I R P S BULLETIN

Newsletter of the International Radiation Physics Society

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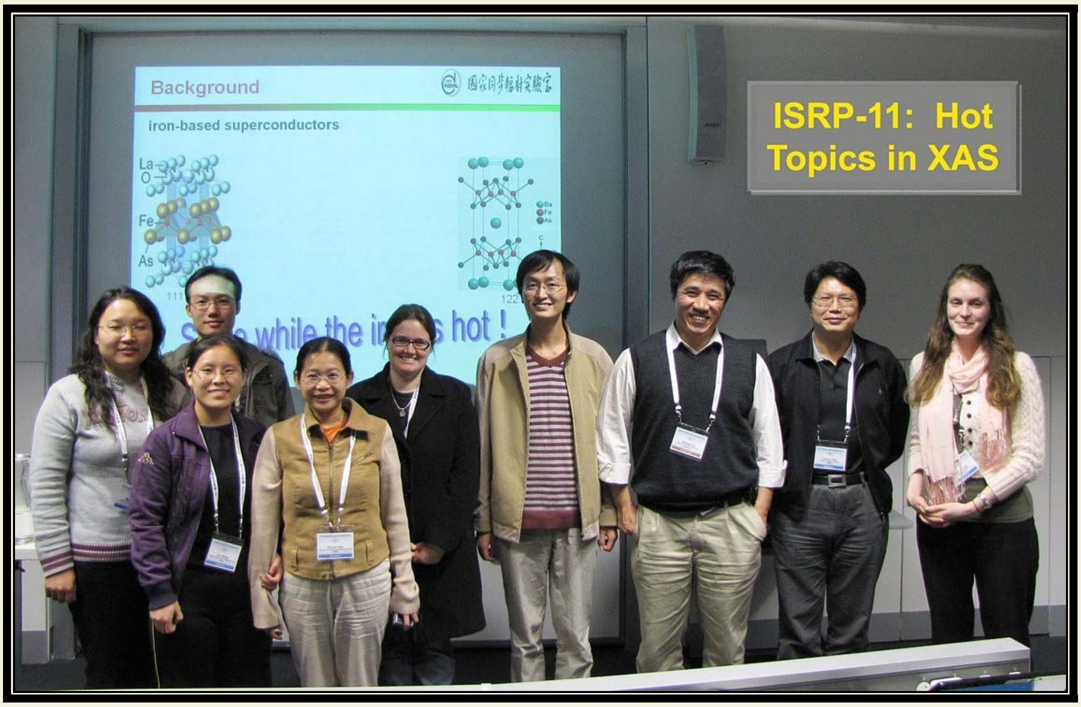
From the Editors

Dear Reader,

Please be encouraged by this first issue of Volume 24 of the Bulletin of your International Radiation Physics Society. In our times the global media are abuzz with societal issues that involve uses of radiation physics. From energy sources to security screening, much information, and much misinformation, are being showered onto an underinformed public. IRPS members are knowledgeable, and in many cases, engaged in the public dialog of today's global issues; it is hoped that this Bulletin is a small enabler in both of these regards.

In this month's issue, one will find articles ranging from the fundamental ("Report on X-Ray Energies, Transition Probabilities, Fluorescence and Coster-Kronig Probabilities" by J. L. Campbell) to the applied ("Nuclear Renaissance: the Brazilian Case" by Odair Dias Gonçalves). In addition to regular features like topical meeting announcements and reports from the Society's regional Vice Presidents, Malcolm Cooper and Dudley Creagh have generously captured for us the technical content of ISRP-11 with a thoroughgoing review of the Society's triennial symposium held last September in Melbourne, Australia. As one can see from the smiling faces below, this was an event to remember and/or read about within. Thanks for all your contributions to this and future issues.

Larry Hudson and Ron Tosh



President's Report

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This is my first message as President of the International Radiation Physics Society since my candidacy was approved in the last elections.

The first thing to say is that I am a little scared about taking this position, which, traditionally, has been occupied by very distinguished scientists, some of them I would regularly reference in my publications, like Didier and Hubbell, just to name those that are no more with us, and Dick Pratt, who was one of my mentors in photon scattering research.

When Mic Farquharson and David Bradley contacted me to ask if I would agree to stand for the presidency, my first thought was, "those guys are kidding me." But on second thought, I realized that it could make some sense, since I am involved in nuclear policy through the Brazilian Nuclear Energy Agency (CNEN) and also as part of the Board of Governors of the International Atomic Energy Agency (IAEA).

And for sure, nuclear energy is rising again as one of the most viable means to produce electric power, and, big surprise, as one of the safest and cleanest forms of producing energy. In a short text in this Bulletin, I will try to justify these opinions, and I think that our Society has an important role to play in this context. New technologies are being developed. Safety and security are the most important aspects and must be regarded as so. Developing countries are rising as important new players. Even the role of international organizations is being reviewed. And the IRPS and the radiation physics community have much to say about these issues.

There is much knowledge to be gathered and lots of problems waiting to be tackled, like, among others, new forms of reprocessing used fuel, long term behaviour of radioactive waste in repositories, and new uses and applications for radioactive materials. Needless to say, this new era of nuclear power will most certainly bring more funds to a very broad field of research.

I would like to thank the confidence of those that voted for my candidacy, the hard and good work done by the former directory board, with a particular reference to Dudley Creagh, former President. I would also like to thank those that are with me in this new term, Dudley, as chairman of the Advisory Board, Mic, as Secretary, Malcom Cooper, taking care of the money, Elaine Ryan, receiving the membership dues, and all the Vice Presidents and members of the Executive Council.

I hope that I will be able to do for the IRPS at least a small part of what has been accomplished by my predecessors.

Odair Dias Gonçalves

Conference Report
11th International Conference on Radiation Physics : ISRP-11

The conference, which attracted nearly 200 participants from all over the world to the University of Melbourne (Australia) on 20-25 September 2009, embraced a wide range of topics in radiation physics - an A-to-Z of topics, ranging from Astronomy and Archaeology to XAFS and Zoology. It grappled with topics as diverse as *WIMPS* (weakly interacting massive particles) in the hunt for dark matter, radiation-based inspection methods for border security, aspects of nuclear power production, radiotherapy, dosimetry, and cell biology. A very large proportion of the papers involve synchrotron radiation research which reflects the very rapid growth and diversification of synchrotron studies worldwide in the spectrum of radiation physics.

This series of triennial conferences was the 11th of a series which was formally set up in 1985 to bring together radiation physicists from developing and developed countries, capitalising on the fact that this subject, more so than any other physics specialism, is a truly worldwide activity. This arises for several reasons: firstly, the plain fact is that not all radiation physics has costs on the scale of big budget accelerators, telescopes and interplanetary probes. Far from it, much good radiation science can still be done with track detectors and scintillation counters. Thus both experimental and theoretical research can be on the agenda in developing as well as developed countries. Secondly, radiation physics development is often driven by mining (often in less developed regions) and nuclear energy production in countries without indigenous fossil fuels. Thirdly, diagnostic and therapeutic healthcare, and border security concern all countries. Finally simple scientific curiosity and ingenuity knows no geographical boundaries ! All this constitutes the rationale for our society, IRPS, and is evidenced by the vitality of our ISRP conference series. At Melbourne it was certainly manifest not only in the diversity of the science presented but also in the origin of the delegates: 80% came from outside Australia, in fact from 37 other countries, and, as the local organiser Chris Chantler pointed out, every continent except Antarctica was represented. There is plenty of radiation physics going on in Antarctica but perhaps travel to and from Antarctica in the Southern Hemisphere winter is somewhat restricted. With almost 200 participants this was the second largest conference in the ISRP series. *Figure 1* shows some of the participants at the conference: others were playing truant in down town Melbourne.

The conference began with the very serious basics: Gordon Drake (University of Windsor, Canada) talked about quantum electrodynamics, which is probably the most successful theory in physics and constitutes the basis for the Standard Model. The feast of equations continued with Richard Pratt (University of Pittsburgh, USA) who talked about quadrupole photoeffect matrix elements in a session dedicated to John Hubbell, a founder of IRPS, and a NBS/NIST physicist, whose comprehensive compilations of x-ray absorption cross sections have underpinned many decades of x-ray science.

The transition from fundamental processes to the experimental world was provided authoritatively by John Rehr's (University of Washington, USA) review of x-ray and electron spectroscopies – XAFS; XANES; EELS etc. The processes underlying these spectroscopies occur in the region of the absorption edges of atoms in crystals. The experimental results mirror the environments of the atoms and their location in materials. XAFS and XANES are techniques extensively used at synchrotron radiation sources in studying the A-to-Z of science. The scope and potential of XAFS spectroscopy was later illustrated by Ronald Frahm (University of Wuppertal, Germany) who talked about Quick-XAFS developments. The adjective 'quick' seemed rather inadequate a description: complete XAFS spectra are taken at millisecond intervals, enabling for the first time studies of the dynamics of physical and chemical systems.

The conference's diverse topics ranged from the study of metal take-up by plants such as nickel as a way of detoxifying soil (Paul Dumas, Soleil, France) to designing detectors to survey the myriad of sunken ammunition or gas canisters still littering sea beds from the Mediterranean to the Pacific (Jasmina Obhodas, Zagreb, Croatia). Soichi Wakatsuki from the Photon Factory *(Figure 2)* discussed some beautiful results on the transport of proteins by transport vesicles: the results show how "one-legged" as well as "two-legged" macromolecules are able to walk-the-walk of life.

