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Multiphoton ionization of aluminum and copper planar target

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The total charge of the emitted ions from the planar metal targets irradiated by the mode-locked ruby laser was measured. The dependence of the total emitted ions on the laser fluence is consistent with the theory of the multiphoton ionization in the low fluence region. However, it shows saturation phenomena at the higher fluence regions. These saturations could be explained by the depletion of the neutral atoms and the level shifting of the intermediate state due to the strong Stark effect.

Multiphoton ionization by an intense laser radiation has been studied theoretically by Bebb and Gold¹ in rare gases and by Silin² in solid, and experimentally by several authors.³⁻⁶ The experimental results indicate that the ionization probability is proportional to the flux raised to a power somewhat less than that predicted by theories. There have been three different explanations for this deviation. First, it was attributed to the depletion of the neutral atoms by the strong radiation.⁴ Later, it was suggested that, in the strong electric field of the laser radiation, the Stark shift of the intermediate state lowered the ionization potential.⁵ Recently, it was suggested that the emitted charges screened the sample from the bias field and thus suppressed the photoemission.⁷ The present letter gives new experimental results on the multiphoton ionization of aluminum and copper solid that support the first explanation. However, the new local saturation effect in the intermediate fluence region seen in this experiment could be consistent with the second explanation.

A passively mode-locked ruby laser pulse has typically a 50-ns *Q*-switched envelope and consists of 10 ps pulses evenly separated by 7 ns. This laser beam of 2 mm diameter is focused by a lens ($f = 50$ mm) to 30 μm diameter on an Al or a Cu target placed in 10^{-3} Torr vacuum. The incident laser energy is controlled by a couple of thin-film polarizers. The emitted ions are collected by a diode configuration, which consists of a copper collector cup and a sample holder. The collector has a hole 5 mm diameter to pass the incident beam. A bias voltage of 500 V was applied on the sample and the collector was grounded through the load resistance. The space between the collector and the sample was 10 mm. In preparation for the experiment, samples were submerged in the etching solution for a minute in order to reduce the generation of the secondary electrons on the collector by the reflected beam.

The time-integrated signal of the emitted ion is the total charge of the emitted ions. This signal was amplified by two Canberra model 816 spectroscopy amplifiers each with gain up to 64×8 and then fed into the computer data-acquisition system through an analog-to-digital converter. Simultaneously the laser energy was monitored by a fast vacuum photodiode.

The log-log plots of the total charge of the emitted ions versus laser fluence are shown in Fig. 1 for the Al sample and

in Fig. 2 for the Cu sample. They show three different regions of fluence. In the low fluence region (up to 15 J/cm^2 for Al, 8 J/cm^2 for Cu), the data can be linearly fitted by the least-square method. The fitted slope is 3.44 ± 0.28 for Al and 5.02 ± 0.42 for Cu. In the intermediate fluence region (up to 80 J/cm^2 for Al, 50 J/cm^2 for Cu) the linear fitting is not adequate because of the local saturation. As clearly seen in the figures, the local saturation is especially distinctive for Cu. In the high fluence region the slopes gradually decrease as the fluence increases and this indicates the saturation phenomena.

In the low fluence region, it can be assumed that very small fractions of the evaporated neutral atoms are ionized during the irradiation of the laser pulses. Thus one would expect the total number of ions created per shot to be propor-

