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Low-energy X-ray standards from hydrogenlike pionic atoms

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We demonstrate the first step of a complete program, which consists in establishing an X-ray energy standard scale with the use of few-body atoms, in the few keV range. Light pionic and muonic atoms as well as one and two-electron ions from Electron-Cyclotron Ion sources are used. The transition energies are calculable from quantum electrodynamics, meaning that only a very limited subset need be measured and compared with theory, while providing a large number of standard lines. Here we show that circular transitions in pionic neon atoms, completely stripped from their electrons, reveal spectral lines which are narrow, symmetric and well reproducible. We use these lines for the energy determination of transition energies in complex electronic systems, like the $K\alpha_{1,2}$ transitions in metallic Ti, which may serve as secondary standard.

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Accurate (below 1 ppm) and *reproducible* X-ray wavelength standards with reasonably dense set of lines would be very valuable for the most widespread application of X-rays: the determination of crystal lattice parameters with diffractometric methods (see e.g., Ref. [1] and references therein.) Other practical applications are found, like the energy calibration of synchrotron radiation beams, monochromators and spectrometers and the determination of the response function of X-ray spectrometers and diffractometers. X-ray standards can thus be useful in many areas of modern science like crystallography, solid state, molecular, atomic and particle physics, chemistry, and biochemistry.

A recent experiment used the ^{57}Fe Mössbauer radiation, excited by synchrotron radiation, improved the energy (wavelength) standard for the energy region of 14 keV by two orders of magnitude in accuracy from 10 ppm to 0.2 ppm [2]. This attempt, while very promising, is very difficult to extend to lower energies, where electron conversion would dramatically reduce the nuclear fluorescence. In the absence of an appropriate excitation source, such as synchroton radiation, this would require unrealistically high source activities with the additional requirement of a sufficiently long life time of the parent isotope. In addition self-absorption of low energy X-rays in the source is very strong. The X-rays can thus only originate from the surface layer, which leads to an upper bound to the maximum effective activity that can be reached by increasing the amount radioactive material. Finally, all these transitions are orders of magnitude narrower than crystal spectrometers resolution. For some applications the extreme narrowness of γ lines is of no use while limiting severely their intensity.

The most widely used X-ray energy standards, at present time, are made by exciting inner-shell transitions in atoms with either electrons or photons. In a number of cases their energies are given with precision close to 1 ppm [3], which does not necessarily mean that these standards can be used to such an accuracy. For X-rays originating from inner-shell transitions in multielectron systems, the center of gravity of the line cannot be attributed unambiguously to a physical transition. Shake-off processes (which create additional vacancies) and open outer shells lead to numerous satellite transitions very close in energy to the diagram line, which cannot be resolved and produce asymmetric line shapes. Moreover, the line shape of transitions in multielectronic systems depends also on the excitation mechanism used to create the inner-shell vacancies. For example the evolution of the K ($1s^{-1}$) Argon spectrum has been studied as function of excitation energy [4, 5], and dramatic qualitative changes were observed. The chemical environment of the atom also plays a strong role as can be seen from the comparison between solids, metallic vapors and theoretical X-ray absorption edges energies [3, 6, 7]. This problem also affects transition energies, particularly when they involve $M_{2,3}$ and $N_{2,3}$ shells.

An inherent problem with current standard X-ray lines is their natural width, which is typically more than 10 times larger than the resolution of the best X-ray spectrometers. Hence, fluorescence radiation is unsuitable to determine the response function of the apparatus.

As an alternative and more general approach to both γ-rays or natural X-rays, we thus propose to profit from recent developments in exotic-atoms research and in heavy-ions sources, and to use two- and three-body systems as photon emitters in the few keV range. In contrast to γ-rays, electronic, muonic and pionic atoms would provide a dense set of lines, that can be supplemented by an even denser set if one can use antiprotonic atoms with beam

intensities comparable to LEAR at CERN [8]. Our program consists first in doing relative energy measurements of transitions in one and two-electron ions, emitted by the plasma of Electron-Cyclotron Ion Sources (ECRIS), of circular transitions in fully stripped pionic atoms and of X-rays from solid fluorescence targets. Modern, commercial, permanent-magnet ECRIS are small and relatively economical to operate, and could be available in a large number of places to provide reference lines.

