High-resolution extreme-ultraviolet spectroscopy of potassium using anti-Stokes radiation

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We report the use of a new extreme-ultraviolet radiation source based on spontaneous anti-Stokes scattering for high-resolution absorption spectroscopy of transitions originating from the $3p^6$ shell of potassium. The region from 546.6 to 536.8 Å is scanned at a resolution of about 1.2 cm^{-1}. Within this region, four previously unreported lines are observed.

High-resolution spectroscopy in the extreme-ultraviolet (XUV) spectral region is limited by the power per spectral bandwidth of traditional radiation sources and by the need to filter the radiation with low-efficiency, highly dispersive monochromators. In this Letter we describe the application of a new type of XUV radiation source to this problem.

The radiation[1–4] source is based on spontaneous anti-Stokes scattering of incident laser photons from excited metastable atoms in an electrical discharge. The frequency of the scattered photons is equal to the sum of the metastable storage frequency and the frequency of the incident laser photon. The linewidth of the scattered radiation is the convolution of the laser linewidth and the Doppler width of the emitting species. The radiation is polarized, has the same time duration as the incident laser, and can be tuned by tuning the incident laser frequency. If the intensity of the tunable anti-Stokes radiation is sufficiently great that it may be distinguished from the background radiation of the plasma, then no monochromator need be used to obtain an absorption spectrum.

Figure 1 is a schematic of the apparatus used to take absorption spectra of K. The He $1s2s\,^1S$ level at 166277 cm^{-1} is used as the storage level for the anti-Stokes-radiation source. Thus common dye lasers in the 6000–5000-Å spectral region enable one to examine features in the region from 182975 to 186769 cm^{-1}, as shown in Table 1. The XUV anti-Stokes radiation passes through a K cell and a thin aluminum filter and is incident upon an electron multiplier.

The He $1s2s\,^1S$ storage population is created in a 60-cm-long hollow-cathode glow discharge. Typical operating parameters are 2-Torr pressure, 190 V, and 30 mA, which, based on previous measurements,[5] implies a population of about 5×10^{11} atoms/cm^3. A Quanta-Ray dye laser provides a maximum input pulse energy of about 50 mJ at a 10-pulse/sec repetition rate.

The K cell consists of a 1.2-cm-diameter stainless steel tube and wick with an active length of 5 cm. Typical operating K density is 10^{15} atoms/cm^3 (300°C). The He background pressure from the discharge region prevents the diffusion of K atoms both into the discharge and to the aluminum filter that separates the He pressure from the 10^{-6}-Torr operating pressure of the electron multiplier. Experiments were performed with K having a maximum of 2% impurities and 0.05% impurities, with the same results.

The output of the electron multiplier is processed with a fast preamplifier and a 10-nsec-wide gated integrator coincident with the laser pulse. A small computer digitizes and records the resultant voltage along with the intensity of the input dye laser. The computer also tunes the laser in 0.15-cm^{-1} steps, recording 10 values at each setting.

The spontaneous anti-Stokes-scattering cross section[6] (see Fig. 2) for visible photons at 6000 Å is 4×10^{-23} cm^2; thus for an incident pulse energy of 50 mJ, about 1.8×10^8 XUV photons are produced in the 60-cm cell length. The solid angle of the detector reduces the effective flux to about 800 photons/pulse or 8000 photons/sec. Thus the ratio of the number of effective XUV photons to the number of visible photons originally present is about 10^{-14}. It is therefore essential to provide excellent discrimination against the visible

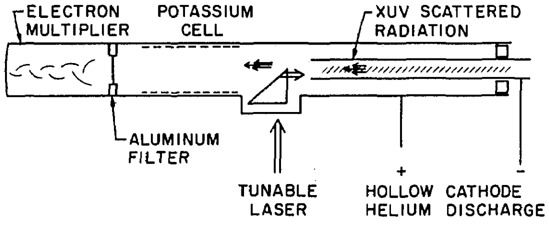

Fig. 1. Schematic of apparatus used for absorption spectroscopy of K.

Table 1. Spectral Regions Studied

Laser Dye	Range of Laser Wavelength (Å)	Range of XUV Frequency (cm^{-1})
Kiton Red 620	5990–5740	182975–183695
Rhodamine 590	5760–5470	183635–184549
Coumarin 500	5220–4880	185435–186769

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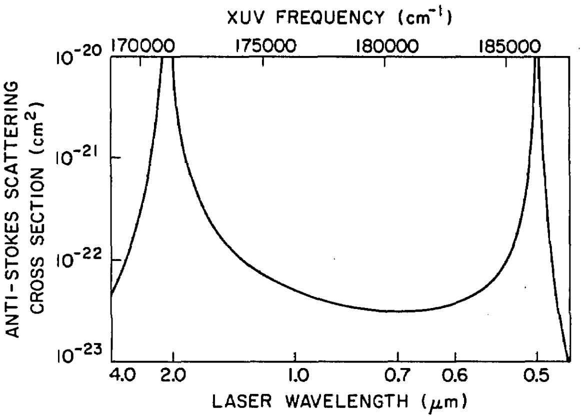

Fig. 2. Plot of anti-Stokes scattering cross section versus tunable-laser wavelength and scattered XUV frequency.

photons. This is accomplished by arranging the visible and XUV radiation to travel in opposite directions, by the $\sim 10^9$ insertion loss of the 1500-Å-thick aluminum filter and by the high work function of the electron multiplier. Since the XUV transmission of the filter is 10%, the expected flux at the detector is 800 photons/sec. Our measured signals correspond to a flux of $\sim$200 photons/sec, assuming a 10% detector quantum efficiency.

