https://doi.org/10.61092/iaea.g8n0-f825

IAEA
International Atomic Energy Agency

INDC(NDS)-0638
Distr. G+SD

# INDC International Nuclear Data Committee

Summary Report of Consultants' Meeting on

## Auger Electron Emission Data Needs for Medical Applications

IAEA Headquarters
Vienna, Austria

9 – 10 May 2013

Prepared by

Roberto Capote Noy and Hyun Kyung Chung
IAEA Nuclear Data Section, Vienna, Austria

Klaus Bartschat
Drake University, Des Moines, IA, USA

Chenzhong Dong
Northwest Normal University, Lanzhou, China

Per Jonsson
Malmö University, Malmö, Sweden

Tibor Kibédi
Australian National University, Canberra, Australia

Filip G. Kondev
Argonne National Laboratory, Argonne, IL, USA

Hooshang Nikjoo
Karolinska Institutet, Stockholm, Sweden

Adriana Pálffy
Max Planck Institute for Nuclear Physics, Heidelberg, Germany

November 2013

**IAEA Nuclear Data Section**
**Vienna International Centre, P.O. Box 100, 1400 Vienna, Austria**

Selected INDC documents may be downloaded in electronic form from
*http://www-nds.iaea.org/publications*
or sent as an e-mail attachment.
Requests for hardcopy or e-mail transmittal should be directed to
*NDS.Contact-Point@iaea.org*
or to:
Nuclear Data Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
A-1400 Vienna
Austria

Produced by the IAEA in Austria

November 2013

**INDC(NDS)-0638**
**Distr. G+SD**

Summary Report of Consultants' Meeting on

# Auger Electron Emission Data Needs for Medical Applications

IAEA Headquarters
Vienna, Austria

9 – 10 May 2013

Prepared by

Roberto Capote Noy
IAEA/NDS
Vienna, Austria

Hyun Kyung Chung
IAEA/NDS
Vienna, Austria

Klaus Bartschat
Drake University
Des Moines, IA, USA

Chenzhong Dong
Northwest Normal Univ.
Lanzhou, China

Per Jonsson
Malmö University
Malmö, Sweden

Tibor Kibédi
Australian National Univ.
Canberra, Australia

Filip G. Kondev
Argonne National Laboratory
Argonne, IL, USA

Hooshang Nikjoo
Karolinska Institutet
Stockholm, Sweden

Adriana Pálffy
Max Planck Institute for Nuclear Physics Heidelberg, Germany

**Abstract**

A summary is given of a Consultants' Meeting on "Auger Electron Emission Data Needs for Medical Applications". Participants assessed and reviewed detailed atomic and nuclear data needs for a number of Auger emitters deemed as potentially suitable for applications in nuclear medicine and radiotherapy. Technical discussions are described in this report, along with recommendations for future work. Presentations by the consultants at the meeting are available at http://www-nds.iaea.org/index-meeting-crp/CM-Auger-2013/.

November 2013

# TABLE OF CONTENTS

# 1. INTRODUCTION

Chemotherapy and radiation therapy are the two major methods to treat cancerous tumours. In radiation therapy this can be done using external radiation beams such as gamma photons, X-rays, protons, and more recently carbon ions. While external radiotherapy is broadly used, there is also an emerging modality that uses ionizing radiation emitted by a radioactive isotope directly delivered to the tumour. Such targeted delivery can be done, for example, by monoclonal antibodies or via an antigentic target. In all forms of cancer therapy, the primary objective is to maximize toxicity of drug/radiation to the tumour cells while minimizing it to the healthy tissue and the organ. In general, radioisotopes emit radiations such as electrons, photons, alpha-particles, and or a combination of these. Currently, many radioisotopes are used either for radiation therapy or for imaging purposes. Among approved radionuclides for use in clinical studies we may mention $^{131}$I (gamma emitter), $^{90}$Y (beta emitter), and $^{211}$At (alpha-emitter).

Among electron emitting radionuclides, there is a particular group of radionuclides which decay by capture of an inner shell electron, which is followed by emission of a shower of low energy electrons. Such radionuclides are generally known as Auger emitters named in honor of Pierre Auger who discovered them in 1925. If these low energy electrons are absorbed completely within the chromosomes they would induce severe DNA damage in the genome to cause cell death. A current list of Auger emitters with existing or potential medical applications is listed in Appendix 1. Among many Auger emitters some, for example $^{125}$I, $^{123}$I, $^{124}$I, $^{111}$In, have been used with different degrees of success in radiation therapy research and applications. However, the decay and emission data of relevant very low-energy Auger electrons (L, M, N, shells) are poorly known.

Selection of radioisotope for radiation therapy requires considerations of their physical and biological aspects. The physical aspects include decay scheme, the physical half-life, binding energies, spectrum of electrons released in the decay, and very important, accurate physical dosimetry including tracing of the isotope by imaging techniques. Biological and medical aspects deal with the biological half-life, mechanism of delivery of the radionuclide to the targeted tumour, imaging, retention and biological dosimetry. There are identified needs for primary physics data for Auger emitters to assist providing better dosimetry and computer simulation.

To fulfill these requirements, the Consultants' Meeting (CM) on "Auger Electron Emission Data Needs for Medical Applications" was held at IAEA Headquarters, Vienna (Austria) on 9-10 May 2013, and attended by seven external consultants. R. Capote Noy (IAEA, Vienna, Austria) served as Scientific Secretary and T. Kibedi (Australian National University, Canberra, Australia) agreed to act as rapporteur. The approved Agenda is attached (Appendix 2), as well as a list of participants and their affiliations (Appendix 3).

