Identification and oscillator-strength measurement of the 109.1-nm transition in neutral Cs

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Received February 10, 1986; accepted April 16, 1986

The 109.1-nm transition in neutral Cs is the prototype of a class of transitions that originate from doubly excited quasi-metastable quartet levels. In this Letter we describe experiments that use tunable VUV radiation to determine the identity of this transition, to measure its oscillator strength, and to estimate its hyperfine splitting.

In recent work[1] it was postulated that particular core-excited quartet levels of alkali and alkalilike ions should have radiative rates that are comparable with their autoionizing rates. This occurs since, by LS selection rules, these levels may couple only to doublet levels, which are themselves prohibited from autoionizing by angular momentum and parity considerations. Levels of this type have been termed quasi-metastable and are specified by the condition that $|J - L| = 3/2$ and that parity and angular momentum be both even or odd; they can occur even in heavy atoms.

The 109.1-nm transition in Cs (Fig. 1) is a prototype of the class of transitions that originate from levels of this type. The upper level of this transition is $Cs(5p^5 5d6s)^4P^{\circ}_{5/2}$ and is expected to radiate to the valence fine-structure levels $Cs(5p^6 5d)^2P_{5/2}$ and $Cs(5p^6 5d)^2P_{3/2}$. Holmgren *et al.*[2] recently observed intense emission at 109.1 nm using a pulsed hollow-cathode discharge and, on the basis of the fine-structure splitting and the intensity ratio of its fine-structure components, ascribed the radiation as originating from this transition. Somewhat earlier, Aleksakhin *et al.*[3] associated radiation at 108.5 nm with the same transition.

In this Letter we describe an experiment to confirm the identity of this transition through accurate measurements of the fine-structure splitting and from estimates of the oscillator strength and hyperfine splitting. A pulsed hollow cathode was used to populate the lower level of this transition, and tunable VUV radiation, generated by four-wave mixing, was used to make absorption measurements at near-Doppler-limited resolution.

Experiment

Tunable radiation near 109.1 nm was generated using two-photon resonant four-wave sum-frequency mixing (4WSFM) using a process in Zn similar to that of Jamroz *et al.*[4,5] The particular scheme, shown in Fig. 2(a), was chosen based on our desire to minimize the linewidth of the VUV source and to use 532-nm pumped dye lasers. Such dye lasers are able to generate only wavelengths longer than 540 nm; thus, in order to generate radiation near 109 nm by using only five dye photons, it was necessary to find a two-photon resonant level between 73 228 and 74 074 cm^{-1}. Referring to Fig. 2(a), the desired level was found by observing degenerate 4WSFM (tripling) through successive levels in the Zn $4sns\ ^1S_0$ Rydberg series. Based on a quantum-defect extrapolation from the (tabulated)[6] lower members of the series, we identify the level at 73 747.7 cm^{-1} as Zn $4s10s\ ^1S_0$. No other two-photon levels were observed within the necessary energy range.

A single Q-switched, frequency-doubled Nd:YAG laser was used to pump two dye-laser systems. The first was operated with an intracavity étalon to narrow the linewidth to 0.05 cm^{-1}, tuned to 542.2 nm, and frequency doubled to provide the two-photon resonant wave. The second dye laser was tuned around a small region near 558 nm and had a linewidth of 0.3 cm^{-1}. The two outputs were combined by using a dichroic mirror and focused into the Zn cell at a power density of about 2×10^{10} W cm^{-2}. The Zn cell consisted of a 2.5-cm-diameter stainless-steel tube (horizontal) with a 6-cm-long hot zone produced by an external (vertical) sodium heat pipe.[7] Typically, the cell was operated at a Zn density of about 10^{17} cm^{-3} along with 50 Torr of He to reduce the rate of Zn diffusion out of the hot zone. We estimate our generation efficiency as 10^{-8}. The source linewidth was measured (using the Xe 108.4-nm transition) at 0.7 cm^{-1}, resulting in a spectroscopic resolving power of 1.3×10^5.

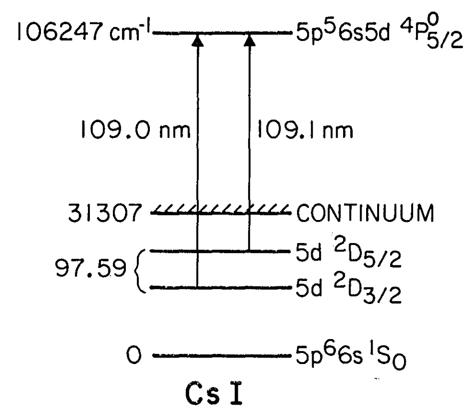

Fig. 1. Selected Cs I energy levels showing the transitions observed.

