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LETTER TO THE EDITOR

Lifetime of a K-shell vacancy in atomic carbon created by 1s → 2p photoexcitation of C^+

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Abstract

Lifetimes for K-shell vacancy states in atomic carbon have been determined by measurement of the natural linewidth of the 1s → 2p photoexcited states of C^+ ions. The K-shell vacancy states produced by photoionization of atomic carbon are identical to those produced by 1s → 2p photoexcitation of a C^+ ion: $1s2s^22p^2\ ^2D$, ^2P, and ^2S autoionizing states occur in both cases. These vacancy states stabilize by emission of an electron to produce C^{2+} ions. Measurements are reported for the lifetime of the $1s2s^22p^2\ ^2D$, ^2P and ^2S autoionizing states of C^+: 6.3 ± 0.9 fs, 11.2 ± 1.1 fs and 5.9 ± 1.3 fs respectively. Knowledge of such lifetimes is important for comparative studies of the lifetimes of K-shell vacancies in carbon-containing molecules, benchmarking theory, and interpreting satellite x-ray spectra from astrophysical sources such as x-ray binaries. Absolute cross sections were measured for both ground-state and metastable-state ions providing a stringent test of state-of-the-art theoretical calculations.

Carbon is ubiquitous in nature and is the building block of life. This atom in its various stages of ionization has relatively few electrons, and is thus amenable to theoretical study. Lifetimes

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of core-hole states (inner-shell vacancy states) by measurement of the natural linewidth of a resonant state are routinely determined by electron spectroscopy in gaseous molecules. The high density of a gas target and the availability of commercial high-resolution electron spectrometers allow this technique to be routinely applied at third-generation light sources. Despite the importance of the neutral carbon atom, there are no detailed measurements of inner-shell photoionization or photoexcitation processes, and the lifetimes of K-shell vacancy states have not been determined experimentally. The major difficulty is that free carbon atoms cannot readily be isolated, and various attempts to produce them generally result in neutral dimers and clusters. In the present investigation a different experimental approach has been followed using a photon-ion merged-beams apparatus at the Advanced Light Source (ALS) to study inner-shell photoexcitation leading to photoionization of the C^+ ion. The first measurements of the lifetimes of the $1s2s^22p^2\ ^2P$, ^2D and ^2S autoionizing states of C^+ are reported, along with absolute cross sections and precise resonance energies, for both ground-state and metastable-state C^+ ions.

An ion beam provides an exceptionally tenuous target. Compared to a gas target its density is limited by its own space charge to $\sim 10^5$ cm^{-3} in the present experiment. Only the exceptionally high brightness of the ALS photon beam and the extended length of the photon-ion merged beam path allowed sufficiently high-resolution measurements to be made in the present experiment.

Understanding the chemical abundances of the universe relies in large measure on spectroscopic observations of cosmic plasmas. Quantitative information about the cosmos is based on observation from ground- and satellite-based observatories. In particular, the recently launched x-ray satellites, *Chandra* and *XMM-Newton*, are currently providing a wealth of x-ray spectra of astronomical objects. There is a serious lack of adequate atomic data needed for the interpretation of these spectra [1].

Atomic spectroscopy in the soft x-ray region (5–45 Å) provides a valuable probe of the extreme environments in active galactic nuclei (AGNs), x-ray binary systems and cataclysmic variables. Limited high-quality theoretical or experimental cross section data are available for inner-shell photoexcited neutral or ionized species of relevance to astrophysical applications [1–6]. Such data are insufficient to meet the needs of the models which require many hundreds of cross sections for several different atomic species. Modellers hence resort to using crude models with all their inherent limitations [7].

The primary goal of the present research is to provide benchmark theoretical and experimental values for the energies and lifetimes of autoionizing states of the C^+ ion in the vicinity of the K-edge. Promotion of the inner-most K-shell (1s) electron to the L-shell (2p) by *photoexcitation of* C^+ produces autoionizing states via the process:

$$h\nu + C^+(1s^22s^22p\ ^2P^o) \rightarrow C^+(1s2s^22p^2\ ^2P, ^2D, ^2S). \quad (1)$$

Similarly, $C^+(1s2s^22p^2\ ^2P, ^2D, ^2S)$ Auger states may be created by direct *photoionization of neutral carbon*:

$$h\nu + C(1s^22s^22p^2\ ^3P) \rightarrow C^+(1s2s^22p^2\ ^2P, ^2D, ^2S) + e^-. \quad (2)$$

The same K-shell vacancy states of C^+ *are produced in both processes*; these states autoionize to produce $C^{2+}(1s^22s^2)$ and a free electron.