The CM was convened to explore what sort of work is reasonable and feasible to produce data for Auger electron production in connection with internal radiotherapy applications. We understand that there is a strictly nuclear side to this problem, but that is not considered here; at this meeting we focused on the atomic and molecular decay process that follows the electron capture nuclear decay.

# 2. REPORTS OF THE PARTICIPANTS

## 2.1 Hooshang Nikjoo (Karolinska Institutet, Sweden)

*Auger electron transport calculations in biological matter*

The talk briefly discussed physical, biophysical, and biological aspects of Auger emitters. A summary of radiationless transition data available in published literature and databases were presented. Data were presented for electron capture (EC), internal conversions (IC), binding energies of some commonly used radionuclides $^{123}$I, $^{124}$I, $^{125}$I, and $^{158}$Gd. For each of these Auger emitting radionuclides some examples of Monte Carlo calculated electron spectra of individual decays were presented. Because most Auger electrons emitted in the decay of radionuclides are short range low energy electrons below 1 keV, a brief discussion was presented on most recent development of physics models for energy loss of electrons in condensed phase and compared with other models and gas phase data. Accuracy of electron spectra calculated in the decay of electron shower by Auger emitting radionuclides depends on availability of accurate physics data. Currently, there are many gaps in physics data as input data to computer codes in need of new evaluation. In addition, comparison should be made between deterministic and Monte Carlo methods to access the accuracy and sensitivity of data to methods and the chosen parameters. It has long been recognized that Auger electron show a high-LET like characteristics when radionuclide is very closely bound to DNA. As most Auger electrons are short range low energy electrons and mostly absorbed with the DNA duplex when in close vicinity to DNA duplex, we believe the physical and biological dosimetry are best achieved by using Monte Carlo track structure simulations able to simulate tracks of low energy electrons below 1keV and in particular sub 100 eV in condensed phase.

## 2.2 Klaus Bartschat (Department of Physics and Astronomy, Drake University, USA)

*Atomic Structure Calculations using the B-Spline R-Matrix Approach*

The B-spline R-matrix (BSR) method [1] and the published Breit-Pauli version of the computer code [2] were mainly developed to allow for an accurate treatment of electron collisions with complex targets. The characteristic features of the method are the use of B-splines as an effectively complete and numerically stable basis to expand the wavefunction of the projectile inside the R-matrix box and the ability to handle term-dependent, and hence non-orthogonal, one-electron orbitals in the calculation of any matrix element of interest.

With a slight change of boundary conditions, bound rather than continuum states can be generated in the frozen-cores approximation, where the close-coupling equations for electron scattering from the ion with one less electron than the target of interest are solved subject to the requirement that the wavefunction vanishes for large distances away from the target nucleus. The method has been applied very successfully to the calculation of energy levels and oscillator strengths [3], as well as photo-induced processes (see [1] for details).

In principle, the BSR method with nonorthogonal orbitals can also be used to calculate Auger emission spectra, although at the present time it may be more appropriate to stick with more standard atomic structure packages, despite their limitations regarding the continuum wavefunction of the ejected electron and the usually enforced orthogonality of the one-electron orbitals. Figure 1, however, shows an example where channel-coupling effects and non-orthogonal orbitals have been used to obtain excellent agreement with experimental Auger shake-off spectra, even in the presence of a laser field that was tuned to a resonance transition in order to enhance a satellite line after creating a hole in the 2s subshell of Na[4,5].

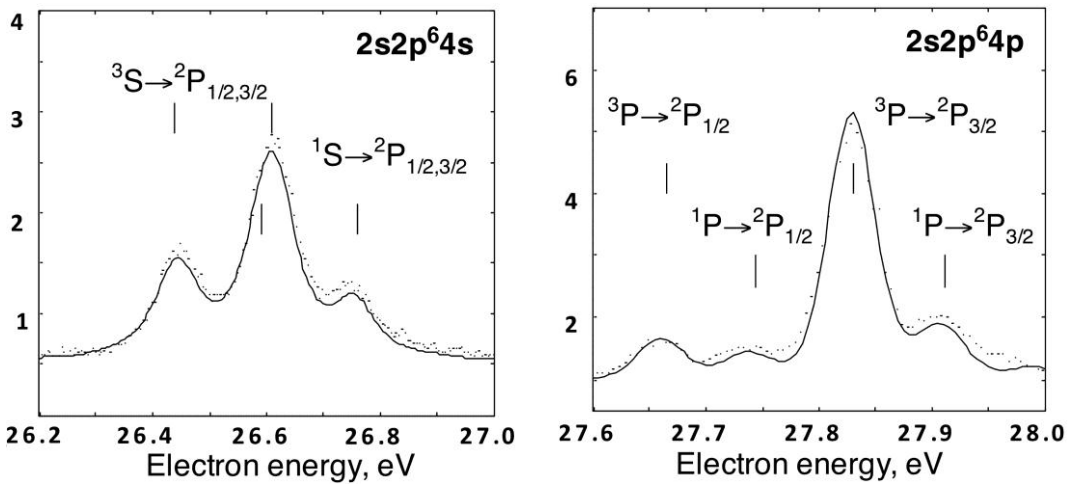


Fig. 1: Ejected electron of sodium vapour at 1.5 keV incident electron energy for the 4s and 4p satellite lines in Na after creation of a 2s hole. Theoretical predictions [4] are compared with experimental data [5]. In the 4p case, a laser was tuned to the 3s-3p resonance transition.

