RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 34, NUMBER 1 JULY 1986

Density-sensitive electric quadrupole decays in Ni-like ions observed in laser-produced plasmas

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(Received 25 March 1986)

The electric quadrupole transition $3p^6 3d^{10}\,{}^1S_0$–$3p^5 3d^{10} 4f$ $(\frac{3}{2},\frac{7}{2})$ $J=2$ has been observed in the spectra of Ta XLVI, W XLVII, Re XLVIII, Pt LI, Au LII, and Hg LIII. The excited-level populations of Ni-like gold are calculated by means of a collisional-radiative model and show that the intensity ratio of the electric dipole $3p_{3/2}$-$4d_{5/2}$ and electric quadrupole $3p_{3/2}$-$4f_{7/2}$ resonance transitions is sensitive to density in the range 10^{19}–10^{21} cm^{-3}, in qualitative agreement with the present observations.

The spectra of multicharged ions produced by laser irradiation of high-Z elements are dominated by the electric-dipole resonance lines of the isoelectronic sequence of nickel. The transitions from the $J=1$ odd levels $3d^9 4p, 4f, 5f$, $3p^5 4s, 4d$ to the ground state $3p^6 3d^{10}\,{}^1S_0$ have been identified below 10 Å in most of the elements from hafnium ($Z=72$) to gold ($Z=79$).[1-7] It has been concluded so far that the laser-produced plasmas, which involve electron densities n_e typically larger than 10^{20} cm^{-3}, are not suitable for the observation of "forbidden" lines. In this article, we report on the experimental and theoretical evidence of electric-quadrupole decays in these spectra.

Since their ground state corresponds to complete filling of the $n=3$ shell, all $J=2$ even levels having one electron in the $n=4$ shell may decay by means of one electric-quadrupole transition with $\Delta n=1$, except for the first excited configuration $3d^9 4s$, to the $J=1, 2$, and 3 odd levels by means of electric-dipole transitions with $\Delta n=0$. It is known[8] that the transition probabilities for these two decays scale, respectively, as Z_c^6 and Z_c (Z_c being the charge of the ion core). As a consequence, the $E2$ transitions lose their "forbidden" character at high-Z_c values. With regard to the closed-shell character of their ground state, neonlike and nickel-like ions behave in the same manner. The identification of the $E2$ transition from the $2s2p^6 3d$ level to the ground state $2s^2 2p^6\,{}^1S_0$ has already been reported for Sr XXIX, Zr XXXI, Mo XXXIII, Rh XXXVI, and Ag XXXVIII[9] in neonlike ions produced by laser irradiation.

The spectra of the six elements tantalum ($Z=73$), tungsten ($Z=74$), rhenium ($Z=75$), platinum ($Z=78$), gold ($Z=79$), and mercury ($Z=80$) have been recorded at the Groupement de Recherches Coordonnées Interaction Laser-Matière (GRECO-ILM) laser facility in Palaiseau, France. The laser wavelength was 0.53 μm with pulse duration of 600 ps. The laser beam was focused on wire, foil, or drop (in the case of Hg) targets, and the laser intensity was about 10^{14} W/cm^2. Further details on the experimental setup, as well as a typical spectrum of gold, were published recently.[6]

A picture of the spectral region of interest is given in Fig. 1. The line labeled B is the transition $3p^6 3d^{10}$–$3p^5 3d^{10} 4d$ $(\frac{3}{2},\frac{5}{2})$ $J=1$. It is surrounded by the $3p$-$4d$ transitions of the neighboring ions, mostly copperlike at longer wavelengths and cobaltlike at shorter wavelengths. At the left-hand side of the six spectra, the unresolved $3d$-$5p$ and $3p$-$4d$ transition arrays (UTA) emitted by ions isoelectronic to zinc and elements of higher Z, merge in complex structures. The lines labeled A form a sequence which cannot be interpreted as an electric-dipole transition in Ni-like and nearby sequences by *ab initio* estimates of wavelengths and transition probabilities provided by the RELAC code using the relativistic parametric potential method.[10,11] The same code was then used for the determination of the electric-quadrupole A_{E2} transition probabilities and it was noticed that the transitions with the highest probabilities, $3p^6 3d^{10}$–$3p^5 3d^{10} 4f$ $(\frac{3}{2},\frac{7}{2})$ $J=2$ did fit quite well (within 0.01 Å) the observed sequence of A lines.

