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Multiphoton processes in an intense laser field. III. Resonant ionization of hydrogen by subpicosecond pulses

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We have calculated photoelectron energy spectra for ionization of H(1*s*) by subpicosecond pulses with wavelengths 608 and 616 nm and with maximum intensity 1.2×10^{14} W/cm^2. The main features of the recently measured spectrum (at 608 nm) are reproduced.

Recent experiments[1-3] have revealed the surprisingly important role played by intermediate resonances in the dynamics of multiphoton ionization by intense subpicosecond pulses. Prominent structure in the "above-threshold" peaks of the photoelectron energy spectrum has been observed and attributed[1] to the enhancement that occurs as Rydberg sublevels shift in and out of resonance during the rising and falling of the pulse. Up to now, no quantitative theoretical comparison with experiment has been given, but this has become possible in view of the experiments on atomic hydrogen recently performed in Bielefeld by Rottke *et al.*[3]

Rottke *et al.* measured the photoelectron energy spectrum for multiphoton ionization of H(1*s*) by linearly polarized subpicosecond pulses that have maximum intensities of up to about 1.2×10^{14} W/cm^2 and a wavelength in the range 596–616 nm. The most prominent feature in the spectrum is an exceptionally wide subpeak whose origin was attributed to an intermediate resonance involving the Rydberg manifold with principal quantum number 4. The energy at which this subpeak occurs is slightly below that expected on the assumption that the Rydberg manifold does not shift relative to the continuum. We have performed Floquet calculations of the shifts and widths of many sublevels of hydrogen for similar intensities and wavelengths as covered in the experiment. We present theoretical data for the lowest above-threshold peaks of the spectra at 608 and 616 nm. Each of the numerous subpeaks can be identified with a particular resonance. In accord with the experimental observations, we find a prominent subpeak whose width is exceptionally large—about 0.15 eV at half maximum, the same as the measured width. This subpeak is due to a 4*f* seven-photon resonance shifted slightly below the "expected" position, and it occurs at a laser intensity where at least eight photons must be absorbed to ionize H(1*s*). We find that not all intermediate resonances enhance the ionization rate; if a Rydberg sublevel is coupled to the continuum too weakly, as it is if the orbital angular momentum quantum number l is too large, or too strongly, as it may be if l is too small, the corresponding resonance does not enhance the rate.

We treat the field classically, and we describe the electron-field interaction $V(t)$ in the dipole approximation and in the velocity gauge: $V(t)=\mathrm{Re}[-(e/\mu c)(\mathbf{A}_0\cdot\mathbf{p})\times e^{-i\omega t}]$, where e, μ, and $\mathbf{p}$ are the charge, mass, and canonical momentum of the electron, and where $\mathbf{A}_0$ is related to the intensity I through $I=\omega^2|\mathbf{A}_0|^2/(8\pi c)$. Our calculation is based on the Floquet ansatz,[4] that is, we approximate the electron state vector by $\exp(-iEt/\hbar)|F(t)\rangle$, where E is the quasienergy of the initial bound level and $|F(t)\rangle$ is periodic in t with period $2\pi/\omega$. Writing $|F(t)\rangle=\sum_n e^{-in\omega t}|F_n\rangle$, the harmonic components $|F_n\rangle$ satisfy a set of coupled equations which, together with complex outgoing-wave boundary conditions, form an eigenvalue problem, with E the eigenvalue. The total ionization rate is $\Gamma/\hbar$, where $\Gamma/2=-\mathrm{Im}(E)$. Note that we have omitted the $\mathbf{A}_0^2$ term from $V(t)$, and therefore the continuum threshold is unshifted by the field, while the initial level (if it is tightly bound) is shifted downward by roughly the ponderomotive energy $P=2\pi e^2I/(\mu c\omega^2)$. We reduce the coupled equations for the harmonic components to a matrix equation by expanding each harmonic component on a basis consisting of products of spherical harmonics and complex radial Sturmian functions,[5] and we solve these equations using the method of inverse iteration.[6]