For an initial $1s^22s2p^2(^3P)\ ^4P$ metastable state of the C^+ ion, $1s2s2p^3\ ^4D^o$, $^4P^o$, $^4S^o$ Auger states can be formed in the photoexcitation/photoionization process. Metastable states of the C^+ ion that are present in the beam may be detected if such states are excited. In fact, the experiment was performed twice: once with an electron–bombardment ion source, for which no metastable ions were detected; a second time with an electron–cyclotron–resonance (ECR) ion source, for which 20% of the beam was found to be metastable.

Photoionization calculations in *LS*-coupling were performed on the C^+ ion using the *R*-matrix method [8] including radiation damping [9, 10]. An appropriate number of C^{2+} target states (135 levels) were included in the *R*-matrix plus pseudo-state close-coupling calculations. An $n = 4$ basis set of C^{2+} orbitals was used to represent the target wavefunctions [11]. Double-electron promotions from specific base configuration sets were used to describe the scattering wavefunction in the calculations. Auger linewidths and fluorescence yields for a K-shell vacancy produced in a free carbon atom may readily be determined theoretically [1, 7, 12, 13]; however, until now no measurements were available. Energies, linewidths and lifetimes for the $1s2s^22p^2\ ^2D$, ^2P, ^2S and $1s2s2p^3\ ^4D^o$, $^4P^o$, $^4S^o$ Auger states of C^+ from the present *R*-matrix work are presented in table 1.

K-shell photoabsorption experiments have been performed elsewhere on various ion species using the dual plasma technique which is useful for obtaining absorption spectra over a wide energy range [14, 15]. However their interpretation can be complicated due to ions being distributed over various charge states in both the ground and metastable states, and the presence of a plasma can affect energy levels, as can post-collision interactions (PCI). C^+ Auger states determined by this technique [16] are given in table 1.

The present experiment used a merged-beam technique which was first employed by Lyon and co-workers [17], and has been applied recently by several groups [18–21]. Experiments were performed at the 10.0.1.2 undulator beamline of the ALS synchrotron radiation facility located at the Lawrence Berkeley National Laboratory. A photon beam was merged with an ion beam from a small accelerator [21] to study the $1s \rightarrow 2p$ photoexcitation processes in C^+ ions for the energy region 286 eV–291 eV. Beams were merged over a path length of approximately 100 cm. The central region is maintained at an elevated potential to energy-label C^{2+} product ions created in this region, allowing measurement of product ions over a known path length [21]. The ion beam was charge-state analysed after the interaction region: the primary C^+ ion beam was collected in a Faraday cup, while the product C^{2+} ions were counted by a calibrated single-particle detector. The photon beam was chopped to subtract background mainly resulting from ions which have changed their charge state in collisions with the residual gas. Typical primary ion-beam currents were 40 nA, the photon flux with a resolving power $E/\Delta E = 2000$ was 4×10^{11} photons/s at 300 eV, and count rates were up to 60 Hz from the energy-labelled interaction region of 29.4 cm length. Absolute cross sections were determined by measuring the spatial overlap of the beams and the efficiency of the single-particle detector for C^{2+} product ions [21].

Experimental cross-section results are shown in figure 1, for $1s \rightarrow 2p$ photoexcitation of C^+ ions with an admixture of 80% ground-state ions and 20% metastable-state ions, for three values of spectral (instrumental) resolution. Data were fit to a Voigt profile, resulting in a Gaussian width (instrumental resolution) and a Lorentizan width (lifetime width) for each peak. The narrowing of the peak widths for the ^2D and ^2P Auger states with increasing instrumental resolution is evident. Lorentzian linewidths for the ^2D, ^2P and ^2S peaks were found to be 105 ± 15 meV, 59 ± 6 meV and 112 ± 25 meV, respectively. The metastable fraction was determined by comparing measured and theoretical cross sections; a metastable fraction of $(20 \pm 5)\%$ was obtained. Absolute cross sections were measured for each peak shown. Uncertainty in the metastable fraction is included in the uncertainty of the absolute cross sections: $\pm 30\%$ for ground-state ions and $\pm 40\%$ for metastable-state ions. Energies were determined by measurements of photoabsorption in CO, and compared with both the absolute energy for the $1s \rightarrow \pi^*$ resonance in CO measured by electron-energy-loss spectroscopy (EELS) [23] and the relative energies for photoexcitation [24]. The energies for the peaks were determined to have an uncertainty of ± 30 meV.