This relative energy scale will then be tied to a few, very bright lines, the energy of which will be measured absolutely, with either a double-flat crystal instrument, or a backscattering spectrometer as in [2] obtained from a Electron-Cyclotron Ion Trap (ECRIT), a device derived from the ECRIS, and optimized for increasing the trapping time of the ions [9], and thus the production of X-rays from highly-charged ions. Intense M1 radiation from the $1s2s\,^1S_0 \rightarrow 1s^2$ transition in helium-like Argon has been observed both in conventional ECRIS [10] and in the first run of the PSI ECRIT in 2002 [11]. The ions energy in such a device ranges from ≈ 0.5 eV to 6 eV depending on the injected RF power[12], giving rise to a Doppler broadening in the 5 to 18 ppm, i.e., 0.07 to 0.28 eV for Ar. This allows for measurements well below 1 ppm, and corresponds also to the expected accuracy (typically 1 meV) of theory for one electron ions in this range of Z. Different energy ranges and diffraction orders can be connected by exploiting the Yrast structure of the exotic-atom cascade, which leads to a strong population of circular states, thus favoring transitions with a change of the principal quantum number n by 1. Hence, successive transitions in the same atom connect different energy scales in ratios $\approx \left(1/(n+1)^2 - 1/n^2\right) / \left(1/n^2 - 1/(n-1)^2\right)$, which are roughly of the order of 0.5 for n around 5. For instance, πNe provides the photon energies of 2.7, 4.5 and 8.3 keV corresponding respectively to the $7 \rightarrow 6$, $6 \rightarrow 5$ and $5 \rightarrow 4$ transitions. With such a comparison method one can transfer energy standards between orders without problems due to changes in index of refraction.

The present program has several advantages. First, once a set of lines has been measured and compared to a given accuracy to quantum-electrodynamics (QED) calculations, other lines from the same source or from neighboring elements can be used as standards without the need of a direct measurement. Transition energies of these simple system can probably be calculated nowadays from first principle to better than 1 meV, benefiting from high-accuracy tests of QED in hydrogen [13]. This is even true for hadronic atoms, if one uses circular transitions, which are not affected by strong interaction. Only particle masses (very well known except for pion), the fine structure constant and the Rydberg constant are needed. Over time the precision of the calculation can be improved by systematically including contributions from higher-order Feynman diagrams. The quality of the calculation can be checked by comparing to the directly measured lines energies as well as to all the relatively measured ones which are directly connected to the direct measurements.

Second, the natural line widths of these transitions, while not as small as γ-ray line, are three orders of magnitude narrower than the ones of multielectronic systems.

Finally, the combination of the exotic-atom and electronic X-ray sources can provide a powerful tool for establishing high-quality X-ray energy standards. As an example, the $5g \rightarrow 4f$ transition in πC, the Lyman-α in hydrogenlike Cl and the Kα fluorescence line in singly ionized Ar, degenerate in energy by few eV, can be easily related by the method presented here.

In the present letter, we present the measurement, with a crystal spectrometer of the characteristic X-radiation from hydrogenlike pionic atoms. We use these transitions as energy standards for the energy determination of the transition energies in a complex electronic systems, the K$\alpha_{1,2}$ transitions in Ti. In this way we intend to show that the pionic transition is in good agreement with a reasonably well measured K$\alpha_{1,2}$ transition. Copper would have made a better case, but the present world average for the pion mass involve the Cu Kα doublet. This issue will however be solved when the final value for the pion to muon mass ratio is released [14].

For this experiment we used the cyclotron trap II [15] attached to the πE5 pion line at the Paul Scherrer Institute (PSI, Switzerland). In this device a 112 MeV/c pion beam is decelerated in a magnetic field using a suitable set of degrader foils. Such a set-up allows the use of dilute targets like gases. Typically 4×10^8 π^-/s are injected in the trap for 1 mA proton current. The target consists of a cylinder of 60 mm diameter and 26 cm length, with 50 μm-thick kapton walls. The pressure in the target was around 1 bar, leading to typically 1.7×10^6 π^-/s stops in the gas.

