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LETTER TO THE EDITOR

A laser-generated plasma as a source of VUV continuum radiation for photoelectronic spectroscopy

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Abstract. The feasibility of using laser-generated plasmas as VUV continuum sources for photoelectron spectroscopy has been demonstrated by measuring the spectral intensity distribution of the VUV continuum in the wavelength region from 79 nm to 43 nm by energy analysis of the photoelectrons ejected from argon atoms. The maximum photon flux obtained after reflection at a gold-coated spherical mirror was of the order of 10^{11} photons nm^{-1} per pulse at 50 nm for a laser energy of 830 mJ. The results show a shift of the emission maximum to lower wavelengths with increasing laser energy.

This letter describes a study of the suitability of laser-produced plasmas as sources of vacuum ultraviolet (VUV) radiation sufficiently reproducible and intense for photoelectron spectroscopy on a shot-to-shot basis. Since the use of laser-generated plasmas as background continuum light sources for absorption spectroscopy was first suggested by Ehler and Weissler (1966), a number of research groups have studied their properties; in particular, Breton and Papoular (1973) investigated the radiance between 1000 and 2000 Å as a function of several different experimental parameters, and Carroll *et al* (1978, 1980) made a systematic study of the effect on the spectrum of the target material over the range 35–2000 Å. Mahajan *et al* (1979) and Carroll *et al* (1980) recently demonstrated that the shot-to-shot reproducibility is good enough ($\pm 15\%$) to allow photoelectric scanning of absorption spectra. In this letter we show that the intensity and the reproducibility, both short-term shot-to-shot and long-term over several thousand shots, also make it suitable for pulsed photoelectron spectroscopy, thus opening up the possibilities of time-resolved experiments on short-lived species such as molecular radicals and laser-pumped atoms and molecules.

The feasibility of a pulsed technique for photoelectron spectroscopy was demonstrated by applying the method in reverse: the energy spectrum of the photoelectrons ejected from argon atoms, together with the known differential photoionisation cross-section of argon, were used to derive the spectrum of the VUV source over the region on the short wavelength side of the ionisation threshold.

Figure 1 shows a schematic view of the apparatus used. A 20 ns pulse of radiation from a Q-switched ruby laser, of energy up to 830 mJ, was focused on to a rotatable solid copper target, forming a plasma spot about 0.3 mm in diameter. This plasma spot was imaged by a gold-coated spherical mirror in the interaction region, with a magnification of 10. In this region the VUV beam of a divergence of ± 50 mrad intersected an intense

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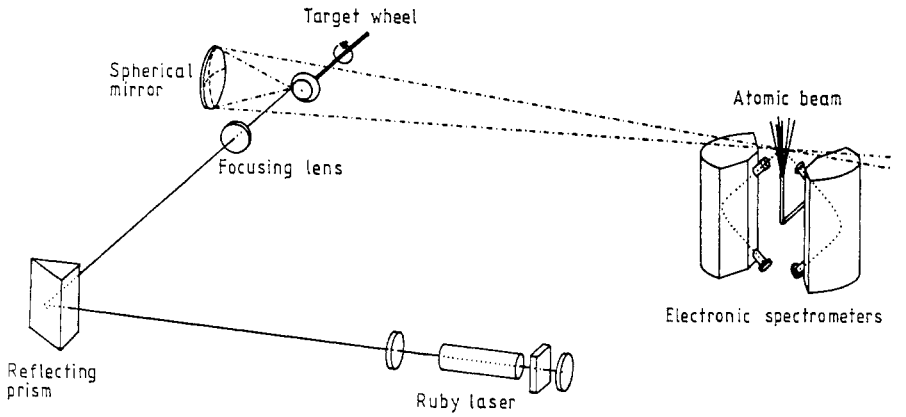

Figure 1. Schematic view of the experimental arrangement.

atomic beam of argon. The resulting photoelectrons were collected by two cylindrical mirror analysers (CMA), one of which was used as a reference detector (Heckenkamp 1981). The entrance system of each CMA contained two spherical molybdenum grids with centre of curvature in the interaction region. By applying a voltage V_g between these grids, electrons could be accelerated or retarded before entering the analyser. Spectra were recorded by keeping the pass energy of the analyser constant at some value E_t and sweeping V_g. Since the energy bandwidth ΔE of the electrons detected depends on E_t, this technique has the advantage of constant resolution over the whole electron spectrum. The central energy, given by

$$E_{el} = E_t - eV_g$$

ranged from 0 to 13 eV in these experiments. The collection efficiency of the CMA is proportional to E_t^2 (Palmberg 1975, Heckenkamp 1981), and in this experiment we used $E_t = 5.2$ eV, giving $\Delta E = 200$ meV (FWHM). Electrons passing the CMA were detected by a microchannel plate electron multiplier, followed by a preamplifier, the signal from which was proportional to the total charge of the photoelectrons passing the spectrometer during the pulse. The outputs from the preamplifiers were amplified, shaped, gated to synchronise with the laser pulse, digitised and stored by a microcomputer system for data acquisition.