[1] Zatsarinny O and Bartschat K 2013 J. Phys. B **46** 112001
[2] Zatsarinny O 2006, Comp. Phys. Commun. **174** 273
[3] Zatsarinny O and Bartschat K 2009 Phys. Scr. **T134** 014020
[4] Zatsarinny O J. Phys. B **28** 4759
[5] Dorn A, Winnewisser C., Wetzstein M., Nienhaus J., Grum-Grzhimailo A. N., Zatsarinny O. and Mehlhorn W. 1995 J. Elec. Spectr. Rel. Phen. **76** 245

### 2.3 Adriana Pálffy (Max Planck Institute for Nuclear Physics, Heidelberg, Germany)

*Theoretical uncertainties for Auger electron cascades*

The presentation comprises of three main points: a brief review of the processes involved in the Auger electron cascades, the discussion of the theoretical transition rates and their uncertainties, and finally a short summary and bottom-line requirements on theory.

The Auger cascades interesting for medical applications usually start with an inner shell vacancy produced in the neutral atom by internal conversion (IC) or electron capture (EC). A typical example is the $^{125}$I isotope, which decays by EC to an excited state of $^{125}$Te, followed by IC of the latter. The inner shell vacancy propagates towards the outer shells and multiplies via Auger and Coster-Kronig (CK) and super-CK decays. The cascades thus consist of many low-energy electrons, in the range of 20-500 eV, with small propagation ranges of 1-10 nm, i.e., macromolecular dimensions. Such electrons are difficult to detect and not easy to be accounted for in dosimetry - their effect is not included in MIRD. However, since Auger emitters are used in diagnostic nuclear medicine and are candidates for photo-activation therapy, the emitted electron spectrum is of paramount interest. Here is where theory can play an important role.

The main problem remains however that the questions around the Auger emitters are touching biology. Thus, the main issue is: what are the *biological* implications of the Auger-electron cascades. The energy imparted to the biological tissue is related to the simultaneous energy deposition by many Auger electrons (questions here: how many are they, what is their energy spectrum?) and to the charge potential remaining with the multiply ionized ion (how does charge neutralization work and what damage does it produce?). The trouble begins here, with a number of unanswered questions. How are the valence electrons to be considered for the

decay cascade? What is the effect of the molecular binding of the radioisotope on the sensitive low-energy electrons emitted in CK and super-CK processes? How should the Auger spectra be calculated in a way relevant to matter in condensed phase, in particular in macromolecules?

Do we need to include vibrational effects due to mechanical nuclear dynamics in molecule, as some studies suggest?

How does charge neutralization occur? How much does it contribute to cellular damage? What cellular damage mechanisms come into play? What is the role of indirect effects such as radical species which arise from water radiolysis? Do we need to take into account the Auger decay from ionized water molecules around the cell DNA?

Most of these issues are so far not included in the traditional modelling of the decay cascades. This is done by either deterministic or Monte Carlo approaches which both require the following input data:

- *Nuclear data* from ENSDF,
- *Fluorescence yields*, the typical data are the Dirac-Hartree-Slater (DHS) radiative rates by Scofield from 1974,
- *Auger and CK transition rates*, here values from the DHS independent-particle model by Chen (1981) are used and
- *Electronic binding energies*.

The fluorescence yields, Auger and CK transition rates and electronic bound energies are available in the Evaluated Atomic Data Library (EADL) compilation. How precise is this data?

A review of the theoretical calculations available so far includes the Dirac-Fock codes by Desclaux [1], which can take into account multiple vacancies; the relativistic calculations by Chen [2] using the DHS independent-particle model (considers only single vacancy) and the higher-shell calculations of McGuire [3]. A study of the theoretical uncertainties for the L shell [4] shows that for theoretical radiative and non-radiative rates these can be on the order of as much as 30 %. Thus even M. H. Chen, who calculated the non-radiative rates available the EADL database, once, mused about using multi-configurational Dirac-Fock (MCDF) calculations as done for highly charged ions. The MCDF method is an extension of the relativistic Dirac-Fock method involving a linear combination of test functions with different inner symmetry to construct the atomic state function. The eigenvalue problem using the variation of the atomic state function coefficients and the variation of the orbital wave functions leads to a set of coupled integro-differential equations that can be then solved numerically.

A well-known code for MCDF calculations is the General Relativistic Atomic Structure Package (GRASP) [5], which can provide bound wave functions and radiative transition rates including Breit and QED corrections. The calculation of Auger rates requires however also continuum wave functions, which are not directly available from GRASP. Back in 1991, an Auger rate module was developed at the University of Giessen in Germany. The AUGR module was included in the GRASP 1 version. This calculates the Dirac spinor of the continuum wave function in the screened potential of the ion using the frozen-orbital approximation. The GRASP results for dielectronic recombination in highly charged ions are in excellent agreement with experimental results. This is possible due to the optimal-level calculations for few-electrons ions, where one can focus on the desired configurations. The

question is whether this would also work for the case of only few vacancies and many electrons in higher shells?

To conclude, on the present wish list is a code which delivers transition energies, electronic bound and continuum wave functions, radiative rates and Auger and CK rates which take into account the real vacancy situation in the ion and the specific molecular environment. Instead, what we have are difficulties to produce accurate values even for isolated atoms!

