Direct Measurement of Radiative Lifetimes of Excited States in Refractory-Metal Atoms Using a Sputtering Technique

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A new technique is reported for measuring atomic lifetimes in uranium and other refractory metals. Selective excitation of low-lying levels in neutral atoms can be obtained by means of a fast heavy-ion–induced sputtering. By pulsing the ion beam, the lifetimes of the 16 900- and 19 885-cm^{-1} levels in neutral uranium have been determined by the delayed-coincidence technique to be 232 ± 25 and 142 ± 15 ns, respectively. New lifetime determinations in Zr I are also reported.

Direct measurements of the atomic lifetimes for the low-lying excited levels in neutral uranium have so far been very limited because of the complex structure of uranium[1] and the difficulties associated with production of uranium vapor. Klose[2] has reported one direct measurement of the 27 887-cm^{-1} level in U I, using electron excitation and delayed-coincidence technique. Carlson *et al.*[3,4] have recently reported some lifetimes for high-lying odd levels and for low-lying even levels in U I by means of multistep photoionization under pulsed-dye-laser excitation. For purposes of a laser isotope-separation process, particularly the low-lying levels are of interest, and hence direct measurements of the atomic lifetimes are desirable. In this Letter, I report a new technique for the determination of atomic lifetimes of low-lying levels in uranium and in other refractory metals like Zr, Nb, Mo, or W. This technique is based upon selective excitation of atoms by heavy-ion sputtering.

Studies of optical emission from solids under ion irradiation have been performed for a long time,[5,6] mainly with ions of rather low energy, and most of them have dealt with continuum radiation from solids. I have observed that heavy-ion–induced sputtering produces rather simple spectra of the sputtered atoms even for a complicated atom like uranium. In all cases studied so far, excitation of only the low-lying states in such a sputtering process[7] explains the simple spectra obtained. A sample spectrum of the sputtered neutral uranium atoms is shown in Fig. 1. The resolution of the spectra is about 1 Å.

A 400-keV Ar^+ beam bombards the target under investigation, and the light from the sputtered particles is observed by a McPherson model 218, 0.3-m monochromator equipped with a dry-ice–cooled EMI 9658 photomultiplier. Typical ion-current density at the target is 40 $\mu A/mm^2$. By means of a set of deflection plates situated about 3 m in front, the ion beam can be pulsed by sweeping the beam across narrow slits placed in front of the target. The signal is subsequently processed by standard electronics used for delayed-coincidence experiments and accumulated in a multichannel analyzer operating in the pulse-height–analyzer mode. Beam pulses of half-widths less than 10 ns at a repetition rate of 50 kHz can be obtained.

The intensity of the discrete emission from the sputtered particles is found to increase with increasing energy of the projectile and, for a given energy, with the mass of the projectile. The simplicity of the atomic spectra from the low-lying levels makes the technique attractive for lifetime determinations, since no cascades from higher-lying levels are seen.

Typical decay curves of the 16 000-cm^{-1} level in U I and the 26 875-cm^{-1} level in Fe I are shown in Figs. 2 and 3. The cascade-free nature of the decay can be inferred from the decay of the 26 875-cm^{-1} level in Fe I. For uranium, a strong

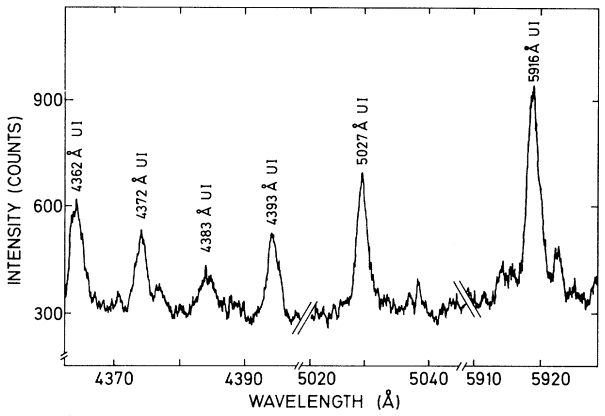

FIG. 1. Sample spectrum of neutral uranium produced by the sputtering technique.

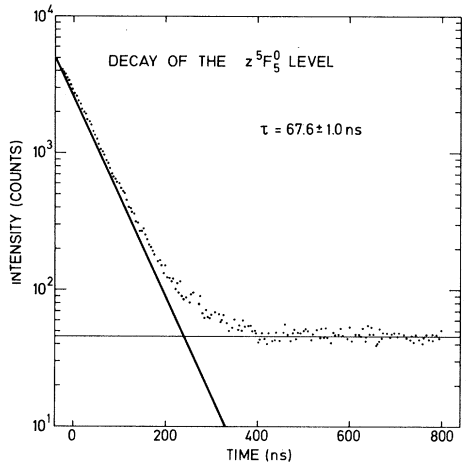

FIG. 2. Exponential decay of the 26 875-cm^{-1} level of Fe I.