Chris Ryan (CSIRO, Australia) described the new 'Maia' detector (384 elements), which is intended to be used at the Australian Synchrotron on its micro-focus beamline. It has been tested at the NSLS, Brookhaven, USA, a partner in its development, for elemental analysis in such diverse fields as geology, biology (heavy metal cancer drugs in cells), and cultural heritage investigations.

A significant part of the conference was concerned with the growing use of radiation (predominantly but not exclusively synchrotron radiation) in investigating aspects of cultural heritage: for example establishing the provenance of objects, understanding their degradation and developing methods of preserving them as authentically as possible for future generations to appreciate and enjoy. Eric Dooryhėe (NSLS II, USA), outlined the broad raft of analytical techniques now available to cultural heritage scientists and described how the long-lost art of the making of 'Maya blue' ceramics was re-discovered. The question of how the organic (indigo) dye could exist in a stable state in a clay host had been a puzzle for many centuries. In a similar vein Annemie Adriaens (University of Ghent, Belgium) described the study of the electrochemical degradation of lead artefacts arising from "acid attack" in humid environments. The most famous victims of this process are lead organ pipes in medieval church organs and she described progress with assessing the efficacy of carboxylate coatings. The general obsession with x-rays was challenged by Mark Dowsett (University of Warwick, UK) who extolled the virtues of using the technique of x-ray induced optical luminescence (XEOL) as a spectroscopic and imaging tool, for example capable of producing micron image resolution from beamlines with mm not µm beams. Amazingly the optical signal mimics faithfully XANES and near edge structure.

The meeting also saw the triennial award by the Elsevier Journal *Applied Radiation Isotopes* of the JARI medal to a senior scientist who has made outstanding contributions to the field of radiation physics. This year it was awarded to David Bradley (University of Surrey, UK), *see Figure 3,* for his long and distinguished research into many aspects of the application of radiation physics. In his lecture he talked about his recent synchrotron studies of biological tissue such as cartilage whilst many of us in the audience wondered how long it would be before our own knee joints were similar to those with which he illustrated his talk ! A veritable army of his research students, originating from a variety of countries worldwide, provided a significant part of the poster sessions and the contributed oral papers: in fact the Didier Isabelle award (Isabelle was an IRPS founding father) for the best student presentation went to Entesar Dalah, one of David's students. The accompanying JARI Enterprise award to a mid-career scientist, went to Larry Hudson (NIST, USA) (shown in Figure 4 with his long-term collaborator John Seely (NRL, USA) and Ned Blagojevic (ANSTO, Australia). Larry has been closely involved in the experimental verification of QED theory as well as the establishment of standards for airport baggage inspection systems. The awards (the JARI Medal and the Enterprise Award) were presented by the journal's Editor-in-chief Richard Hugtenburg (University of Swansea, UK). Our society should be proud of the fact that the JARI awards have regularly been bestowed on IRPS stalwarts such as Richard Pratt, David Bradley, Chris Chantler and Larry Hudson.

This meeting saw an increase in the number of papers using synchrotron radiation and other radiation techniques for the solution of biomedical and biophysical problems. Martin Feiters (Radbough University, Nijmegen, The Netherlands) commenced proceedings with his presentation on two- and three- dimensional geometry of metal coordination inorgano-metallic and bio-inorganic catalysts probed by x-ray absorption spectroscopy. As it happens the study of organo-metallic compounds and the understanding of coordination number is an integral part of obtaining an understanding of how cancer-forming agents can attack cells on the one hand, and how anti-cancer drugs protect cells on the other. Later, Isabella Ascone (Ecole Nationale Superieure de Chimie de Paris, France) described how anti-tumoral metallo-organic drugs can be studied using XAS. Don McNaughton (Monash University, Australia) spoke on his research using synchrotron infrared micro-spectroscopy for the study of cells and tissues. This work is strongly related to fertility in mammals. Another aspect of his work deals with the development of bio-models for the study of multiple sclerosis.

In other talks of a medical nature Elaine Ryan (University of Sydney, Australia) discussed the differentiation of malignant and non-malignant kidney tissues using Compton scattering, and of course, the JARI lecture was given by David Bradley (University of Surrey, UK) on his research into cartilage in joints. Away from Medical Physics Bill Dunn (Kansas State University, USA) talked about methods for detecting improvised explosive devices, the IEDs whose effects in current conflicts we read about with depressing frequency in our newspapers. Odair Gonçalves, our new President, talked about the need for nuclear power generation and Professor Jose Rodenas (University Politecnica de Valencia, Spain) discussed the modelling of radioactivity in control rods.

The theme of linking from basics to applications continued to bridge the gap to the larger SRI meeting taking place the following week also in Melbourne, and prevent delegates from contracting pneumonia over the coldest Melbourne spring weekend on record. The two day *Workshop on Analytical Techniques* on

26-27 September was underpinned by comprehensive introductions to x-ray fluorescence (Gary Pritchard, PanAnalytical); x-ray diffraction (Brendan Kennedy, University of Sydney, Australia); infrared (Paul Dumas, Soleil, France); infrared-Raman (Ken Williams, Renishaw); XAFS (Ronald Frahm, University of Wuppertal, Germany) and imaging modalities (Dudley Creagh, University of Canberra, Australia). All of this underpinned some intriguing examples of forensic analysis in the fullest meaning of the term.

In this vein Debra Lau (National Gallery of Victoria, Australia) explained how the treatment of silk cloth with tin, which was motivated largely by greed (cloth sales price depended on weight) led to current conservation problems with high fashion (e.g. Balenciaga's Infanta Gown in the National Gallery of Victoria). Koen Janssens (University of Antwerp, Belguim) described a variety of techniques for the study of easel paintings including micro-XANES and elemental mapping of pigments in purported Rembrandts using the Maia detector described earlier in the symposium by Chris Ryan (CSIRO, Australia). Vincent Otieno-Alego (Australian Federal Police, Canberra, Australia) explained how a multitude of techniques (including SEM-infrared Raman) were necessary to show whether ships that should have 'passed in the night' but reportedly collided, were making legitimate claims, not to mention whether particles found on suspects really were from gunshot discharge and not overactive use of power drills for home do-it-yourself as they often claimed! Dudley Creagh, and Ned Blagojevic (ANSTO, Australia) each explained how x-ray Compton scattering underpins the formation of images for most of Australia's and everyone else's border security at acceptable combinations of dose and speed. Chan Tranh (Latrobe University, Australia) and Andrew Stevenson (CSIRO, Australia) continued the theme of x-ray imaging but in the laboratory and at synchrotron sources using phase contrast techniques. *Figure 5* shows a selection of the speakers at the symposium: some international, some local.

The conference organizers, Chris Chantler and his committee, produced a vibrant and diverse program for both the symposium and the workshop. All the participants appreciated both the scope and quality of the lectures given. But not all was intense science. That all was not "noses to the grindstone" can be seen in *Figure 6*. Clearly no one was suffering at the conference dinner which was held in the impressive surroundings of the Melbourne Museum.

Whilst synchrotron radiation and its uses were featured strongly in this conference the trend towards the blending of many diverse complementary techniques moves on apace. It may be that the solutions to many problems will lie in the application of appropriate admixtures of synchrotron radiation, neutron scattering, ion-beam scattering, and so on. This is great news for our society as it emphasises the relevance of radiation physics in all its manifestations.

The next meeting, ISRP-12, will be held at Salvador Bahia, Brazil in September 2012. It is likely that the associated workshop will be held either in Rio de Janiero or Sao Paolo, immediately preceding the main meeting. Attention is also drawn to the related IRRMA meeting (Industrial Radiation and Radioisotope Measurement Applications), which is scheduled to be held in Kansas City 26th June - 1st July 2011: despite its name IRRMA's scientific agenda is very close to that of our symposia as could be seen from the contributions of IRMMA's organisers to this Melbourne meeting: David Bradley will be the Technical Program chair for their

2011 meeting and Bill Dunn its General Chair.

Some of the more synchrotron radiation-related aspects of this meeting and its workshop will be reported in the next edition of Synchrotron Radiation News (Ed. Ronald Frahm). Those who came to Melbourne will have left with the knowledge that the subject and our Society are in very good health and those of you who could not come to Melbourne should make plans to be in Brazil in 2012.

