RIKEN-RAL MUON FACILITY

FOURTH INTERNATIONAL ADVISORY COMMITTEE REVIEW REPORT



7-8 April 2014



Caption to Figures on cover page (clockwise from top left): candidate muon sites in La_2CuO_4 calculated using DFT techniques; ordering pattern driven by dipolar interactions for $Fe_{19}(m)$ (from F.L. Pratt et al, Phys. Rev. B, 89 144420 (2014)); magnetic and diffusive properties of LiFePO₄ investigated using μ SR, with predicted muon sites shown in the Figure (from J. Sugiyama et al, Phys. Rev. B, 84 054430 (2011)); RIKEN-RAL and ISIS joint facility workshop held at The Cosener's House, Abingdon, April 2014.

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The Fourth RIKEN-RAL International Advisory Committee (L to R): Dr Stephen Cottrell (Secretary), Prof Yasuyuki Matsuda, Dr Jun Sugiyama, Prof En'yo, (Director of the RIKEN Nishina Centre), Dr Andrew Taylor (Chair), Dr Jean-Michel Poutissou, Prof Dr Klaus Jungmann, Prof Roberto De Renzi

1. Executive Summary

The RIKEN-RAL International Advisory Committee (IAC) met 7-8 April 2014 at STFC Rutherford Appleton Laboratory in the UK. The aim of this fourth meeting, building on the findings of the first three IAC reports, was to evaluate the response of the RIKEN-RAL facility to the previous IAC recommendations, review scientific activities of RIKEN-RAL since the last report; consider whether the facility is world leading in its field; evaluate the direction of RIKEN-RAL through RIKEN's fourth five-year term and recommend any aspect of the RIKEN-RAL facility that needs strengthening. The committee consisted of both international and Japanese experts – members are listed in Appendix 1.

The committee heard a variety of reports on activities at the facility, together with documentary evidence relating to facility work, including publications and annual reports. The committee's agenda is given in Appendix 2. The following sections of this report give more detailed findings and recommendations for each of the various science areas covered by facility activities. The broad findings and recommendations are summarised below.

1.1. Findings and Recommendations of the Fourth IAC Meeting

The IAC continues to endorse its conclusions and recommendations from previous reports, in which two key science pillars for the facility – condensed matter and molecular science, and ultra-slow muon generation, were highlighted. Key findings from this fourth IAC meeting are as follows:

The *condensed matter and molecular science programme (pillar 1)* continues to be a very strong area for RIKEN-RAL and a central pillar of its activity. A *diverse science programme* has continued, studying a *broad range of topics* including magnetism, superconductivity, ionic diffusion, and spin and charge dynamics. The *unique capabilities of the facility*, providing high pressure and laser stimulation in combination with a pulsed beam, enhance this programme. The facility is *very productive*, with over 24 high quality journal papers published in the 2011-2014 period. There is strong evidence from the portfolio of highly cited work presented to the committee for *scientific excellence* in the output of the facility that is *clearly world leading in this field*.

The *ultra-slow muon project (pillar 2)* has progressed significantly with the development of a high intensity Lyman- α laser system coupled with the discovery of enhanced muonium production from fine-drilled laser-fabricated room-temperature silica aerogel targets. Together, these improvements can realistically be expected to provide almost an order of magnitude increase in slow muon flux, and it is possible to envisage a science programme based upon this source.

The new initiative for the *measurement of the hyperfine-splitting in muonic hydrogen* promises to become a high profile project. The determination of the proton radius has become an increasingly interesting topic. The IAC notes that for rapid progress of this high profile work, *the project will require a sufficient number of dedicated personnel* with expertise to collaborate across the different scientific and technical areas needed for its success. The IAC also recommend that the impact of this activity on the existing programme pillars should be considered, and that a detailed proposal be developed for the experiment which can be subject to external review.

The IAC are enthusiastic about *chip irradiation experiments* which are a new opportunity since the previous IAC meeting, and see significant potential to grow this activity in the future. The committee suggest links with industry are explored, and see great benefit in promoting this type of facility at ISIS where there is complementarity with the existing neutron programme.