A typical absorption scan of K is shown in Fig. 3. The level of the background emission from the plasma with the tunable laser blocked is shown to the left of the dashed line. In most cases the absorption of the observed K features brought the transmission back down to this background level, indicating that any other laser-induced emission from the plasma was not of significant amplitude. (The amplitude of the Stokes signal is about 15 times weaker than that of the anti-Stokes signal.) The scans are computer-generated plots in which each point represents the sum of integrated signals from 5 laser settings (a total of 50 pulses). The XUV signal at each frequency setting was normalized to the relative dye-laser intensity, which has the effect of increasing the statistical fluctuation in regions of lower laser intensity.

Table 2 summarizes the energies and linewidths of the observed K absorption features. The broader features were observed earlier by Mansfield,[7] Mansfield and Ottley,[8] and Kavei *et al.*[9] The narrower features have not been reported previously. The absorption linewidths were a function of the K vapor pressure. The widths of Table 2 are the minimum full widths at half maximum obtained by reducing the cell pressure to a value at which negligible change in width is seen with further reduction. The narrowest observed absorption feature (at 185806 cm^{-1}) has a width of 1.9 cm^{-1}, which is not much larger than the 1.2-cm^{-1} theoretical linewidth of the XUV radiation, i.e., the convolution of the 0.3-cm^{-1} laser linewidth and the $\sim$1-cm^{-1} Doppler width of the emitting He atoms.

The narrow absorption lines are probably the result of transitions from the $3p^6 4s$ ground level to levels that, in the approximation of LS coupling, are forbidden by selection rules to autoionize, such as odd-parity doublet series levels with even angular momentum, and levels in the quartet series. The narrow features at 184076 and 184321 cm^{-1} may correspond to transitions from ground to the $3p^5 4d4s\ {}^4D_{1/2}$ and to the $3p^5 4d4s\ {}^4D_{3/2}$ levels, respectively. If we assume that the linewidth of the narrow features is about equal to that of the anti-Stokes source, than a 50% absorption corresponds to an oscillator strength of $f = 3 \times 10^{-4}$.

There are several engineering improvements that could increase the intensity and utility of this radiation source. The discharge could be pulsed to increase the storage-state density; the tunable laser could be multipassed through the discharge, increasing XUV intensity linearly with distance; grazing incidence or a collecting mirror could increase the usable solid angle.

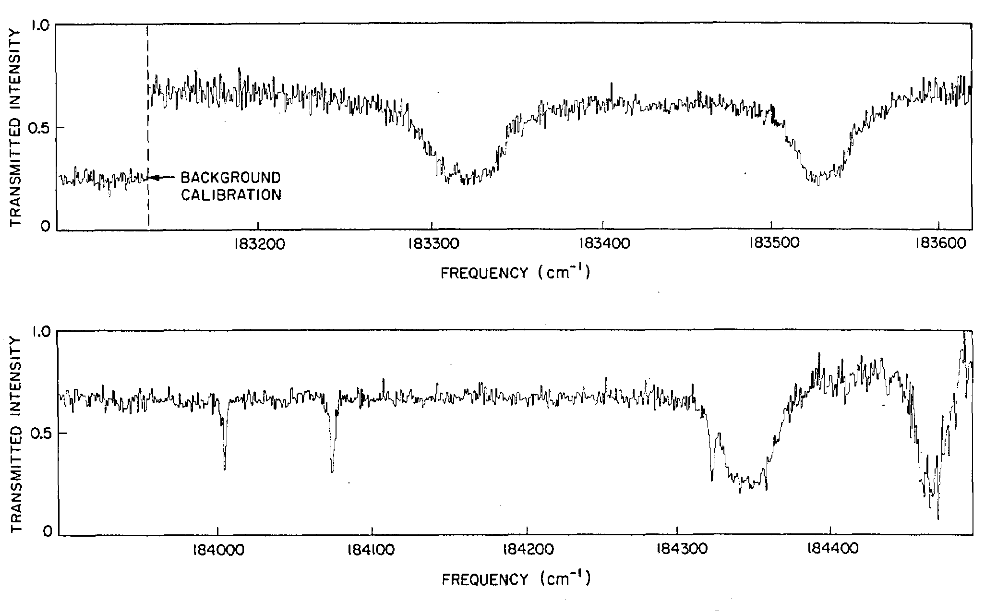

Fig. 3. Absorption scans of K. Vapor pressure is 10^{15} atoms/cm^3, and cell length is 5 cm.

Table 2. Potassium Absorption Features

Energy	Linewidth (cm^{-1})	Designation	Previously Observed Energy (cm^{-1})
183320 ± 1 cm^{-1}	8.4	$3p^5 3d(^3P)5s\ ^2P_{1/2}$	183322[a]
183530	10.5	$3p^5 3d(^3P)5s\ ^2P_{3/2}$	183532[a]
184008	2.6	–	–
184076	3.4	–	184076[b]
184321	2.5	–	–
184344	15.0	$3p^5 4s(^1P)5s\ ^2P_{3/2}$	184342[a]
184465	7.8	$3p^5 4d(^3P)5s\ ^2P_{1/2}$	184471[a]
185806	1.9	–	–
186659	5.0	$3p^5 4d(^1D)4s\ ^2D_{3/2}$	186656[a]

[a] Ref. 7.
[b] Ref. 9.

It seems likely that with these improvements, 20000 cm^{-1} of tunability should be accessible from a particular storage level. Extension of this radiation source to spectral regions centered at 199 Å in Li^+ and 103 Å in Be^{2+} seems probable.

Because of its high spectral intensity for short periods of time, this radiation source is particularly well suited for experiments of the McIlrath and Lucatorto[10] type in which inner-shell absorption is studied with the outer electron first promoted to a higher orbit. In light elements, this permits access to levels at which autoionization is prohibited (for example, Li $1s2p3p\ ^2P$) and at which near-unity fluorescent yields are expected.

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