In Fig. 1 we depict the variation with intensity of the real parts of the quasienergies E_j of various sublevels j of atomic hydrogen for the wavelength 616 nm and a maximum intensity of 1.2×10^{14} W/cm^2. We have plotted $\mathrm{Re}(E_j-N_j\hbar\omega)$, where each sublevel j is specified by the atomic configuration that is dominant in the *zero-field* limit[7] when this is known; two curves are unlabeled because we do not know the zero-field configurations from which the sublevels originate. As the intensity varies the configuration mixing changes, of course. We determined the configurations by analyzing the composition of the eigenvectors on the basis. We have chosen N_j so as to display the crossing of eigenvalue j with the 1*s* eigenvalue, corresponding to an N_j-photon resonance; the parity of sublevel j is $(-1)^{N_j}$. The 1*s* level undergoes both real and avoided crossings[8-11] with Rydberg levels, but the gaps at the avoided crossings are very small, and would not be visible in Fig. 1. In fact, we have drawn the *diabatic* 1*s* level, along which the 1*s* character is preserved. The diabatic 1*s* level follows the curve -0.4997 a.u. $-1.03P$ with $P/I=0.35\times10^{-13}$ eV (cm^2/W). At weak intensities the minimum number, N_0, of photons required to ionize H(1*s*) is 7, but at 1.35×10^{13} W/cm^2, N_0 increases to 8 and at 6.88×10^{13} W/cm^2 N_0 increases to 9. Note that Rydberg sublevels with large l values ($l\geq4$) barely shift relative to the continuum. Note also that the 3*p* and 3*d* sublevels at first shift downward and, at rela-

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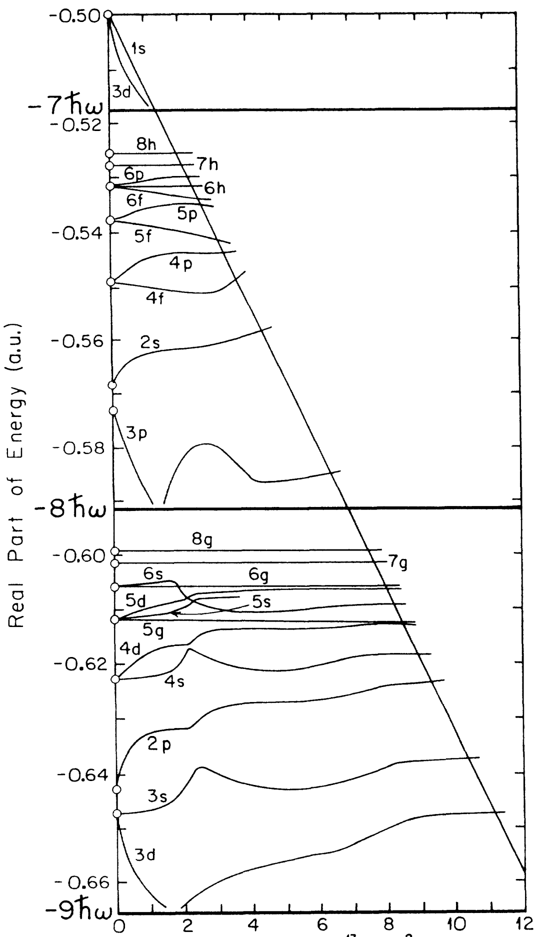

FIG. 1. Real parts of eigenvalues $E_j - N_j \hbar\omega$ at 616 nm for various levels j of atomic hydrogen, each of which in the weak intensity limit corresponds predominantly to the atomic configuration indicated next to the curve. The values of N_j are as follows: $N_{1s} = 0$; $N_{2s} = 6$; $N_{2p} = 7$; $N_{3d} = 6$ (upper curve) or 8 (lower curve); all other sublevels drawn above eight-photon threshold: $N_j = 7$; all other sublevels drawn below eight-photon threshold: $N_j = 8$.

