this is because the percolation probability p(E) has been taken as unity, with the C(E) of  $EC \sim 0.5$ . The point of this note is to display the dependence of  $\mu$  on  $(\partial \rho / \partial P)_{T \approx T_{\rho}}$ . Equation (3) becomes

$$\mu = \mu_{c1} \cdot \frac{b'}{\delta} (kT)^{1/4} \int_0^\infty dy \, \frac{y^{1/2} e^{-y}}{y^{1/2} + b'} \bigg/ \int_0^\infty dy \, y^{3/4} \, e^{-y} ,$$
(4)

where  $b' = 4\pi c \rho \sigma (3\hbar^2/2mkT)^{1/2}$  and  $\delta$  is defined in Eq. (2). Equation (4) states  $\mu$  is proportional to  $(\partial \rho / \partial P)_T^{-1/2}$  [in contrast to Eq. (1)]. Thus, on ap-

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# Effect of Local Laser-Beam Intensity Fluctuations in Multiphoton Ionization\*

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Local laser-beam intensity fluctuations were observed and related to the scattering in the data of multiphoton ionization of atoms and molecules.

In the recent publications on multiphoton ionization of atoms reported by Chin, Isenor, and Young<sup>1</sup> and of molecules reported by Chin, <sup>2</sup> the reason for the rather large scattering in the experimental data on the log-log plots was not mentioned. We would like to point out that this was largely due to the local intensity fluctuation of the laser beam as is evident from the experiment reported here. The result of this work also gives further support of the same observation (i.e., local intensity fluctuations) recently reported by Abbi and Mahr.<sup>3</sup>

The method is similar to that of Abbi and Mahr<sup>3</sup> in that the time evolution of the power of the whole laser beam is compared with that of a small portion



FIG. 1. Experimental setup for observing local fluctuations in a laser beam.

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ments in neon could directly reveal information about the critical point. Since the average electron energy is  $\sim \overline{V}$ , the electron is a probe with a typical dimension  $\sim 30$  Å. The dip in  $\mu$  is analogous to the phenomenon of critical opalescence. Care will have to be taken in analyzing experimental data to ensure that any observed dips are due to critical-point fluctuations, and not to stable bubble formation.

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of the laser. However, instead of employing different laser shots in the comparison,<sup>3</sup> a simultaneous comparison of the whole beam and a portion of it was performed. The laser was the same one used by the author in several experiments on multiphoton ionization.<sup>1,2,4</sup> It consisted of a ruby oscillator Q switched by a saturable dye (cryptocyanine in methanol). The back reflector was a total reflecting prism while the output reflector was a resonant reflector so that a single axial mode was obtained.<sup>5</sup> The oscillator was followed by a ruby amplifier. This oscillator-amplifier system provided a peak power output of the order of 100 MW. The experimental setup is shown in Fig. 1. The steel plate S was placed in the focal plane of a 103-cm-focal-length lens.<sup>6</sup> S could be adjusted across the laser beam so that a small part of the beam could be selected via an opening of 1-mm diameter in S. The total beam and the beam transmitted through S were monitored by two fast photodiodes. The signals from the detectors were relatively delayed and fed into the same fast rise oscilloscope (Tektronix 519). Figure 2 shows some examples of the traces. For any position of S, the ratio r of the peak height of the transmitted pulse to that of the total pulse gives the normalized local intensity. S was placed at several arbitrary positions. A series of 16 laser pulses such as those shown in Fig. 2 were photographed for each fixed arbitrary position of S. It was found in some cases that

$$(r_{\rm max} - r_{\rm min})/r_{\rm min} \simeq 3$$
 (1)

This means that local fluctuation can be as high as a few hundred per cent. In the experiments of multiphoton ionization, <sup>1,2</sup> the laser flux was measured according to the smooth pulse of the whole laser beam. As a result, the effect of the local fluctuation could be such that the maximum local intensity would create either an unusually large number of ions or a local saturation, while the number created by the minimum local intensity could be much less than one. This can be seen from the following simplified example. Suppose that the laser beam had a uniform spatial distribution across the focal area except at two equal local areas where there were a maximum and a minimum  $% \left( {{{\mathbf{n}}_{{\mathbf{n}}}}_{{\mathbf{n}}}} \right)$ intensity, respectively. Let us assume further that the average of the two local fluctuations  $F_{max}$  and  $F_{\min}$  was equal to the measured uniform flux  $F_0$ across the whole focal area; i.e.,

$$F_0 = \frac{1}{2} (F_{\max} + F_{\min}) \quad . \tag{2}$$

If Eq. (1) was valid in this case, using the definition  $r_{\text{max}} = F_{\text{max}}/F_0$  and similarly for  $r_{\text{min}}$ , the two equations (1) and (2) combined would give

$$F_{\text{max}} \simeq 1.6 F_0$$
 and  $F_{\text{min}} \simeq 0.4 F_0$  .



