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# Observation of multiple-harmonic radiation induced from a gold surface by picosecond neodymium-doped yttrium aluminum garnet laser pulses

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Illuminating a gold surface by strong (5 GW/cm$^2$) picosecond neodymium-doped yttrium aluminum garnet laser pulses at grazing incidence, we observed generation of coherent beams of both even and odd harmonics up to fifth order in the reflected direction with efficiencies $10^{-10}$–$10^{-13}$. The observed decrease of the harmonic efficiencies with increasing harmonic order is much weaker than predicted by perturbative theories.

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Great successes have been achieved in the understanding of the newly discovered production of laser-induced very-high-order harmonics in rare-gas atoms [1–5], exhibiting spectacular characteristics at high laser intensities: extremely wide spectra consisting of a high number of odd harmonics up to as high as 53, with a wide plateau region terminated with a sharp cutoff. This phenomenon is governed by the fundamental laws of the intense field QED, therefore the obvious question arises: Do these fundamental laws produce analogous phenomena for solids characterized by the rectangular-well model potential? The general aim of this work is to try to answer this question experimentally using metal surface as a model.

Multiple harmonic generation may be treated generally by both perturbative (at low laser intensities) and nonperturbative theoretical methods (at arbitrary intensities). Both treatments are elaborated exhaustively and correctly for atoms [3,4]. The situation is different for metals [6–9]. As for the perturbation (low-intensity) approach, all works dealing with metal surfaces are restricted to low-order (second, third) harmonic generation only. The procedure followed in these calculations was to determine the corresponding order current induced by the incoming laser as the light-emitting source term either by classical or quantum (low-order perturbation) nonlinear phenomenology (for a summary, see the references in Refs. [7,9,10]). Then the emitted harmonic field in question is obtained by a coherent summation of all these currents over the manifold of electrons of the illuminated area of the metal surface and applying the Maxwell equations. Although the experimental verifications of these predictions are quite satisfactory, these early theories and experiments [6,7] may not be relevant to answer our general question exposed above.

As for the very-high-intensity range, the nonperturbative generalization of the interaction process in question up to arbitrary high intensity and harmonic order has been published only recently [10–12]. The work [10] tackles the problem as a single-particle-quantum-mechanical process, by using a rectangular single-step model potential. Consequently, it cannot be applied directly to a realistic experiment, considering that a single-particle treatment cannot furnish macroscopic quantities as, e.g., the correct (specular) harmonic emission angle, which has to be determined by the collective coherent contributions of all electrons. Nevertheless, it does predict the appearance of both even and odd harmonics as the consequence of very general principles relating to these kind of models [10–12].

Therefore it seemed to be reasonable to perform experiments for the eventual analogy between the multiple harmonic generation in gas atoms and metal surfaces.

In the present work we report high-order harmonics (up to the fifth order, both odd and even) from a gold surface; gold was chosen as an appropriate metal to realize the rectangular-model potential case.

The experimental arrangement is shown in Fig. 1. Linearly polarized, single, bandwidth-limited pulses were selected from the leading part of an actively mode-locked pulse train of a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser (L) (Quantel YG501C, pulse duration, ~35 psec; intensity, ~5 GW/cm$^2$ in the cross section of the beam; repetition rate 10 Hz; wavelength, 1.064 $\mu$m). The light beam was directed through the slit (D)

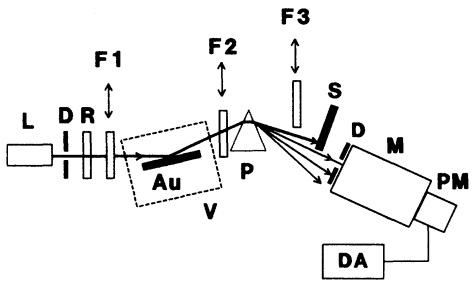


FIG. 1. Experimental arrangement. L: passive mode-locked Nd:YAG laser; D: diaphragms; R: polarization rotator, $\lambda/2$ plate; F1,2,3: spectral filters; Au: polished gold target; V: vacuum vessel; P: quartz prism; S: beam stop for blocking the fundamental; M: monochromator; PM: photomultiplier; DA: data acquisition system.