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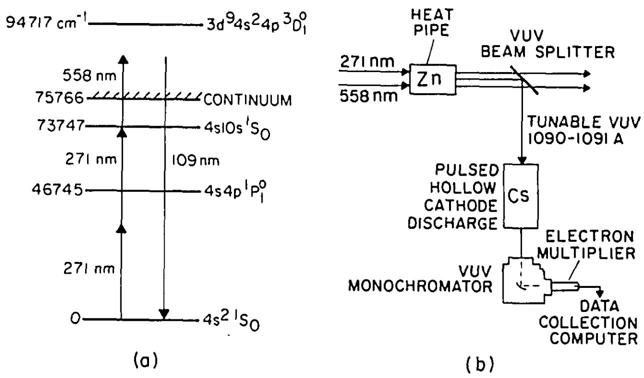

Fig. 2. (a) Energy levels for resonant VUV generation in Zn vapor; (b) simplified diagram of the experimental apparatus.

Because of a small residual sensitivity in our detector at 271 nm, it was necessary to filter 271 nm from the beam leaving the Zn cell. An uncoated quartz plate was put in the beam oriented at Brewster's angle for the 271-nm radiation, which thus passed through it. The orthogonally polarized 109.1-nm radiation, however, experienced a 30% reflection,[8] and the reflected beam was directed through the Cs absorption cell and into a 0.2-m VUV monochromator, which provided further discrimination. A schematic of the optical path is shown in Fig. 2(b); LiF windows were used to isolate the beam splitter, spectrometer, and detector volumes. Approximately 10^3 photons of 109.1-nm radiation per shot were incident upon the EMI D233 electron multiplier.

The Cs pulsed hollow cathode has been described in Ref. 9. The active region consists of a stainless-steel cathode 2 cm in diameter and 30 cm long operated at peak currents from 0.6 to 300 A with pulse lengths ranging from 20 to 1 μsec, respectively. For these measurements both the Cs vapor pressure and the He buffer-gas pressure were set at 1 Torr.

Results and Discussion

Figure 3 shows VUV absorption scans of the pulsed hollow-cathode discharge, both with the discharge on and with the discharge off. Absorption on both fine-structure components of the $5p^65d\ ^2D$–$5p^55d6s\ ^4P^\circ_{5/2}$ transition is clearly seen. The fine-structure splitting measured by us is 97.3 ± 0.3 cm^{-1} and is in agreement with the published value of 97.59 cm^{-1}.

Absolute calibration of the 109.1-nm wavelength was accomplished by comparing the wavelengths of the dye lasers used to generate the VUV radiation with emission lines of Kr and Hg. The result for the $^2D_{5/2}$ absorption is 109.111 ± 0.0035 nm. The uncertainty is due to errors introduced by the drive in the spectrometer used for the intercomparison.

The oscillator strength of each of the fine-structure components of the 109.1-nm transition was measured by using the curve-of-growth method.[10] For these measurements the hollow-cathode discharge current was reduced until each of the components was optically thin. In this regime the frequency-integrated VUV absorption depends on NfL, i.e., on the product of population, oscillator strength, and distance, and is insensitive to linewidths or hyperfine structures.

The density–length product NL of the $(6p^65d)^2D$ fine-structure levels was measured with a laser tuned between the $(5p^65d)^2D_{5/2}$ and $(5p^68f)^2F^\circ$ transition at 662.9 nm and the $(5p^65d)^2D_{3/2}$ and $(5p^68f)^2F^\circ_{5/2}$ transition at 658.6 nm. These transitions have oscillator strengths of 2.02×10^{-2} and 2.11×10^{-2}, respectively,[11] which are thought to be accurate to 10%. Under our operating conditions, i.e., an electron density of about 10^{14} electrons/cm^3, the chosen valence transitions are sufficiently Stark broadened that they do not become opaque at line center. This procedure resulted in measurement of NL of the valence-level atoms to an accuracy of about 20%.