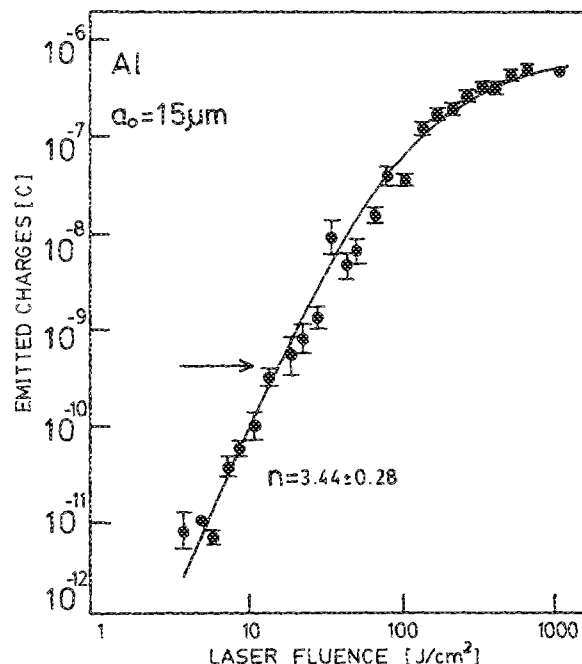


FIG. 1. Dependence of charge amount of the emitted ions on the laser fluence for Al. The solid line is the theoretical curve for $A = 4.49 \times 10^{-34} \text{ cm}^6 \text{ s}^3 \text{ J}^{-4}$ for the value of the evaporated neutral atom density 10^{19} cm^{-3} . The slope n is obtained by the linear fitting in the range below the arrow.

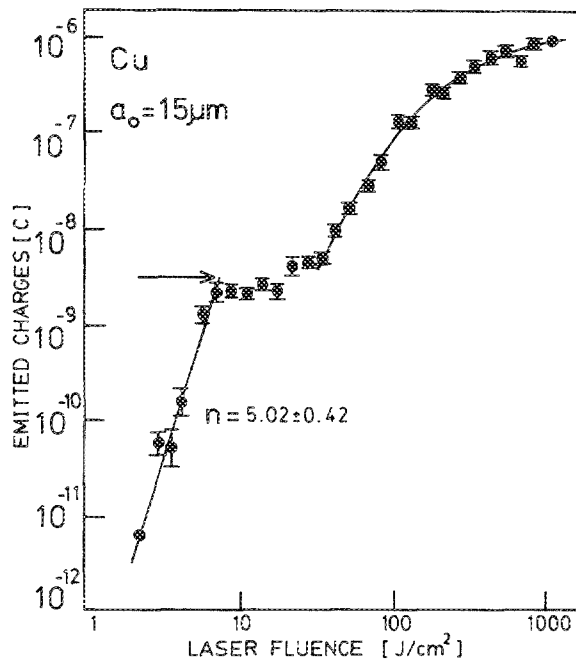


FIG. 2. Dependence of charge amount of the emitted ions on the laser fluence for Cu. The solid line in the low-power region is a linearly fitted line for $A = 9.58 \times 10^{-47} \text{ cm}^{10} \text{ s}^4 \text{ J}^{-5}$ and in the high-power region the line is the theoretical curve for $A = 1.48 \times 10^{-49} \text{ cm}^{10} \text{ s}^4 \text{ J}^{-5}$ for the value of the density of 10^{19} cm^{-3} . The slope n is obtained by the linear fitting in the range below the arrow.

tional to the ionization rate. The ionization rate is proportional to the n th power of the average laser fluence, where n is the number of photon quanta absorbed during the ionization. The number n must be equal to $\langle I/\hbar\omega + 1 \rangle_{\text{int}}$, where I is the ionization potential of the atom and $\hbar\omega$ is the laser quanta energy. Since the ionization potential is 5.985 eV for Al and 7.726 eV for Cu, n is 4 for Al and 5 for Cu at the ruby quanta energy ($\hbar\omega = 1.785 \text{ eV}$). In the case of Cu, the experimental number 5.02 agrees with the theoretical value. However, the experimental number 3.44 for Al is lower than the theoretical value of 4. The reason for this discrepancy can be explained by the assumption that the quasiresonating or resonating state of the integral multiple of the photon quanta energy contributes to lower the ionization potential of the atom.⁸ In the Al atom, the resonance level may be the $6^2P_{3/2}$, whose energy, relative to the ground state, differs by 15 meV from the energy of three ruby quanta.

In the intermediate fluence region, the stationary state which is close to the integral multiple of photon quanta is overshifted by the Stark effect in the strong electric field of the incident laser ($E \sim 2 \times 10^7 \text{ V/cm}$). Thus the cross section of the multiphoton ionization is degraded, and the local saturation results from this degradation. The detailed calculation of the level shifting due to the Stark effect has not been performed theoretically except for the hydrogenlike atoms. In the potassium atom case, $4f$ level shift is about 1000 cm^{-1} at $2 \times 10^7 \text{ V/cm}$ electric field.⁹ The local saturation is more significant for Cu, that is, in the nonresonating case in common with the result of Delone *et al.*⁵

