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Energy spectrum of photoelectrons produced by picosecond laser-induced surface multiphoton photoeffect

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Photoelectrons were produced from a gold surface using strong picosecond Nd:glass laser pulses of 10^{10}-W/cm^2 intensity. Unexpectedly high (up to 600 eV) energies were observed in the electron energy spectrum only when space-charge effects were completely eliminated. This result is compared with the well-known electron energy spectra of the multiphoton ionization of gases.

Multiphoton light-matter interaction processes are analogous in the cases of individual atoms (ionization of gases) and solid targets (photoelectric effect of metals), respectively.[1,2] Good agreement with theories for both gases and metals was found for nearly all characteristics of the multiphoton processes (e.g., intensity dependence, polarization dependence, wavelength dependence, etc.). However, in spite of the interesting new results found recently for the energy spectrum of the electrons appearing in the ionization process of the atom, which is called "above threshold ionization" (ATI),[3,4] less attention has been paid to the energies of the photoelectrons emitted from the metal. Even the only two early experimental observations are in contradiction: One showed[5] the existence of electrons with higher energy than $E=n_0h\nu-A$ (n_0 is the minimum number of photons necessary to overcome the work function of the metal A and $h\nu$ is the quantum energy of light); the other[6] gave the electron-energy distribution corresponding to the above-mentioned "multiphoton Einstein equation."

The studies of electron-energy spectra initiated a new experimental activity in the field of multiphoton ionization of gases.[7] Very detailed experimental[8-11] and theoretical[12-14] investigations of these spectra revealed a series of new basic physical phenomena: ATI,[3,4,8-10] ponderomotive acceleration of electrons,[9] the role of the ac Stark shift,[11] etc.; however, each of them is for the case of gases. The following obvious questions arise: Does ATI really exist in the case of the photoeffect of metals? What is the reason for the mentioned disagreement?[5,6] And, in general, how does the electron-energy distribution depend on the various parameters of the interaction?

Therefore, in our recent experiments we analyzed the kinetic energies of photoelectrons produced by high-intensity laser light pulses on a metal (gold) surface. We have found in a preliminary work[15] unexpectedly high —up to 600 eV—electron energies, and in this Rapid Communication we describe our experiment and report in detail on the results obtained.

Meanwhile, a further, interesting piece of experimental work has been published on the subject in question[16] which, as we shall see below, has partly confirmed our results and partly disagreed with them—probably due to the different experimental arrangement used.

In our experimental work we also bore in mind the above-mentioned earlier experimental evidences, relating to the energies of the photoelectrons from the multiphoton photoeffect of metals: At low ruby-laser intensities (<10 MW/cm^2) and using various metal targets (Au, stainless steel), no electrons with energy higher than $E=n_0h\nu-A$ were found;[6] at somewhat higher ruby-laser intensities (50–100 MW/cm^2) and with a Ag target, electrons having about 10-eV energy were observed.[5] It should be stressed here that the key point of the undisturbed observation in all laser-induced metallic photoeffect experiments is the complete elimination of the space-charge effect caused by the high photocurrent densities, as has been observed in many experiments[17,18] and confirmed theoretically.[19] In the cases of Refs. 5 and 6 these inherent space-charge effects were not completely eliminated, which might wash out the effects to be observed at high intensities, even if they existed. The same seems to be partly true for the experiment described in Ref. 16.

Therefore, our aim was to determine the energies of the multiphoton photoelectrons emitted from metals at high laser intensities in the absence of space charge. To realize this, the basic features of our experiment were the following: application of high intensity (≤ 25 GW/cm^2), ultrashort (8 psec) Nd-laser pulses ($h\nu=1.17$ eV); use of the surface photoeffect of gold as the multiphoton interaction process, i.e., using grazing incidence and p polarization of light to ensure the exclusion of the volume photoeffect and thermal electron emission;[20] application of high (>10 kV/cm) extracting electrostatic fields to eliminate the space-charge effect; use of an appropriate form of energy analysis (retarding field method with meshes, small solid angle for electron collection, and an electromultiplier for detection, as suggested in Refs. 21 and 22).