The results of the VUV oscillator strength measurements are summarized in Table 1 and are compared with values that are calculated by using the RCN/RGG atomic-physics code.[12]

Figure 4 shows the measured equivalent width of the 109.1-nm absorption as a function of population in the lower state. The solid curve, denoted by B, is the calculated equivalent width based on the measured oscillator strength including both Doppler and Stark broadening. This curve does not fit the experimental data. To fit the data at the largest populations, a Stark width about 10 times larger than that which we calculate must be assumed, and the resulting curve deviates from intermediate experimental points.

We believe that this discrepancy results from neglecting the hyperfine splitting of the $(5p^55d6s)^4P^\circ_{5/2}$ level. The nuclear spin of $I = 7/2$ causes the transition to be split into nine components. (The hyperfine structure of the lower level of the 109.1-nm transition can be neglected.[13]) The relative frequency spacing is given by the Landé rule with a scale factor A. Curve A of Fig. 4 shows the calculated equivalent width assum-

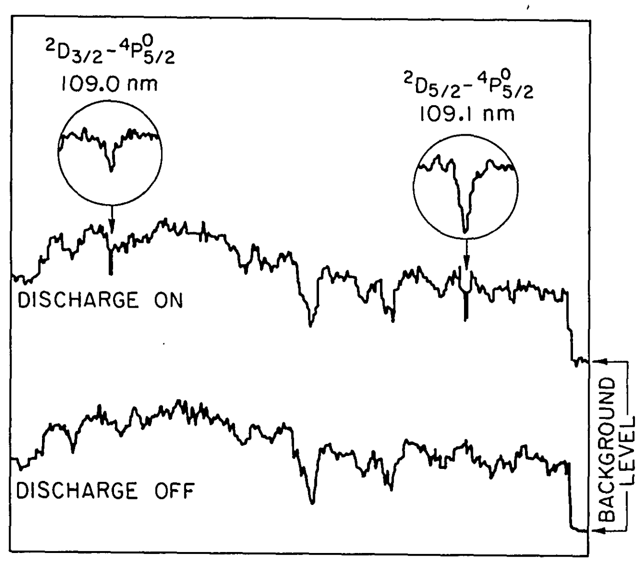

Fig. 3. VUV absorption scan showing the two fine-structure absorptions from Cs $(5d)^2D$. The data were taken with a lower-level population of 3×10^{13} cm^{-3}; the insets show the absorptions at full instrumental resolution.

Table 1. Summary of Results from Absorption Measurements on the 109-nm Transitions

Transition	λ (nm)	$f_{exp.}$	$f_{calc.}$[a]
$Cs(5p^6 5d)^2D_{5/2}$–$(5p^5 5d6s)^4P^{\circ}_{5/2}$	109.111	$(7.2 \pm 4) \times 10^{-3}$	6.95×10^{-3}
$Cs(5p^6 5d)^2D_{3/2}$–$(5p^5 5d6s)^4P^{\circ}_{5/2}$	108.998	$(6.5 \pm 2) \times 10^{-4}$	4.8×10^{-4}
Fine-structure splitting: 97.3 ± 0.3 cm^{-1}			
Hyperfine splitting constant: $A = 0.025$ cm^{-1}			

[a] Calculated using the RCN/RCG atomic-physics code.[12]

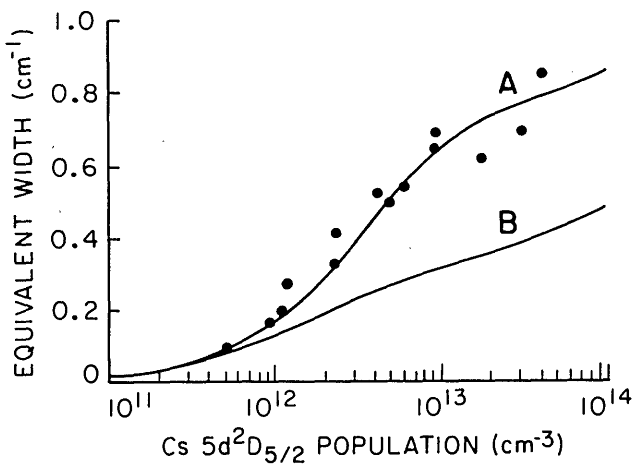

Fig. 4. Equivalent width versus lower-level population for the 109.1-nm absorption, assuming a 30-column length. The points are experimental data. The solid lines are calculated values from computer-modeled curves of growth assuming Doppler, Stark, and collision broadenings of 0.14, 1.3 $\times$ 10^{-3}, and 1.5 $\times$ 10^{-3} cm^{-1}, respectively. Curve A also includes hyperfine splitting with a constant of A = 0.025 cm^{-1}.

ing a scale factor of A = 0.025 cm^{-1} and Stark widths determined by the measured electron density.