Malcolm Cooper
Dudley Creagh

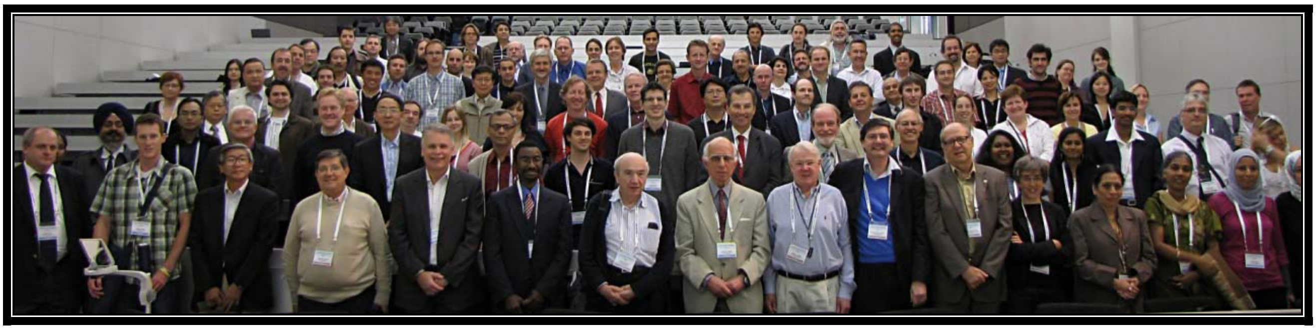

Figure 1. Some of the conference participants.

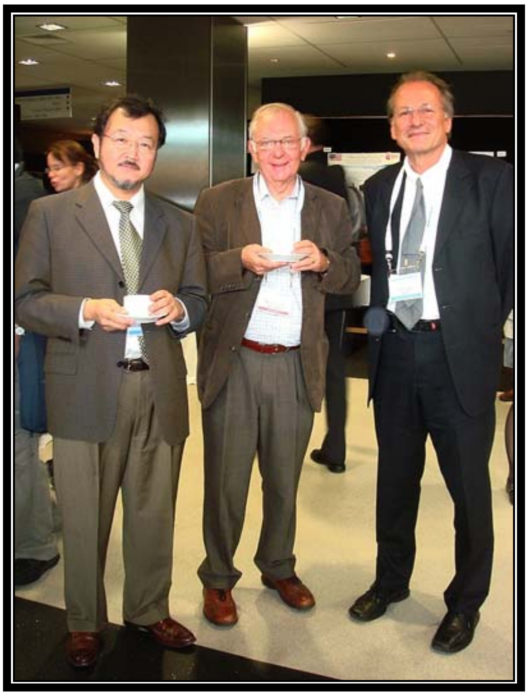

Figure 2.

Soichi Wakatsuki
(left, Director of IMMS, KEK, Japan) talks to
Malcolm Cooper (centre, University of Warwick, UK),
and Paul Dumas (Synchrotron Soleil).

Figure 3.

David Bradley
(University of Surrey, UK) showing
the JARI medal which was
presented by the Editor in Chief of
Applied Radiation and Isotopes,
Richard Hughtenberg.

Figure 4

The JARI Enterprise Award winner, Larry Hudson, (NIST, USA) centre of picture, talks with John Seely (NRL, USA) right, and Ned Blagojevic (ANSTO, Australia).

Figure 5.

Speakers and delegates at the Workshop on Advanced Analytical techniques :
Left to right:
Paul Dumas (Soleil, France)
Debbie Lau (CSIRO, Australia)
Alana Treasure (Australian War Memorial, Australia)
Dudley Creagh (University of Canberra, Australia)
Petronella Nel (Ian Potter Gallery, University of Melbourne, Australia)
Koen Janssens (University of Antwerp, Belgium).

Figure 6.

Conference dinner at the Melbourne Museum.

Vice President's Report, Africa and the Middle East

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The following is a summary of the current activities in the regions of Africa and the Middle East.

1. January Activities

1-1 Africa regional congress

The President of the International Radiation Protection Association, Ken Kase, visited Nairobi and met the local organizing committee of the 3rd All Africa Regional Radiation Protection Congress and Dr. A. Mustafa, Chairman of the Congress committee. The regional congress will be held 19-23 September, 2010. You are invited to attend this congress. Please learn more at

http://irpa-egypt.com/2010.pdf

1-2 Cairo Radiation Protection Workshop

This workshop proceeded with the help of several supporting organizations: International Commission for Radiological Protection (ICRP), International Labor Organization (ILO), Arab Atomic Energy Authority (AAEA) and National Radiation Physics Network of Egyptian Atomic Energy Authority (EAEA), IRPA Egypt, ESNSA and MERRCAC.

The speakers for this workshop included:

J-F Lecomite, a member of Committee 4 of the ICRP;
from ILO, Senior Physicist Niu;
from AAEA, Dr. Mosbah;
from Tunisia, Dr. Hannou;
from Sudan, Dr. Osman;
from Lebanon, Mr. Dihini;
from UAE, Ms Bushra;
from Mobinil, Mr. Issa.

Including myself and others, there were 10 participants from outside Egypt and 40 Egyptians.

The workshop was aimed to inform workshop participants about two new ICRP recommendations; these are

The 2007 ICRP Recommendations (ICRP-10-3)
Radiation Protection in Medicine (ICRP-105).

Among the outcomes of the workshop came the following:

1. The French translation of ICRP-103 is now available on the internet.
2. Arabic translation of ICRP-105 is in progress by an African and Middle Eastern group headed by Mr. Dihini. Translation costs and printing expenses shall be covered by AAEA.
3. Encouragement for African medical physicists to form medical physics societies and to join the African Federation of Medical Physics (AFOMP). AFOMP is part of International Organization of Medical Physics.

4. Acknowledgement of the role MEFOMP plays in improving the status of medical physics in the Middle East.

5. Encouragement of radiation physicists and radiation protection experts to form radiation protection societies in Africa and Middle East and to join ARPA International.

6. Planning for workshops on the following topics: Safety Culture, Radiation and Nuclear Emergencies, Protection Against Non Ionizing Radiation, the Handling of Batteries and Computer Waste Products.

Below is a photo of workshop participants.

2. Other activities

2-1 IRPA : To encourage radiation experts to go through draft 3.0 documents of IAEA Safety Standards for protecting people and the environment for comments.

www-ns.iaea.org/downloads/standards/drafts/ds379

2-2 Medical physicists were invited to participate in the radiation medicine conference held in Riyadh, Saudi Arabia. The International Conference on Radiation Medicine (ICRM) was organized by the King Faisal Specialist Hospital and Research Centre (KFSH&RC) in collaboration with King Abdulaziz City for Science and Technology (KACST), Saudi Food and Drug Authority (SFDA) and International Atomic Energy Agency (IAEA). ICRM aims to enhance the development of health professionals in various aspects of radiation medicine in support of providing state-of-the-art, safe and quality healthcare. For more information about ICRM 2010, see :

www.radmed.org

Nuclear Renaissance - The Brazilian Case

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Editors' Note: At the 11th International Symposium on Radiation Physics in Melbourne this past September, Dr. Odair Gonçalves gave a talk on the rise of nuclear energy, particularly in Brazil. Several persons suggested that these topical ideas be recapitulated for the Bulletin of the Society as part of the ongoing societal dialog. Odair has graciously provided the following white paper at our request.

The last five years have seen a renewed interest in nuclear energy as a source of electric power. Countries until now declared as anti nuclear energy are rethinking their options, and even planning significant investments in this form of electric generation. Prestigious environmentalists like Patrick Moore and James Lovelock revised their positions based mainly on the consequences of each energy source on the environment. Nuclear, from an evil reputation, turns up as a clean and environment-friendly energy source. But the concerns with waste and used fuel, as well as proliferation of nuclear weapons, still remain and have to be carefully addressed.

We intend to discuss the issue, look to the world scene and then address with more detail the Brazilian case as an example.

There are a number of reasons that justify the new investments in nuclear energy:

1. Safety

One of the oldest arguments used by the opponents of nuclear energy is related to safety of nuclear power plants (NPPs), usually associated with the sad memories of Chernobyl and Three Mile Island. Both accidents were due to human failures. The Three Mile Island accident caused no fatalities. In Chernobyl, some tens of people died from immediate exposure to radiation and, as a consequence of the explosion, a few hundred in the actions taken to control the burning of the nucleus. A number of people that varies from hundreds to thousands, following different methods of estimation, is expected to die in the next 30 years due to propagation of radioactive materials through the air or contaminated food (the correct number is still under discussion, in the academic community as well as in expert forums like UNSCEAR).

Two important points must be stressed. First, the Chernobyl NPP was an old model, built in a time when safety aspects were not so important. There was no containment building, for example. That kind of accident is virtually impossible in pressurized water reactors (PWR). Second, since World War II, when nuclear science gave rise to the nightmare of nuclear bombs, the use of nuclear energy and NPPs has become one of the most controlled, tested and strictly regulated activities. NPP operation is probably the only activity where some failure detected in one machine is rapidly communicated and, once the ways to prevent such failure are found, the measures are usually adopted as mandatory in regulations around the world.

The safety measures used in the "modern" generation II PWR comprise, for example, the necessity of three barriers to avoid radiation leakage in case of an accident. The external building must be hardened against natural events like earthquakes, tsunamis and floods and even to plane crashes.

Finally, even considering the highly safe status, new generations of reactors are being developed within a concept to make them safer by requiring less need for human interaction.

2. Economics

The energy price of specific sources and technology varies a lot, depending on the cost of production and also on the availability of that source, but on average nuclear energy is the second or third least expensive after hydro, coal, and sometimes oil. Wind energy is about 2 to 3 times more expensive than nuclear, and solar costs about 10 times more.