According to their transition probabilities, other $E2$ transitions should be observable at lower wavelengths ($3p$-$4f$ and $3s$-$4d$ types), but they occur in the dense spectral region of the $n=3$ to $n=5$ transitions and could not be identified unambiguously. We recall that the two $E2$ transitions from the $3d^9 4s$ levels in Mo XV[12] and in Xe XXVII[13] have already been observed in tokamak plasmas at much lower electron densities. In the sequence from tantalum to mercury presented here, their transition probabilities are two orders of magnitude lower than for the A line now identified, and they are not detectable in our spectra.

Because of the good signal-to-noise ratio, an isoelectronic sequence of faint lines, labeled a in Fig. 1, could be measured at about 0.02 Å from the A lines. Two possible interpretations of this satellite spectral feature have been examined. First the Ni-like transition $3p^6 3d^{10}\,{}^1S_0$-$3p^5 3d^{10} 4f$ $(\frac{3}{2},\frac{5}{2})$ $J=2$ for which the transition probability is about 1% of that for the A line, proved unsatisfactory with regard to experimental and theoretical wavelength comparisons. The fact that the wavelength differences $\lambda_{\rm exp}-\lambda_{\rm theor}$ are similar

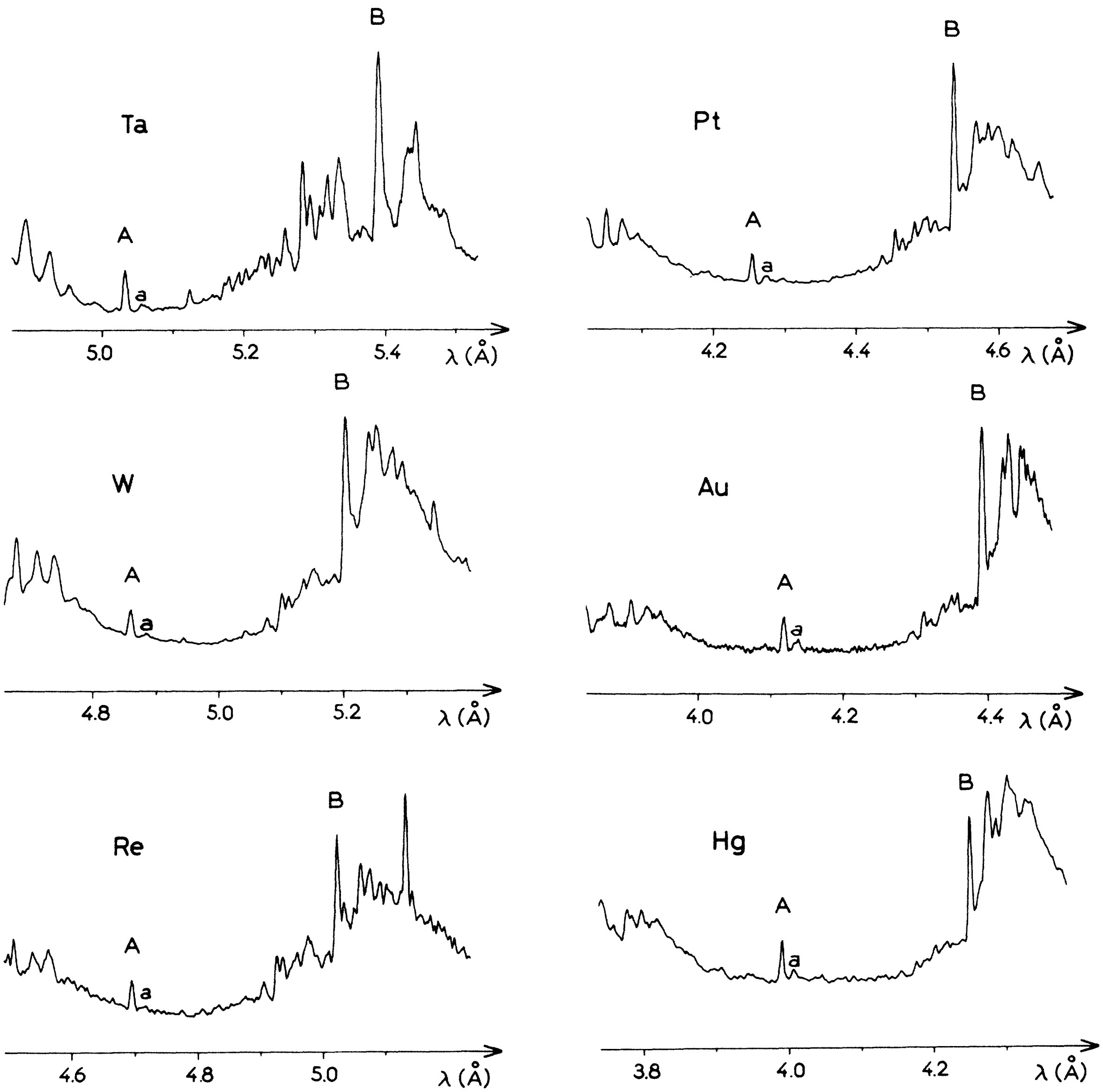

FIG. 1. Spectra of laser-irradiated tantalum, tungsten, rhenium, platinum, gold, and mercury. The lines labeled A and B are, respectively, the transitions from $3p^5 3d^{10} 4f\ (\frac{3}{2},\frac{7}{2})\ J=2$ ($E2$ type), and from $3p^5 3d^{10} 4d\ (\frac{3}{2},\frac{5}{2})\ J=1$ ($E1$ type) to the ground state of nickel-like ions. The long-wavelength companion (labeled a) of the A line is its Cu-like satellite.

for A and a lines supports the second interpretation that the a lines are $E2$ emissions from the $3p^5 3d^{10} 4fnl$ level in Cu-like ions. Furthermore, the larger width of the a lines is indicative of a satellite structure associated with the parent A line. To check this, and since the UTA formalism has not yet been developed for $E2$ transitions, a detailed evaluation of the two arrays $3d^{10}4s$-$3p^5 3d^{10} 4s4f$ and $3d^{10}4p$-$3p^5 3d^{10} 4p4f$ was performed and we found that all strong lines of these arrays occur in a range of 0.005 Å. The ratio of the intensity of the A line to that of its recombination satellite is several times larger than