For light elements ($Z \leq 10$) the cascade that follows leads quickly to the formation of an hydrogenlike exotic atom in a circular state. All the electrons are ejected by Auger effect in the early stage of the cascade, in a process similar to internal conversion in nuclei, because of the large mass of the pion ($\approx 273 \times m_{e^-}$). Accordingly transition energies are 273 times larger than electronic ones between states of identical quantum numbers. Because the atoms are formed in a low-pressure gas the time it takes to recapture electrons from molecules in the gas is much longer than the pionic atom lifetime, hence, the exotic atoms stay in an hydrogenlike state for the rest of the atomic cascade. It has been shown that less than 2 % of the X-ray observed are affected by the presence of an extra electron [16]. When using solid targets the undefined status of the electron shell is the principal limitation in high-precision experiments using exotic-atom X-rays [17].

X-rays emitted by the exotic atoms at the center of the

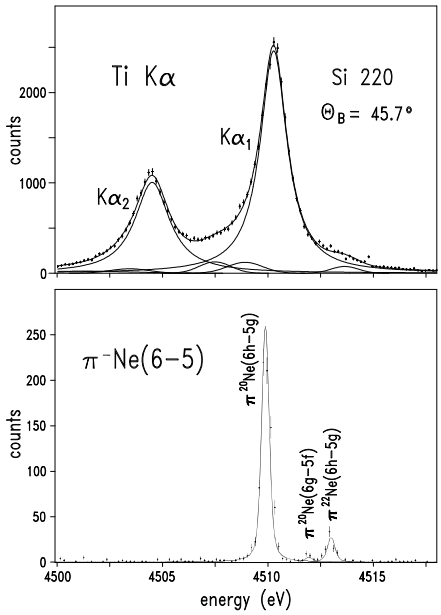

FIG. 1: Upper : Ti Ka doublet fitted by a sum of six Voigt functions. Lower : πNe 6 $\rightarrow$ 5 transitions showing fine-structure and isotopic effects. Comparison of the two spectra demonstrates the energy coincidence of the Ti Kα_1 and of the π^{20}Ne $6h \rightarrow 5g$ lines.

trap are analyzed using a Johann-type Bragg spectrometer that was developed for applications requiring very high luminosity and excellent resolution in noisy environments, in the 1.7 to 10 keV range. It was equipped with a spherically bent Si(220) crystal having a radius of curvature of 2.9854 m. A detailed description of the apparatus is given in [8, 16, 18] .

For this proof-of-principle measurement we relate the energy of the $6h \rightarrow 5g$ transition in pionic neon to energy of the K$\alpha_{1,2}$ transitions of Ti. The Ti Kα_1 differs only by a fraction of an eV from the strong $6h \rightarrow 5g$ transition in π^{20}Ne. Hence, the measurement could be performed by exchanging only the neon-filled target cell with a 30×20 mm^2 plate of metallic Ti, without any other change of the experimental set-up. The fluorescence X-rays were excited by means of an X-ray tube with a Cr-anode. The consecutively recorded πNe and Ti spectra are shown in Fig. 1.

The pionic transition energies are calculated from the world average pion mass $m_\pi = 139.57018 \pm 0.00035$ MeV as given by the particle data group [19] and fundamental constants [20] with the Klein-Gordon equation for a spherical nuclear charge distribution (to improve numerical stability although direct effect on energy is small). They include the Uehling potential for vacuum polarization to all order, the Källèn and Sabry as well as the Wichman and Kroll correction, and include nuclear recoil and relativistic recoil. The nuclear masses for ^{20}Ne and ^{22} Ne are deduced from atomic masses in [21] and [22] respectively, by subtracting the mass of the 10 missing electrons. These energies can be calculated with high precision since the strong interaction plays no role for such high-lying circular states. We obtain 4509.894 eV and 4512.948 eV for the $6h \rightarrow 5g$ transition in π^{22}Ne and π^{20}Ne, respectively. There is an uncertainty of 11 meV on these energy values which originates exclusively from the pion mass.