The experimental setup is shown in Fig. 1. Single-bandwidth-limited pulses (8 psec, 1 mJ, 1-Hz repetition rate) of a passive mode-locked Nd:phosphate glass oscillator-amplifier laser system were directed onto a properly polished and cleaned[23] thick polycrystalline gold plate T (reflectivity $\sim$99%) at grazing incidence $\sim$85°. After slight focusing the maximum light intensity at the metal surface was of $I_0=25$ GW/cm^2 related to the cross section perpendicular to the beam axis. The electric field of the laser was p polarized, i.e., perpendicular to the gold surface. In front of the gold plate a mesh M, kept at +15 kV extracting potential (V_{ext}) was placed at a distance of 15 mm to eliminate the space charge. Behind this mesh a

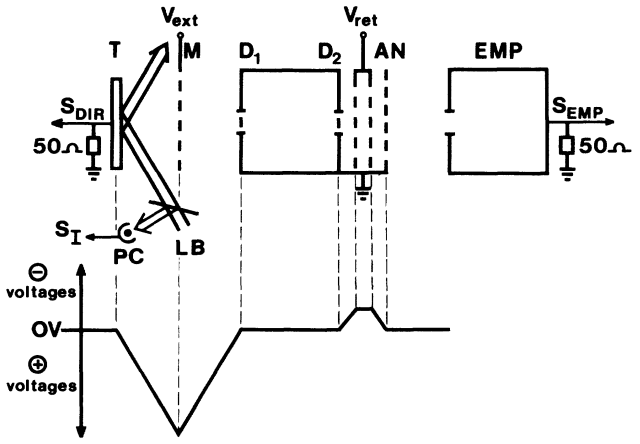

FIG. 1. Experimental setup: laser beam (LB), monitoring photocell (PC), gold target (T), accelerating mesh (M), diaphragms with diameter of 3 mm ($D1$, $D2$), retarding field analyzer (AN), and electron multiplier (EMP). The T-M distance is 15 mm. The curve at the bottom shows the variation of the electrostatic potential along the electron path to the electron multiplier.

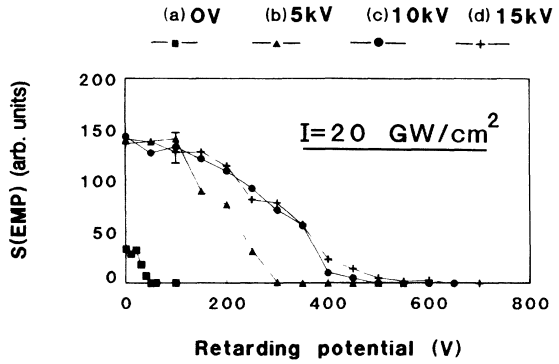

FIG. 2. Integral electron-energy distribution curves at various V_{ext} extracting electric potential values: (a) $V_{ext}=0$ V, (b) $V_{ext}=5$ kV, (c) $V_{ext}=10$ kV, (d) $V_{ext}=15$ kV. S(EMP) is the electron multiplier signal. The laser intensity is ~ 20 GW/cm^2.

usual retarding field energy analyzer system (AN) with 0.01 sr observing solid angle—determined by diaphragms $D1$ and $D2$—was used. The entrance diaphragm D1 of the analyzer was kept at the same potential (i.e., ground) as the gold target to reobtain the original, space-charge free-electron energy spectrum. In this way the accelerating high-voltage electrode eliminates the space-charge effect only, and it has no effect on the electron-energy distribution.

The incoming laser intensity I was monitored by the vacuum photocell (PC) giving signal S_I. The total number of electrons emitted from the gold plate was measured directly on a 50-Ω load resistance of an oscilloscope (signal S_{DIR}), while the analyzed electrons were detected by an electron multiplier (EMP) (signal S_{EMP}). The overall interaction system was situated in a vacuum vessel pumped down to 5×10^{-8} mbar by a turbomolecular pump.

To obtain the electron-energy distributions we measured the multiplier signal S_{EMP} as a function of the retarding potential V_{ret} (it was varied between 0 and -1000 V), which gave the integral energy distribution. Owing to the considerable fluctuation of our laser (signal S_I), energy resolution with a precision of $\Delta V_{ret}\sim h\nu$ (i.e., precision of ATI peaks) was not possible; only the distribution envelope was measured. To prove that the space-charge effect seriously alters the electron-energy distribution up to its complete elimination, this procedure was repeated at various V_{ext} values (Fig. 2). The energy distribution curves changed in a very sensitive way with V_{ext} below ~ 10 kV, while above this value no change was observed: This feature really shows the key role of the elimination of the space charge.