MCDF calculations for multi-vacancy scenarios are in principle possible but rather time-consuming due to the difficulty to achieve convergence for many-electrons few-vacancies systems. Furthermore, the question arises whether it is worth the trouble and time to perform MCDF calculations for isolated atoms, since this is not the real case. The audience says a firm "yes!". Anything is better than the present state of affairs with single-vacancy independent-particle model calculations and a comparison will make clear what difference do better theoretical rates make for the Auger cascades modelling.

[1] J. P. Desclaux, Comput. Phys. Commun. 9, 31 (1975).
[2] M. H. Chen et al., At. Data Nucl. Data Tabl. 19, 97 (1977), ibid. 24, 13 (1979).
[3] E. J. McGuire, Research Reports SC-RR-710835 (1972) and SAND-75-0043 (1975), Sandia Laboratories.
[4] J. L. Campbell, At. Data Nucl. Data Tabl. 85, 291 (2003)
[5] P. Jönsson et al, Comp. Phys. Comm. 177, (2007) 597

### 2.4 Tibor Kibedi (Australian National University, Australia)

*Data needs for Auger electron cascade simulations*

T. Kibédi [1], B.Q. Lee [1], A.E. Stuchbery [1], F.G. Kondev [2], K.A. Robertson [1]

1 Department of Nuclear Physics, The Australian National University, ACT 0200, Australia
2 Nuclear Engineering Division, Argonne National Laboratory, Argonne, IL 60439, USA

A brief description was given of the current progress in developing a new computational model to evaluate the complete energy spectra of Auger electron emission in nuclear decay. It is widely accepted that, due to their short range, low energy Auger electrons have the potential to be used for radiotherapy. Accurate knowledge of Auger yields is needed both to evaluate the dose to healthy cells when radioisotopes are administered for diagnostics, and to design radioisotope use in the targeted cancer therapy.

Comparatively, progress in theoretical modelling of the Auger cascade has been slow and leading researchers have expressed their concerns regarding the accuracy of the existing Auger-electron spectra calculation [1]. Available computer programs and methodologies used to calculate the energy spectra and emission probabilities for such transitions are often limited to only K and L shells, and they also neglect the presence of secondary vacancies and use binding energies for neutral atoms. Experimental data for such processes are scarce.

A new model aimed at improving the low-energy electron data is being developed which uses the most up-to-date nuclear and atomic input data [2]. Nuclear structure data is extracted from the ENSDF, electron capture rates are taken from Schönfeld compilation [3] and internal conversion coefficients from BrIcc [4].

The propagation of the primary vacancies is treated using a full Monte Carlo approach. Atomic transition rates are obtained from the Evaluated Atomic Data Library [5] and theoretical transition energies from the RAINE Dirac-Fock code [6].

Representative energy spectra, charge state distributions and Auger yields for $^{99m}$Tc (IT), $^{111}$In (EC) and $^{131m}$Xe (IT) were presented and compared with available experimental data. In summary, the new model has thus far proved to be a useful tool in calculating Auger energy spectra and charge distributions of residual ions following nuclear decays. We do not expect it will achieve perfect agreement with all experimental results. For example, the disagreement between the model and experiment for KLL Auger electrons signifies that our work is not complete. However we understand the reasons for these discrepancies between theory and experiment. The pilot model will be the cornerstone for our future work using GRASP2K [7] and RATIP [8] to evaluate more accurate atomic transition rates and electron configuration energies.

[1] H. Nikjoo et al, Int. J. Radiat. Biol. 84, (2008) 1011.
[2] B.Q. Lee et al., Comp. Math. Meth. Med., Art. ID 651475 (2012) doi:10.1155/2012/651475.
[3] E. Schönfeld, Appl. Radiat. Isot. 49, (1998) 1353.
[4] T. Kibédi et al, Nucl. Instr. and Meth. in Phys. Res. A589, (2008) 202.
[5] S.T. Perkins et al, Lawrence Livermore National Laboratory, UCRL-50400 30, (1991).
[6] I.M. Band et al, At. Data and Nucl. Data Tables 81, (2002) 1.
[7] P. Jönsson et al, Comp. Phys. Comm. 177, (2007) 597.
[8] J. Nikkinen et al, Comp. Phys. Comm. 175, (2006) 348.

### 2.5 Chenzhong Dong (Northwest Normal University, Lanzhou, China)

*Theoretical Calculations of Auger Electron Spectrum*

Hollow atoms and ions with various vacancies can be produced by electron impact, photoionization, ion-atom collision, electron capture (EC), internal conversion (IC), and so on. These states are unstable and therefore may decay either radiatively by emitting photons or non-radiatively by emitting Auger electrons. Usually the final states of the first step decay are also unstable, and they can create various cascades involving X-ray and Auger as well as Coster-Kroning. As most of these Auger electrons have very low energies with ranges of the order of subcellular dimensions in tissue, so the biological effects of such electrons can be significantly higher than expectations based on the average absorbed dose (MIRD method) to the organ.

On the basis of the well-known GRASP92/2K [1] and the component AUGER of the RATIP packages [2] as well as the FAC packages in the frame of Multi-configuration Dirac-Fock method, a series of works on the Auger decay processes have been carried out by our group in recent years [3]. Firstly, Auger widths, radiation widths, Auger yields and fluorescence yields of the hollow atoms and ions with high Z have been calculated. Secondly, on the basis of a previous model on radiative-Auger cascade for calculations of final-charge-state distributions of hollows ions, a new model of radiative-Auger cascade plus shake off has been constructed, and was applied to study the final-charge-distribution of some initial hollow ions. Thirdly, the Auger decay spectrum of K-shell ionized Np ions have been studied theoretically using FAC code. At last, the angular distribution of Auger electron following inner-shell electron-impact excitation has also been calculated by our group recently. In all of these calculations, the Breit interaction, the QED corrections and the nuclear finite mass effects can be considered.