3. Storage

One important factor to consider is the sustainability of the provision, which depends on the storage capacity. Usually it is not the energy that is stored but the energy source, hence the necessity to build large water dams or big depots to keep stocks of oil and coal. This is one disadvantage of wind and solar energies, since such sources are not suitable for storage. The advantage of nuclear in this respect is dramatic, since 10 g of ^{235}U are equivalent, in energy, to 700 kg of oil or to 1200 kg of coal.

4. Environmental effects

This is another big advantage of nuclear. After being considered the environment's most important enemy, lately nuclear is being considered a clean energy, because the total emission of green-house-effect gases from a NPP is only 4 g per kWh compared to 818 g from a thermal-oil plant and to 955 g from a thermal-coal plant. Even hydropower produces gases through the methane produced in the dams. The quantity in this case varies with the pre existing vegetation in the area, and its determination is still the object of a number of studies, but the amount is known to be significant.

5. Waste management

Since the beginning of the 21st century, when the necessity of new sources of energy reinstated nuclear as an option that should be considered, it was clear that the management of used fuel and contaminated residues, like machine parts, filters and tools, usually called nuclear waste, should be addressed in an urgent way. A lot of investments and research are being allocated in order to find "ultimate" solutions to the problem.

It is necessary to clarify what "ultimate" means. All kind of electrical energy production generates waste and interferes with the environment.

When we say that nowadays there is no economically sustainable "definite solution" to the nuclear high level radioactive waste, we are referring to millions of years without monitoring and guarding. For about 1 million years, it is well known how to SAFELY and

SECURELY deal with the problem, but this period is not considered "ultimate".

Another point to stress is that nuclear waste is usually divided in two categories, according to the amount of radiation produced:

- High level: this is the used fuel element. It must be stored in water pools inside the NPP for ten years or more, and then removed to final storage or reprocessing, since it still has a lot of potential energy (around 40 % in PWRs). This is the most problematic waste, since it takes hundreds of years to decay before being released to the environment;

- Low and Medium: this refers to the material that was in contact with radioactive elements, consisting of clothes, filters and decommissioning of facilities. This is easy to control and to store, but needs much more space.

But how much space are we talking about? 40 years of operation of a power plant (1000 MW) produce about 1000 m^3 of irradiated uranium, which could be stored in a depository measuring 20 x 10 x 5 m^3; an equivalent coal power plant produces 28 000 000 m^3 of waste, and 60 years of operation of 20 nuclear power plants produce 30 000 m^3 of irradiated uranium, which can be stored in a volume of 30 m x 20 m x 5 m, equivalent to half of a soccer field (100 m x 50 m). It must be recognized that this is not impossible to be controlled until an "ultimate solution" is achieved.

Those are the reasons why nuclear energy is being taken into consideration in many countries that are beginning to build new NPPs, like the USA that has 144 NPPs in operation, stopped new constructions after Three Mile Island, and now has around 15 new license applications and the government is even lending funds to construct them. Other countries are reviewing their positions, like Italy; and others, like China and India, that never gave up the nuclear option, are accelerating their programs (see the IAEA site for exact figures). Those are, in general, also the reasons why developing countries like Brazil decided to go nuclear.

THE BRAZILIAN NUCLEAR PROGRAM

Brazil has been conducting research in nuclear sciences since just after the Second World War, when the whole world was hoping for the benefits of the recently discovered potential of nuclear energy. In the early 1950s, Brazil had an amazing team of scientists who participated in the debates about creating an international scenario in which access to the benefits of the new discoveries would be granted, together with a strong non-proliferation commitment. As an example, Brazil's first research reactor, the first in South America, began operating in 1958. In 1971, the contract for the first 630 MW power plant was signed with Westinghouse. It was finished and commissioned in 1986.

In 1975 Brazil signed a broad program of cooperation with Germany, which envisioned building eight 1300 MW nuclear power plants, manufactured by KWU, to be located at several sites around the country. The program was not successful, resulting in just one operating reactor (Angra 2, 1995), with most of the equipment for a second one stored in special and proper containers, in Brazil, costing about US$ 20 million per year, just to keep the equipment in perfect condition. One ironic positive side effect is that we developed expertise in the storage of technical equipment.

During the 1980´s and part of the 1990´s, as in other parts of the world, Brazil decided to restrain its Nuclear Program, reducing the scope of the Brazilian-German nuclear agreement to just maintenance and supply. The government also decreased its financial support to levels that almost reached a situation of undermining the effectiveness of regulatory actions. The construction of the first German NPP, Angra 2, was slowed and completed only in 2002.

Moreover, with very little to do, the staff of the institutions involved in the program was not strengthened. The remaining human resources, particularly scientists and senior engineers, began to take their skills to other fields, such as materials science, fusion and other energy programs. Nevertheless, the basic capacity was preserved. The average age of the staff is now about 54 years, but they are still prepared to face the new prospect of a resurgent Brazilian Nuclear Program with a lot of energy and knowledge.

In 2003, it was clear to policymakers that a decision about Angra 3 was necessary. The CEOs of the Brazilian Nuclear Energy Commission (CNEN), Eletronuclear (ETN), operator of the NPPs, Indústrias Nucleares Brasileiras (INB) (responsible for the nuclear fuel cycle, including mining, enrichment and fuel assembly, and Navy Technical Center (CTM-SP), owner of the enrichment centrifuging technology, proposed a broad Program based on four pillars: safety and security, exclusively peaceful uses, respect for international commitments and development of national capacities.

Guided by these principles and by the recognition that Brazil had the human resources, the necessary knowledge and know-how, and that Brazil has the 6th largest uranium reserves in the world, with only 30% of its territory prospected, a new program was proposed.

The discussion that followed was quite productive and informative. It involved several Ministries and the discourse was open and transparent, accompanied by significant attention by the media. The consequence was that, when the government announced in 2007 the launch of the construction of Angra 3, a newspaper public opinion poll indicated 65% approval of nuclear energy. A similar result was obtained by an academic survey (Milanez et al., *Revista Ciências do Ambiente* online, vol. 2, number 1, pp. 1-10).

After three years of debate, the Brazilian Nuclear Program is now a strategic national plan, comprising:

1. Increasing the nuclear energy share of Brazil's electrical power matrix. Aside from the third reactor (Angra 3), between 4 and 8 new NPP of 1000 MW should be installed until 2030, increasing the nuclear portion of Brazil's energy matrix from the present 2.5% to about 5 or 6%.

2. Ensuring self-sufficiency in the production of nuclear fuel, providing 100% of the country's needs. This involves beginning the search for new uranium deposits, expanding both mining activities and the existing enrichment plant, and adjusting the fuel assembly complex.

3. Investing in all applications of nuclear energy and radiation devices in medical, industrial and agricultural activities that use ionizing radiation, such as radio pharmaceutical production and building of high-dose irradiators.

4. Strengthening regulations and creating a new regulatory agency.

5. Developing a broad and consistent educational and training program, joining the universities and the research institutes of the CNEN and the Ministry of Science and Technology, to address all areas of the Brazilian Nuclear Program, followed by a strategy of hiring highly skilled personnel in order to face the challenges.

Naturally, all these initiatives will require revision of the pertinent legal instruments, which is already under way, while retaining the essence of the whole plan, which is to maintain international commitments under the non-proliferation regime. Brazil is perhaps the only country that establishes in its Constitution that nuclear activities must be devoted solely to peaceful uses. Brazil is also subject to three independent safeguards systems: the Brazilian Nuclear Energy Commission (CNEN, which is the national regulatory body), the Brazilian-Argentine Accounting and Control Agency (ABACC), and the International Atomic Energy Agency (IAEA), thus completely satisfying the requirements of the Nuclear Non-Proliferation Treaty (NPT).

The Brazilian government is convinced that its plan for the expansion of nuclear power generation is feasible and mature. The country's nuclear scientists, engineers, technicians and operators are determined to fully participate in its implementation, in order to ensure its success.

NOTES: For details about nuclear programs around the world, see INTERNATIONAL STATUS AND PROSPECTS OF NUCLEAR POWER, INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 2008 and other sources in the IAEA homepage, where many other issues related to nuclear energy and applications could be found; for details about Chernobyl see the UNSCEAR home page :

http://www.unscear.org/unscear/en/publications.html

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The poster, which originally was designed by Dan Jones, is now the responsibility of Leif Gerward. Suggestions for improving the promotional material are most welcome. Please, send your comments to the e-mail address :

gerward@fysik.dtu.dk

X-Ray Energies : Transition Probabilities, Fluorescence and Coster-Kronig Probabilities

Editors' Note: Jorge Fernandez (U. of Bologna), our IRPS Vice President for Western Europe, recommended that we capture some of the technical content of the "Fundamental Parameters" meeting held in Berlin last May. It was thought that of particular interest to Society members were a couple of reports from their Working Group 4. In this issue of the Bulletin, J. L. Campbell of the University of Guelph, Ontario, Canada presents his "Report on X-Ray Energies, Transition Probabilities, Fluorescence and Coster-Kronig Probabilities." In the next issue of the Bulletin, Pierre Caussin of Bruker-AXS will report on “A Comparison of Five Available X-ray Absorption Tables.”

By way of background, the International Initiative on X-ray Fundamental Parameters is a collaborative effort of the Laboratoire National Henri Becquerel (Paris), the Physikalisch-Technische Bundesanstalt (Berlin), and the Technical University of Vienna. Its objective is to improve the quality of the database which underlies the fundamental parameters approach to X-ray fluorescence analysis of materials. At present it has seven working groups studying different issues.