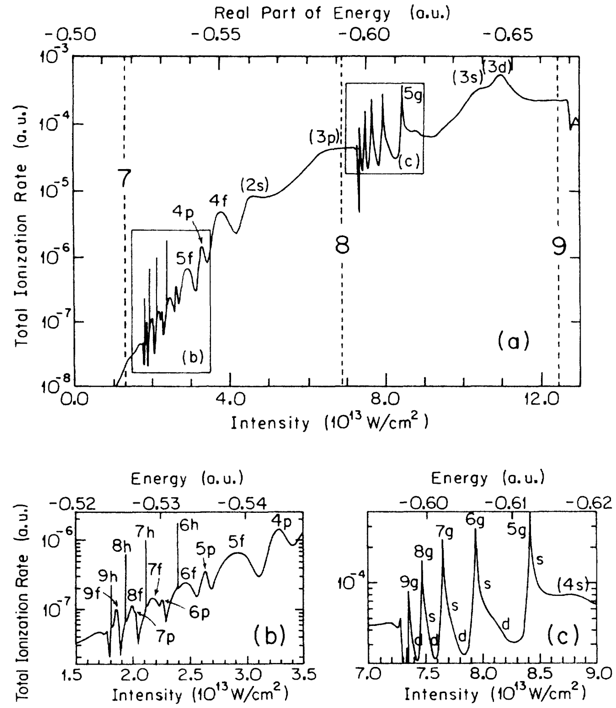

FIG. 2. Total rate for ionization of H(1*s*) by 616-nm light. (b) and (c) are magnifications of the structure inside the boxes (b) and (c) of (a).

tively low intensities, cross ionization thresholds. Subsequently these sublevels encounter a myriad of true and avoided crossings with high Rydberg sublevels (not shown) that accumulate just below the thresholds. The two unlabeled curves that rise above ionization thresholds, close to where the 3*p* and 3*d* sublevels intersect these thresholds, might be sections of the continuous adiabatic curves that join to the 3*p* and 3*d* curves, respectively. (A similar feature, but involving the 2*s* and 2*p* sublevels, was found[11] at 1064 nm.) We note that Crance[12] has calculated some of the curves shown in Fig. 1; the discrepancies, mostly minor, occasionally significant, may be due to the smaller number of basis functions used in her work.

In Fig. 2 we show the *total* rate (that is, $\Gamma_{1s}/\hbar$) for ionization of H(1*s*) by 616-nm light versus intensity. This is the rate for ionization from the *diabatic* 1*s* state. For subpicosecond pulses the intensity sweeps through the crossings so rapidly (on the time scale set by the Rabi frequency) that the system simply jumps across the gap of an avoided crossing, from one adiabatic curve to another, and therefore the atom remains in the diabatic state.[13] Each of the peaks (or bumps) in the rate can be correlated with one (or more than one) crossing of the 1*s* level and another level. We have labeled each peak (and bump) by the dominant configuration of the sublevel that crosses the 1*s* level if this is the dominant configuration both at zero intensity and at the crossing intensity. We have enclosed the configuration label in parentheses, or have omitted the principal quantum number, if there is some ambiguity. Thus, in Fig. 2(a), the peak labeled (2*s*) is due to an intermediate six-photon resonance corresponding to the (true) crossing of the 1*s* level and the level that originates from the 2*s* level, but at the crossing the dominant configuration of the excited-state level is no longer 2*s*. A similar remark applies to the peak labeled (3*s*), and also to the peaks labeled (3*p*) and (3*d*) except that these latter peaks correspond to the crossings of the 1*s* level with the unlabeled curves in Fig. 1, whose origin we are unsure of. Focusing on Fig. 2(b), all of the peaks are due to intermediate seven-photon resonances. The tall spikes (their widths are less than 1 meV) correspond to *avoided* crossings. Note that seven photons can excite sublevels with l values of 1, 3, 5, and 7, in lowest order. It turns out that the l value of the dominant component of the Rydberg sublevel involved in each spike is 5; hence, there are no spikes associated with Rydberg sublevels having a principal quantum number less than 6. There are no $l = 7$ spikes simply because the $l = 7$ Rydberg sublevels are too weakly coupled to the continuum; the widths of these sublevels are smaller than the $1s$ width. On each side of the $6h$ and $7h$ spikes are two broad peaks which correspond to true crossings and involve Rydberg sublevels that contain two strongly mixed components, with l values of 1 and 3, one of which is dominant; the dominant components are $l=3$ ($l=1$) for the peaks on the high- (low-) intensity sides of the $6h$ and $7h$ spikes. Peaks on the high-intensity sides of the $8h$ and $9h$ spikes are also visible, and they too are due to true crossings with Rydberg sublevels whose dominant components are $l=3$; no additional $l=1$ peaks are visible—they have merged into the $l=1$ peaks. The $5p$ and $5f$ peaks also correspond to true crossings; so do the $4p$ and $4f$ peaks seen in Fig. 2(a). Focusing now on Fig. 2(c), we see narrow peaks (the widths at half maximum are about 20 meV) which are due to intermediate eight-photon resonances. The maxima occur at avoided crossings, and involve Rydberg sublevels with (dominant) l values of 4. Note that each peak falls off sharply on its low-intensity side, but more gradually on its high-intensity side. Two effects contribute to this gradual falloff: (i) the ionization rate drops off rapidly with increasing energy of the emergent photoelectron—this energy is lower at higher intensity; (ii) on the high-intensity side there are one or more true crossing resonances at the positions indicated by the (dominant) l value of the appropriate Rydberg sublevel. The absence of peaks corresponding to these true crossings is due to the fact that the peaks are so broad that they merge into the background.[11]