 

and a polarization rotator (R) onto the appropriately polished and treated [13] surface of a 2-mm-thick polycrystalline gold sample (Au) kept under $\sim 10^{-7}$ torr in a vacuum vessel (V). A grazing incidence of 70° was used—at which the laser intensity was roughly $5\cos(70°) \approx 2$ GW/cm$^2$ on the surface—to prevent the heating and plasma effects and to favor the predicted [6] surface excitations. (The sample was prepared in the same traditional way as in all our previous multiphoton electron emission experiments [13–19].) The reflected light passed through a quartz prism (P) which deviated and separated the direction of the different harmonics. The first one of them, i.e., the high-intensity fundamental laser light was blocked by the stop (S). Then, by turning the prism P in the appropriate direction, the further harmonic light components entered the slit (D) of a monochromator (M) (capable of measuring down to 200 nm) one by one and were detected by a photomultiplier (PM). The spectrum was taken by the wavelength scan of the monochromator M simultaneously rotating the prism P into the appropriate direction. The most crucial disturbing effects which may occur in this type of experiment [7] had been controlled and taken into account. For example, the possible hazard lights which might be induced in elements other than the gold surface (diaphragms, windows, prism, etc.) were filtered and completely eliminated by inserting different density color filters (F1, F2, F3) in appropriate combinations between the different elements of the setup. A further similar control was made realizing the configuration, in which the light beam passed the whole setup without touching the gold surface. The signal of the photomultiplier was analyzed by a data processing system (DA) containing a boxcar average and an acquisition and detecting setup.

In the following we summarize the experimental results. Figure 2 shows that the second, third, fourth, and fifth harmonics, which were emitted in the specularly reflected laser beam in solid angles roughly equal to the 0.7-mrad laser divergency, appeared at the correct expected harmonic wavelengths, i.e., at 533, 355, 266, and 213 nm within the ~1-nm instrumental resolution of the monochromator. The amplitude values of the spectrum lines present immediate records only. The real relative values of them were calculated taking into account the overall spectral response of the whole measuring system and are presented in Table I. The appearances of the laser monitor pulses and those of the harmonic light pulses were simultaneous—within the 5-nsec time resolution of our detection system. Special care was taken during the whole experiment to avoid any plasma formation on the surface by keeping the laser intensity value lower than 10 Gw/cm$^2$ in the beam cross section, i.e., $I < 3$ GW/cm$^2$ on the surface. On the other hand, in contrast with those other previous works, in which long pulses (nanosecond) or long pulse trains of picosecond pulses were used (which may cause accumulation heating effects), the plasma creation threshold in our case was sufficiently increased by applying single, selected, transform limited picosecond pulses.

As can be seen in Fig. 2, the second and the third harmonic lines appeared in a clear form, without any additional lines or considerable background in this part (700–300 nm) of the spectrum. To detect the weaker fourth and fifth harmonics, we had to increase the detection sensitivity in the 300–200 nm spectral range. This is shown in the right side of Fig. 2, where we see the fourth and fifth harmonic lines in the somewhat increased background noise level. Besides the harmonic lines, with less reproducibility, sharp additional weaker lines appeared also in time coincidence with the laser pulse, however, only in this latter shorter wavelength spectral range (*A* and *B* in Fig. 2). They correspond to the $5d^{10}6p_{3/2} \rightarrow 5d^{10}6s_{1/2}$, and $5d^9 6s6p_{5/2} \rightarrow 5d^9 6s^2_{3/2}$ transitions of the neutral Au atom, as it was identified from table data [20]. In contrast to the similar appearance of additional lines in noble-gas experiments which are always performed in inherent plasma background [2,3], here any plasma process can be excluded as a possible origin.

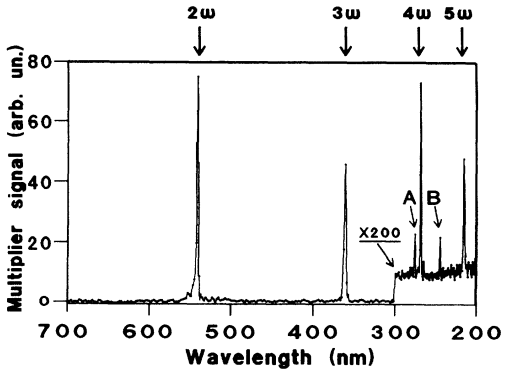


FIG. 2. Spectrum of the light emitted from the gold surface illuminated by picosecond Nd:YAG laser pulses at $I \sim 5$ GW/cm$^2$ intensity. Shown are the second, third, fourth, and fifth harmonics and the satellite lines *A*,*B*. (Direct readings without the overall spectral response calibration.) Note the enhanced sensitivity of the detecting system in the 300–200-nm wavelength region.