Table 1. C^+ Auger states for the $1s2s^22p^2$ and $1s2s2p^3$ configurations; experiment and theory, energies E (eV), linewidths Γ (meV) and lifetimes τ (fs) of K-shell vacancy states for the $1s \rightarrow 2p$ inner-shell photoexcitation processes in C^+.

State	E (eV)[a] (ALS)	E (eV)[b] (beam-foil)	E (eV)[c] (dual-plasma)	E (eV)[d] (R-matrix)	Γ (meV)[a] (ALS)	Γ (meV)[d] (R-matrix)	τ (fs)[a] (ALS)	τ (fs)[d] (R-matrix)
$1s2s^22p^2\ ^2D$	287.93 ± 0.03	288.68 ± 0.7	287.91 ± 0.2	287.96	105 ± 15	103	6.3 ± 0.9	6.4
$1s2s^22p^2\ ^2P$	288.40 ± 0.03	288.73 ± 0.7	288.59 ± 0.2	288.63	59 ± 6	62	11.2 ± 1.1	10.6
$1s2s^2\ 2p^2\ ^2S$	289.90 ± 0.03	290.98 ± 0.7	290.53 ± 0.2	289.97	112 ± 25	93	5.9 ± 1.3	7.1
$1s2s(^3S)2p^3\ ^4D^o$	287.25 ± 0.03	–	288.11 ± 0.2	287.29	110 ± 40	84	6.0 (+3.4, −1.6)	7.8
$1s2s(^3S)2p^3\ ^4S^o$	–	–	287.13 ± 0.2	287.73	–	25	–	26.3
$1s2s(^3S)2p^3\ ^4P^o$	289.42 ± 0.03	–	289.80 ± 0.2	289.46	55 ± 25	52	12.0 (+10, −4)	12.7

[a] Advanced Light Source, present work.
[b] Rødbro and co-workers [22].
[c] Jannitti and co-workers [16], assignment of states is based on HF + relativistic correction calculations using the Cowan code.
[d] R-matrix with radiation damping, present work.

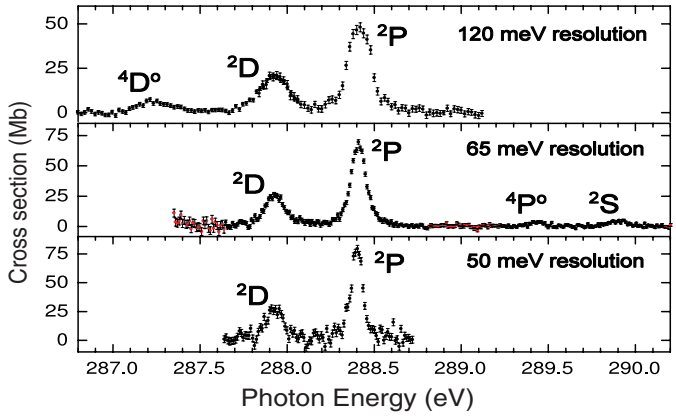

Figure 1. Cross sections for 1s → 2p photoexcitation of C^+ ions for an admixture of 80% ground-state and 20% metastable-state ions, measured at three values of spectral resolution.

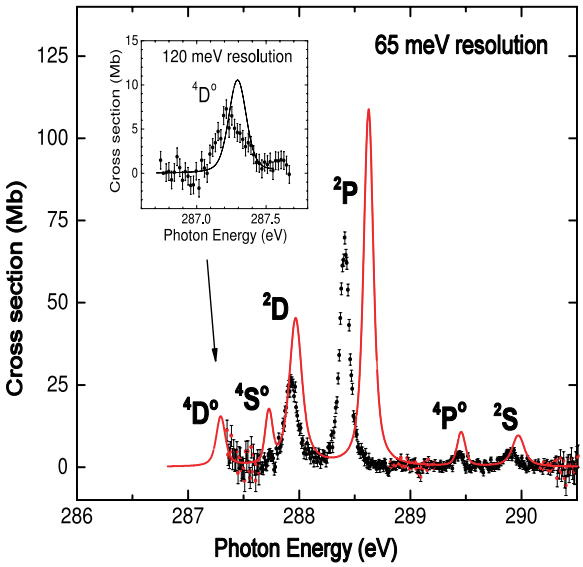

Figure 2. Experimental and theoretical cross sections for 1s → 2p photoexcitation of C^+ ions for an admixture of 80% ground-state and 20% metastable-state ions. Measurements are for a spectral resolution of 65 meV; theory is convoluted with a FWHM Gaussian of 65 meV for comparison with experiment. The $^4D^o$ resonance was measured with a spectral resolution of 120 meV; the inset shows the comparison with theory convoluted with a FWHM Gaussian of 120 meV.

