Observation of Lifetime-Limited X-Ray Linewidths in Cold Highly Charged Ions

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The temperature of Cs^{45+} ions interacting with a 7500-eV electron beam in an electron-beam ion trap was reduced to as low as 59.4 ± 9.9 eV. This decoupling of the electron and ion energy greatly advances the achievable precision of spectral measurements and allowed the first observation of the lifetime-limited linewidth of an x-ray transition in a highly charged ion, enabling the measurement of a radiative lifetime in the femtosecond regime. Using the uncertainty principle, we infer $1.65^{+0.24}_{-0.05}$ fs for the lifetime of the upper level of the $3d_{5/2} \rightarrow 2p_{3/2}$ electric-dipole transition in the neonlike Cs ion. [S0031-9007(96)02034-0]

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Radiative lifetimes less than a few femtoseconds are common among dipole-allowed resonance transitions in highly charged ions and are found, for example, among the K-shell transitions of heliumlike ions above argon ($Z = 18$) and among the L-shell transitions of neonlike ions above krypton ($Z = 36$). Dipole-allowed resonance transitions form the dominant lines in a given x-ray spectrum and play an important role in the density and temperature diagnostics of high-temperature plasmas, such as those found in inertial-confinement fusion, tokamaks, and astrophysical sources [1–4]. Because fast transition rates correspond to large absorption oscillator strengths, resonance transitions dominate the Planck mean opacity of a high-temperature plasma, and accurate knowledge of their radiative rates is important for plasma opacity and line transfer determinations [5–7]. Predictions of the radiative rates of these transitions have not been subject to experimental scrutiny, as existing techniques for measuring fast transition rates fail to yield results for lifetimes shorter than a few picoseconds [8,9]. This experimental void cannot be filled by atomic structure measurements alone, because unlike lifetime measurements transition-energy measurements do not test the long-range behavior of atomic wave functions.

In this Letter, we report on the first measurement of the femtosecond radiative lifetime of a dipole-allowed resonance transition in a highly charged ion. Our measurement is based on reducing the ion thermal motion and thus increasing the spectral precision to a point where it is possible to observe the energy uncertainty associated with the radiative lifetime. The energy uncertainty ΔE of an excited state with lifetime Δt decaying to the ground state results in a lifetime-limited, Lorentzian-shaped "natural" linewidth given by [10,11]

$$\Delta E = \hbar/\Delta t\,. \tag{1}$$

Direct observation of the natural linewidth has been precluded in standard spectroscopic sources, such as tokamaks, laser-produced plasmas, heavy-ion accelerators, or the Sun, by source-specific line broadening mechanisms. These include density or opacity broadening, blending with satellite transitions, and broadening by the thermal or directional ion motion associated with the energy required to produce and excite x-ray transitions in highly charged ions [12,13]. Using the trapped-ion approach, we have overcome these limitations. We produced and excited Cs^{45+} ions ($Z = 55$) in a low-density environment ($n_e < 5 \times 10^{12}$ cm^{-3}; $n_i < 10^9$ cm^{-3}) that have kinetic energies more than 2 orders of magnitude lower than the electron energy needed for excitation. This decoupling of the ion and electron motion sets the stage for measuring the line emission of highly charged ions with unprecedented precision, limited only by the uncertainty principle. It allows us to infer the radiative lifetime of the $2p$-$3d$ resonance transition from the observed width, and we find $1.65^{+0.24}_{-0.05}$ fs. This time is more than 3 orders of magnitude faster than that accessible with current measurement techniques. A variety of novel methods have been developed in recent years for the measurement of radiative transition probabilities of highly charged ions that are slower than those accessible with the original and highly successful beam-foil technique [14], e.g., the probing of excited levels by resonant recombination in storage rings [15], modulation of the electron-ion interaction energy [16], or capture of excited ions into ion traps [17]. The present method for the first time determines lifetimes that are considerably faster than the standard method.

