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Witzel, B.; Poppadogiannis, N. A.; Uiterwaal, Cornelis J.; Schröder, H.; and Charalambidis, D., "Multiphoton ionization of V, Cr, Y, La, and Ir through 0.5-ps ultraviolet laser pulses" (1999). *C.J.G.J. Uiterwaal Publications.* Paper 11.
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Phys. Rev. A 60, 3311 - 3312 (1999)
[Issue 4 – October 1999]

Multiphoton ionization of V, Cr, Y, La, and Ir through 0.5-ps ultraviolet laser pulses

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Received 29 April 1999

The two-photon ionization of the metal atoms V, Cr, Y, La, and Ir sputtered by an Ar-ion gun from solid targets has been investigated using linearly polarized laser pulses of 500-fs duration and 248.6-nm (KrF) wavelength in combination with mass spectroscopy that is free of volume effects. A near-resonant ionization process has to be attributed to all five atoms. By fitting analytical solutions of a rate equation model describing the temporal evolution of the ionization process, we can determine absolute values of the one-photon cross sections and saturation intensities.

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URL: http://link.aps.org/abstract/PRA/v60/p3311
DOI: 10.1103/PhysRevA.60.3311
PACS: 32.80.Rm

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PACS number(s): 32.80.Rm

Quantitative ion mass spectroscopy subsequent to multiphoton ionization (MPI) allows the identification of the ionization process as a resonant or nonresonant one, the determination of the saturation intensity, and generalized cross sections. Quantification of a technique employing MPI involves complications arising from the nonlinear character of the process and from the three-dimensional intensity distribution around the focus of the ionizing laser. Because of this intensity distribution and the nonlinearity of the process, the ionization rate is strongly position dependent. As a result, measured spectra usually include what is called volume effects. In our experiments the laser–secondary neutral mass spectroscopy (L-SNMS) [1] or surface analysis by laser ionization (SALI) [2] is employed: particles sputtered by an Ar-ion gun from the solid target are postionized with a fixed frequency laser and further mass analyzed. Quantification of our results is enabled by an advanced mass spectroscopic technique [3] that detects only ions originating from a small fixed spatial volume in which the intensity distribution of the radiation is practically constant. In this way volume effects are eliminated from the measured spectra.

In a recent publication, employing the above technique the MPI of a series of metals has been systematically investigated using 0.5-ps laser pulses of 5-eV photon energy [4]. In the present work we report measurements of five additional metals (V, Cr, Y, La, and Ir) and thus expand the database of the MPI of metal atoms under the given radiation parameters of the KrF laser system used [5,6]. At these laser parameters, our completely volume-effect-free technique is unique for the study of resonant or near-resonant two-photon ionization processes. In a log-log representation of the ion yield as a function of the laser intensity in the nonsaturated regime, the slope of the line is between 1 and 2. For a Gaussian beam profile, volume effects result in a slope of $\frac{3}{2}$ [7] above the saturation intensity. These slope values are so close that saturation (change of slope) is effectively not observable. Elimination of volume effects is an efficient way to avoid this problem.

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The experimental setup and method employed in the present work have been described elsewhere [4]. What is measured here in a volume-effect-free manner is ion yield versus laser intensity for the V, Cr, Y, La, and Ir atoms. The ionization thresholds of the investigated elements range between 5.58 and 9.1 eV. Thus for all of them the ionization involves a two-photon absorption. Figure 1 shows the measured ion yields in a log-log representation. For four of the above metals, observation of absolute saturation was possible within the range of intensities available in the experiment. In the case of La, absolute saturation is not observable for an averaged intensity $I_{av} \leqslant 2\times10^{12}$ W/cm^2. For all five elements, double ionization is observed which (except for the case of La) almost saturates for the highest intensities used here. Triply ionized Y, V, and Cr could also be observed. In the case of La, LaO ions are strongly present and depict saturation. This oxide could perhaps be related to the production mechanism of La^+, which does not show saturation. Furthermore, for all curves in Fig. 1, the observed slope for the lowest intensities employed (far from the saturation intensity) is significantly smaller than 2 (1.5–1.7), which is the expected value for a completely nonresonant process.

Since the coherence time of the laser is about 10 times shorter than its pulse duration, we apply a rate equation model, describing the temporal evolution of the ionization

TABLE I. Experimental ($\sigma_1,\sigma_2,I_{sat}$) values of the resonantly enhanced MPI cross sections for V, Cr, Y, and Ir. For each of the listed pairs (σ_1,σ_2) there exists another pair ($\frac{1}{2}\sigma_2,2\sigma_1$) that gives an equally good fit to the experimental data. The errors given are one standard deviation, and include the experimental uncertainty in the laser intensity. The error values for the given saturation intensity I_{sat} are approximately 30%. For the definition of the symbols used and the fitting procedure, see Ref. [2].

