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A vacuum double-crystal spectrometer for reference-free X-ray spectroscopy of highly charged ions

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H I G H L I G H T S

- First use of a double crystal spectrometer to directly observe X-ray transitions in highly charged ions.
- Accuracies of a few parts per million could be reached that provides test of quantum electrodynamics.
- We have developed an *ab initio* simulation code that allows us to obtain accurate line profiles.
- It can reproduce experimental spectra with unprecedented accuracy.
- The details of the newly developed *ab initio* simulation code is presented.

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We have built a vacuum double crystal spectrometer, which coupled to an electron-cyclotron resonance ion source, allows absolute measurements of low-energy X-ray transitions in highly charged ions with accuracies of the order of a few parts per million. We describe in detail the instrument and its performance. Furthermore, we present a few spectra of $n=2 \rightarrow n=1$ transitions in Ar^{14+} , Ar^{15+} and Ar^{16+} . We have developed an *ab initio* simulation code that allows us to obtain accurate line profiles. It can reproduce experimental spectra with unprecedented accuracy. The quality of the profiles allows the direct determination of line width.

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1. Introduction

The measurement of X-ray transition energies of highly charged ions (HCIs) is one of the main methods to test bound-state quantum electrodynamics (BSQED) effects in strong fields. BSQED has been intensively tested experimentally, yet recent measurements of the proton size in muonic hydrogen, which disagree by 7σ from

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measurements in normal hydrogen (Antognini et al., 2013; Pohl et al., 2010), show that its short distance behavior may not be as well understood as was believed up to now. Highly charged ions can be a very good testing ground for improving our understanding of BSQED.

Highly charged ions can be created, e.g., using high-energy accelerators, Electron Beam Ion Traps (EBIT), or Electron-Cyclotron Resonance Ion sources (ECRIS). Transitions between excited states and the $n=1$ ground state in few-electron atoms or ions have been measured in a number of elements ranging from hydrogen to uranium. For medium atomic number elements, relevant to X-ray reflection Bragg spectrometry (transition energies in the 2–15 keV range), accuracies in the few tens of parts per million range have been obtained. Beam-foil spectroscopy has been employed to provide measurements in hydrogenlike and heliumlike ions like phosphorus, sulfur, argon (Briand et al., 1983a) (80 ppm), iron

(Briand et al., 1983b, 1984) (90 ppm) (Indelicato et al., 1986a) (25 ppm), germanium (Chantler et al., 2009) (14 ppm) and krypton (Indelicato et al., 1986b; Tavernier et al., 1985) (20 ppm). The main limitation to obtain high accuracy in Beam-foil spectrometry is due to the Doppler effect. Correction for the Doppler shift requires precise determination of the ions speed and angle of observation of the X rays emitted in flight. To get rid of this uncertainty, argon was also studied by X-ray spectrometry of recoil ions with an accuracy of 5 ppm (Beyer et al., 1985; Deslattes et al., 1984) relative to an X-ray standard. The uncertainty then was due to the presence of satellite lines associated with electron capture in the target gas. Another method to reduce the Doppler effect was to decelerate the beam after stripping it at high energy by capturing electrons from a gas cell, in the so-called “accel–decel” method. Hydrogenlike nickel was studied by this method (Beyer et al., 1991) with an accuracy of 13 ppm.

Devices like EBITs have been used to measure X-ray transitions in HCLs. Because the ions in EBITs have only thermal motion, no correction for Doppler shifts is required. Transition energies have been measured in hydrogenlike magnesium (Hölzer et al., 1998), hydrogenlike chlorine, hydrogenlike and heliumlike argon (Bruhns et al., 2007) and vanadium (Chantler et al., 2000). In Hölzer et al. (1998) and Bruhns et al. (2007), the hydrogenlike magnesium and chlorine Lyman α lines are measured without the use of X-ray reference lines, with an accuracy of 24 ppm and 10 ppm, respectively. The accuracy of the w line in He-like given to 2 ppm relative to the Lyman α was later improved to 1.5 ppm without the use of a reference line (Kubiček et al., 2012). In this work, the spectrometer used is made of a single Bragg crystal coupled to a CCD camera, which can be positioned very accurately with a laser beam reflected by the same crystal as the X-rays. More recently, a measurement of X-ray transition energies in heliumlike titanium was performed at an EBIT using a calibration based on neutral X-ray lines emitted from an electron fluorescence X-ray source. The value of the Lyman α was acquired with an accuracy of 15 ppm (Chantler et al., 2012).