for any $E1$ resonance line observed in our spectra (see, for example, Fig. 1 of Ref. 6) and this can be understood qualitatively. In Cu-like ions, the odd and even levels of the $3p^5 3d^{10} 4fnl$ level can decay by means of $E1$ transitions with $n=1$, and the Z_c^5 factor which favors some $E2$ transitions in the branching ratios of nickel-like ions is then changed in Z_c^2.

The wavelengths and transition probabilities have been calculated by taking into account the mixing of all configurations having 17 electrons in the $n=3$ shell and one electron in the $n=4$ shell, the potential parameters being fitted for such upper configurations. The discrepancies with experimental wavelengths reported in Table I are somewhat larger than for $3d$-nl resonance transitions. In order to understand this, the repulsive effect of the $3p^6 3d^8 4f$ level on the $3p^5 3d^{10}$ level was evaluated in Co-like ions. It assumes that the wavelengths of the resonance lines from the $3p^5 nl$ levels in Ni-like ions should be shifted by 0.002 Å if the $3p^6 3d^8 4fnl$ levels are explicitly introduced in the energy matrices. It was found also that potential parameters fitted on $3p^5$-type configurations only can further reduce the wavelength discrepancies by another 0.002 Å.

Space-resolved intensity measurements of the A and B lines in Au show that their intensity ratio varies significantly

TABLE I. Wavelengths (in Å) and weighted transition probabilities gA (in 10^{13} s^{-1}) for the transition $3p^6 3d^{10}$ $J=0-3p^5 3d^{10} 4f$ $(\frac{3}{2},\frac{7}{2})$ $J=2$ in Ta XLVI, W XLVII, Re XLVIII, Pt LI, Au LII, and Hg LIII, and the same for the strongest associated satellites in Ta XLV, W XLVI, Re XLVII, Pt L, Au LI, and Hg LII.

Ion	J_{upp}	J_{low}	Tantalum λ_{theor}	Tantalum gA	Tantalum λ_{expt}	Tungsten λ_{theor}	Tungsten gA	Tungsten λ_{expt}	Rhenium λ_{theor}	Rhenium gA	Rhenium λ_{expt}
Ni-like	2	0	5.019	0.253	5.028	4.848	0.278	4.857	4.686	0.296	4.695
Cu-like	$\frac{5}{2}$	$\frac{1}{2}$	5.042	0.285		4.870	0.319		4.707	0.357	
(4s spectator)	$\frac{3}{2}$	$\frac{1}{2}$	5.044	0.199		4.872	0.222		4.709	0.247	
Cu-like	$\frac{3}{2}$	$\frac{1}{2}$	5.046	0.197	5.054	4.874	0.334	4.881	4.710	0.246	4.717
(4p spectator)	$\frac{5}{2}$	$\frac{1}{2}$	5.046	0.299		4.874	0.334		4.710	0.371	
	$\frac{5}{2}$	$\frac{3}{2}$	5.047	0.273		4.875	0.304		4.711	0.337	