The IAC *applauds the success of the RIKEN-RAL facility in establishing a broad range of national and international collaborations*. In particular, the committee noted the benefit of reaching out to potential user communities in Asia, such as in Malaysia, Indonesia and more recently in Korea.

The IAC recognises the need for *detailed planning for the future of the RIKEN-RAL facility beyond 2018*, and encourages discussion with ISIS as soon as possible.

The IAC reiterates its *concern with respect to the staffing level* at the RIKEN-RAL facility; in some areas, particularly condensed matter and molecular science, there is a chronic shortage of staff to maintain the experimental programme. In the absence of permanent staff positions being made available by RIKEN, the committee recommends the facility explores ways of attracting long term visitors or staff to support the experimental programme. The committee is convinced that the proton radius experiment will attract new resources and *recommends that support for the \muSR programme is developed in collaboration with the ISIS facility*.

2. Introduction

2.1. Aims of the Review

Based on terms of reference provided by RIKEN President Prof. Noyori, the charge given to the IAC by Prof.z En'yo, Director of the RIKEN Nishina Centre, was to:

- 1. Evaluate the response of the RIKEN Facility at RAL to the recommendations made by the last RAL-IAC.
- 2. Are the research outputs from the RIKEN Facility at RAL up to international standards? Is the RIKEN Facility at RAL a world-leading facility in its field and also in interdisciplinary research?
- 3. Evaluate the direction of the RIKEN Facility at RAL towards RIKEN's 4th Five-Year Term.
- 4. Recommend any items that need to be further strengthened in the RIKEN Facility at RAL.

The structure of this report is based on these terms of reference.

This report is presented to the President of RIKEN through the RIKEN Nishina Centre Advisory Committee.

3. Evaluation of Findings and Responses to Recommendations made by the Third IAC

The IAC evaluated the detailed response to the recommendations made at the previous IAC review meeting, together with evidence relating to facility work. The outcome is summarised below with the previous recommendations summarised at the start of each section in italics.

3.1. Condensed Matter and Molecular Science – Pillar 1

The IAC continues to find that this is a successful, diverse and productive programme, with many collaborating groups from Japan and overseas. It welcomes the provision of a new spectrometer (Chronus) for μ SR studies.

However, the Committee was concerned about the staffing level, the temporary nature of personnel associated with this part of the programme and the rapid staff turnover. There will soon be two spectrometers available for the condensed matter and molecular science user programme once Chronus is fully commissioned. Based on the ISIS model for providing support for instruments within its neutron and muon user programme, the Committee would expect to see four scientific staff available to support the science on the RIKEN-RAL spectrometers, in addition to technical support. The Committee therefore urges RIKEN to provide staff to support the programme at a level appropriate to ensure its ongoing success.

The IAC find that the condensed matter and molecular science programme continues to be a *very strong* area for RIKEN-RAL – a central pillar of its activity. The group is delivering a *diverse and compelling* science programme across a broad range of topics (including magnetism, superconductivity, ionic diffusion, and spin and charge dynamics). The publication record is good, with a total of over 250 journal papers from work in this area – a record that is comparable to that of the ISIS European muon facility. One example of high interest science is the recent study of URu₂Si₂ heavy fermion superconductor (using a sample that cannot easily be measured at J-PARC); results are currently being prepared for publication in Physical Review Letters.

The IAC noted that the RIKEN-RAL facility provides ISIS with a unique capability, complementary to that available on the ISIS European muon beamlines. Both pressure studies and μ^- experiments are made possible because of the availability of the decay beam. Laser stimulation experiments were developed at the facility, and this work has been the foundation for a large grant awarded to Alan Drew (Queen Mary University of London) to develop the technique on the high field instrument (HiFi) at the ISIS European muon facility.