A report prepared for the International Initiative on X-ray Fundamental Parameters

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Abstract:

A preliminary examination is conducted of several components of the X-ray fluorescence database, in each case with reference to the approach taken by Elam et al. in 2002. For the energies both of X-ray absorption edges and X-ray diagram lines, improvement in database accuracy should be possible, based upon the extensive project conducted over 20 years at NIST. However, attention needs to be paid to the issue of chemical bonding shifts in X-ray energies. For K X-ray relative intensities (RIs) there is strong experimental evidence to support theoretical Dirac-Fock predictions over those of the Dirac-Hartree-Slater model; nevertheless, new MCDF calculations in limited regions of atomic number would be useful. For the L sub-shells, the quantity of evidence is less, and experimental data exist predominantly for medium and high atomic numbers; for this region the DF approach appears to be superior. A new effort to compile and compare experimental data with these RI predictions would solidify the L-shell database, while new experimental work is needed for low atomic numbers.

The situation is more difficult for fluorescence and Coster-Kronig probabilities. There appear to be some differences between the ω_K values adopted by Elam et al. and those that we consider to be the best values based upon theory and compilations. A small number of new, carefully selected experiments would be helpful. The K-shell situation appears quite good except for the region of low atomic number, where

experimental data are sparse and are influenced by chemical bonding effects; experimental work is needed. For the L2 and L3 fluorescence yields at medium-high Z, DHS theoretical values are supported by experiment, but there are large uncertainties for all three L1 subshell decay parameters and the issue of methodological dependence arises. More experimental and theoretical work is needed, with the former demanding thoughtful design. Alternatively, a special set of fluorescence and CK parameters, derived from XRF experiments and consistent with a particular adopted set of photo-ionization cross-sections, may prove more relevant to immediate needs in the applied field of XRF analysis.

1. Introduction

This paper attempts to review the history and the current situation for several atomic physics quantities that together comprise a large portion of the database for X-ray fluorescence (XRF) analysis of materials. There is no intent to probe to great depth in this effort. The objective is a simple one, viz. a first look, with perhaps low "resolving power", at the issues. It is hoped that this approach will provide a basis for progressively deeper examination of the various areas that are discussed. No attempt is made to prioritize among these areas.

In an ideal situation, one would carry out such an exercise using only carefully selected experimental data from the literature, thus remaining independent of theoretical predictions. In practice, one sometimes finds that the quality of some of the data is questionable, or that the quantity is insufficient, or both. One also notes a tendency for multiple data values to exist where the experimental conditions are straightforward; concomitantly, there is an opposing tendency for a lack of data where the conditions are difficult. This can lead to the need for interpolation. Such interpolation requires justification that is scientifically well-founded. (It might be accomplished for a given quantity by studying the Z-dependence of the ratio between experimental and theoretical values of that quantity).

We (and others) therefore deem it necessary on occasion to employ theoretical predictions as well as compilations of measured data. In some situations, the theory merely assists interpolation and the filling of empty gaps in our knowledge. Other situations arise where a particular theory appears to describe the existing data, and one then simply accepts this theory as a representation of the data. This acceptance does not imply that the theory is perfect in all its assumptions. It merely reflects that the theory provides a "fit" to the real data, and one can thus avoid the labor of fitting functional relationships to the data.

Papp (2009) privately points out that if a reliance on theory cannot be avoided, then there is merit in employing the same theoretical approach as far as possible across the entire database, as any shortcomings may cancel out. In other words, consistency is a good thing.

Returning to the experimental data, it is important to address reasons for inconsistencies. An important issue is that of ascertaining whether systematic deviations are due to methodological differences. While there is no intent to do that in this overview, such an effort is a vital component of any "deeper" examination of any one segment of the XRF database. It may even be the case that observed deviations are not due to systematic errors associated with a particular methodology, but instead are due to the fact that different methodologies are in fact measuring slightly different things.

Finally, we need to be aware that many scientists regard the field of data that we are addressing here as "closed". They assume that all these quantities have been accurately determined long ago, and that there is not a need for new effort. While we can demonstrate that this view is incorrect, we cannot lose sight of the need to develop very coherent and persuasive arguments if and when we seek support for the "deeper" work that we hope will follow this preliminary overview.

2. Edge energies and X-ray diagram line energies

In their widely-cited paper "Atomic database for X-ray spectroscopic calculations", Elam et al. (2002) used the X-ray absorption edge energies compiled by Williams (1986); they found that thirty-seven of these edge energies disagreed by 1 -6 eV with the values used by Berger and Hubbell (1987) in their XCOM code for X-ray cross-sections.

More recently, a twenty-year project at NIST has provided experimental K and L edge energies, and the corresponding K and L X-ray energies. No data are explicitly provided for M and N edges and lines, but determination of the M and N edge data is implicit in this work and so we deduce that tabulations must exist from which M X-ray energies could be determined. The NIST authors (Deslattes et al., 2003) observed that most of the previous tabulations essentially reproduce the data of the very extensive early work of Bearden and his colleagues: Bearden (1967) first dealt with X-ray wavelengths, and Bearden and Burr (1967) re-evaluated energy levels (edges). Deslattes et al. discerned some problems in this work, one example being the use of interpolation approaches that neglected shell structure. Recognizing the near impossibility of a huge effort to make many precise X-ray spectroscopy measurements within whose results interpolation would pose less difficulty, Deslattes et al. relied instead upon a rather small number of very well characterized measurements of K and L diagram lines from various expert laboratories, and then accomplished the interpolation using theoretical edge energies. As the work proceeded, the sophistication of these theories reached a high level. Absorption edge energies were estimated by combining measured electron binding energies for outer shells with the energies of emission lines in which the transition terminates in that outer shell. By drawing upon the large existing database of outer electron binding energies that is available from photo-electron spectroscopy, Deslattes et al. were able to achieve a degree of redundancy in edge determination.

The Williams (1986) tabulation of K, L, M, N and O sub-shell edge energies which was used by Elam et al. (2002) does not specifically state its main source of K and L edge values; however, it does cite two well-known photo-electron spectroscopy references as its source for many outer-shell values. Our random examination of its K and L values suggests that they are those of Bearden and Burr (1967).

Another post-Bearden, pre-NIST tabulation is the Lawrence Berkeley Laboratory X-Ray Data Booklet (Thompson, 2001). Its edge energies are taken from the Bearden and Burr tables together with extensive photo-electron data for outer shells. Its X-ray energies are taken from the Bearden table and are restricted to the major lines Kα1, Kα2, Kβ1, Lα1, Lα2, Lβ1, Lβ2, Lγ1, Mα1; we consider this selection insufficient in the context of present-day resolution of energy-dispersive X-ray detectors

While the NIST tabulation of 2002 provides

K, L1, L2 and L3 absorption edge energies and both K and L X-ray energies that cover diagram transitions having final states out to N7, it does not provide M X-ray energies, which are needed in any XRF database. Presumably these could be developed in a reasonably consistent manner by combining M sub-shell edge energies deduced directly from the Bearden tables with updated literature photo-electron edge energies of N, O and P sub-shells.

The energies discussed above are for elements in their natural state. One might reasonably express the view that the question of highly accurate energies for XRF is somewhat academic because of the chemical shifts that occur when the element in question is chemically bound in a molecule. At first sight, the effort to catalogue such chemical shifts appears impossibly large. However Papp (2009) has a proposal that might offer a practical means of accomplishing this.

3. X-ray relative intensities (RIs)

Elam et al. (2002) took their X-ray relative intensities for K and L X-rays from the review paper of Salem et al. (1974). This paper was published just at the beginning of the widespread use of energy-dispersive Si(Li) and Ge detectors in atomic physics measurements. It relies heavily, therefore, on experimental data from older wavelength-dispersive (WD) experiments and from the earliest data of the high-resolution energy-dispersive era. Detection efficiency corrections are more accurate in the energy-dispersive case. In the 35 years that have elapsed since the 1974 Salem et al. paper, there has been a great deal of experimental work based on energy-dispersive spectroscopy, and increasing sophistication of that work has been evident.

3.1 K lines

There are two widely used sets of theoretical prediction, both due to Scofield in the 1970s. The first (Scofield, 1969 and 1974A) is based on the Dirac-Hartree-Slater version of the independent-particle model; it covers essentially all elements in the periodic table. The second (Scofield, 1974B) is based on the more sophisticated Dirac-Fock model. The later, extensive tabulation of Perkins et al. (1991), based upon the Lawrence Livermore Evaluated Atomic Data Library, adopted the RIs from the Scofield DHS calculation. The DHS and DF predictions differ. Salem's 1974 review suggested strongly that the best fit to experimental data for the $K\beta/K\alpha$ ratio differs systematically from the DHS prediction all across the periodic table. The review of Khan and Karimi (1980) showed the same trend, and it also showed that the experimental $K\beta/K\alpha$ data favor the DF predictions over the DHS ones. Scofield himself also made this observation. An extensive tabulation of K-shell experimental data was assembled by Schönfeld and Janβen (1995, 1996). These authors fitted 329 out of the entire 585 $K\beta/K\alpha$ ratios and found good agreement with the Scofield DF predictions above $Z = 30$. They adopt Scofield's DF predictions also for $K\alpha_2/K\alpha_1$ ratios.