Ion	J_{upp}	J_{low}	Platinum λ_{theor}	Platinum gA	Platinum λ_{expt}	Gold λ_{theor}	Gold gA	Gold λ_{expt}	Mercury λ_{theor}	Mercury gA	Mercury λ_{expt}
Ni-like	2	0	4.246	0.457	4.254	4.113	0.497	4.119	3.985	0.544	3.993
Cu-like	$\frac{5}{2}$	$\frac{1}{2}$	4.263	0.492		4.129	0.546		4.001	0.604	
(4s spectator)	$\frac{3}{2}$	$\frac{1}{2}$	4.265	0.335		4.130	0.370		4.002	0.407	
Cu-like	$\frac{3}{2}$	$\frac{1}{2}$	4.266	0.336	4.274	4.132	0.372	4.137	4.004	0.410	4.008
(4p spectator)	$\frac{5}{2}$	$\frac{1}{2}$	4.266	0.505		4.132	0.558		4.004	0.615	
	$\frac{5}{2}$	$\frac{3}{2}$	4.267	0.453		4.133	0.498		4.005	0.546	

along the laser axis. One can expect that the ratio of these two lines, which have very different transition probabilities, varies with increasing electron densities, when the collisional processes have to be considered in the population and/or depopulation processes of the quasimetastable upper level of the A transition. The chain of programs developed by the last three authors at Jerusalem was used to solve the collisional-radiative problem of evaluating the excited-level populations in Au LII. We assumed a plasma with an electron temperature of 800 eV consisting of 50% Ni-like ions in their ground state. The configurations $3p^6 3d^9 4s$, $4p$, $4d$, $4f$ and $3p^5 3d^{10} 4s$, $4p$, $4d$, $4f$ were included in the calculations to evaluate the radiative transition probabilities and the collisional rates connecting all the excited levels among themselves and with the ground state. Lines were assumed to have a Doppler profile at an ion temperature of 400 eV. Self-absorption in a 30-μm-thick plasma, corresponding approximately to the observation conditions, was also taken into account.

Figure 2 shows the computed intensity ratio I_A/I_B for several electron densities up to 10^{22} cm^{-3} together with typical experimental results. Error bars are evaluated from the uncertainties in the film calibration and the residual inaccuracies in subtracting the underlying continuum of the B line. In the experiments, space resolution was kept to a minimum value of 25 μm in order to improve the signal-to-noise ratio. At low electron densities, the I_A/I_B ratio is unaffected by collisions up to $n_e=10^{19}$ cm^{-3} when collisions begin to play a part in the depopulation of the upper levels. Above $n_e=10^{21}$ cm^{-3}, opacity effects on the B line saturate the density variations of the I_A/I_B ratio.

A good qualitative agreement is found between theory and experiment, in view of the fact that the measurements are time integrated and the model simple. In particular, cascades from upper levels with $n=5$, autoionization processes, and recombination from Co-like ions were neglected in the present study. However, these results show that the

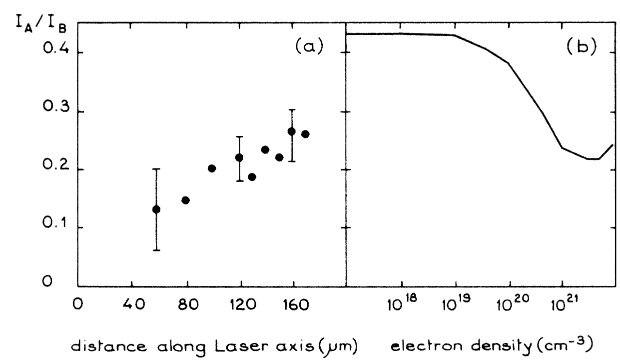

FIG. 2. (a) Experimental intensity ratio I_A/I_B of the lines labeled A and B in Fig. 1, as a function of the distance from the laser target. The origin of the distance scale is arbitrary. (b) *ab initio* evaluation of the same ratio as a function of the electron density in the collisional-radiative model.

electron density can be estimated to be in the range 10^{21}–10^{22} cm^{-3} at a distance of 100 μm in Fig. 2(a), but the origin of the scale cannot be well defined. This distance corresponds to the spatial peak of $3d^{10}$-$3d^9 4f$ emissions in previous experiments using Au-Al alloy targets with electron densities in the same range.[14]

In conclusion, the present wavelength measurements of electric-quadrupole transitions in several Ni-like ions for elements in the range $Z=73$–80 are in satisfactory agreement with *ab initio* calculations. Satellite spectra of these $E2$ decays have also been measured and identified as originating from copperlike ions. Density-sensitive $E2$-to-$E1$ line ratios in high-density plasmas had already been predicted in the neon isoelectronic sequence.[15] The present work completes a similar theoretical analysis in one element of the nickel sequence with experimental checks. This opens the way to promising electron density diagnostics in high-density, high-Z plasmas.

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