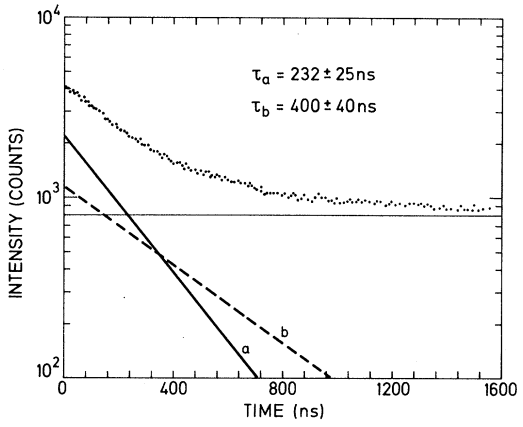

FIG. 3. Decay of the 16 900-cm^{-1} level of U I. Solid line indicates decay of the uranium line and the dashed line that of the continuum. A constant background of 800 counts is also indicated.

continuum was also observed. Similar continua have been observed for other metals and can be attributed to the presence of oxygen.[8] Thus the intensity of the continuum depends on the nature of the surface and on the vacuum inside the chamber. Typically, the vacuum has been about 6 $\times 10^{-7}$ Torr.

By pulsing the ion beam and using a delayed technique, the decay of the continuum around 5850 Å, off the 16 900-cm^{-1} line, was measured. It was possible to fit this decay ($\chi^2 = 0.96$) to one exponential with a lifetime of 400 ± 40 ns and a constant background, which presumably arises from much longer-lived states of the continuum. The decay of the U I, hence, have been computer fitted by two exponentials and a constant background. The data for Zr I represent the first direct lifetime measurements in this element. No continuum was observed for Zr in the wavelength region where the lifetimes were measured.

Table I lists some of the observed lifetimes. It can be seen that the agreement with other techniques is excellent where a comparison is possible.

TABLE I. Lifetimes measured using the sputtering technique.

	Level (cm^{-1})	Transition	Wavelength (Å)	Present (ns)	Others (ns)
Pb I	45 443	$^3F^\circ \rightarrow {}^3P_2$	2873	28 ± 2	25.8 ± 1.3[a]
Fe I	26 875	$z\,^5F_5 \rightarrow a\,^5D_4$	3720	67.6 ± 1.0	63.2 ± 3.6[b]
					61.5 ± 1.4[c]
					61.9 ± 1.5[d]
	27 167	$z\,^5F_4 \rightarrow a\,^5D_3$	3737	67.4 ± 1.0	63.3 ± 3.8[b]
					67.1 ± 1.7[c]
					62.6 ± 1.5[d]
	25 900	$z\,^5D_4 \rightarrow a\,^5D_4$	3860	86.8 ± 2.0	89.0 ± 4.8[e]
					68.5[f]
					74.6 ± 4.7[b]
	29 469	$z\,^5P_2^\circ \rightarrow a\,^5D_3$	3441	53.7 ± 1.5	36.0[f]
U I	16 900	$^7M \rightarrow {}^5L_6^\circ$	5916	232 ± 25	255 ± 25[g]
					330[h]
	19 855	$^5M \rightarrow {}^5L_6^\circ$	5027	142 ± 15	200[h]
Zr I	21 849	$a\,^3F_2 \rightarrow z\,^3G_3^\circ$	4576	440 ± 25	
	22 144	$a\,^3F_3 \rightarrow z\,^3G_4^\circ$	4634	380 ± 15	
	22 564	$a\,^3F_4 \rightarrow z\,^3G_5^\circ$	4688	450 ± 25	

[a]Ref. 10. [c]Ref. 12. [e]Ref. 14. [g]Ref. 3.
[b]Ref. 11. [d]Ref. 13. [f]Ref. 15. [h]Ref. 16.

The longest lifetimes that can be measured by the sputtering technique are limited only by the physical removal of the particles from the observation region because of their finite velocity. It has been proposed[7] that excited atoms from metallic targets are due to binary collisions at the surface of the target, and only those with velocities large enough to escape the radiationless transition region contribute to the observed spectrum. The typical velocity of the sputtered particles has been determined to be about 3000 m/sec by measuring the decay of the 30 656-cm^{-1} level of Cd I, which has a well-determined lifetime of 2.4 μsec. This velocity determination agrees well with the value calculated for a simple binary collision at the surface. In the present case, the velocity effect was overcome by a suitable choice of the optical system.

In conclusion, I report a new technique to measure atomic lifetimes, using the process of sputtering excitation and delayed coincidence. The advantages of this method are the relatively simple spectra arising only from low-lying configurations and the completely cascade-free determination of lifetimes. Since it is known that pulses of about 1-ns half-widths are obtainable by bunching an ion beam, measurements of short lifetimes should be possible with this technique. Determinations of lifetimes and transition possibilities in refractory metals of astrophysical interest are thus possible and can lead to a better estimate of their abundances in the sun and other stellar objects.

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