[1] P.Jonsson, X.He, C.F.Fischer and I.P.Grant, Comput. Phys. Comm. 177, 597(2007).
[2] S.Fritzche, Comput. Phys. Comm. 141, 163(2001).
[3] X.L.Wang, C.Z.Dong,et.al.Chin.Phys.Lett.29,43201(2012); C.Z.Dong, et.al. J. Phys. B. 39 3121(2006); H.W.Hu, C.Z.Dong, Acta.Phys.Sin. 55,6326 (2006); H.W.Hu, C.Z.Dong, Acta.Phys.Sin.55,6326(2006); X.L.Wang, C.Z.Dong, et.al. Acta.Phys.Sin. 58,5297 (2009); X.L.Wang, C.Z.Dong, Chin. Phys. Lett. 29, 103201(2012) ;X.B.Ding, C.Z.Dong, Chin.Phys.Lett. 29, 63201 (2012).

### 2.6 H.-K. Chung (Atomic and Molecular Data Unit, IAEA Nuclear Data section)

*Studies of inner shell excitation and ionization and the resulting cascades by x-ray free electron lasers*

An X-ray free electron laser (XFEL) provides a unique opportunity to study extreme states of matter. The lasers of sub-nano second pulses and high photon energies ranging from 800 eV to 10 keV make it possible to probe ultrafast transitions occurring in sub-nanometre ranges. An XFEL called Linear Coherent Light Source (LCLS) is currently operated by SLAC National Accelerator Laboratory and located in Palo Alto, USA. The European XFEL under the construction at DESY, Hamburg in Germany is scheduled to start an operation in 2015. Studies in the novel XFEL facility are already making high impacts on physical, chemical, biological and material sciences.

In addition to scattering to probe target structures, X-ray free electrons lasers can be applied to drive ionization, excitation of bound and free electrons, particularly, inner-shell electrons. Once an electron is removed from an inner-shell state by photons, the hole is filled either by a radiative decay or by a radiation-less Auger decay. While radiative decay rates of inner-shell holes increase with the atomic number Z, Auger decays dominate for low Z elements with the Auger yield of ~98% for Neon. When the XFEL photon energy greater than K-shell ionization energy (~ 870 eV) is applied to a neon gas from a gas jet, neutral atoms are ionized with an K-shell hole ($Ne^{1+}$), which rapidly decay to the next charge state ($Ne^{2+}$) since the Auger decay time scale is 2.4 femto-seconds. Therefore, an asymmetry in charge state distribution is observed to show that the doubly ionized charge state ($Ne^{2+}$) is more observed than the singly ionized charge state ($Ne^{1+}$) [1].

When the XFEL photons interact with solid targets, however, the asymmetry disappears since the collisional processes by free electrons are comparable to Auger processes. Due to the electron heating by high energy photons, electron and ion temperatures increase rapidly during the laser pulse, electron collisions dominate ionization processes compared with photo-ionization and cascades of Auger electrons [2]. A time-dependent collisional and radiative model or NLTE (non-local thermodynamic equilibrium) model taking an account of collisional and radiative processes in the atom gives a good description of atomic processes and agree well with spectroscopic observations. It is noted that the ionization potentials of bound electrons change due to interactions with neighbouring atoms in a dense medium [3].

EUV-free electron lasers can be employed to study short-time electron dynamics for low Auger electrons by ionizing higher inner-shell electrons (that is, L-shell electrons instead of K-shell electrons). Spectroscopic observations bear signatures of Auger electrons and Monte-Carlo simulations including electronic band structures have predicted the cascades of Auger electrons and the time-dependent electron energy distributions [4].

X-ray and EUV free electron lasers can readily create Auger states by photoionization and hence provide a good benchmark experiment to test Monte Carlo methods or any model to

explain the cascades of Auger electrons as a function of time and energy.

[1] L. Young et al., Nature 466, 45 (2010).
[2] S. Vinko et al., Nature 482, 59 (2012)].
[3] O. Ciricosta et al., PRL, 109, 065002 (2012).
[4] N. Medvedev et al., PRL 107, 165003 (2011)].

# 3. SUMMARY OF THE DISCUSSIONS AND RECOMMENDATIONS

Extensive discussions held at the meeting can be summarized in four working directions as stated below:

## 3.1 Benchmarking of new calculations of atomic data vs. atomic data tabulated by Perkins et al 1991 (EADL) needed as input for Auger-cascade calculations.

The creation of a vacancy in an inner atomic subshell leads to a series of transitions as the vacancy moves to outer subshells and the atom relaxes back to a stable configuration. There are two types of transitions; radiative and non-radiative.

**Radiative transitions**

In a radiative transition a vacancy in one subshell is filled by an electron from an outer subshell and it is followed by X-ray emission. The rate and X-ray photon energy for such a transition can be obtained from relativistic multiconfiguration Dirac-Fock calculations or by a combination of multiconfiguration and perturbation approaches. Both the energies of the initial and final states should be computed as well as the transition matrix element between the states. To obtain accurate values for both the rate and the transition energy several effects must be carefully handled. These are: nuclear size effects, electron-correlation effects involving Coulomb and Breit interaction, QED effects (R. Deslattes et al., Reviews of Modern Physics, vol 75, 35-99, 2003). A special challenge is to balance the correlation effects for the initial and final state wave function in the transition to obtain accurate transition energies. For radiative transitions involving a state with a vacancy it is also important to describe the initial and final state wave functions by different and non-orthogonal one-electron orbital sets.

