PHYSICAL REVIEW A VOLUME 32, NUMBER 1 JULY 1985

Analysis of the $3d$-$5f$ subarrays in the spectrum of highly ionized tantalum

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(Received 4 February 1985)

The spectrum of a laser-produced plasma of tantalum in the range 4.4—4.8 Å is interpreted as spin-orbit-split arrays of the $3d$-$5f$ transitions for ${}_{73}$Ta XLII—${}_{73}$Ta XLVI. The excellent agreement between model and experiment indicates the possibility of use for plasma diagnostics.

I. INTRODUCTION

The emission spectrum of a laser-produced plasma of tantalum has been recently recorded in the wavelength range 3.5—8.5 Å at Groupe de Recherches Coordonnées de l'Interaction Laser-Matière (GRECO-ILM). Its complete analysis will be published later. The present paper is devoted to the identification of the features which appear between 4.4 and 4.8 Å, in terms of subarrays corresponding to the $3d$-$5f$ transition.

Transitions in this range have already been observed for heavy ions (e.g., see Refs. 1 and 2). In the Ta spectra, Boiko[3] made the first tentative identifications.

In the case where the numerous lines of the transition array are very close to each other, they cannot be resolved because of the various broadening mechanisms in the plasma and the finite resolution of the spectrometer. For this reason a model has been developed[4] describing the unresolved transition array (UTA) as a whole. The formulas in Ref. 4 give the first and second moments of the wave-number distribution, i.e., the mean wavelengths and the spectral widths of the UTA's between whole configurations. However, in many cases of highly ionized heavy atoms, the spin-orbit splitting of one (or several) of the electrons may be the dominant feature of the configuration structure, so that the lines are grouped in nearly-pure-*jj*, well-separated, subarrays. Now, the formulas of Ref. 4 are still valid in this case, since it was shown that the results are independent of the coupling, but they are not useful for identifying the spectra, since the appearance of one theoretical peak is quite different from that of the experimentally distinctly separated subarrays. For these cases, new formulas have recently been established,[5] which give the first and second moments of each of the spin-orbit-split subarrays (SOSA's), assuming now pure *jj* coupling.

Features of this type have been encountered in ${}_{42}$Mo XXXI—${}_{42}$Mo XXXIV (Ref. 6) for the $3s$-$3p$ and $3p$-$3d$ transitions, cases where all the lines have been calculated individually. They were also recently recognized in the $3d$-$4f$ transitions of highly ionized gold,[7] but the peaks were superimposed on a continuous spectrum and it was not possible to measure their individual widths. The present case, which is much clearer, is presented here as a typical illustration of the SOSA formulas, which allow readily a *quantitative interpretation* in terms of subarrays.

On the spectrum displayed in Fig. 1, there appear isolated peaks and blends, associated with the ${}_{28}$Ni-like $3d^{10}$-$3d^{9}5f$ transitions (isolated lines) and their ${}_{29}$Cu-like, ${}_{30}$Zn-like, etc., satellite transitions, with one or several electrons added as "spectators." Actually, each satellite peak is the superposition of several subarrays. For instance, the Cu-like satellite of a given $3d_j$-$5f_{j'}$ Ni-like line is a blend of the $3d^{10}nl$-$(3d^9)_j5f_{j'}nl$ subarrays, with different nl's. Indeed, the average wave number of such a subarray depends very little on the nl electron, because it is an outer electron.

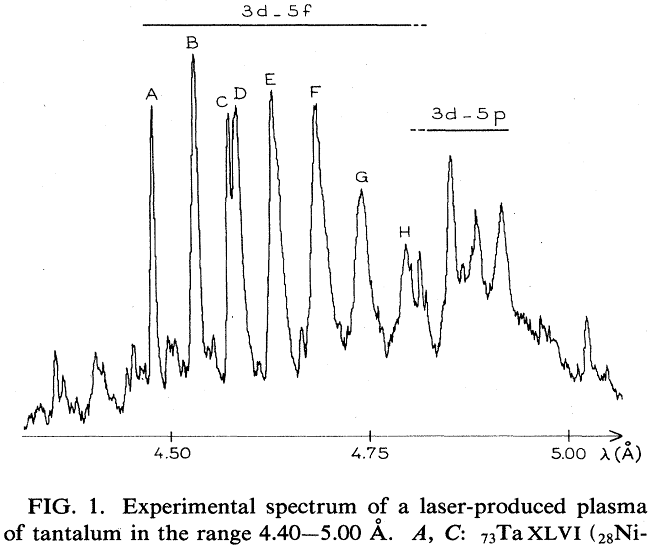

FIG. 1. Experimental spectrum of a laser-produced plasma of tantalum in the range 4.40—5.00 Å. *A*, *C*: ${}_{73}$Ta XLVI (${}_{28}$Ni-like) $3d^{10}$-$3d^{9}5f$. *B*, *E*: ${}_{73}$Ta XLV (${}_{29}$Cu-like) $3d^{10}$-$3d^{9}5f$ + one $n=4$ spectator electron. *D*, *F*: ${}_{73}$Ta XLIV (${}_{30}$Zn-like) $3d^{10}$-$3d^{9}5f$ + two $n=4$ spectator electrons. *E*, *G*: ${}_{73}$Ta XLIII (${}_{31}$Ga-like) $3d^{10}$-$3d^{9}5f$ + three $n=4$ spectator electrons. *F*, *H*: ${}_{73}$Ta XLII (${}_{32}$Ge-like) $3d^{10}$-$3d^{9}5f$ + four $n=4$ spectator electrons.