Present day X-ray standards, as can be found in Deslattes et al. (2003), even though they are known with accuracies in the ppm range, are based on neutral elements with a K vacancy created by electron bombardment or photoionization. The shape and peak position of those lines depend on many factors like the excitation energy (see, e.g., Deslattes et al., 1982, 1983; Deutsch et al., 1996), the chemical composition and the surface contamination of the sample. Physical effects like shake-off, Auger and Coster–Kronig effects lead to multivacancies, which distort and broaden the line shape. Examples of the complex structure of $K\alpha$ lines in transition elements can be found, e.g., in Deutsch et al. (2004, 1995) and Hölzer et al. (1997). It is thus very difficult to use these standard lines with their quoted accuracy. Therefore, it was recently proposed to use either exotic atoms (Anagnostopoulos et al., 2003b) or combination of exotic atoms and relatively cold, highly charged ions, produced in an ECRIS (Anagnostopoulos et al., 2003a) to provide reliable, reproducible, narrow, X-ray standard lines.

The first observation of strong X-ray lines of highly charged argon ions (up to He-like) in an ECRIS was made in 2000 (Doussset et al., 2000). This experiment led to the description of the mechanisms for the production of the different lines in the plasma (Costa et al., 2001; Martins et al., 2001). Since then, several experiments have been performed at the Paul Scherrer Institute (PSI), using a spherically curved crystal spectrometer and an ECRIS, (Anagnostopoulos et al., 2005; Indelicato et al., 2007, 2006; Le Bigot et al., 2009; Trassinelli et al., 2007) leading to improved understanding of the ECRIS plasmas for sulfur, chlorine and argon (Martins et al., 2009; Santos et al., 2010, 2008, 2011). Such lines can be used, e.g., to characterize X-ray spectrometers response functions (Anagnostopoulos et al., 2005). Yet specific techniques

are required to measure their energy without the need for reference lines. The technique of Bruhns et al. (2007) and Kubiček et al. (2012), using a single flat crystal, is well adapted to the EBIT, which provides a very narrow ($\approx 100 \mu\text{m}$), but rather weak X-ray source. The ECRIS plasmas have been shown to be very intense sources of X-rays, but have diameters of a few cm. They are thus better adapted to spectrometers that can use an extended source. At low energies, cylindrically or spherically bent crystal spectrometers and double-crystal spectrometers (DCSs) can be used, but only the latter can provide high-accuracy, reference-free measurements.

Precision spectroscopy with double-crystal X-ray spectrometers has a long history. The first DCS was conceived and employed independently by Compton (1917), Bragg et al. (1921) and Wagner and Kulenkampff (1922) to measure absolute integrated reflections of crystals. Davis and Stempel (1921) used the DCS to study the width of the reflection curve. These experiments showed that the DCS was an instrument of high precision and high resolving power. They were followed by several others (see, e.g., Refs. Allison, 1932; Allison and Williams, 1930), and were instrumental in establishing the dynamical diffraction theory of Darwin (1914a,b) and Prins (1930). The capability of the dispersive mode to reach high-resolution was found by Davis and Purks (1927, 1928). The DCS was then used to obtain the K-line widths of some elements (Allison, 1933; Allison and Williams, 1930). A theoretical description of the instrument was provided by Schwarzschild (1928). A detailed technical description was given by Compton (1931) and Williams and Allison (1929). Williams (1932) introduced the vertical divergence correction in 1932, allowing for an improved accuracy for energy measurements. Bearden (1931a) provided an absolute measurement of copper and chromium K lines with the use of ruled gratings and calcite crystals in a DCS and deduced a value for the calcite lattice spacing, (Bearden, 1931b) leading the way to absolute X-ray wavelength measurements (Bearden, 1932). Detailed description of the instrument can be found in classic textbooks (Compton and Allison, 1935; James, 1948).

In a DCS, the first crystal, which is kept at a fixed angle, acts as a collimator, defining the direction and the energy of the incoming X-ray beam, which is analyzed by the second crystal. The first peak is obtained by scanning the second crystal angle when the two crystals are parallel (non-dispersive mode). Wagner and Kulenkampff (1922) were the first to show the absence of dispersion in the parallel mode. The peak shape depends only on the reflection profile of the crystals and provides the response function of the instrument. The second peak is obtained when both crystals deflect the beam in the same direction (dispersive mode). The peak shape is then a convolution of the line shape and of the instrument response function. The position of the first crystal is the same in both modes. The difference in angle settings of the second crystal between the non-dispersive and the dispersive modes is directly connected to the Bragg angle.

The DCS can be used in reflection (low-energy X-rays) in which case the energy that is being measured depends only on the Bragg angle, on the crystals lattice spacing d , on the crystal index of refraction and on the geometry (distance between the entrance and exit slits and height and width of the slits) of the instrument. In this case the reflecting planes are parallel to the surface of the crystal. In transmission (high-energy), there is no index of refraction correction, and the reflecting planes are perpendicular to the surface.

The DCS in both modes was used for many years to measure X-ray energies relative to standard lines, as the crystal lattice spacing was not known. This changed dramatically when high-purity Si and Ge high-purity single crystals became available, as they were needed for the fabrication of transistors. Interferometric

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