Modern atomic structure theory has come far in meeting these challenges. There are a number of freely available computer codes that can be used to compute both the transition energies and the rates with high accuracy. There are many codes that can be used, four of them are

- GRASP2K (http://dx.doi.org/10.1016/j.cpc.2013.02.016),
- MCDFGME (https://dirac.spectro.jussieu.fr/mcdf/mcdf_welcome/mcdf_homepage.html),
- Dirac (http://diracprogram.org/), and
- RATIP (http://dx.doi.org/10.1016/j.cpc.2012.02.016).

**Non-radiative transitions**

In a non-radiative transition, the initial vacancy is filled by an electron from an outer subshell, the available energy given to the removal of an electron from the same subshell or from one further out. The rates of the non-radiative transitions, or Auger rates, involve the matrix element of the electron-electron interaction Hamiltonian. The latter comprises of two terms, the Coulomb electron-electron interaction and the Breit term which corresponds to the interaction between the two electron currents. Especially for highly-charged ions, the Breit interaction can have a significant contribution to the total matrix element. Two difficulties arise in the calculation of Auger rates: (i) the final state contains a free electron, hence continuum electronic wave are required. These should be calculated taking into account the screened Coulomb field of the nucleus, which is not trivial for complicated many-electron bound electronic configurations. (ii) The traditional multiconfiguration Hartree–Fock or Dirac-Fock and configuration interaction methods are based on a single orthonormal orbital basis. The challenge is here to compute the two-particle Auger matrix elements between states that are separately optimized and built from different orbital sets (see for instance Tulkki et al. Phys Rev A 48, 1277-1291, 1993).

## 3.2 Intercomparison of Auger emission spectra calculations for $^{123}$I and $^{125}$I vs. published data

The knowledge of the full energy spectrum of the Auger-electrons is required for the applications of the radioisotopes for medical applications. For low energies (<1 keV) on one hand only a handful of experimental data is available, on the other hand the existing calculations often produce incomplete or contradicting information. Table 2.2 compares the total yield of Auger-electrons for selected radioisotopes. The (large) differences can be attributed to differences in the nuclear and/or atomic data used or to the treatment of the vacancy propagation. For a more detailed analysis see Lee et al. (2009).

**Table 1** Total yield of Auger-electrons for selected radioisotopes. The yield is given for a single nuclear decay event.

| | **RADAR** | **DDEP** | **Eckerman & Endo (2007)** | **Howell (1992)** | **Stepanek (2000)** | **Pomplun (2012)** | **Lee (2012)** |
|---|---|---|---|---|---|---|---|
| **$^{99m}$Tc (6.007 h)** | 0.869 | 0.13 | 4.363 | 4.0 | | 2.5 | 3.37 |
| **$^{111}$In (2.805 d)** | 1.136 | 1.16 | 7.215 | 14.7 | 6.05 | | 5.749 |
| **$^{123}$I (13.22 h)** | 1.064 | 1.08 | 13.71 | 14.9 | | 6.4 | 6.806 |
| **$^{125}$I (59.4 d)** | 1.77 | 1.78 | 23.0 | 24.9 | 15.3 | 12.2 | 10.91 |
| **$^{201}$Tl (3.04 d)** | 0.773 | 0.614 | 20.9 | 36.9 | | | 11.9 |

**Recommendation:**

To carry out detailed calculations with the available models (Lee et al. 2012, Wang et al. 2012, and others) and compare the calculation results with each other and the existing theoretical and experimental data on Auger-electrons. Based on their importance, the electron capture decay of the $^{123}$I and $^{125}$I isotopes are recommended. These calculations should use the same nuclear data (decay schemes, electron capture rates, photon emission energies and intensities and conversion coefficients).

**References:**


| | |
|---|---|
| RADAR | RAdiation Dose Assessment Resource (RADAR),<br>http://www.doseinfo-radar.com/RADARHome.html<br>M. G. Stabin and L. C. Q. P. da Luz,<br>*"Decay data for internal and external dose assessment"*<br>Health Physics, vol. 83, no. 4, pp. 471–475, 2002. |
| DDEP | Decay Data Evaluation Project,<br>$^{99m}$Tc: C. Morillon, M.M. Bé, V.P. Chechev, A. Egorov (2012);<br>$^{111}$In: V.P. Chechev (2006); $^{123}$I: V. Chisté, M. M. Bé (2004);<br>$^{125}$I: V. Chisté, M. M. Bé (2010); $^{201}$Tl: E. Schönfeld, R. Dersch (2005),<br>http://www.nucleide.org/DDEP WG/DDEPdata.htm |
| **Eckerman & Endo (2007)** | K. F. Eckerman and A. Endo,<br>*"MIRD: Radionuclide Data and Decay Schemes"*<br>Society of Nuclear Medicine, Reston, Va, USA, 2007. |
| **Howell (1992)** | R. W. Howell,<br>*"Radiation spectra for Auger-electron emitting radionuclides: report No. 2 of AAPM Nuclear Medicine Task Group No. 6"*<br>Medical Physics, vol. 19, no. 6, pp. 1371–1384, 1992. |
| **Stepanek (2000)** | J. Stepanek,<br>*"Methods to determine the fluorescence and Auger spectra due to decay of radionuclides or due to a single atomic-subshell ionization and comparisons with experiments"*<br>Medical Physics, vol. 27, no. 7, pp. 1544–1554, 2000. |