The IAC welcomed completion of commissioning of a second high data rate spectrometer, Chronus, which is now becoming available to users. Use of a standard ISIS DAE-II system, which will greatly facilitate on-going technical and user support, was appreciated. The instrument is located in Port 4 of the facility and will take muons in parallel with the existing ARGUS spectrometer, effectively doubling the capacity of the facility for condensed matter experiments. There was concern, however, that this will put even more pressure on the supporting staff, limiting its optimal use for outstanding science. The IAC reiterates its concern with respect to the staffing level at the RIKEN-RAL facility. In the absence of permanent staff positions being made available by RIKEN, the committee recommends the facility explores ways of attracting long term visitors or staff to support the experimental programme.

3.2. Ultra-Slow Muon Development – Pillar 2

The IAC notes the continued activity in this area, for development of a low energy muon source for materials studies and g-2 measurements. The activity at RIKEN-RAL provides a necessary stepping-stone to enable future implementation at J-PARC. The Committee recommends putting in place an action plan, with milestones and resource allocation, to ensure continued future development. Staffing levels for the development activities seem adequate; however, the committee recommends identification of a science leader for the future materials programme.

Collaborative work with groups within RIKEN has enabled the development of a new highintensity Lyman- α (VUV) laser system through the synthesis of a novel ceramic-based Nd:YGAG crystal. Experiments at TRIUMF have investigated muonium production and, in a recent breakthrough, huge efficiency gains have been demonstrated through the use of finedrilled laser-fabricated room-temperature silica aerogel targets.

The IAC recognise that both systems require optimisation, but already a viable ultra-slow muon source (900 USM per sec) with superb emittance can be envisaged. The group has attracted collaborators and has developed a plan to implement an ultra-slow muon source on Port 3 over the next two years. The facility is expected to be complementary to that at PSI, offering a smaller spot size and improved timing resolution.

3.3. Muon Catalysed Fusion

The IAC notes ongoing work to develop μ CF measurements under pressure. However, the Committee felt that, at present, there are not sufficient resources available for the considerable technical and theoretical challenge that this aspect of the programme will require, and recommends continuing to focus on the two programme pillars discussed above.

No further work has taken place on muon catalysed fusion in Port 1 since last IAC. Plans are in-hand for a safe and proper termination of the project as advised by the previous IAC, with removal of the tritium gas handling system from Port 1 likely early 2015.

The IAC wishes to thank the RIKEN-RAL group, and particularly Tei Matsuzaki, for the necessary work that has been carried out to decommission the tritium handling system from Port-1. The committee was pleased to note that the system will go to JET (Culham, UK) for continued use, providing a cost-effective option that will continue the RIKEN-RAL legacy in this area.

3.4. Other Science

The IAC recommends that nuclear transmutation activities with muons are not taken forward.

It was reported to the IAC that this recommendation was followed, and that activities in this area have not been taken forward.

3.5. Project Planning

The IAC notes that the extension of the RIKEN-RAL agreement to 2018 provides a good timescale for a planned and timely transition to J-PARC of muon activities, as facilities at J-PARC allow.

The IAC welcomed initiatives designed to facilitate an eventual transition of muon activities to J-PARC in 2018. The development of ultra-slow muon production, particularly work focussed on lasers and aerogel targets, promises an excellent exploitation of the RIKEN-RAL facility to provide key technologies for a future ultra-slow muon facility at J-PARC. The RIKEN focus on fundamental science will be developed through a new programme studying hyperfine splitting of muonic hydrogen. The committee remains concerned that adequate staffing is available for these activities; the reassurance of work sharing between J-PARC and RIKEN-RAL in this area is therefore welcome, however, the committee also sees the need to attract new researchers to the project.

The IAC considered the present work to expand collaborative research with Asian countries is of strategic importance for building a vibrant future user community. The committee noted the analogy with a similar outreach programme at CERN by Prof. SFJ Cox in earlier decades; this successful work was instrumental to the timely development of the ISIS European muon facility.

The IAC applauded the collaboration between RIKEN-RAL and J-PARC that enabled a number of J-PARC experiments to be carried out at the UK facility following the 2011 Earthquake of East Japan.

3.6. Collaborative activity

The IAC continues to note that the RIKEN-RAL muon facility is an extremely successful international collaboration in its own right. In addition, a wide variety of collaborations within RIKEN, within Japan, internationally, and at ISIS and TRIUMF, were noted. It recommends further publicising of RIKEN-RAL activities widely within RIKEN to develop further collaborations with other RIKEN centres.