The intensity profile of a typical Fourier-transform-limited pulse has the form

$$I(\rho,z,t)=[R^2/r^2(z)]I_0 e^{-2\rho^2/r^2(z)} e^{-(t-z/c)^2/t_p^2}, \quad (1)$$

where I_0 is the maximum intensity, ρ is the cylindrical radius, t_p is the characteristic pulse duration, R is the spot size at the laser focus, and $r^2(z)=R^2[1+(\lambda z/\pi R^2)^2]$, with λ the wavelength. Assuming that the electron follows the diabatic "eigenvalue" curve, the ionization yield Y is

$$Y=2\pi\int \rho\, d\rho \int dz \left[1-\exp\left(-\int_{-\infty}^{\infty} dt\, \Gamma(I(\rho,z,t))/\hbar\right)\right], \quad (2)$$

where $\Gamma(I(\rho,z,t))/\hbar$ is the ionization rate at each space-time point, assuming cylindrical symmetry with respect to the z axis. The spatial integration is over the volume of atoms, whose linear dimensions are large compared to R and to R^2/λ. The characteristic bandwidth $\Delta\omega$ is of order $1/t_p$, and this must be very small compared to the frequency ω if the Floquet estimate of the ionization rate is to be sensible; hence the pulse duration must be more than a few cycles. In terms of the variables $\rho'=\rho/R$, $z'=(\lambda/R^2)z$, and $t'=(t-z/c)/t_p$, the intensity profile is independent of R, λ, and t_p. Hence Y scales with R and, if we neglect depletion, with t_p. If we divide the intensity interval $0\leq I\leq I_0$ into small segments, and calculate the yield produced in each segment, the envelope of the resulting histogram is a yield-intensity profile. For a Fourier-transform-limited pulse with specified I_0 the pattern of resonance peaks in the yield-intensity profile is independent of R and (if depletion is negligible) of t_p, provided that the passage through the resonances occurs sufficiently rapidly to be considered diabatic, and provided that $\Delta\omega$ is small compared to both ω and the peak widths. Of course, this pattern changes as I_0 varies.