Highly charged Cs^{45+} ions have been produced in the Livermore electron beam ion facility [18], which employs a monoenergetic electron beam to generate and excite ions in a cylindrical trap. The energy of the electron beam was set to 7500 eV. This energy is 400 eV below the ionization potential energy of Cs^{45+} and well above the 4740 eV required to excite characteristic x-ray transitions between levels with principal quantum numbers $n = 2$ and $n = 3$ so that no satellite transitions were excited that could blend with the lines of interest. Double excitation is insignificant because of collisional excitation rates below 10^3 s^{-1}. The ions are confined in the trap by an externally applied axial potential $V_{\rm ax}$ and the space charge potential of the electron beam. Interaction with the electron beam heats the ions until their kinetic energy is larger than the

potential well and they are able to leave the trap. Loss of Cs^{45+} ions, however, can be prevented by providing a constant source of low-charge ions, such as Ne^{10+} employed in our measurements. The low-charge ions are in thermal equilibrium with the high-Z ions, but are less deeply trapped and thus leave before the highly charged ions, carrying with them the heat deposited by the electron beam [19,20]. The temperature of the high-Z ions is thus limited by this so-called "evaporative," light-ion cooling.

The mechanisms of production and trapping suggest the ability to produce highly charged ions with much lower temperature than that of the electrons by reducing the potential well of the trap. To test this hypothesis, we have studied the two x-ray transitions from upper levels $(2p^5_{3/2}3d_{5/2})_{J=1}$ and $(2p^5_{1/2}3s_{1/2})_{J=1}$ that terminate in the $(2p^6)_{J=0}$ closed-shell neonlike ground state. The two lines are situated within 13 eV of each other at energies $E = 4740$ and 4753 eV, respectively, thus allowing observation in a single spectrum. Moreover, the lines emanate from upper states with disparate lifetimes predicted in single-configuration calculations to be 1.39 and 91 fs. These lifetimes correspond to natural linewidths of 0.47 and 0.0072 eV, respectively. The natural line broadening of the $2p$-$3d$ transition should become obvious provided the ion temperature is less than about 200 eV, i.e., for temperatures where the Doppler-broadened linewidth is less than the natural width. By contrast, the predicted natural width of the $2p$-$3s$ transition is an order of magnitude smaller than the instrumental broadening and cannot be observed. In our measurements, the $2p$-$3s$ line serves instead as an indicator of the temperature of the Cs^{45+} ions. The observed width was typically $\Delta E \geq 0.25$ eV with a Gaussian line shape indicative of thermal Doppler broadening, from which the ion temperature T_i was determined from the Doppler relation

$$T_i = m_i c^2 (\Delta E/E)^2 / 8 \ln 2 , \qquad (2)$$

where m_i is the ion mass and c is the speed of light.

Three spectra of the $2p$-$3d$ and $2p$-$3s$ transitions obtained under different operating conditions are shown in Fig. 1. The measurements were carried out with a high-resolution crystal spectrometer arranged in the von Hámos geometry [21,22]. The spectrometer employed a quartz crystal cut to the (203) reflection plane with a lattice spacing of 1.375 Å and intrinsic resolving power of 10^5. The crystal was cylindrically bent with a radius of curvature of 240 cm. The instrumental resolving power was 68 000 at the nominal Bragg angle $\theta = 71.8°$ of our measurements and corresponded to an energy resolution of 0.07 eV. The wavelength scale of the spectra was established from the splitting of the two lines. The splitting was determined in a separate measurement to be 13.08 ± 0.03 eV by utilizing the four $n = 2 \rightarrow n = 1$ K-shell transitions in heliumlike Ti^{20+} as reference lines. The wavelengths of heliumlike titanium lines are theoretically well known [23] and fall into exactly the same wavelength range as the Cs^{45+} lines.

Lowering the potential well successfully reduced the temperature of the ions interacting with the 7500-eV electron beam. As shown in Fig. 2(a), the temperature drops by a factor of 5, from well above 1000 to 250 eV, as the externally applied potential trapping the ions in