The IAC applauds the success of the RIKEN-RAL facility in establishing a broad range of national and international collaborations (shown in Figure 1). In particular, the committee noted the benefit of reaching out to potential user communities in Asia, such as in Malaysia, Indonesia and more recently in Korea. The agreements that have been signed will provide access to students and young researchers, and RIKEN-RAL should be the perfect training ground for future µSR users in these communities.

The IAC welcomed collaborations developed within RIKEN, and particularly those that have led to the development of laser technologies for muonium ionisation and on-going work with the Condensed Molecular Materials laboratory. The IAC encouraged the group to explore further collaborations, and would be keen to see links developed with RIKEN's recently established Centre for Emergent Matter Science to promote interdisciplinary research.

The IAC commends the strong links that have developed between RIKEN-RAL and ISIS. Through technical collaborations, ISIS supports the Chronus data acquisition system (now using DAE-II), software for instrument control (SECI) with convergence towards a compatible data format, sample environment (including cryostats and pressure cells) and maintenance of the RIKEN-RAL facility. Science collaborations have been formed through collaborative studies, while there is also significant sharing of resources through the use of RIKEN-RAL by UK/European researchers and of HiFi by Asian groups. Meetings have developed to underpin these activities, with the annual joint workshops and site calculation meetings proving effective in stimulating dialog and collaboration.



Figure 1: Collaborations formed within Japan (top) have increased to 70 groups in the period 2011-2013 (compared to 60 groups to 2010). The number of international collaborations has also grown, with 34 groups involved during a similar period (compared to 13 groups to 2010).

4. Evaluation of Research Outputs from the RIKEN-RAL Facility

4.1. Condensed Matter and Molecular Science

The condensed matter and molecular science programme continues to be a very strong area for RIKEN-RAL – a central pillar of its activity. A diverse science programme has continued, studying a broad range of topics including magnetism, superconductivity, ionic diffusion, and spin and charge dynamics. The unique capabilities of the facility, providing high pressure and laser stimulation in combination with a pulsed beam, enhance this programme. The facility is very productive, with over 24 high quality journal papers published in the 2011-2014 period. There is strong evidence from the portfolio of highly cited work presented to the committee for scientific excellence in the output of the facility that is clearly world leading in this field. That this has been achieved despite an acute shortage of manpower and constraints on the operating budget reflects extremely well on the facility and its staff; the facility is clearly highly cost effective.

The IAC welcomed highlights from the condensed matter programme presented by the RIKEN-RAL group, which included:

- Discovery of dipolar ordering in a molecular nanomagnet
- Study of lithium ion diffusion in lithium-transition-metal oxides
- Investigation of internal fields in the superconducting state of URu₂Si₂
- Study of chemical reaction theory by the reaction of Mu with vibrationally excited H₂
- Study of superconductivity and fluctuating magnetism in a molecular superconductor

In addition, the IAC welcomed the recent chip irradiation studies, considering a potential for developing an important new area of work at the facility.

4.2. Ultra-Slow Muons

The ultra-slow muon project has progressed significantly with the development of a high intensity Lyman-a laser system coupled with the discovery of enhanced muonium production from fine-drilled laser-fabricated room-temperature silica aerogel targets. Together, these improvements can realistically be expected to provide almost an order of magnitude increase in slow muon flux, and it is possible to envisage a science programme based upon this source.

The formation of muonium at rather high yields inside aerogels is well-known. However, the fabrication of SiO₂ aerogel targets with 500 μ m spaced 5 cm deep holes brings a new era for the production of cold muonium in the vacuum, with yields of ~9% being demonstrated. Furthermore, the overlap of the laser light frequency distribution with the Doppler-broadened lineshape for cold muonium is much improved, enhancing the number of slow muons after photo-ionisation.

The IAC also appreciated the development of a proposed new laser system based on Nd:YGAG lasers and CLBO nonlinear crystals. This system can be expected to have no

timing jitter issues, to operate at the full 50 Hz repetition rate and to provide (per pulse) a factor of four increase in intensity.