We can roughly translate the yield-intensity profile into the photoelectron energy spectrum for ionization by a subpicosecond pulse as follows. The pulse is sufficiently short that we can neglect the ponderomotive scattering of the electron as it leaves the atom and traverses the focal region of the laser. We assume that at each intensity ionization occurs only into the lowest open channel, that is, the N_0-photon channel, with $N_0=7$ to 9 (the main contribution to the spectrum is in this channel and it is not significantly affected by branching into higher channels). We calculate the energy with which a photoelectron is emitted at a particular intensity from a knowledge of the shift of the $1s$ level, neglecting both the width of this level and the laser bandwidth (the bandwidth of a 100-fsec pulse is about 18 meV). Photoelectron energy spectra are shown in Fig. 3 for ionization of H($1s$) by 608- and 616-nm pulses, with $I_0=1.2\times10^{14}$ W/cm^2. We have labeled some of the peaks by the dominant configuration of the significant Rydberg sublevel at resonance. The spectra are independent of t_p when depletion is neglected [which is justified for $2t_p<0.05/\sqrt{\ln(2)}$ psec]. For 608 nm we show spectra both when depletion is neglected and included, in the latter case for a pulse duration $2t_p=0.5/\sqrt{\ln(2)}$ psec; this may be compared with the measured[3] spectrum at the same wavelength, t_p, and I_0. As noted above, an exceptionally prominent, wide peak was identified in the experiment as being due to either a $4p$ or $4f$ seven-photon resonance (or both). At 608 nm our theoretical $4f$ peak is the most prominent (the signal is the largest), it has about the same width (0.15 eV) as the observed peak, and, as in the experiment, it is energy shifted slightly toward the zero-energy threshold, more so when depletion is included

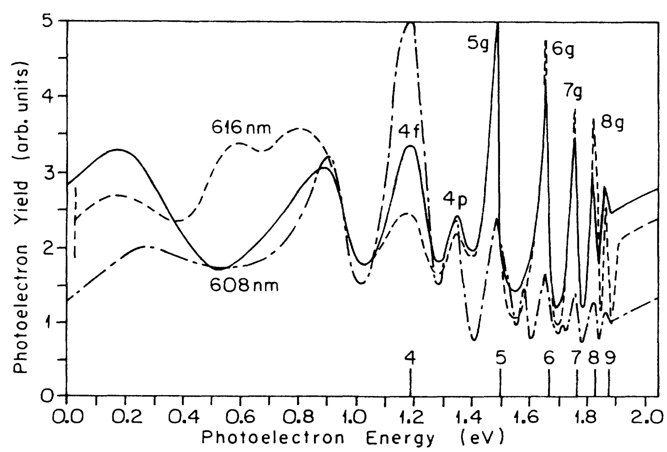

FIG. 3. Yield in photoelectrons vs photoelectron energy for ionization of H($1s$) by a subpicosecond pulse whose peak intensity is 1.2×10^{14} W/cm^2 and whose wavelength λ is 608 or 616 nm. Curves are as follows: ——, $\lambda=608$ nm, depletion neglected; $-\cdot-\cdot$, $\lambda=608$ nm, depletion included, for $2t_p=0.5/\sqrt{\ln(2)}$ psec; $---$, $\lambda=616$ nm, depletion neglected (latter two curves shown only when different from solid curve). For convenience, we have displaced the 616-nm spectrum to the right by 0.03 eV.

(but not quite as far as in the experiment). The theoretical 4*p* and 4*f* peaks are well separated and so it is likely that the observed peak is the 4*f* peak. The 4*p* resonance is much less prominent and it is shifted somewhat further from the expected position, away from the zero-energy threshold.[14] When depletion is neglected, the 5*g* to 8*g* eight-photon resonances, which occur at very high intensities, are prominent, but when depletion is included these resonances are less important than the 5*f* to 8*f* seven-photon resonances. The broad hump on the low-energy side of the 4*f* peak is due to the contributions from the (2*s*), (3*p*), (3*s*), and (3*d*) peaks seen in Fig. 2(a) (mostly 2*s* when depletion is included). Note that at 616 nm the 4*f* peak is less prominent relative to the 5*g* peak than at 608 nm; this occurs because at 616 nm the peaks are shifted, each by the same amount, to slightly lower intensities, but the *relative* drop in intensity is greater for the 4*f* than for the 5*g* peak.

In conclusion, we have performed calculations of the substructure in the lowest peak of the photoelectron energy spectrum for ionization of H(1*s*) by subpicosecond pulses. The results confirm the main features seen in the Bielefeld experiment, as reported[3,14] so far.

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