Our experimental setup being roughly the same as used in our multiphoton electron emission experiments [17–19], the absence of plasma and any surface damage might be routinely checked (as we do it always [13–18]) by observing the multiphoton photocurrent, which is sensitive in high order for any disturbing effects. In any case, the appearance of the weak *A*,*B* lines is irrelevant from the point of view of our original objective.

The observation of higher than fifth order was not possi-

TABLE I. Experimental exponents of the power dependences and relative intensities of the harmonic lines found experimentally by us and predicted (Ref. [10]).

| Harmonic order, $n$ | $n_{expt}$ | Relative intensities Expt. | Ref. [10] |
|---|---|---|---|
| 2 | $1.8 \pm 0.2$ | 1 | 1 |
| 3 | $3.2 \pm 0.2$ | $5 \times 10^{-1}$ | $10^{-5}$ |
| 4 | $4.1 \pm 0.2$ | $5 \times 10^{-3}$ | $10^{-8}$ |
| 5 | $4.7 \pm 0.5$ | $10^{-3}$ | $10^{-10}$ |

ble with the monochromator available in this experiment.

The maximum harmonic signal was obtained at the $p$ polarization of the incoming laser light. The produced harmonics also were always $p$ polarized. (On the contrary, the light of the satellite lines was randomly polarized.) Measuring the yields of each harmonic as a function of the laser polarization we obtained an unambiguous proof that only the laser electric field component $E_{\perp}(\omega)$, which is perpendicular to the gold surface causes the effect. This fact proves that the effect is predominantly of surface origin, i.e., it comes from the laser-induced nonlinear current of surface electrons [6,7,10]. We measured the laser light polarization dependence curves of all detected harmonics by changing the angle $\phi$ between the laser field strength and the plane of incidence. Figure 3 shows a typical polarization dependence curve for the third harmonic ($n=3$) and the expected $(\cos^2\phi)^3$ curve with the fitted $n_{\text{exp}}=3.2\pm0.2$ value. The measured curves for $n=2,3,4,5$ correspond to the expected $\cos^{2n}\phi$ dependences. (This polarization measurement furnished a basic control: In that geometrical configuration, in which the laser beam did not touch the gold surface, no effect was found.)

From the polarization dependence we estimated that the harmonic intensity $I(n\omega)$ depended on the incoming laser intensity $I(\omega)\propto|E_{\perp}(\omega)|^2$ in the form of $n$th order power law, i.e., as $I(n\omega)\propto I^n(\omega)$ or $|E(n\omega)|^2\propto|E_{\perp}(\omega)|^{2n}$, where $E(n\omega)$ is the field of the $n$th harmonic (see Table I). It was found that the polarization of $E(n\omega)$ was lying also in the plane of incidence. The polarization dependences of the additional satellite lines were also measured and exhibited stronger, $n_{\text{expt}}\sim5\pm1$ values. The experimental data led to typical harmonic production efficiencies of $10^{-10}$ for $n=2$ at $I\sim5$ GW/cm$^2$.

The experimental results obtained cannot be compared directly to the existing theories. The first class of them, i.e., the early semiclassical theories [6,7], are restricted to the determination of the characteristics of the low- (second- and, rarely, the third-) order harmonic production only, but they use a very realistic, collective-electron model. They account for the basic features observed by us: the polarization dependence, the laser intensity dependence, and the specular reflection direction of the harmonic light. Their $\sim10^{-10}$ efficiency value predicted for

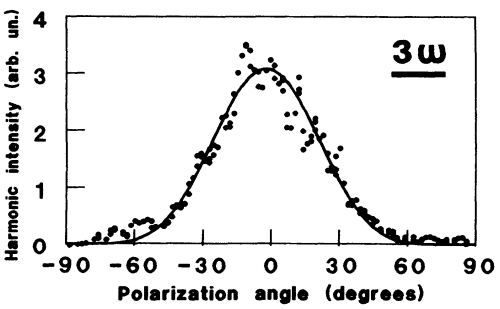


FIG. 3. Harmonic light intensity dependence on the polarization angle, relative to the plane of incidence ($\phi=-90°$ and $90°$ correspond to the $s$ polarization; $\phi=0°$ corresponds to the $p$ polarization) for the $3\omega$ case.