With energy-dispersive X-ray detectors used to study atoms having intermediate and high Z values, one can measure not only the gross $K\beta/K\alpha$ ratio but also ratios such as $K\alpha_2/K\alpha_1$, $K\beta_2/K\beta_1$, $K\beta_3/K\beta_1$, etc. This enables a more stringent test of the two predictive approaches; one can emulate the early high-resolution work that was based on WD spectroscopy, but with more accurate knowledge of detection efficiency corrections. Examples of this work are: Barreau et al. (1982), Maxwell and Campbell (1984), Kasagi et al. (1986), Campbell et al.

(1986). These works find that the more sophisticated DF approach gives good agreement with experiment, while the DHS approach is systematically slightly different from experiment. At medium atomic numbers, more recent studies (Martins et al., 1989; Campbell, 2001) reach the same conclusion. Some of these works are missing from the Schönfeld and Janβen tabulation.

At lower atomic numbers, we note that the Scofield DF approach breaks down in its prediction of Kβ/Kα for the special cases of chromium and copper. In the region $22 \leq Z \leq 29$, the 3d shell is filling and in all but two cases the 4s shell is empty; however one 4s electron appears in Cr and two appear in Cu. In these two cases, the predictions fall below the quite well-established monotonic trend of the data, and so Schönfeld and Janβen (1995, 1996) prefer to interpolate between neighboring theoretical values. Polasik (1998) explored this region of atomic number theoretically using multi-configuration Dirac-Fock calculations, from which he demonstrated quantitatively the dependence of the Kβ/Kα ratio on the particular electronic 3p4s configuration. Further work of this type has been done by Jonnard et al. (2002).

Another issue arises in this region of the periodic table, where there is angular momentum coupling between the final 3p hole and the open 3d shell. This results in intense satellites below the $K\beta_{13}$ line, with the largest one (Kβ') displaced by up to 15 eV and ranging in intensity as high as 40% of $K\beta_{13}$. The question arises – should one try to include Kβ' in any new XRF database? The energies and intensities have been much studied: there are many papers from the groups led by Deutsch and by Sorum: examples are Holzer et al. (1997) and Sorum and Bremer (1982); more recent work by Jonnard et al. (2002) is instructive. However, the experimental numbers still are not fully established across the atomic number region of interest, and they are sensitive to chemical bonding, rendering this issue even more complex. A theoretical prediction of these satellite intensities would need a multi-configurational Dirac-Fock approach of the type reported by Jonnard et al. (2002).

There is also the issue of including the radiative Auger satellite predictions of Scofield (1974A), which amount to a few percent for KMM and a fraction of 1% for KLL and KLM.

Finally in this section, chemical bonding effects are known to cause X-ray energy shifts for the lightest elements, so presumably they must also affect intensities: this general issue is different from the specific 3d issue just described. At very low Z values (< 20) there are very few experimental data.

All these issues – chemical effects, radiative Auger satellites, the need for more sophisticated calculations in the $22 < Z < 29$ region, etc are discussed also by Schönfeld and Janβen (1995, 1996).

3.2 L lines

Again, the most recent and extensive theoretical work comes from Scofield. The DHS predictions are tabulated in Scofield (1974A) and the DF in Scofield (1974C and 1975). The results of the latter are given for only 21 elements, and so Campbell and Wang (1989) devised a simple interpolation scheme to provide "DF-equivalent" results for $22 \leq Z \leq 94$. Again, the Perkins (1991) EADL tables reproduce the Scofield DHS values. In the L case, radiative Auger contributions are negligible.

A difference between the DHS predictions and experiment was observed at an early stage in X-ray coincidence experiments with radioactive sources carried out by several groups in the 1970s and 1980s. These works, whose objectives were fluorescence yields and Coster-Kronig probabilities, measured the X-ray intensity ratios L3NOP/L3M, and L2NOP/L2M as a "by-product". Drawing upon these and other results, Salem et al. (1974) showed convincingly that the L3N45/L3M5 ratio departs systematically from the DHS prediction, but at that time the DF prediction was not available for comparison. There was too much scatter in their L2N4/L2M4 data to draw conclusions about a similar departure.

The superiority of DF over DHS was again suggested in a more sophisticated experiment by Papp et al. (1993), who were the first to fit pure L3 and L2 energy-dispersed X-ray spectra using Voigtian lineshapes in order to take account of the natural width of L X-ray lines. (The Voigtian is the convolution of the Gaussian detector response with the Lorentzian natural lineshape). The Voigtian lineshape is only very slightly broader than the widely-used Gaussian approximation, and so the change in line width is a trivially small issue. But the flared shape of the Voigtian far to the left and right of the peak centroid is very different, and this strongly influences the fitting of the background continuum and hence the fitted intensities of weak peaks. The Papp experiment confirmed that the DF model was very good for the two cases Z = 64 and Z = 92. Unfortunately, we are not aware of further L subshell experiments of this type.

Schönfeld and Janβen (1995, 1996) did not undertake a tabulation of L-shell RIs.

The great majority of experimental data on L-shell RIs is for medium and high atomic numbers. Thus the above remarks apply to these regimes. An indication that new studies may be required in the low Z regime is found in the measurements of Müller et al. (2009) on nickel. Their value for the intensity ratio L3M1/L3M4,5 exceeded Scofield's value by 30% and Elam's value by 20%.

3.3 Initial recommendations for RIs of K and L lines

Our view is that the RI portion of the Elam database could be improved. For the K-shell, some minor updating of the Schönfeld and Janβen (1995, 1996) tables could be done. But given that experiments largely (with noted exceptions) support the Scofield DF predictions for the two ratios Kβ/Kα and $K\alpha_2/K\alpha_1$, and given that high-Z experiments also support the Scofield DF values for $K\beta_3/K\beta_1$, we suggest that the Scofield DF predictions could be adopted (again with noted exceptions) for all of the K diagram lines, without resorting to combining these into subgroups such as Kα and Kβ. Modern Si(Li) and SDD detectors have sufficiently good resolution to justify doing this.

For the L-shell, one could perform a modern review which takes the Salem et al. (1974) paper as a starting point and which then seeks out all extant energy-dispersive measurements. One aspect of such a review would be a comparison of wavelength-dispersive with energy-dispersive results, seeking to determine if systematic differences exist and what is their magnitude. The effort required to do this and to conduct a comparison of the Elam RIs with this more modern database is significant but not excessive. Present indications are that the Scofield DF predictions of RIs for the K and L shells, using the Campbell and Wang (1993) interpolation for the L subshells, suffice rather well as an RI database at medium and high atomic numbers. Systematic comparison of these predictions with experimental data from Salem et al. (1974)

and for the subsequent 36 years would enable a comparison that would illuminate the degree of error that is incurred by use of the Elam RI database. While we have not attempted a literature search at low atomic numbers, the new experimental work of Müller at al (2009) suggests that significant experimental efforts may be needed here.

3.4 M lines

In this case, because Salem et al. (1974) do not provide any information, Elam takes a rather empirical approach. We would instead recommend interpolation in the DHS theoretical predictions of Chen and Crasemann (1984).

4. Fluorescence and Coster-Kronig probabilities

These quantities ω_i and f_{ij} were computed for the K, L, M and N subshells in several papers by McGuire (1969, 1970, 1971, 1972) using the non-relativistic Herman-Skillman approximate self-consistent field potential. Later theoretical work on the K, L and M subshells by Chen et al. (1980A[K], 1981 [L], 1980B [M], 1983 [M]) was based on the DHS version of the independent-particle model. For the L case, Chen et al. (1981) provided results for only 22 elements. Therefore, Puri et al. (1993) introduced an interpolation scheme to extend the non-radiative transition rates of Chen to all other atomic numbers, and from these they were able to provide a full set of "Chen-equivalent" values.

Chen (1990) later conducted a limited exploration of what changes might be expected in the L2 and L3 subshells by using the DF approach. He found that ω_2 and ω_3 were changed only very slightly, whereas f_{23} decreased by about 10% at high Z values; in addition, the need for MCDF calculations where dominant Coster-Kronig transitions lay close to energy cut-offs was demonstrated.

4.1 K-shell

The most recent compilation of experimental ω_K values is that of Hubbell et al. (1994). In this work they collect together the results of two previous reviews. One of these is the well-known and widely used review of Krause et al. (1979); the other is the slightly later work of Bambynek (1984), which superseded earlier work by the same author. Figure 1 shows us that there is a smooth Z-dependent difference between the recommended values from the Krause (1979) and Bambynek (1984) reviews, reaching as much as 5% in the region of Z ~ 25. One might argue that the second Bambynek review, with access to more data, would be more accurate. A second reason for preferring that review is the very close agreement of its recommended values with the theoretical predictions of Chen et al. (1980A); in the $20 < Z < 30$ region this agreement is within 1%, and at higher Z the agreement improves to within 0.25%. We observe that the Chen values were obtained by combining DHS non-radiative rates with DF radiative rates: while the DF radiative rates are indeed likely to be more accurate than their DHS counterparts, this blending violates the idea of using a single theory across the database. However, the close agreement with the Bambynek compilation suggests that it has merit. Schönfeld and Janβen (1995, 1996) also adopt the Bambynek (1984) tabulation.