The performance of the pulsed system was assessed by using it to obtain the spectrum of the VUV source, i.e., plasma spot plus gold mirror. In the case of argon, the only significant ionisation thresholds are 3p $^2P_{3/2}$, $^2P_{1/2}$ at 15.76 and 15.94 eV, since the cross-section at the next threshold, 3s at 29.20 eV, is an order of magnitude smaller (Adam *et al* 1978) and corresponds to a wavelength ($\sim$40 nm) at which the reflectivity of gold is very low. The photon energy corresponding to a photoelectron energy E_{el} is, therefore, $h\nu = E_{el} + 15.8$ eV, taking the weighted mean of the ^2P values. The spectral distribution of the ionising VUV radiation can then be found from the known differential cross-section of argon over the range 30 eV to threshold (Marr and West 1976, Dehmer *et al* 1975).

Figure 2 shows the results for three different laser energies. Each point is the mean of 20 shots. The error bars represent a standard deviation, shot-to-shot, of about 8%, plus a contribution for the $\pm 5°$ angular acceptance of the spectrometer and the uncertainty in the differential cross-section. It must be emphasised that these distributions refer not to the plasma itself but to the source after reflection of the radiation by a mirror

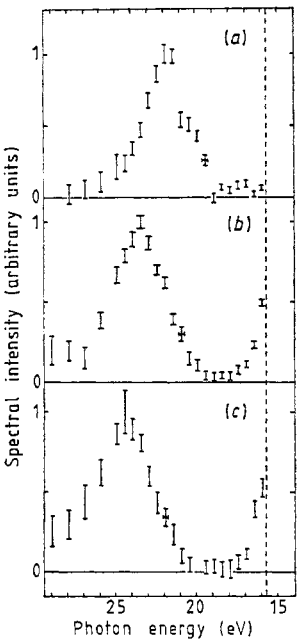

Figure 2. Spectral intensity distribution of the VUV radiation after reflection at the spherical mirror for: (*a*) 450 mJ; (*b*) 620 mJ; (*c*) 830 mJ laser energy.

initially coated with gold and effectively over-coated with copper from the target; the variation of reflectivity with wavelength is therefore not well known. However, the same derived spectrum could be reproduced with the same laser energy after a sequence of some 30 000 shots. The changes in derived spectrum with increasing laser energy (shift and increasing width of peak) are therefore real effects, and, more importantly for our purpose, the properties of the source are seen to remain constant over this large number of shots. The photon flux at the atomic beam estimated from direct measurement of the output of the preamplifier with an oscilloscope is about 10^{11} photons nm^{-1} or 10^{20} photons $nm^{-1}\ sr^{-1}\ mm^{-2}\ s^{-1}$ at the maximum of the distribution, in reasonable agreement with the measurement by an entirely different technique of Mahajan *et al* (1979). (Note that the latter paper contains a misprint: the photon flux should read $4.6\times10^{18}\ s^{-1}\ sr^{-1}\ Å^{-1}$, not 4.6×10^{8}.)

The principal interest of this pulsed technique lies in its application to photoelectron spectroscopy on atoms or molecules selectively excited by pulsed dye lasers. The main advantage of this VUV source is its high photon flux, especially if the photoelectron spectroscopy experiment can be done without an optical monochromator. The requirement of energy dispersion is, as shown in this letter, then fulfilled by analysis of the energy distribution of the photoelectrons produced. This method of 'photoelectron conversion' is a well known technique in x-ray spectroscopy as pointed out by Krause *et al* (1972). It can be used in cases in which only one ionisation threshold in the continuous spectrum of the source has to be taken into account or in which the photoionisation cross-section shows a very pronounced resonance structure as a function of the photon energy, e.g. in auto-ionisation resonances (as shown for Hg by Schönhense 1981). This second case should be especially important for applications of the source in photoionisation experiments on laser-excited metal atoms.

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