There exist two further proofs of the elimination of the space-charge effect by V_{ext}. The first one, which has been performed traditionally by us and others[2,5,6] in the course of multiphoton experiments is the following. The measured dependence of the total emitted electron current S_{DIR} on the light intensity I must follow the theoretical I^{n_0} power function law in the intensity range (<10 GW/cm^2) where the perturbative approach is valid;[20–24] however, only in the space-charge-free case. For our gold plate[25] $A=4.679$ and $h\nu=1.17$ eV for the Nd-laser light, in our case the theoretical $n_0=[A/h\nu+1]_{integer}=4$, and we measured $n=n_0=4$ only above $V_{ext}\sim 10$ kV (i.e., space charge eliminated) and $n<n_0=4$ for $V_{ext}<10$ kV (influenced by space charge). The second one is the comparison of the actually measured maximum current density $j_{max}\sim 60$ A/cm^2 (being the total emitted charge $\sim 2\times10^{-12}$ C and the laser spot size on the surface ~ 3.2 mm^2), and the theoretically predicted (e.g., Ref. 19) space-charge-limited current density j_{lim}, which is 75 A/cm^2 for our case. In the course of the experiments the condition $j_{max}<j_{lim}$ was always ensured.

After these checks we measured the integral electron-energy distributions at $V_{ext}=15$ kV. As for the laser intensity range used, we had to bear in mind that the electron number arriving to the multiplier through the analyzer was about 10^{-3} times lower than the total emitted electron number. Therefore, to obtain detectable signals even at high V_{ret} voltages we were obliged to apply higher incident light intensities of 13–25 GW/cm^2; such intensities are higher than the mentioned <10-GW/cm^2 values for which the perturbative approach exactly holds. On the other hand, special care was taken to avoid any kind of heating, melting, or plasma creation at the surface, which began to appear above 30 GW/cm^2. The results are summarized in Fig. 3. The scattering of the experimental points, caused by the laser instability, did not allow straightforward numerical differentiation of these integral distributions to obtain distribution maxima. However, it is clear that for each distribution an interval can be found around a certain $V_{ret,0}$ value in which the decrease of the electron number is significantly stronger than in the other regions (i.e., a maximum corresponds to it in the differential electron-energy distribution at $E_0=eV_{ret,0}$). Table I summarizes the energy values E_0 around which the maximum electron number was created. It can clearly be seen that E_0 increases monotonically with the laser intensity I up to as high as 600 eV at $I=25$ GW/cm^2.

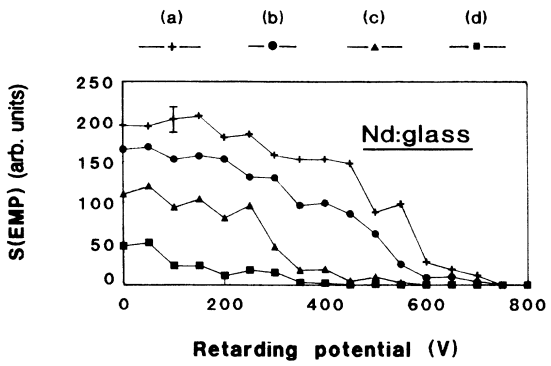

FIG. 3. Integral electron-energy distribution curves obtained with 8-psec Nd:glass laser pulses at various laser intensities: (a) $I=25$ GW/cm^2, (b) $I=22$ GW/cm^2, (c) $I=18$ GW/cm^2, (d) $I=13$ GW/cm^2. The extracting electric potential $V_{ext}=15$ kV.

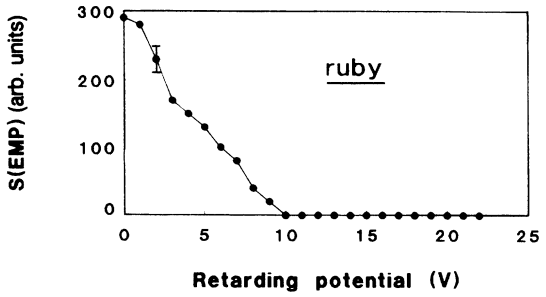

FIG. 4. Integral electron-energy distribution curves obtained with 20-nsec, low-intensity (2 MW/cm^2) ruby-laser pulses. The extracting electric potential $V_{ext}=15$ kV. Note the much lower energy scale on the horizontal axis compared with Fig. 3.