As a theoretical estimate of the splitting,[14] we used RCN[12] to compute $|\psi_{6s}(0)|^2$ for Cs $5p^6 6s$ and Cs $5p^5 5d6s$. (The $5d$ electron and the $5p^5$ subshell contribute little to this hyperfine interaction.) If we use the known splitting constant for the Cs ground state of A_{ground} = 0.076 cm^{-1} and account for an angular-momentum factor of 1/5, this predicts $A_{quasi\text{-}metastable}$ = 0.021 cm^{-1}.

The hyperfine splitting reduces the gain cross section for lasers operating on the 109.1-nm transition. If hyperfine structure is neglected, the calculated cross section is 4.0 $\times$ 10^{-14} cm^2, based on our measured oscillator strength and assuming a Doppler width of 0.15 cm^{-1}. If, however, we assume that the upper level's population is distributed among its hyperfine components according to their degeneracies and sum the contributions of the various Doppler-broadened components, the peak value of the cross section is reduced to 1.2 $\times$ 10^{-14} cm^2. If the hyperfine splitting were much larger than the Doppler width, then the gain cross section on the largest component would be 1.08 $\times$ 10^{-14} cm^{-1}.

In summary, the 109.1-nm transition in Cs is an example of a transition that originates from slowly autoionizing, quasi-metastable levels of alkalilike atoms and ions. In this work we have confirmed the identity of this transition, measured its oscillator strength and fine-structure splitting, and estimated its hyperfine splitting.

This research was supported jointly by the U.S. Air Force Office of Scientific Research and the U.S. Army Research Office.

The authors acknowledge helpful discussions with A. J. Mendelsohn and J. K. Spong. D. P. Dimiduk acknowledges support from the U.S. Air Force Institute of Technology.

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References

1. S. E. Harris, D. J. Walker, R. G. Caro, A. J. Mendelsohn, and R. D. Cowan, Opt. Lett. **9,** 168 (1984).
2. D. E. Holmgren, D. J. Walker, and S. E. Harris, in *Laser Techniques in the Extreme Ultraviolet,* S. E. Harris and T. B. Lucatorto, eds. (American Institute of Physics, New York, 1984), p. 496.
3. I. S. Aleksakhin, G. G. Bogachev, I. P. Zapesochnyl, and S. Yu. Urgin, Sov. Phys. JETP **53,** 1140 (1981).
4. W. Jamroz, P. E. LaRocque, and B. P. Stoicheff, Opt. Lett. **7,** 617 (1982).
5. Initial 4WSFM experiments in Kr showed that the negative dispersion limit precludes generation of wavelengths shorter than 109.45 nm: K.D. Pedrotti, "Atomic spectroscopy for soft x-ray lasers," Ph.D. dissertation (Stanford University, Stanford, Calif. 94305).
6. C. E. Moore, *Atomic Energy Levels, Vol. II* (National Bureau of Standards, Washington, D.C., 1949).
7. H. Scheingraber and C. R. Vidal, Rev. Sci. Instrum. **52,** (1981).
8. R. W. Falcone and J. Bokor, Opt. Lett. **8,** 21 (1983).
9. R. W. Falcone and K. D. Pedrotti, Opt. Lett. **7,** 74 (1982); R. W. Falcone, D. E. Holmgren, and K. D. Pedrotti, AIP Conf. Proc. **90,** 287 (1982).
10. S. S. Penner and R. W. Kavanaugh, J. Opt. Soc. Am. **43,** 385 (1953).
11. M. Fabry and J. R. Cussenot, Can. J. Phys. **54,** 836 (1976).
12. R. D. Cowan, *The Theory of Atomic Structure and Spectra* (U. California Press, Berkeley, Calif., 1981); Mail Stop B212, Los Alamos National Laboratory, Los Alamos, New Mexico 87545 (personal communication).
13. K. Fredriksson, H. Lundberg, and S. Svanberg, Phys. Rev. A **21,** 241 (1980).
14. I. I. Sobel'man, *An Introduction to the Theory of Atomic Spectra* (Pergamon, Oxford, 1972), pp. 205–216.