This research is expected to make a key contribution to the g-2 experiment at J-PARC, while also providing a new facility for surface and interface studies. The committee noted that this provides a great opportunity for new condensed matter physicists to come into the field, both in Japan as well as in Europe. The facility promises higher timing resolution and a smaller spot size compared to PSI, and is therefore better suited for small sample sizes.

4.3. Impact of RIKEN-RAL Science

The IAC noted that the number of publications from the facility in the field of condensed matter and molecular science continues to grow. One measure of the impact of RIKEN-RAL science comes from citations of research papers published by facility users (although this does not capture all aspects of impact, something which is not fully apparent for many years). Analysing the impact from RIKEN-RAL output in terms of citations reveals the following scientific highlights:

- Highest cited work: *Studies of new iron arsenide superconductors*: Nature Materials 8, 310 (2009) – 161 citations to date Phys. Rev. Lett. 101, 097010 (2008) – 86 citations to date Groups involved include Fribourg, London, Oxford, PSI, Hefei, RIKEN.
- Highly cited work: *Cuprate superconductors*: Phys. Rev. B 67, 014514 (2003) – 64 citations to date Groups involved include Kyoto and RIKEN. Phys. Rev. B 69, 184507 (2004) – 48 citations to date Groups involved include Sendai, RIKEN, Tsukuba.
- Highly cited work: *Excitations in polymers*: Phys. Rev. Lett. 79, 2855 (1997) – 48 citations to date Groups involved include RIKEN, Oxford, Tsukuba, Durham.
- Highly cited work *Fundamental magnetism:* Nature 471, 612 (2011) – 40 citations to date Groups involved include ISIS, Oxford, J-PARC, Nagoya, PSI, Tokyo, RIKEN, Kyoto.

The IAC recognises the impressive scientific output of the RIKEN-RAL facility during this period, with a large number of highly-cited publications on diverse topics ranging from material science to fundamental studies in superconductivity and magnetism. The committee consider this particularly noteworthy since, after renormalising for the number of available instruments, this output is comparable with that of the world leading ISIS European muon facility.

5. Evaluation of direction of RIKEN-RAL towards RIKEN's 4th Five-Year Term

5.1. New Activities: Proton Radius Experiment

The new initiative for the measurement of the hyperfine-splitting in muonic hydrogen promises to become a high profile project. The determination of the proton radius has become an increasingly interesting topic in science: recent laser spectroscopy of the Lambshift in muonic hydrogen at PSI suggests that the proton size measured with muons is significantly smaller than that obtained with laser spectroscopy of electronic hydrogen or deduced from electron scattering.

At this early stage, the approach of measuring the hyperfine-structure in muonic hydrogen appears a very promising method for developing this area. The IAC recommends working on a well-structured project towards a precise measurement of the proton Zemach radius. *In view of its scientific importance and anticipated impact, the committee strongly encourages the development of a full proposal with high priority. The proposal can then go forward to be scrutinised by the responsible PAC. This would also enable the impact of this experiment on the other two programme pillars of condensed matter and molecular science, and low energy muon production, to be evaluated.*

The IAC welcomes the development of this exciting experiment that fully exploits the existing infrastructure of RIKEN-RAL. The committee noted that the experiment could use Port 1 to advantage after timely removal of the tritium handling equipment to minimise impact on the ongoing μ SR programme.

The IAC notes that for rapid progress of this high profile work, the project will require a sufficient number of dedicated personnel with expertise to collaborate across the different scientific and technical areas. However, the IAC believe this project provides an excellent opportunity to recruit and maintain talent in the field of muon instrumentation and science.

5.2. New Activities: Chip Irradiation

Recent interest from the electronics industry has led to the inception of chip irradiation experiments at the RIKEN-RAL facility. These are designed to investigate and evaluate the potential problem caused by single event errors being generated by muons originating from cosmic rays. Initial experiments have been led be two academic groups, working in collaboration with members of the RIKEN-RAL and European muon groups at ISIS. The RIKEN-RAL facility is particularly suited to this work as the momentum of the muon beam can readily be tuned to penetrate circuit layers and probe inside electronic modules, while negative muons can be useful for the analysis of the circuits themselves.