In the course of these experiments we noticed that the characteristic slope values $n'=\partial\log(S_{EMP})/\partial(\log I)$ (derived from the $I^{n'}$ power form) showed a continuous increase up to values as high as $n'\sim 15$ when we increased V_{ret} up to the detection limit of $S_{EMP}\sim 0$.

In order to be sure that the high-energy electrons came only from the pure interaction processes of the laser light and the metallic electrons, we wanted to exclude any kind of artifacts which might also create electrons with the same energies, e.g., from the meshes kept at 15-kV voltage (producing secondary electrons, photons, etc.). For this purpose we repeated the measurements with the same electric conditions ($V_{ext}=+15$ kV, $V_{ret}=0$–1000 V), but instead of the psec Nd-laser system we used a giant pulse ruby laser, of which the pulse duration, the pulse energy, and the intensity could easily be varied. In this way the same electron number was achieved with the following parameters: $h\nu=1.78$ eV, the multiphoton order $n_0=3$, the pulse duration 20 nsec, the intensity $I\leq 2$ MW/cm^2, which is 4 orders of magnitude less than in the case of Nd laser. The result, represented in Fig. 4, shows less than 10-eV energies for the emitted electrons (and $S_{EMP}\propto I^3$ intensity dependence, i.e., the interaction was pure multiphotonic). This proves that none of the mentioned side effects of the high-voltage electrodes occurred. (Similar results supported this check: $\sim$10-μsec normal spikes of the ruby laser were used, whereupon long thermionic electron pulses were created with even lower electron energies.)

If we compare these results with those mentioned earlier[6] where the intensity was the same and the space charge was partly eliminated, we can state that the observed effect corresponds to the lowest-order perturbative interaction. As for the other[5] early ruby experiment at $I\sim 100$ MW/cm^2, in spite of the inherent presence of the space charge—due to the simple two parallel cathode-anode system used—$\sim$10-eV electron energies were found. This fact also supports our present results obtained in the high-intensity range.

TABLE I. Energy values around which the photoelectron energy distribution has its maximum.

Laser intensity, I (GW/cm^2)	E_0 (eV)
13	100
18	300
20	350
22	500
25	600

In conclusion, we measured the photoelectron energy distribution induced by the multiphoton photoeffect of gold. High-energy electrons (up to 600 eV) (appeared even at unexpectedly low laser intensities (in contrast to the case of atoms[22]). Our results strongly differ from those obtained recently also for metals,[16] where the energies of the emitted electrons fell in the region of 0–10 eV, showing ATI-like structure. The same low-energy spectrum, apart from the 1-eV resolution, was observed at similar light intensities in our measurements also, when the space charge was not eliminated (see the curve at the bottom of Fig. 1, $V_{ext}=0$). Our apparatus was not able to resolve peaks even if they existed.

The theoretical interpretation of our results is not yet obvious. The basic nonperturbative theories[26,27] and their variants, particularly applied to metals,[24,28] do imply the existence of ATI. However, these theories give undetectable low yield for the ATI peaks with respect to the n_0th-order "normal" multiphoton current. For example, the ratio of the yields corresponding to the first- and zeroth-order ATI peaks is 10^{-4} at $I=2.5\times 10^{10}$ W/cm^2. The other possible effect, the ponderomotive potential,[9,22] $\Delta=e^2E^2/4m\omega^2$ would give about 2 meV in our case. The laser-induced thermal effects[29] would have produced energies less than the order of eV; however, they were completely eliminated in our experiment by using grazing incidence, psec pulses, etc.

Because the observed effect cannot be explained by the simple application of the theories mentioned here, further generalization seems to be necessary to take into account such fundamental facts as the macroscopic character of the metal as a third body, the handling of the interaction in nondipole approximation (due to the grazing incidence), and the dynamic population of the infinite number of empty states of the metal between the Fermi and vacuum levels.

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