Figure 2 shows experimental and theoretical cross sections for 1s → 2p photoexcitation of C^+ ions, for a C^+ admixture of 80% ground-state ions and 20% metastable-state ions. Theory has been convoluted with a 65 meV Gaussian for comparison with experiment (120 meV for the $^4D^o$). Theory and experiment are independently absolute (although theory was used to determine the metastable fraction of the beam). Cross sections, energies and linewidths all show reasonable agreement between theory and experiment. The $^4S^o$ peak is predicted theoretically but is likely obscured by the ^2D resonance. The two resonances have either similar experimental energies and cannot be individually resolved, or the $^4S^o$ resonance is

Table 2. Measured linewidths and lifetimes of a K-shell vacancy in carbon-containing molecules and in atomic carbon.

	Molecule				Carbon : $1s2s^22p^2$ vacancy states		
	CO	CF_4	CH_4	CO_2	^2D	^2P	^2S
Γ (meV)	95 ± 5[a]	77 ± 6[a]	95 ± 2[b]	99 ± 2[c]	105 ± 15[d]	59 ± 6[d]	112 ± 25[d]
τ (fs)	6.9 ± 0.4	8.5 ± 0.6	6.9 ± 0.1	6.6 ± 0.1	6.3 ± 0.9	11.2 ± 1.1	5.9 ± 1.3

[a] Carroll and co-workers [29].
[b] Carroll and co-workers [30].
[c] Carroll and co-workers [31].
[d] Present Advanced Light Source experimental results on atomic carbon.

weaker than predicted. Any effect of the $^4S^o$ resonance on the ^2D resonance values is included in the uncertainties presented.

Experimental and theoretical values for energies and linewidths of all the peaks found in the spectra shown in figures 1 and 2 are presented in table 1. Two significant findings emerge from this investigation. Firstly, overall agreement of theoretical and experimental energies is evident, although theoretical values occur at slightly higher energies. Secondly, values for Auger linewidths show excellent agreement between theory and experiment. In particular the linewidth of the $1s2s^22p^2\ ^2P$ autoionizing state of C^+ at 288.40 eV is 59 ± 6 meV, in accord with the theoretical value of 62 meV determined from the *R*-matrix method (56 meV without radiation damping), and with previous theoretical estimates: 52 meV [12, 25], 56 meV [26, 27] and 53 meV [28]. The $1s2s^22p^2\ ^2D$ and $1s2s^22p^2\ ^2S$ autoionizing resonances of C^+ found experimentally at 287.93 eV and 289.90 eV have respective experimental linewidths of 105 ± 15 meV and 112 ± 25 meV in agreement with the corresponding theory estimates of 103 meV and 93 meV. The difference between theory and experiment for the resonance energy positions we attribute to the limitations of our present $n = 4$ basis set representing electron correlation effects. The position of the ^2P resonance is much more sensitive to electron correlation effects than the other resonances, so greater discrepancy with experiment can be expected for this symmetry. It is highly probable that an enhanced scattering basis would be required to account for the electron correlation deficiencies in the present calculations. The linewidths converted to lifetimes τ using the Heisenberg uncertainty principle are given in table 1.

Theoretical and experimental linewidths (Γ) for a K-shell vacancy in a wide variety of carbon-containing molecules are available in the literature [26, 29–31], from which lifetimes (τ) can be obtained via the uncertainty principle. A notable absence is that of an experimental value for the free carbon atom. It can be seen from our experimental results in table 2 that the lifetime τ (fs) for the $1s2s^22p^2\ ^2P$ vacancy state in atomic carbon is significantly larger ($\sim$ a factor of two) than for a corresponding K-shell vacancy in carbon-bearing molecules. This increase in lifetime can be attributed to there being fewer L-shell electrons to fill the K-shell vacancy in atomic carbon compared to the case of a carbon-containing molecule.

In summary, lifetimes of the K-shell vacancy states in atomic carbon; $1s2s^22p^2\ ^2P$, ^2D and ^2S have been measured by photoion spectroscopy for $1s \rightarrow 2p$ photoexcitation of C^+ ions. This is the first time that lifetimes of an inner-shell vacancy in photoexcited C^+ ions have been measured, and hence lifetimes of a K-shell vacancy determined in atomic carbon. Suitable agreement is found between theoretical estimates determined using the *R*-matrix method and the ALS experimental work. Finally, it would be interesting to see what effect these resonance structures have on the relative abundances obtained from steady-state ionization when soft

x-ray radiation illuminates low-density cold matter, where photons are efficient at K-shell ionizing light elements such as carbon in its different ionized stages [32, 33].

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