| | |
|---|---|
| **Pomplun (2012)** | E. Pomplun,<br>*"Monte Carlo-simulated Auger electron spectra for nuclides of radiobiological and medical interest—a validation with noble gas ionization data"*<br>International Journal of Radiation Biology, vol. 88, no. 1-2, pp. 108–114, 2012. |
| **Lee et al. (2012)** | B.Q.Lee, T. Kibédi, A.E.Stuchbery,and K.A.Robertson,<br>Atomic *"Radiations in the Decay of Medical Radioisotopes: A Physics Perspective"*<br>Computational and Mathematical Methods in Medicine, Volume 2012, Article ID 651475 |
| **Wang et al. (2012)** | Wang Xiang-Li, Dong Chen-Zhong, Xie Lu-You, Shi Ying-Long, Saber Abdalla, Zhou Wei-Dong, "The Radiative and Auger Decay Properties of K-Shell Ionized Np Ions", Chinese Physics Letters, 29, 103201, 2012 |

### 3.3. Compilation of references and experimental data

The development of new Auger-electrons computational tools would require extensive comparison (benchmarking) with available experimental data on both Auger-electron energies and emission probabilities for a range of decaying radionuclides. The survey of the experimental data available in the nuclear structure and decay databases (ENSDF – www.nndc.bnl.gov/ensdf and DDEP - http://www.nucleide.org) showed that the relevant experimental Auger-electron data are not included (evaluated) in those databases. Instead, the DDEP database provides only limited calculated values for the Auger-electron energies and emission probabilities, which are determined from the recommended decay schemes. Similar information can be obtained using the ENSDF analysis program RADLIST. The bibliographical information in the NSR database (www.nndc.bnl.gov/nsr) is also incomplete. There is no keyword on "Auger electron emissions" and if one search on a general "Auger" in the text, 270 entries were found, but only a handful of those referred to published experimental data, with many know references missing.

An extensive compilation of bibliographical data of relevance to the medical research community is available at the Harvard Medical School Website that was developed and maintained (1925-2007) by Prof. Kassis - http://www.hms.harvard.edu/kassislab/.

**Recommendation:**
A dedicated effort to compile relevant bibliographical and experimental data on Auger-electron energies and emission probabilities is required. The IAEA-NDS needs to address the issue, by organizing a group of nuclear structure and decay experts, who can complete such a survey. In addition, this effort would lead to an improved quality of both the NSR and ENSDF databases, which development and maintenance are under auspices of IAEA-NDS.

### 3.4 Motivation of new experiments to address experimental data needs for high resolution Auger electrons

A direct measurement of Auger electrons following the internal conversion (IC) is critical to benchmark current models of Auger electron transport in biological matter. However, such comprehensive experiments are very scarce and the most recent experiments date back to 1980s [Ref: Aksela H, et.al. Phys.Rev.A. 39, 3401(1989), I. Ahmad et al., Phys. Rev. C 27, 2239 (1983)] though resulting charge state distributions and fluorescence radiation spectra have been measured somewhat later [Ref: P. N. Johnston, Nuclear Instruments and Methods in Physics Research B 56, 57 (1991).]. In addition, little is known for Auger transition probabilities and fluorescent yields from the states with vacancies in the outer-shells such as M-shell or N-shell vacant autoionizing states. It is urgent to validate those data to increase the fidelity of comprehensive simulations of

Auger cascades.

The 4th generation light sources of free electron lasers (FEL) offer high intensity and ultrashort (< 1ps) photon sources with variable energies from tens of eVs (EUV) to tens of keV (X-rays). An electron beam interacts with matter by excitation, ionization and recombination and scattering processes and produces a matter of states extremely complicated to explain. However, photon sources with a narrow bandwidth have a merit to preferentially ionize electrons from an inner-shell of interest to produce a sample primarily undergoing Auger cascades or radiative decay. In the example of $^{125}$I, an ionization of M-shell electrons (3s, 3p, 3d) requires photon energies of 600-1100 eV and that of N-shell electrons (4s, 4p, 4d) requires 50-190 eV. The energy ranges are readily available at current FEL facilities and the high repetition rate makes it possible to explore cascades as a function of photon energies as different initial vacancies are produced at different photon energy. The FEL facilities such as LCLS (SLAC, USA), FLASH (DESY, Germany) are equipped with ion and electron time-of-flight (TOF) spectrometers or COLTRIMS (Cold Target Recoil Ion momentum Spectroscopy) and REMI (Reaction Microscopes) to characterize ions and electrons leaving the reaction volume. In addition to particle detection, scattered and fluorescent photons are routinely measured with large-area, low-noise photon cameras (pnCCD) in EVU and X-ray energy ranges.

Data needs in Auger electron transport modeling in biological matter may be addressed by well- designed experiments at FEL light sources, which can offer a comprehensive picture of cascades of Auger electrons by measuring ions, electrons and photons after the creation of inner-shell vacancies. Experiments can provide data to benchmark currently available models and data used within. Other light sources such as synchrotron radiation sources may be considered with careful designs of experiments. It would be necessary to motivate atomic physics community with the importance and applicability of Auger cascades experiments.

## CONCLUSIONS

The main goal of this meeting was to establish collaborations between Auger (nuclear) data users and (atomic) data producers and stimulate long-due developments in Auger electron emission calculations. Low energy Auger data are needed in internal-therapy applications, and clear deficiencies in existing data have been identified and were addressed. The contents of this report constitute a subjective selection of relevant radionuclides based on our knowledge of Auger emitters used in medical applications. Participants assessed and reviewed detailed atomic and nuclear data needs for a number of Auger emitters deemed as potentially suitable for applications in nuclear medicine and radiotherapy. Technical discussions are described in this report, along with recommendations for future work. A follow-up IAEA meeting to check adopted recommendations and actions is foreseen.