The natural width of the πNe $6h \rightarrow 5g$ transition is 12 meV. The fact that the calibration line is almost a δ function allows for precise determination of the spectrometer response function. The measured instrumental response function width of 440$\pm$20 meV is close to the theoretical limit of 330 meV as predicted from the Monte-Carlo simulations for the chosen geometry. The πNe line shape is fitted sufficiently well by using Gaussian line profiles.

To deduce the Ti Kα line shape we fitted a sum of six Voigt profiles, following [18] using the Gaussian response function obtained from the πNe spectrum. The peak positions, K$\alpha^0_{1,2}$, are obtained from the zeros of the derivative of the fitted function. The spectrometer dispersion, necessary to transform the position information of the detector to energy, is obtained in a self-consistent way from the πNe spectrum itself (Fig. 1). For the very small energy difference, for the case of πNe 6 $\rightarrow$ 5 and Ti Kα, the energy-dependent corrections originating from the imaging properties of crystal spectrometer (refraction index, crystal bending and size, rocking curve) are almost invariable and cancel out.

The results are displayed in Table I, together with presently known values. The experimental error in the energy of the Kα_1 line is practically given by the statistical uncertainty on the Kα_1 and π^{20}Ne $6h \rightarrow 5g$ lines. In the case of the Kα_2 line the error is dominated by the uncertainty on the dispersion value, which is due to the limited statistics of the π^{22}Ne $6h \rightarrow 5g$ transition. Both contributions could be drastically reduced by increasing the statistics of the measurement.

Narrow transitions from exotic atoms allow to characterize very precisely the response function of a curved crystal set-up. For the described experiment the accuracy of the extracted natural widths of the electronic systems (Table I) reaches the one obtained with ultimate resolution devices like double-flat crystal spectrometers [7, 23].

In the present letter we have demonstrated that narrow lines from hydrogenlike pionic atoms are able to serve as energy standards in the few keV range. By using this method the energy uncertainty is limited primarily by the knowledge of the charged pion mass, whenever transitions close in energy can be found. We have proven that an hydrogenic pionic line, the energy of which has been calculated from QED can be used to establish the energy of a previously well measured line with comparable accuracy. A research program is underway to improve on the precision of the pion mass to the order of about

TABLE I: Peak position energies and natural widths of the $K\alpha_{1,2}$ transitions in metallic Ti in comparison with previous measurements (in eV). For this work the errors on the energies from the experiment (first parenthesis) and from the calibration standard, i. e., the uncertainty of the pion mass (second parenthesis), are given separately.

Element	Line	Energy (this work)	Energy (Refs. [3])	Width (this work)	Width
Ti	$K\alpha_1^0$	4510.903(19)(11)	4510.869(49)	1.6(1)	1.5(3)[a]
	$K\alpha_2^0$	4504.942(40)(11)	4504.887(49)	2.1(1)	2.1(4)[a]

[a]Ref. [23]

1 ppm [14]. An alternative is to use muonic transitions as the muon mass is known to 0.05 ppm [19]. However, intensities achievable are about two orders of magnitude smaller than in the case of pionic atoms. Much higher intensity will be available from X-ray sources like the super-conducting Electron-Cyclotron Ion Trap (ECRIT) developed at the Paul Scherrer Institute, in which few-electron atoms up to hydrogenlike systems are produced [9]. Such electronic two-body systems will be used as calculable energy standards in the same way as exotic atoms.

In a separate experiment we have recently used the technique presented here to measure the energy of the Sc $K\alpha$ lines, which are known only from interpolation [24], with an accuracy improved by a factor of 12 [25].

By using X-ray lines, both from hydrogenlike exotic and electronic atoms a relative energy scale is established that can easily range up to 30 keV. Around 60 lines will be available if electronic, pionic and muonic atoms are used, hundred more if antiprotonic atoms are available. The energy of these lines will depend only on the fine structure constant α, and on the pion, muon and electron mass, and they can also be connected to low-energy γ-ray standards.

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