The IAC are enthusiastic about chip irradiation experiments and see significant potential to grow the activity in the future. The committee suggest links with industry are explored, and see great benefit in promoting this type of facility at ISIS where there is complementarity with the existing neutron programme. With a new neutron instrument, ChipIR, about to be commissioned for similar measurements, ISIS is potentially a very attractive location for chip investigations.

5.3. The Muon Economy

The IAC considered that the two pillars (condensed matter and ultra-slow muons) already make significant demands on sharing muon beams as well as support staff. The committee were therefore concerned about the introduction of new activities, particularly the proton radius experiment, and their impact on the existing programme. *In particular, the committee encourages the location, effort and duration of the proton radius experiment to be considered as part of a future proposal.*

With limited user support, the committee strongly suggests that exploitation of the advanced Chronus spectrometer will offer the most cost effective method of maintaining a strong μ SR programme at the facility.

5.4. Transition to J-PARC

The IAC recognises the need for detailed planning for the future of the RIKEN-RAL facility beyond 2018, and encourages discussions with ISIS as soon as possible.

The committee emphasises the need to develop new strategies for staffing in the run up to the transition, and note that RIKEN-RAL operation should provide an excellent training ground for new and young scientists in the field of muon physics. Staff trained at RIKEN-RAL will provide immediate expertise for future activities at J-PARC.

The UK interest in continuing a condensed matter programme at the facility post 2018 should be explored in tandem with a detailed assessment of the decommissioning liability for the facility in case decommissioning should be necessary.

6. Recommendation of Items to be Strengthened at the RIKEN-RAL Facility

The IAC were enthusiastic about developments planned for low energy muons and the measurement of the proton radius for the period to 2018. The committee considered that these, coupled with the on-going condensed matter programme, are likely to provide an excellent springboard for future operations at J-PARC. The committee, however, encourages RIKEN to ensure these activities are appropriately staffed, noting the benefit of developing key Japanese expertise in muon instrumentation and science at RIKEN-RAL for future operations after the transition to J-PARC.

The IAC noted the on-going work promoting the condensed matter μ SR technique across Asian counties, considering it an important step in developing the user community across Asia. The committee strongly endorses this activity and encourages this development, but notes that a commitment of effective and dedicated user support personnel will be necessary for success in this area.

The IAC welcomed the recent developments reported for the ultra-slow muon programme and are keen to see the enhanced laser and target technologies installed and demonstrated at RIKEN-RAL. The committee considered that the predicted ultra-slow muon rates should lead to a viable source, and encourages the facility to consider this a development centre for ultra-slow muon techniques. Potential European interest in this area was noted, and discussions are suggested to explore future collaboration in this area.

The IAC noted the potential benefit of establishing a programme of interdisciplinary research. The committee encourages RIKEN to explore ways of developing this area, with condensed matter research perhaps benefitting from collaboration with RIKEN's recently established Centre for Emergent Matter Science.

Appendix 1: List of International Advisory Committee Members

Dr Andrew Taylor (Chair)

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Appendix 2: Agenda for the Fourth RIKEN-RAL International Advisory Committee

Monday 7 April 2014			
Opening address	H. En'yo		
Closed Session			
Present status and highlights Present status of the RIKEN-RAL muon facility Recent µSR highlights of RIKEN group Recent µSR highlights of UK group	P. King I. Watanabe A. Hillier		
Recent status of ultra-slow muon beam project p Lyman- α laser	K. Ishida, Y. Oishi		
New Project Mu-p HFS (plan and target) Mu-p HFS (laser) Closed Session	M. Sato Y. Oishi		
Tuesday 8 April 2014			
<i>Future plan of RIKEN-RAL</i> From RIKEN From RAL Decommissioning plan of Tritium Gas Handling System	M. Iwasaki R. McGreevy T. Matsuzaki		
Closed Session			
Closed discussion			
Summary talk	IAC Chairman		