Z	Atom	σ_1 (10^{-17} cm^{-2})	σ_2 (10^{-17} cm^{-2})	I_{sat} (W/cm^2)
23	V	0.4±0.12	7.9±2.4	3.6×10^{11}
24	Cr	1.7±1	3.0±2.4	1.5×10^{11}
39	Y	0.6±0.2	8.7±3	2.7×10^{11}
77	Ir	1.1±0.3	16±4.8	1.5×10^{11}

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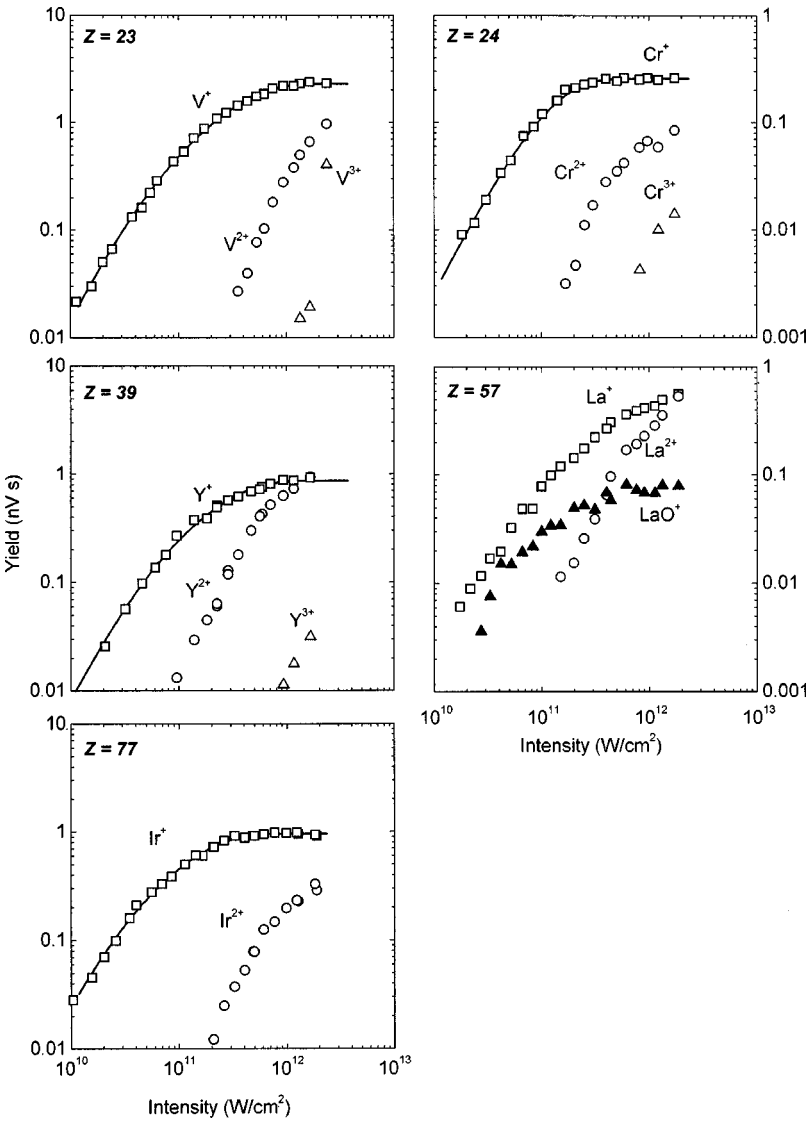

FIG. 1. Measured ion yields as a function of laser intensity for V ($Z=23$), Cr ($Z=24$), Y ($Z=39$), La ($Z=57$), and Ir ($Z=77$).

process, in order to extract quantitative results from the measured data. Details of the model are described in Ref. [4].

An analytical solution of the rate equation model [Eq. (15) of Ref. [4]] has been fitted in the measured single ionization data. The fitted curves are shown in Fig. 1. The resulting fit parameters shown in Table I are the cross sections σ_1 and σ_2 and the saturation intensity I_{sat} of the near-resonant ionization process, as defined in Ref. [4].

The extracted cross sections indicate that saturation intensities for the resonantly enhanced processes of V, Cr, Y, and Ir will be in the range between 10^6 and $10^7\,\mathrm{W\,cm^{-2}}$ when using a common KrF ns laser system, and thus experiments are tractable with such a common excimer laser system too.

This work was carried out in the Ultraviolet Laser Facility (ULF) operating at FORTH-IESL (TMR Contract No. ERB-FMGE-CT950021). Two of us (C.J.G.J.U. and B.W.) gratefully acknowledge the Alexander von Humboldt–Stiftung for support during part of this work. We would like to thank A. Eglezis for his skillful technical assistance.

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