kagomé networks of quantum spins $S = 1/2$ (Ti^{3+}), 1 (V^{3+}), $3/2$ (Cr^{3+}) with local anisotropy for the latter. Characteristic plateau of magnetization were measured in all the compounds signaling the occurrence of incompressible phases of quantum nature [M. Goto *et al.*, Phys. Rev. B94, 104432 (2016) & M. Goto *et al.*, Phys. Rev. B95, 134436 (2017)]. The magnetic orders are inferred from magnetization data. It would be strongly relevant to complement the study by directly probing the magnetic orders and excitation spectra, since single crystals are this time available.

Although in the first instance I mostly discussed with Prof. Yoshimura about topics belonging to the field of condensed matter physics we soon widened our discussions to transverse topics. In particular he showed a strong interest for my involvement in the OSQAR experimental project developed at the CERN - the European Organization for Nuclear Research - in Geneva, to search for weakly interacting ultra-light particles predicted in many extensions of the Standard Model of Particle Physics. The implemented methodologies make use of laser propagation in magnetic field, exploiting the coupling of the searched particles with photons. To a certain extent they consist in measuring the magnetic birefringence, magnetic dichroism and even magnetic phosphorescence of the quantum vacuum or to produce magnetic photoregeneration beyond an optical barrier [R. Ballou *et al.* Phys. Rev. D92, 092002 (2015)]. A strong motivation to search for these ultra-light particles is that, produced through the misalignment mechanism, they might exist with a large density and thereby significantly contribute to the Dark Matter content of the Universe. According to Prof. Yoshimura the topic might interest a wider audience. I followed his suggestion to add it as second part of my talk at the IURAFS.

We opened fruitful discussions on the concept of entanglement of quantum states. It imposes itself naturally in our fields of investigations by generating subtle distinctions in the large variety of the spin liquids awaited in spin networks with or even without geometric frustration and by providing much deeper insights about their specific features. It is especially of relevance for those phases now named quantum spin liquids that distinguishes themselves through states incorporating long-range entanglement, what leads to emergent topological orders and potentially to fractional excitations with braiding statistics. We wished to explore it in a wider context in some of its numerous puzzling avatars that continue to challenge our perceptual understanding of the quantum world and to raise foundational philosophic issues. It is far from evident to provide an intuitive picture of its substance. As a matter of fact it would turn out that even the quantum vacuum would be an entangled state the concrete signatures of which might be found for instance in the Unruh-Davies radiation for accelerated observers in Minkowski space-time [Phys. Rev. D **14**, 870 (1976)] or Gibbons-Hawking radiation in de Sitter space-time [J. Phys. A **8**, 609 (1975), Phys. Rev. D **15**, 2738 (1977)]. It would follow that the shape of the universe might then be “heard through its quantum (tensorial) noise” [A. Kempf, Found. Phys. **44**, 472–482 (2014)] and that the vacuum might show a topological order out of which fermions statistics and gauge interactions would emerge similarly than in quantum spin liquids with long-range entanglement [M. Levin and X.-G. Wen, Phys. Rev. B67 (2003) 245316]. We chose to focus more on

its temporal aspects. A sequence of non commuting operations on a given component of a maximally entangled pair produces, in case the entanglement were to be preserved, a composite state on the second component that can be obtained from exactly the same sequence of operations, but in a reversed order. It follows that time protocols can be implemented whereby future actions might affect past events what naturally raises issues of retrocausality. Entanglement itself can take place across the time what was experimentally demonstrated, for instance through its swapping between two photons that have never coexisted [E. Megidish, Phys. Rev. Lett. 110, 210403 (2013)]. It appears that it has so far not been analyzed as such extensively probably because the tensor product structure is then hardly useful. It must be dealt with in alternative formalisms in order for instance to determine if it shows the monogamy property or to derive Tsirelson bounds [M. Nowakowski, arXiv:1701.08116]. Even more strange protocols or phenomena might then be expected such as quantum teleportation across the time or even quantum resonance across the time. No logical paradox should ensue since teleportation requires a classical signal to be sent from the past to the future whereas by the no-signaling theorem no physical signal can be sent through an entanglement so that it cannot be used to communicate with the past. Nevertheless, an action on the future component of an entanglement will necessarily have an effect on its past component whereas a quantum resonance creates a past-future bridge. Although incoherent noise and intrinsic decoherence generally limit the potentialities of these phenomena to occur at our macroscopic levels, though not systematically, we wondered how philosophes, psychologist, ... or even poets and musicians would perceive this fascinating world of entanglement. In return we might certainly learn from them to get from it a perceptually deeper vision.

I had the opportunity to meet at a dinner the famous musician Stomu Yamashita. I learned at this occasion from Prof. Yoshimura that he is playing on stones percussion instruments what appeared to me unprecedented. The stones are volcanic rocks called Sanukite (“Kankan Ishi”) the sound richness of which is impressive and is ascribed to the high velocity of its P-wave despite a moderate density. Quite surprisingly the literature is limited to their geological origin, X-ray fluorescence analyses [T. Higashimura and T. Warashina, J. of Archaeo. Sc. 2, 169-178 (1975) & T. Warashina, Y. Kamaki and T. Higashimura, J. of Archaeo. Sc. 5, 283-291 (1978)] and a numerical study by the finite element method of the percussion instrument “hokyo” [K. Kishi, H. Meda, M. Sugai, Acoust. Sci. & Tech. 22, 3 (2001)]. We believe it is essential to complement these studies through additional characterizations of the materials (textures by scanning electron microscopy, etc.) and above all through studies of the influence of the shape of the instrument on the sound structure, which are worth attempting even if they finally reveal themselves so complex that we fail to unveil the sound magic of the sanukite stones.

2. Outputs and Prospects

The scientific discussions I had with the team of Prof. Yoshimura on the physics associated with geometric frustration and topological non-trivial phases were fruitful and should certainly result in active collaborations on common experimental studies beyond those anticipated above on the Ir-based oxides with honeycomb network of spins and on the fluorides with distorted kagomé network of spins.

I also considered with Prof. Yoshimura the present theoretical and experimental status of metallic magnetism, unfortunately perceived as old fashioned sometimes. It appeared in contrast to us that interesting physics should be learned from the exploration of the spin fluctuations spectra in the widest ranges of energy. A possible common experimental work in this direction is not completely excluded.

We hope to continue our discussions about quantum entanglement. We shall keep its informal nature, though we do hope to concretely investigate it in its long-range avatar by finding out a quantum spin liquid. An experimental investigation of the sanukite stones is under consideration perhaps in collaboration with experts of sound propagation in heterogeneous solids and hopefully with a musician.