$n=2$ agrees well with our measured experimental value of $\sim10^{-10}$. From this qualitative agreement, however, unambigious conclusions cannot be drawn for the relative contributions connected with the surface or the bulk properties, the conduction or the interband valence electrons, the presence or the lack of the inversion symmetry, etc. Nevertheless, the observed higher-$n$-order polarization and power-law dependences strongly suggest that the generalization of these low-order theories up to higher $n$ orders is possible, similarly to the case of the atoms [3,4].

As for the nonperturbative theories [10–12], at sufficiently low laser intensities they are equivalent with the second-order perturbative calculations. Therefore for $n>2$, these might be considered as the above-mentioned generalizations of the second-order perturbative process (apart from the summation of the collective contributions of each individual electron). Although they are not capable by nature of accounting directly for the correct emission angle, our experiment verified their prediction for the possibility of production of all harmonics above the second up to the fifth. However, they predict a stronger decrease of the efficiencies with the harmonic order $n$ than found experimentally by us (see the right columns in Table I). This fact seems to be surprising at first, being that the perturbative parameter $x$, defined in [10], is $x=3\times10^{-3}$ in our experiment; i.e., we were in the perturbative range. It is well known, however, that theories—elaborated especially for multiphoton electron emission from macroscopic metal surfaces using a modified perturbation parameter and based on the above-mentioned correct coherent summation (e.g., [21])—predict the breakdown of the perturbation approach at considerably lower ($\sim10$ GW/cm$^2$) laser intensities. This was confirmed also experimentally (e.g., [17,18]). Therefore, a similar decrease of the upper limit of the validity region of the perturbation calculation cannot be excluded also in the case of the laser-induced high-order harmonic generation processes at metal surfaces.

As a complementary remark, we stress, that [10] (and indirectly [11,12]) simultaneously determines both the multiharmonic spectrum and the energy spectrum of electrons emitted above the threshold of the multiphoton electron emission process for metals [15]. Therefore, the latter effect (above-threshold photoeffect) which was recently observed by us [16,22] and others [23,24] and the multiphoton photoeffect in general [25] seem to have an interesting physical connection with the multiharmonic generation.

In conclusion, we demonstrated laser-induced multiharmonic generation from a metal surface up to fifth order. The laser intensity dependences of the harmonics showed approximatively a power-law (i.e., perturbative) character; the form of the polarization dependence followed the $\cos^{2n}(\phi)$ law around the $p$-polarization direction. The comparison of the theoretical considerations and our experimental results suggests that our observed multiharmonic generation arises mostly from the Sommerfeld conduction electrons at the surface of the metal. The rough estimation shows that the measured efficiency ($10^{-10}$ for $n=2$ at 5 GW/cm$^2$) coincides quite well with predictions of the theories based on collective many-electron models

[6]. Although the measured efficiencies decrease much weaker with the harmonic order $n$, than predicted by the single-particle perturbative theory [10], other kinds of realistic theories for metals [21] and their experimental verifications [17,18] furnish explanations, which may support our observations.

As a possible future application, the multiharmonic generation from a metal surface may be the base of a simple and very effective vacuum ultraviolet source due to these high efficiency values found experimentally. The harmonic production efficiencies found here at already 5 $GW/cm^2$ can be achieved in noble gases [1-4] only at about 4 orders of magnitude higher, inducing laser intensities ($\sim 10^{13}$ $W/cm^2$).

It is worthwhile to note that there is a further interesting general fundamental consequence of the multiharmonic generation: a rigorous phase matching of all harmonic components follows from the theories both for gases and surfaces [4,5,10]. It follows from this fact that a near-attosecond duration light-pulse sequence is expected to be present in the beam due to an inherent natural mode locking (in the case of equal spectral amplitudes), when it is not decomposed by prism or grating. Our preliminary numerical study [26] based on the experimental data of the work [2] performed for noble gases exhibited the occurrence of $\sim 100$ asec ($\sim 10^{-16}$) duration light-pulse sequence.

*Note added.* After this work was completed another theoretical paper—using a nonperturbative, non-single-particle model, computer calculation method—was published [27] calling attention to the realizability of the multiharmonic generation at solid surfaces in the extreme high-laser-intensity range.

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