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II. EXPERIMENTAL

The spectra were produced by the impact of a 25-*J*, 600-psec Nd-glass laser with frequency doubling ($\lambda = 0.53$ μm), focused onto a flat Ta foil. For wavelength calibration, Ta foils with deposits of sulfur or sodium chloride were also used as targets. The emitted light was analyzed by a pentaerythritol (PET) crystal spectrograph in first order, and recorded on SB2 Kodak films. The intensities have been corrected for the optical-density curve of the film and for the densitometer sensitivity.

III. CALCULATIONS

The calculation of the positions of the different relativistic subconfigurations (average energies) involved has been made with the RELAC code.[8] The values of the relativistic Slater integrals have been obtained with the same potential, and their nonrelativistic averaged approximations,[9] necessary for the use of the formulas, have been computed. Using the angular coefficients given in Ref. 5, we have calculated the mean value and the variance of the wave-number distribution corresponding to each subarray.

(i) $_{28}$Ni-like transitions: $_{73}$Ta XLVI $3d^{10}$-$3d^9 5f$. For this stage of ionization the spectrum consists of three lines which can be labeled $3d_{3/2}$-$5f_{5/2}$, $3d_{5/2}$-$5f_{7/2}$, and $3d_{5/2}$-$5f_{5/2}$. These are the only *individual lines* which were computed in the present work. Their theoretical strengths are like 14, 20, and 1 in the nonrelativistic limit. The first two are denoted *A* and *C* on Fig. 1, the third one not being observable on the recorded spectrum. The width of the *A* peak is considered to be the experimental width for all the lines.

(ii) $_{29}$Cu-like spectrum: $_{73}$Ta XLV $3d^{10}nl$-$3d^9 5fnl$. For this ionization stage and the following, no individual line was computed, but only subarrays. We have only considered spectator electrons with $nl = 4s$, $4p$, $4d$, $4f$. The results for the strongest two peaks are presented in Table I. Each peak is thus the superposition of seven subarrays. The width of each of the subarrays quoted in the table includes the experimental width, as the result of a convolution of two Gaussian curves. Thus the theoretical width for the first three lines of the table is much smaller than the experimental one, but it is the resultant width which was used for the normalization of the relative intensities. These were obtained assuming local thermal equilibrium (LTE), the area of each subarray (height × width) being taken proportional to its total strength (see Ref. 5, Appendix A) multiplied by the Boltzmann factor. The value $T_e = 350$ eV leads to a calculated width in excellent agreement with experiment, but this width is not a sensitive function of T_e.

TABLE I. Results of the calculations of the positions and widths of the subarrays belonging to the $_{73}$Ta XLV ($_{29}$Cu-like) spectrum. Seven subarrays have been considered, corresponding to the various $4l_j$ spectator electrons added.

	$3d_{3/2}$-$5f_{5/2}$		$3d_{5/2}$-$5f_{7/2}$	
	λ (Å)	FWHM[a] (Å)	λ (Å)	FWHM[a] (Å)
$3d^{10}4s_{1/2}$-$3d^9 5f 4s_{1/2}$	4.532	0.007	4.627	0.007
$3d^{10}4p_{1/2}$-$3d^9 5f 4p_{1/2}$	4.533	0.008	4.629	0.007
$3d^{10}4p_{3/2}$-$3d^9 5f 4p_{3/2}$	4.530	0.008	4.625	0.009
$3d^{10}4d_{3/2}$-$3d^9 5f 4d_{3/2}$	4.532	0.025	4.630	0.008
$3d^{10}4d_{5/2}$-$3d^9 5f 4d_{5/2}$	4.533	0.009	4.627	0.027
$3d^{10}4f_{5/2}$-$3d^9 5f 4f_{5/2}$	4.535	0.023	4.634	0.009
$3d^{10}4f_{7/2}$-$3d^9 5f 4f_{7/2}$	4.537	0.010	4.631	0.025
Resultant peak	4.532	0.010	4.628	0.011

[a]Full width at half maximum; the experimental width (0.006_5 Å) is included.

(iii) $_{30}$Zn-like spectrum: $_{73}$Ta XLIV $3d^{10}nln'l'$-$3d^9 5fnln'l'$. All the possibilities with $n,n' = 4$ were taken into account, so that each peak is the superposition of 28 subarrays.

