Multiphoton Ionization of Hg and Xe†

S. L. Chin,* N. R. Isenor, and M. Young
Department of Physics, University of Waterloo, Waterloo, Ontario, Canada

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Observations of laser-induced ionization of Hg and Xe are consistent with existing theory. Effects of the depletion of neutral atoms were observed in both cases. It is proposed that the experimental results of others may have deviated from theoretical results because of this saturation effect, and not because of level shifting as suggested.

Multiphoton ionization of rare gases by an intense ruby- or nydemium-glass laser radiation has been studied theoretically by Bebb and Gold,[1] and experimentally by several authors.[2–4] The experimental results indicate that the ionization probability is proportional to the flux raised to a power somewhat less than that predicted by the theory, that is, the next integer (k) greater than the ionization energy divided by the photon energy. The explanation given was that Stark shifts and broadening of the energy levels in the strong field of the laser radiation effectively lowered the ionization potential. The present paper presents results on the multiphoton ionization of Xe and Hg that do not support the above explanation. These results, rather, indicate that saturation of the ionization is the reason for the discrepancies.

A Q-switched ruby-laser oscillator/amplifier system capable of producing peak power of ~100 MW and beam divergence of 5 mrad was focused into a vacuum system containing the gas under study. The ions formed were separated and identified by a time-of-flight method. An electron multiplier of sensitivity < 10 ions per pulse was used as a detector.

The experimental results are shown in Fig. 1. N is the total number of ions created per pulse, and τ is the pulse width at half-power. F_0 is the peak photon flux in the focal region. One can see that there are significant changes in the slope of the log-log plot of the data at high laser powers.

At low power, it can be assumed that a very small fraction of the atoms are ionized during a laser pulse. Thus one would expect the value of N/τ to be proportional to the ionization rate and thus (according to Bebb and Gold[1]) to be proportional to $F_0{}^k$. However, when the density of ions becomes appreciable compared with the original atom density, the plot of N/τ versus F_0 would no longer follow this relationship. In other words, for higher values of F_0, we would expect the slope to be less than k.

With this saturation effect taken into account, we derived the expressions

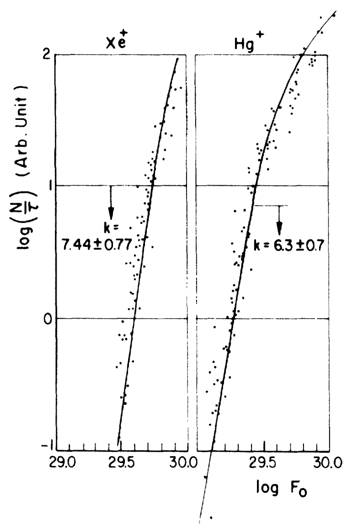

FIG. 1. Log-log plot of the ion-production rate versus laser peak flux. Curves are obtained from Eq. (1). Slopes indicated are based on the experimental points below the arrow.

$$N=\sum_{n=1}^{\infty}\frac{(-1)^{n-1}}{n!}(W_k F_0{}^k \tau_k)^n V_{nk} , \qquad (1)$$

where τ_k, the effective laser pulse width for a kth-order process, is given by

$$\tau_k = \int P^k(t)\,dt ,$$

where $P(t)$ is the normalized power in the laser pulse, and V_{nk}, the effective focal volume for a kth-order process, is given by

$$V_{nk} = \int F^{nk}(r)\,dV ,$$

where $F(r)$ is the normalized peak photon density.

In Table I, we compare the results for the

TABLE I. Values of k and W_k for multiphoton ionization of Xe and Hg.

		Bebb and Gold	Voronov and Delone	This work
Xe	k	7	6.23 ± 0.14	7.4 ± 0.8 (low power) 5.8 ± 0.4 (entire range)
	W_k	$10^{-205.5}$	$10^{-206.3 \pm 3.2}$	$10^{-203.7 \pm 2.8}$
Hg	k	6	···	6.3 ± 0.7 (low power)
	W_k	···	···	$10^{-171.5 \pm 2.3}$

theory of Bebb and Gold, the experiment of Voronov and Delone,[2] and this work. It should be pointed out that Bebb and Gold assumed the exciting radiation to be single mode. Spatial and temporal fluctuations related to the mode characteristics of a laser beam would give higher values for W_k. It has been pointed out that for thermal light, one should obtain a value of $k!\,W_k$ instead of W_k.[5,6] It would seem reasonable to assume that for a real laser beam, a value between W_k and $k!\,W_k$ should be obtained.

The value of k obtained for Xe by Voronov and Delone lies outside that obtained from our low-power data, but is consistent with a slope which could be obtained by using a larger portion of our curve. The higher peak flux used by Voronov and Delone was $\sim 10^{30.25 \pm 0.25}$ cm^{-2} sec^{-1}, and that, as can be seen from Fig. 1, corresponds within the region where saturation is important. It is also important to point out that in our work shorter pulses were used, and that this would have the effect of reducing saturation effects.

With Hg, the larger value of W_k enabled us to show the saturation effect to a much greater degree.

Using the values of W_k determined from this experiment and $k = 6$ and 7 for Hg and Xe, respectively, in Eq. (1), one finds that it fits nicely the experimental points within the error of the experiment (Fig. 1). This gives a further verification of the saturation effect.

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