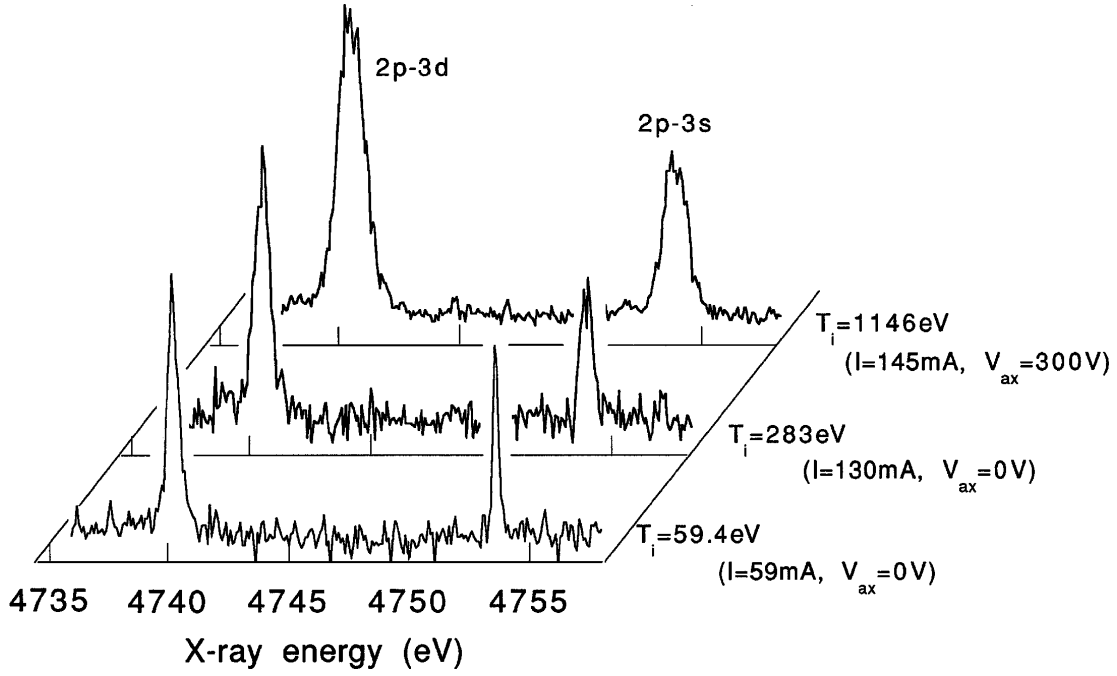

FIG. 1. Spectra showing the two x-ray transitions from upper levels $(2p^5_{3/2}3d_{5/2})_{J=1}$ and $(2p^5_{1/2}3s_{1/2})_{J=1}$ to the $(2p^6)_{J=0}$ closed-shell neonlike ground state in Cs^{45+}. The ion temperature T_i inferred from the width of the $2p$-$3s$ line drops from 1146 to 59.4 eV as the electron beam current I and axial trapping potential V_{ax} are reduced to the values indicated.

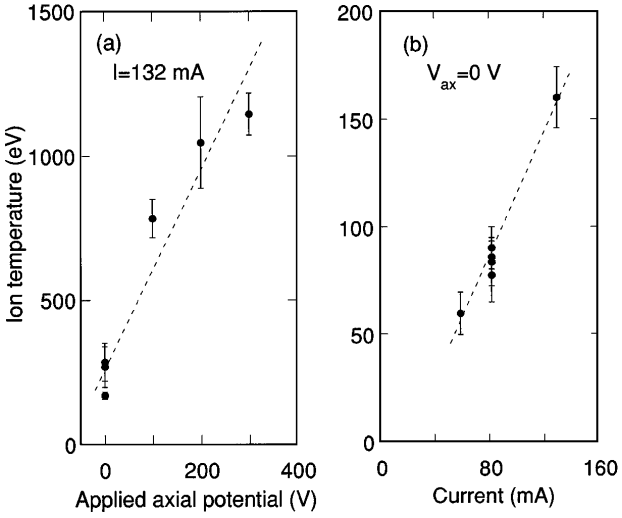

FIG. 2. Dependence of the temperature of Cs^{45+} ions interacting with a 7500-eV electron beam on (a) the externally applied potential and (b) the electron beam current. The dashed lines represent least-squares fits to the data and serve only to guide the eye.

the axial direction is reduced from 300 to 0 V. The temperature drops further [Fig. 2(b)] as the electron beam current, and thus the associated space charge potential, is reduced. Choosing a beam current of 59 mA and an axial trap of 0 V, the lowest ion temperature achieved was 59.4 ± 9.9 eV. For this set of operating parameters, the temperature of the ions was decoupled from the 7500-eV energy of the electrons by more than 2 orders of magnitude.