(iv) For $_{73}$Ta XLIII ($_{31}$Ga-like) and $_{73}$Ta XLII ($_{32}$Ge-like) involving, respectively, 82 and 196 subarrays, the spectator electrons were also assumed to be on the $n = 4$ shell, but simplifying assumptions were made to shorten the computations (for instance, the same values of the radial integrals were introduced in the calculation of the variances of all the subarrays). In the latter three cases, the

TABLE II. Comparison between theory and experiment for the different peaks of the spectrum.

Ions	Key	Transitions j-j'	Calc. λ (Å)	Expt.[a] λ (Å)	Calc. FWHM (Å)	Expt. FWHM (Å)
$_{73}$Ta XLVI ($_{28}$Ni-like)	*A*	3/2-5/2	4.477	4.478		0.006_5
$_{73}$Ta XLV ($_{29}$Cu-like)	*B*	3/2-5/2	4.532	4.530	0.010	0.010
$_{73}$Ta XLVI ($_{28}$Ni-like)	*C*	5/2-7/2	4.574	4.574	0.014	0.020
$_{73}$Ta XLIV ($_{30}$Zn-like)	*D*	3/2-5/2	4.585	4.584		
$_{73}$Ta XLV ($_{29}$Cu-like)	*E*	5/2-7/2	4.628	4.629	0.011	0.016_5
$_{73}$Ta XLIII ($_{31}$Ga-like)		3/2-5/2	4.636	4.635	0.020	
$_{73}$Ta XLIV ($_{30}$Zn-like)	*F*	5/2-7/2	4.683	4.685	0.016	0.022
$_{73}$Ta XLII ($_{32}$Ge-like)		3/2-5/2	4.690		0.024	
$_{73}$Ta XLIII ($_{31}$Ga-like) + ?	*G*	5/2-7/2	4.736	4.742	0.023	0.024
$_{73}$Ta XLII ($_{32}$Ge-like) + ?	*H*	5/2-7/2	4.793	4.799	0.028	Blended

[a]The experimental uncertainty in the calibration is 1.5×10^{-3} Å.

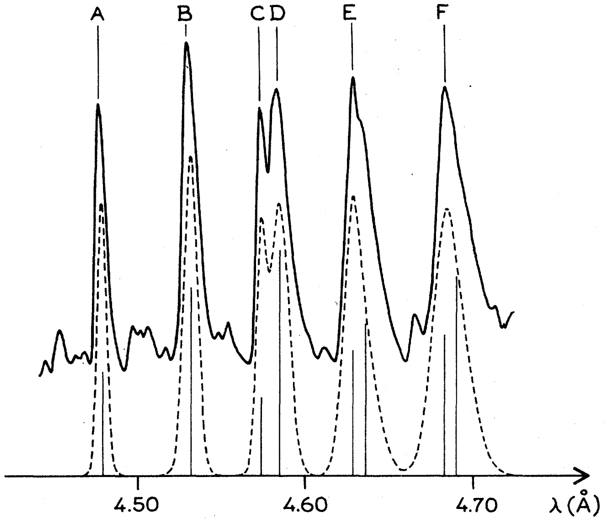

FIG. 2. Comparison between experimental spectrum (solid line) and calculated spectrum (dashed line). Mean wavelength and width of each peak have been computed individually (see Table II). Relative intensities have been adjusted to obtain the best fit. Calculated spectrum is shifted downwards for clarity.

same value $T_e = 350$ eV was used to evaluate the relative contribution of each subarray, as in the case of $_{73}$Ta XLV.

IV. DISCUSSION

All the results are gathered in Table II. It can be seen that agreement with experiment is excellent, for the *positions* as well as for the *widths* of the peaks. A computed spectrum is shown in Fig. 2. In this spectrum, no attempt has been made to compute the relative intensities of the different ionization stages, and these were taken as free parameters to obtain a good fit with the experimental results. Indeed, the photographic spectrum is a time and space average of the plasma; the different ionization stages are not at their maximum abundance for the same density and temperature, and a one-temperature LTE model would be too rough an assumption for intensity calculations. Here, estimating the relative contribution of each subarray to a given peak with the same T_e value (350 eV) was the only possible approximation.

V. CONCLUSION

In the present work, we demonstrate the usefulness of the new SOSA formalism and of the related formulas developed in Ref. 5. The previous, whole-configurations UTA model would have predicted here a structureless spectrum, in disagreement with experiment. Moreover, the full computation of all the individual lines would have been a formidable task, and a waste of efforts.

As is always the case in plasma spectroscopy, identification of the spectrum is a preliminary condition for performing diagnostics. In this respect, the $3d$-$5f$ transition seems an interesting case, since the different ionization stages are well separated (at least lines *A*, *B*, *C*, *D*), in contradistinction to $3d$-$4f$ and $3d$-$4p$.[10] However, the full achievement of spectroscopic diagnostics of a laser-produced plasma requires, of course, space and time resolution on one hand, and a collisional-radiative model, on the other hand.

ACKNOWLEDGMENTS

One of the authors (M.K.) gratefully acknowledges the Université Paris—Sud, for financial support during his stay at Laboratoire Aimé Cotton.

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