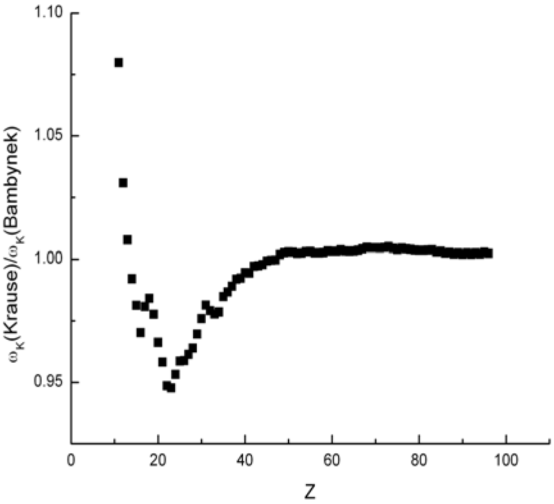

Figure 1: **Ratio between the recommended ω_K values of Krause (1979) and Bambynek (1984)**

Some well-designed and highly accurate new experiments around Z ~ 25 are desirable if we wish to confirm the impression that the Bambynek/Chen recommendations are indeed more accurate than those of Krause.

Elam (2002; private communication, 2009) took his ω_K values from a set of recommendations made by Hubbell et al. (1994); these were based upon a fit to ω_K values which had been extracted from "selected X-ray production cross-section measurements… by both photons and charged particles from the period 1978-1993". At first sight, this selection appears as a good strategy because it might assure consistency between an adopted set of photo-electric cross-sections and the K fluorescence yields. However, as Figure 2 shows, these values differ significantly from the Bambynek and Chen values in a manner that is a smooth function of Z; the difference ranges from -5% around Z=35 to +3% around Z=65. This is a disturbing observation, which leaves us to choose between two significantly different sets of recommended ω_K values.

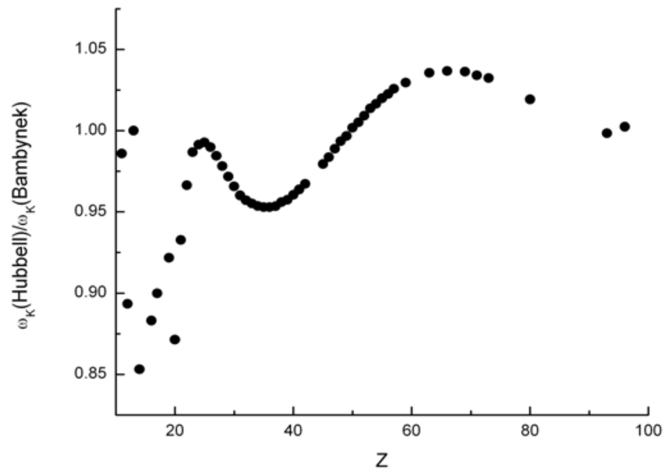

Figure 2. **Ratio between ω_K values proposed by Hubbell et al. (1994) and those of Bambynek (1984)**

In our view, there is a problem with the proposed values of Hubbell et al. (1994). Note once again that these were deduced from measurements of X-ray production cross-section $\sigma(X)$, by charged particle and photon beams. Deduction of ω_K values from such data demands knowledge of the ionization cross-section $\sigma(I)$, which has to be taken from theory. For charged-particle ionization, theoretical hydrogenic cross-sections (Cohen, 1985) were used by Hubbell et al. (1994); but these have been shown to be inferior to cross-sections derived later on the basis of the DHS atomic model, which suggests introduction of systematic error. For photo-ionization, theoretical cross-sections of Scofield (1973) were used; since 1973, these values have effectively had a monopoly but now it would be worthwhile to ascertain if the results are changed by using the more recent tables of Yeh and Lindau (1985) or of Chantler (1995).

Ours is certainly not the first observation of this problem with the recommendations of Hubbell et al. (1994) for ω_K. In response to correspondents, Hubbell et al. issued an Erratum (Hubbell et al., 2004), in which they recommended that their proposed recommendations be set aside in favor of those of Bambynek (1984). Obviously, this Erratum was published after the work of Elam et al. (2002) on the XRF database. We recommend that the proposed new database should revert in the interim to the K fluorescence yield values of Bambynek (1984). Nevertheless, as stated above, a small number of new, sophisticated experiments to measure ω_K in the area of Z=25 would be valuable, as would new investigations at very low values of Z – see below.

There are further ramifications here, which we judge important. The cross-section for K-shell ionization $\sigma(I)$ is related to the cross-section for K X-ray production by the relationship

$$\sigma(X) = \sigma(I)\,\omega_K$$

For electrons and light ions there has been continued interest in measuring $\sigma(I)$ for comparison with theory – which has seen many improvements. These $\sigma(I)$ values are almost always obtained from the above relationship by measuring $\sigma(X)$ and assuming a value of ω_K from some literature database (most often Krause, 1979). Other workers, whose interest lies in the K fluorescence yield, measure $\sigma(X)$ and then use the relationship to determine ω_K, assuming a value of $\sigma(I)$ from the literature. The danger of circular argument is apparent.

We should ask ourselves this question: which is the simpler of the two quantities - K fluorescence yield or K ionization cross-section? We argue that the former is the simpler because it depends on only the atom concerned (with the exception of the next paragraph). In contrast the latter depends on the atom *and* on the projectile (electron, proton, photon), and on details of the interaction between them, and on the complex details of the perturbation of non-participating electrons by the projectile. It might be useful to update the Bambynek (1984) review, with a restriction to experimental data which are as free as possible from assumptions about other quantities, especially theoretical or experimental ionization cross-sections. A second useful exercise would be to compile critically those determinations that are made via photo-ionization, using Scofield's PE cross-sections. One would then test for consistency between the two approaches. Experiments based on charged particle ionization would not be used.

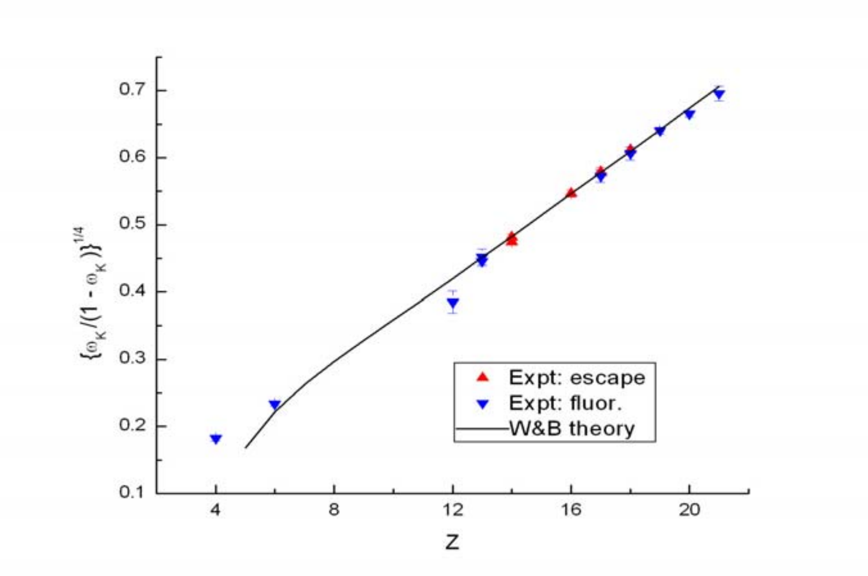

Figure 3. Theoretical and experimental ω_K values at low atomic number: "fluor." Is the method based on detecting fluoresced X-rays

(In passing, we note that the literature is replete with measurements of X-ray production cross-sections from which something is derived assuming something else, sometimes forwards, sometimes backwards. A likely cause of this is simply that these experiments are easy to do. That is not a good reason to do experiments. What is needed are new approaches and clever experiments which attack the problem in a manner that minimizes the need to make assumptions.)

Now we must consider the special situation for low atomic number. Unfortunately, Chen et al. (1980A) provided their DHS predictions only for Z ≥ 18. One theoretical possibility available to us below Z=18 is to accept the non-relativistic HS predictions of Walters and Bhalla (1971); the lack of a full relativistic treatment should not be a serious deficiency for these light elements. There is good agreement between these two theoretical treatments in the region above Z = 18. In *Figure* 3, we follow Bambynek's suggestion of plotting the quantity $\{\omega_K/(1-\omega_K)\}^{1/4}$ in order to present the comparison of the experimental data and the Walters and Bhalla predictions. The agreement is good, apart from the magnesium point. The deviation of the beryllium point from the obvious extrapolation must be set against the fact that Walters and Bhalla did not extend their calculations below Z = 5.

There are further issues at low atomic number, some of which are discussed by Elam et al. (2002) and by Schönfeld and Janβen (1995, 1996). The theoretical predictions are for isolated atoms. At low Z in condensed matter these values may not be appropriate. Krause (1979) gave two tables of recommended values. The larger table was based on experimental data, with only gaseous element data used for Z ≤ 10. A smaller table offered condensed matter values for the range 3 ≤ Z ≤ 15. For sodium the two tables differ by 10%. Krause points out that chemical bonding effects must be important at low Z values, and indeed these have been demonstrated by Campbell et al. (1997) at the 7% level for Mg, Al and Si versus their oxides. Many of the low-Z data points in the literature are from chemical compounds, adding confusion as to the interpretation of the overall data set.