In the high fluence region, the density of ions becomes appreciable compared with the evaporated neutral atom

density. Thus we would expect the slope to be less than n . With this saturation effect taken into account, we make a rate equation of N_i ,

$$\frac{dN_i}{dt} = \beta(N_0 - N_i) - \frac{N_i}{\tau_R}; \quad \beta = AF^n(r,t), \quad (1)$$

where N_i is the density of the ions, N_0 is the neutral atom density, β is the multiphoton ionization probability, F is the laser fluence, and τ_R is the recombination time to the neutral atom. Since the light pulses are very short (10 ps), we neglect the cascade ionization in this equation. We solve the rate equation under the assumption that the mode-locked pulses have seven equal rectangular pulses with 10 ps width and are separated by 7 ns. Since the interpulse separation time (7 ns) is very short compared with the recombination time which is approximately 500 ns, the decay during the interpulse separation may be neglected. We approximate the converging beam as a cone and spatially integrate the solution of the rate equation assuming a Gaussian intensity profile.¹⁰ Then, the total number of ions is given by

$$(N_i)_{\text{tot}} = N_0 \frac{\pi a_0^3}{n} \frac{2f}{D} \sum_{k=1}^{\infty} \frac{(-1)^{k+1} [7A(F_0/7t_p)^n t_p]^k}{(2nk-3)kk!}, \quad (2)$$

where a_0 is the focusing radius at the target surface, F_0 is the laser fluence at the focal point, f is the focal length of the lens, D is the incident beam diameter, and t_p is the width of the light pulse (10 ps).

In Eq. (2) $(N_i)_{\text{tot}}$ is proportional to F_0^n in the low fluence. However, it shows the tendency to saturation as the fluence increases. The asymptotic form of Eq. (2) is

$$(N_i)_{\text{tot}} \approx N_0 \frac{\pi a_0^3}{n} \frac{2f}{D} \frac{1}{3} \ln \left[7A \left(\frac{F_0}{7t_p} \right)^n t_p \right]. \quad (3)$$

For Al, the experimental data can be fitted by Eqs. (2) and (3) in the whole fluence range, and the fitted value of A is $4.49 \times 10^{-34} \text{ cm}^8 \text{ s}^3 \text{ J}^{-4}$ for the value of the evaporated neutral atom density 10^{19} cm^{-3} . For Cu, the above fitting scheme is not adequate because of the local saturation by the strong Stark effect in the intermediate fluence region. Therefore, the data in the low fluence region were linearly fitted with $A = 9.58 \times 10^{-47} \text{ cm}^{10} \text{ s}^4 \text{ J}^{-5}$, and the data in the high fluence region were fitted by Eqs. (2) and (3) with $A = 1.48 \times 10^{-49} \text{ cm}^{10} \text{ s}^4 \text{ J}^{-5}$ for the value of the evaporated neutral atom density 10^{19} cm^{-3} . We deduce the multiphoton ionization cross section from these values of A . For Al the cross section of four-photon ionization is $10^{-97.14} \text{ cm}^8 \text{ s}^3$. For Cu the cross section of five-photon ionization is $10^{-139.11} \text{ cm}^{10} \text{ s}^4$ in low fluence and $10^{-141.9} \text{ cm}^{10} \text{ s}^4$ in high fluence. It shows that the multiphoton ionization cross section is degraded by the order of 3 due to the strong Stark effect.

One may indicate that the density of the evaporated atoms increases as the fluence increases in contrast to the constant N_0 in the case of gases. But with the evaporated neutral atom, the debris of metal is ejected in the form of

small fragments. This phenomenon is easily seen in crystalline Si, and the debris with high momentum etches the Si surface in all directions. Since such debris is not ionized, the density of the neutral atoms may increase very slowly. Furthermore, in the log-log plot of N_i vs F_0 , the tendency of N_i is not affected appreciably by this slow increase.

In conclusion, our measurement of the total charge of the emitted ions shows three different regions of fluence. In the low fluence region, the ionization probability is proportional to F_0^n . In the intermediate fluence region, the local saturation becomes significant due to the Stark effect. The cross section of five-photon ionization is degraded by the order of 3 for Cu. In the high fluence region, the large depletion of the neutral atoms leads to saturation of the ion generation.

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