The reduction in the width of both lines as the temperature is lowered is seen in Fig. 1. Unlike for the $2p$-$3s$ line, the reduction in width nearly ceases for the $2p$-$3d$ line at the lowest temperatures, and the Lorentzian line shape becomes evident. A detailed view of the line shape of the $2p$-$3d$ transition recorded at an ion temperature of 110 eV is given in Fig. 3. Fitting the line with a Lorentzian trial function provides an excellent fit. The goodness of the fit is indicated by the reduced residuals shown in Fig. 3, which are defined as the difference between data and fit normalized to the square root of the fit value at each point. Residual contributions of thermal broadening on the line shape are accounted for by fitting the $2p$-$3d$ line with a Voigt profile [12], which represents a Lorentzian convolved with a Gaussian profile. For this, the temperature parameter for the Gaussian profile is taken from the width of the $2p$-$3s$ line. A value of 0.398 eV averaged over all observations was obtained for the natural width of the $2p$-$3d$ line. The statistical uncertainty is ± 0.012 eV. A systematic uncertainty is introduced by the possibility that natural broadening may also affect the width of the $2p$-$3s$ line. Because of the close vicinity of the upper level of each line, configuration interaction might decrease the lifetime of the $(2p_{1/2}^5 3s_{1/2})_{J=1}$ level while increasing that of the $(2p_{3/2}^5 3d_{5/2})_{J=1}$ level. Within the counting statistics of our measurement, no lifetime-limited line broadening of the $2p$-$3s$ line was detected. However, the minimum amount detectable introduces a systematic uncertainty as large as -0.049 eV in the width of the $2p$-$3d$ line, which combines with the statistical uncertainty and yields a natural line width of $0.398^{+0.012}_{-0.050}$ eV for the $2p$-$3d$ line. From this we infer a radiative lifetime of the excited level of 1.65 fs with an uncertainty of $+0.24$ and -0.05 fs. This is significantly larger than the 1.39 fs predicted by single-configuration calculations. By contrast, it is smaller than the value of 1.98 fs computed in a multiconfiguration Dirac-Fock calculation in the extended average level mode described by Grant *et al.* [24] that allows for configuration interaction by including all 36 excited levels with a vacancy in the $n = 2$ shell and an excited electron in the $n = 3$ shell. This disagreement with theory shows that measurements are needed even for very fast electric dipole transitions in order to guide atomic calculations, especially when configuration interactions play a dominating role.

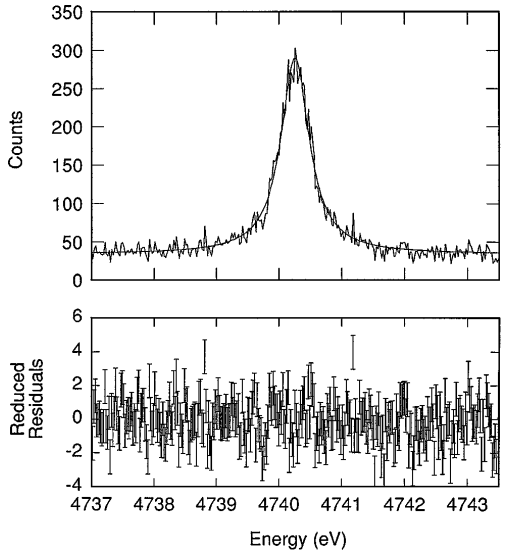

FIG. 3. Observed line shape of the $2p$-$3d$ transition at an ion temperature of 110 eV. The result of a least-squares fit of a Lorentzian trial function is superimposed for comparison. The reduced residuals of the fit are shown below.

The decoupling of the energy of the ions and electrons was achieved by reducing the potential well of the trap and allowing the hottest ions to leave. The decoupling is crucial for precision measurements, since in the absence of x-ray or gamma-ray lasers, probing by energetic electrons is the method of necessity for studying the structure of highly charged ions. Although the total estimated number of ions in the trap dropped by an order of magnitude as the potential well was reduced, about 10^4 ions remained producing about 50 counts/h in the $2p$-$3d$ line. Because fewer photons are needed to map out a narrower line, the peak line intensity, and thus the signal-to-noise ratio, remained nearly constant relative to the background level, as the potential well and the ion energy were reduced. Further decoupling appears

possible, although increasingly longer counting times will be needed if the same total number of counts is desired to fully exploit the higher level of precision. In the present measurements, relative x-ray linewidths as low as $\Delta E/E = 5 \times 10^{-5}$ have been achieved. This in principle enables determinations of x-ray transition energies on the order of one part per million or better. Cooling, i.e., the reduction in the translational motion, has been applied effectively to atoms and singly charged ions to increase the precision and range of fundamental atomic and nuclear physics measurements [25,26]. The present measurements show that a controlled reduction in the translational energy is now possible for highly charged ions as well.

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