The low-Z data in *Figure 3* are from experiments where elemental targets were fluoresced and from experiments employing gaseous X-ray emitters (some of them chemical compounds) within proportional counters. Carefully designed experimental work is needed in order to: (i) improve the accuracy of pure element ω_K values and to compare these with theory; (ii) understand the effects of chemical bonding.

4.2 L subshells

The Elam database relies upon the review of Krause (1979), which produced a set of recommended values for the six quantities ω_i and f_{ij} (i, j = 1, 2, 3). Of the various modifications to these values that were suggested by Jitschin (1990), Elam accepted some and rejected others. The Krause review was a widely cited work, but now it is 30 years old. Since 1979, the volume of experimental data has tripled and all the new measurements have been done with high energy resolution, which was not the case in the data available to Krause. Moreover, the Chen et al. (1981) DHS calculations of theoretical ω_i and f_{ij} values came later than 1979 and obviously were not available to Krause, who had to work with earlier, less sophisticated theories in reaching his recommendations. The Schönfeld and Janβen (1995, 1996) work confines itself to compiling the mean fluorescence yield over the three subshells.

For all these reasons, Campbell (2003) published a new review, giving recommendations for ω_i and f_{ij} in the region Z > 60, based upon a critical examination of the expanded available data set, and assisted by the DHS and (very limited) DF predictions of Chen (1990). Because of the observed agreement between the Chen et al. DHS predictions for ω_3 and the experimental values from a variety of methods, Campbell recommended using the Chen et al. DHS predictions as a convenient representation of ω_2 and ω_3. However, he took the view that for f_{23} these predictions are too high and should be replaced by DF predictions. Figure 4 shows the differences between the Krause (1979) values and the Campbell (2003) values for ω_2 and ω_3.

Papp (2009) has pointed out privately to Campbell that the agreement between the overall trend of ω_3 values and the DHS theory may be fortuitous: the experimental data are dominated by those from KX-LX coincidence experiments, in some of which (depending on the detector geometry) a correction for angular correlation is necessary. New experiments that focus on ω_3 with minimal assumptions might clarify this possibility.

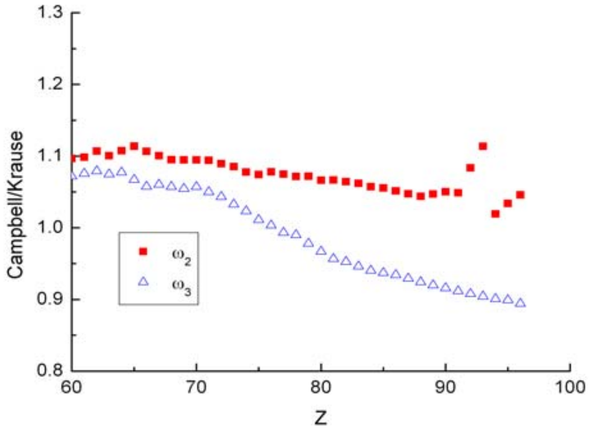

Figure 4.

Ratio between Krause (1979) and Campbell (2003) recommendations for L2 and L3 fluorescence yields

The L1 case was very difficult, due to the high level of scatter in much of the data and to the well-known inability of theory to cope accurately with the challenge of predicting L1 level widths (Campbell and Papp, 2001). This led to a detailed re-examination of the L1 situation (Campbell, 2009). But even after that effort, the L1 case still is troubling. Large uncertainties have to be associated with the recommended values – uncertainties which are, ironically, as large as those recommended by Krause thirty years earlier. Papp et al. (1994) had already attributed these problems in part to neglect of the Lorentzian contribution to X-ray linewidth and neglect of multiple ionization satellites. In addition, evidence is coming to light (Papp et al., 2005) that issues of electronic efficiency may not have been properly addressed in Si(Li) spectroscopy in recent decades, and this may also be a source of error.

Despite the problems, we believe that the Campbell database is preferable to the older Krause database, but Campbell declined to provide recommendations at Z < 60 for the L subshells because of the very low amount of experimental data. So it is an open question what to do for Z < 60.

It needs to be mentioned that the Perkins et al. (1991) EADL tabulation includes all the radiative and non-radiative L subshell transition probabilities and the corresponding fluorescence yields; the Coster-Kronig values may be deduced. The radiative transition probabilities are all based upon the DHS work of Scofield (see above) and the Auger and CK probabilities are based upon the DHS work of Chen et al. (see above). Given our above discussion of Chen's ω_i and f_{ij} values, there appears no need for discussion of the equivalent EADL tabulation. It is worth noting, however, that Perkins et al. (1991) modified the non-radiative transition probabilities, in particular the CK ones, in order that the derived fluorescence yields would agree better with power series formulas used by Hubbell et al. (1989). No details are given as regards these modifications and their magnitude, leading to our view that it is safer to base the present discussion on the original predicted values of Chen et al.

In private communication, Papp (2009) raises an important point. What matters for practical X-ray fluorescence analysis is not having a set of ω and f parameters that have respectability from the viewpoint of basic physics. What is needed is a pair of databases that are mutually consistent – (i) a set of photo-ionization cross-sections; and (ii) a corresponding set of ω and f parameters. In the context of the FP Initiative, he argues for the importance of synchrotron-based experimental effort in that direction. This is a big effort but it merits serious consideration.

4.3 M Sub-shells

Elam adopted the theoretical predictions of McGuire in this case. The experimental data are not capable of supporting any particular theoretical model. One could just as easily adopt the predictions of Chen et al. (1980B, 1983). In either case, the predictions must be inadequate for the higher M subshells due to many-body effects that the theoretical models neglect.

5. Concluding remarks :

The preceding survey indicates potential avenues for XRF database improvement which range from straightforward to difficult and time-consuming. No attempt is made to prioritize, because priorities are best judged by those who are immersed in practical XRF analysis on a day-to-day basis.

Acknowledgements :

This paper has benefited greatly from many discussions with T. Papp, and these are deeply appreciated. Important references were provided by M.-M. Bé, and correspondence with T. Elam was valued. Members of Working Group 4 of the FP Initiative provided helpful comments. The support of the conveners of the FP Initiative is greatly appreciated, as is their approval of the publication of this (slightly updated) paper. Financial support was provided by the Natural Sciences and Engineering Research Council of Canada.

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Announcement of New Editor in Chief and Associate Editors for the journal Radiation Physics and Chemistry

by José Stoop (Publisher, Radiation Physics and Chemistry, Elsevier)

As of February 1st 2010, Professor Christopher Chantler (University of Melbourne) has succeeded Dr. Paul Bergstrom as the Editor in Chief of Radiation Physics and Chemistry.

Dr. Paul Bergstrom has stepped down after 5 years of service to the journal, and we wish him all the best in his future endeavours. We also would like to welcome Professor Christopher Chantler to his new role.

Professor Chantler is an Associate Professor and Reader at the School of Physics, University of Melbourne. He is a Fellow of the Australian Institute of Physics and is Secretary on the Steering Committee for the IUCr International Commission on XAFS, and a Vice-President of the International Radiation Physics Society.

His fields encompass experiment and theory, technical developments and new fundamental insights across physics and chemistry, atomic and condensed matter science.

Professor Chantler has produced the first absolute polarization studies performed on an EBIT, the first investigations of radiative electron capture to test QED and worked on laser resonance spectroscopic tests of QED. He has also been a key developer of X-ray investigations at the NIST EBIT with long term experience in investigating and using EBIT sources. He has extensive experience with investigations at accelerators in Oxford, GSI, Lawrence Berkeley Laboratory and Argonne. He has developed new experimental techniques (XERT), one hundred times more accurate than prior methods in the X-ray regime, for the determination of attenuation and absorption coefficients and the imaginary component of the atomic form factor.

Furthermore, we would also like to welcome our three new Associate Editors for Radiation Physics and Chemistry:

Stephen Best, University of Melbourne
Michael Farquharson, McMaster University
Stefaan Vynckier, University of Louvain

Lastly, we are honoured to welcome Dr. David Bradley on board as Consulting Editor for Radiation Physics and Chemistry.

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Second Announcement

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In 2010, the European Conference on X-Ray Spectrometry (EXRS 2010) will be held on June 20-25, in the Portuguese "Silver Coast" beach resort Figueira da Foz, near the historic city Coimbra that hosts one of the oldest universities of Europe.

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In "Social Events" you can see details on the Conference Dinner. It takes place in the "Grand Casino of Figueira da Foz" http://www.casinofigueira.pt/, in the main room, the "Salão Caffé", which has unique frescos painted on the ceiling. The menu is detailed in the conference website.

For the conference Excursion you may choose the visit to Coimbra and its 700-year old University or the visit to the Fatima Sanctuary and Monastery of Batalha, which is considered UNESCO World Heritage.

IMPORTANT DATES

Abstract submission: 10 April 2010

Abstract acceptance: 15 April 2010

Early registration: 1 May 2010

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