TIME

FIG. 2. Typical oscilloscope traces (retraced) of the laser pulses of the total laser beam (right) and of the beam of the same laser pulse passing through a 1-mm opening (left).

Let us apply this to the case of six-photon ionization of mercury.<sup>1</sup> In the region where saturation was not important, a typical laser flux  $F_0$  at the focal region was around  $10^{29,30}$  ruby photons sec<sup>-1</sup> cm<sup>-2</sup>. The local flux  $F_{max}$  would then become  $10^{29,50}$  ruby photons sec<sup>-1</sup> cm<sup>-2</sup>. This value is around that of the flux at which saturation begins (Fig. 1, Ref. 1). On the other hand,  $F_{min}$  becomes  $10^{28,90}$  ruby photons sec<sup>-1</sup> cm<sup>-2</sup>. At this flux, the number of ions created is well below the detector sensitivity (less than 10 ions). Hence, the rather random local intensity fluctuation could be a major reason responsible for the rather large scattering in the data of multi-photon ionizations.

It is interesting to note that according to Eq. (1) of Ref. 1 (a detailed derivation of this equation was given in Ref. 7), only the effective focal volume  $V_k$  for a kth-order process depends on the spatial distribution of the laser flux, under the assumption that the laser flux  $F(\vec{\mathbf{r}}, t)$  can be separated into a function of space and one of time,

$$F(\mathbf{\vec{r}},t) = F_0 F(\mathbf{\vec{r}}) P(t) \quad ,$$

where  $F_0$  is the peak flux and

$$V_k = \int_{\text{focal volume}} F^k(\mathbf{\hat{r}}) d\mathbf{\hat{r}}$$
 .

Hence, spatial fluctuations would not affect the nonlinear dependence of the number of ions created N on  $F_0$ . That is, N would still be proportional to  $F_0^k$  in the case where there is no saturation. What would be changed would be the absolute value of N. Since the fluctuation is rather random, caused probably by the gain saturation in either or both the oscillator and amplifier ruby, <sup>8</sup> the mean slope k of the experimental plot of  $\log N$  vs  $\log F_0$ would not be altered, as long as a sufficiently large number of measurements were taken. This was verified by the results of multiphoton ionization of mercury and xenon<sup>1</sup> in which there were about a hundred experimental points in each plot.

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<sup>†</sup>Experiment done at the Department of Physics, University of Waterloo, Ontario, Canada. The results were reported in the Ph.D. thesis submitted by the author to the University of Waterloo, 1969.

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<sup>3</sup>Satish C. Abbi and Herbert Mahr, Phys. Rev. Letters 26, 604 (1971).

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<sup>5</sup>M. Hercher, Appl. Phys. Letters 7, 39 (1965).

<sup>6</sup>The purpose of using a long-focal-length lens is to scale up the small focal area of the 4-cm-focal-length lens used in the multiphoton ionization experiments (see Refs. 1, 2, and 4), so that a direct investigation of fluctuation at the focal region is experimentally more convenient.

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## ERRATA

## Distribution of Excitation Transfer in Helium.

John D. Jobe and Robert M. St. John [Phys. Rev. A 5, 295 (1972)]. The correct form of Table VI is at the right.

M-Shell Auger and Coster-Kronig Electron Spectra, Eugene J. McGuire [Phys. Rev. A 5, 1052 (1972)]. The correct date of receipt of this paper is 20 October 1971.

TABLE VI. Distribution of excitation transfer gain  $Q^*(j)$  among various levels at 63 mTorr and 100 eV. Significant cascade terms  $Q_c(j)$  are given in parenthesis. Units are  $10^{-20}$  cm<sup>2</sup>.

n j	$n^{1}P$	nD	nF	$\geq nG$
3ª	0	0(101)		
4	- 55	28(24)	35(29)	
5	-47	25(4)	20(12)	$2 \pm 15$
6	- 33	14(1)	6.2(3)	$13 \pm 8$
7	-21	7.8	3.2(1)	$10 \pm 5$
8	-14	3.7	1.2	$9.1 \pm 3.5$
9	- 9	2.0	0.9	$6.1 \pm 2.7$
10	-7	0.8	0.5	$5.7 \pm 2.5$
>10 <sup>b</sup>	- 30	0.5	0.5	$29 \pm 12$

<sup>a</sup>Transfer on the n=3 level is not a significant part of the apparent cross sections involved and is zero to within experimental error.

<sup>b</sup>Extrapolated values.