# APPENDIX 1 List of Auger emitters with potential medical applications

| Radionuclide | Half-life ($T_{1/2}$) |
|---|---|
| $^{51}Cr$ | 2.77 d |
| $^{64}Cu$ | 12.7 h |
| $^{67}Ga$ | 3.36d |
| $^{71}Ge$ | 11.43d |
| $^{73}Se$ | 39.8m |
| $^{75}Se$ | 1.2d |
| $^{77}Br$ | 2.38d |
| $^{80m}Br$ | 4.42h |
| $^{94}Tc$ | 4.42h |
| $^{99m}Tc$ | 6.01h |
| $^{103}Pd$ | 16.991d |
| $^{111}In$ | 2.8047d |
| $^{114m}In$ | 4.95d |
| $^{123}I$ | 13.2h |
| $^{124}I$ | 4.18d |
| $^{125}I$ | 60.1d |
| $^{140}Nd$ | 3.37d |
| $^{159}Gd$ | 18.5 h |
| $^{167}Tm$ | 9.25d |
| $^{178}Ta$ | 2.36h |
| $^{193m}Pt$ | 4.33d |
| $^{195m}Pt$ | 4.02d |
| $^{197}Hg$ | 64.14h |
| $^{201}Tl$ | 3.0421d |

## APPENDIX 2 Meeting Agenda

**Consultants' Meeting on**

***"Auger electron emission data needs for medical applications"***

IAEA Headquarters, Vienna, Austria

9 – 10 May 2013

Conference Room A0531

IAEA scientific secretary: Roberto Capote Noy

**AGENDA**

## Thursday, 9 May

| | |
|---|---|
| **08:30 - 09:30** | **Registration** (IAEA Registration desk, Gate 1) |
| **09:30 - 10:00** | **Opening Session**<br>Welcoming address and introductory Remarks (R. Capote Noy)<br>Election of Rapporteur<br>Adoption of Agenda |
| **10:00 - 13:00** | **Session 1: Presentations and Discussions** (*coffee break as needed)* |
| | ***13:00 – 14:00 Lunch*** |
| **14:00 – 18:00** | **Session 1 (cont'd)** |
| ***19:00*** | ***Dinner at a restaurant in the city*** |

## Friday, 10 May

| | |
|---|---|
| **09:00 - 12:00** | **Session 2: Discussions and drafting of the report** (*coffee break as needed)* |
| ***12:00 - 13:00*** | ***Lunch*** |
| **13:00 – 15:00** | **Review and Approval of the Summary Report**<br>**Closing of the Meeting** |

## APPENDIX 3 List of participants

**Consultants' Meeting**

***"Auger electron emission data needs for medical applications"***


**AUSTRALIA**
Tibor KIBEDI
The Australian National University
Canberra ACT 0200
Tel. +61 2 6125 5111
E-Mail: Tibor.Kibedi@anu.edu.au

**CHINA**
Chenzhong DONG
Northwest Normal University
Department of Physics
967 East Anning Road
Lanzhou 730070
E-Mail: dongcz@nwnu.edu.cn

**GERMANY**
Adriana PALFFY
Max Planck Institute for Nuclear Physics
P.O.Box 103980
Heidelberg 69029
Tel. +49 6221 516-171
E-Mail: Palffy@mpi-hd.mpg.de

**SWEDEN**
Hooshang NIKJOO
Karoliska Institutet
Department of Oncology-Pathology (OnkPat)
SE-171 77 Stockholm
Tel. +46 8-524 800 00
E-Mail: Hooshang.Nikjoo@ki.se

**SWEDEN**
Per JONSSON
Malmö University
Malmö högskola, Teknik och samhälle
205 06 Malmö
Tel. 040-66 57251
E-Mail: per.jonsson@mah.se

**USA**
Filip G. KONDEV
Argonne National Laboratory
Nuclear Engineering Division
9700 South Cass Avenue
Argonne, IL 60439
Tel. +1 (630) 252 4484
E-Mail: kondev@anl.gov

**USA**
Klaus BARTSCHAT
Department of Physics and Astronomy
Drake University
Des Moines, Iowa 50311
Tel. +1-515-271-3750
E-Mail: klaus.bartschat@drake.edu

**IAEA**
Roberto CAPOTE NOY
NAPC Nuclear Data Section
Vienna International Centre
PO Box 100
1400 Vienna, Austria
Tel. +43-1-2600-21713
E-Mail: r.capotenoy@iaea.org

**IAEA**
Hyung K. CHUNG
NAPC Nuclear Data Section
Vienna International Centre
PO Box 100
1400 Vienna, Austria
Tel. +43-1-2600-21713
E-Mail: H.Chung@iaea.org

## APPENDIX 4 Meeting Photo

**From the left**
**Standing:** H. Nikjoo, R. Capote, T. Kibédi, F. Kondev, K. Bartschat, C. Dong, P. Jonsson
**Sitting :** H.-K. Chung, A. Pálffy

Nuclear Data Section
International Atomic Energy Agency
Vienna International Centre, P.O. Box 100
A-1400 Vienna
Austria

E-mail: NDS.Contact-Point@iaea.org
fax: (43-1) 26007
Telephone: (43-1) 2600-21710
Web: http://www-nds.iaea.org