Above-Threshold Ionization of Cesium under Femtosecond Laser Pulses: New Substructure due to Strongly Coupled Bound States

W. Nicklich, H. Kumpfmüller, and H. Walther
Max-Planck-Institut für Quantenoptik and Sektion Physik der Universität München, W-8046 Garching, Germany

X. Tang, Huale Xu, and P. Lambropoulos
Department of Physics, University of Southern California, Los Angeles, California 90089-0484
(Received 4 June 1992; revised manuscript received 6 August 1992)

First above-threshold-ionization (ATI) spectra on cesium atoms obtained with laser pulses as short as 70 fs are presented. Up to 7 ATI peaks exhibiting new substructure related to the extremely large Rabi coupling between the ground and first excited states have been detected. The angular and intensity dependence of the ATI signal has been studied. It is shown that the ATI peaks shift to higher electron energies with rising laser intensity. The obtained results are compared with a perturbative as well as a nonperturbative theory which predicts the substructure.

PACS numbers: 32.80.Rm, 42.50.Hz

In order to observe above-threshold ionization (ATI), which means the absorption of an additional number S of photons over the minimum required to ionize an atom [1], it is necessary that the atoms experience sufficiently high intensity before the electron leaves the atom. Up to now this has been achieved by choosing photon energies significantly smaller than the ionization energy [2–6], so that the minimum required photon number N was larger than 6. Under these conditions the transition from the ground to a Rydberg state corresponds to a process with an effective Rabi frequency much smaller than the single-photon ionization rate of the Rydberg state. The relatively large value of N avoids saturation (complete ionization) during the rise of the pulse. Attempts to observe ATI on alkali atoms were therefore not very successful and have produced at most two ATI peaks [7,8].

Since the probability of saturation increases with the pulse duration, the use of ultrashort pulses should make it feasible to observe extensive ATI spectra even when N is as small as 2 or 3. If such an experiment would be realized, one would expect significantly different dynamical behavior because the Rabi frequency describing the coupling of the states below threshold would be comparable to or larger than the ionization rate. In contrast to previous experiments [1–4] the process cannot be understood as a weak multiphoton coupling to highly excited states followed by strong ionization, but represents the ionization of strongly coupled bound states exhibiting features reminiscent of ac Stark splitting with a rapidly varying Rabi frequency as the pulse rises to its peak value. Moreover, femtosecond pulses allow one to avoid ponderomotive effects which the electron experiences as it leaves the laser focus [3]. Despite the interesting phenomena expected, femtosecond pulses have been used only in a few ionization experiments so far [5,9], none of those experiments having been performed on alkali atoms. In this paper we present the first observation of ATI spectra under the described conditions and provide the theoretical interpretation.

The pulses are generated in a standard colliding-pulse mode-locked ring dye laser with chirp compensation by four prisms [10]. A two-stage dye amplifier pumped by a copper-vapor laser raises the energy to about 12 μJ with a pulse duration of 70 fs. The design is similar to those described by Hirlimann *et al.* and Fork *et al.* [11]. The pulses are focused down to a beam diameter of about 10 μm which results in a peak intensity of 10^{14} W/cm^2 at a repetition rate of 6.2 kHz.

The electron spectrometer uses time-of-flight analysis. The atoms of an atomic beam are ionized by the laser pulses. The photoelectrons then travel through an electrically and magnetically shielded region and are detected by a microchannel plate at right angles to the incident laser radiation with a collecting angle of 5°. Besides the signal electrons, the energy of the laser pulses is also registered for each pulse by a photodiode. Thus the influence of the intensity fluctuations on the signal can be eliminated. The background pressure in the spectrometer was held at 2×10^{-6} Pa by means of a cryopump.

In view of the variety of the features of the interaction studied in the experiment, the theoretical interpretation necessitated time-dependent (TD) nonperturbative as well as perturbative (lowest nonvanishing order amplitude for each ATI peak) calculations. For TD calculations, we have employed finite difference techniques for the direct solution of the Schrödinger equation on a numerical grid. Details of the method [12] as well as our implementation (in terms of a model potential) to the problem of high-order harmonic generation have been discussed elsewhere [13]. Using TD theory, we have been able to produce ATI photoelectron spectra of high order such as those observed in the experiment. This method, on the other hand, places prohibitive computational demands on the integration over the spatial distribution of the laser intensity which is useful for the determination of saturation intensities and the investigation of the limits

of the validity of perturbation theory (PT).

In order to explore the validity of PT we have obtained generalized cross sections for the first two ATI peaks using a Green's-function technique [14] for the exact multiple summations based on single-channel quantum defect theory. With the same method we have also obtained the photoelectron angular distributions and the ac Stark shifts employed later on. Using these cross sections we can readily perform the spatiotemporal integration. Because of computational limitations, our calculation does not yield reliable values above the third ATI peak at the present time.

The ionization potential of cesium is 3.89 eV, and the photon energy about 1.96 eV. We therefore expect two-photon ionization very close to the threshold. The one-photon step is away from any resonance and about half way between the $6p$ and $7p$ states. In order to explain the measured results we have to assume a small contact potential of about 0.2 eV in the electron spectrometer. Since the height and position of the lowest peak are strongly affected by the closely lying threshold and the contact potentials in the time-of-flight setup, it is omitted in the discussion.

The measured electron spectrum in Fig. 1 (curve *a*), a result of 5×10^5 counted electrons, shows ATI peaks up to 14 eV, the highest one corresponding to a nine-photon process. The peaks are separated by the photon energy, and their height decreases with energy above threshold.

The spectrum calculated through TD theory is shown in Fig. 1 (curve *b*). The general structure is remarkably similar to that of the experiment including the substructures (side peaks). One reason for the difference in the ratio of the ATI peak heights between theory and experiment may be the fact that in the experiment the measured electron signal is in the direction of the polarization, while the theory produces angle integrated spectra. The origin of the side peaks, which are found to grow and shift with the peak intensity, can be traced to Rabi-like oscillations due to the off-resonance strong coupling of the ground state to the first excited state. The relevant Rabi frequency is $1.06\times10^8\sqrt{I}$ rad/s, where I is the intensity in W/cm^2. This causes an ac Stark splitting of a rapidly varying magnitude as the intensity rises and falls during the pulse. Although only one laser is involved, the energy analysis of the photoelectrons serves as a probe of that splitting as well as of the shift of the ground state. The enormous $6s$-$6p$ Rabi frequency induces substantial population deposition to the $6p$ state, in spite of the large detuning. This is equivalent to populating one of the components of the doublet created by the off-resonant ac Stark splitting of the $6p$. Because of the significant laser bandwidth, photon absorption from the $6p$ populates and subsequently ionizes higher bound states [$7d$, $9s$, $10s$, $8d$, and $9d$, see peak (1) in Fig. 1], thus creating substructure, some of which is not obliterated by the spatial distribution of the radiation. This effect is different from the

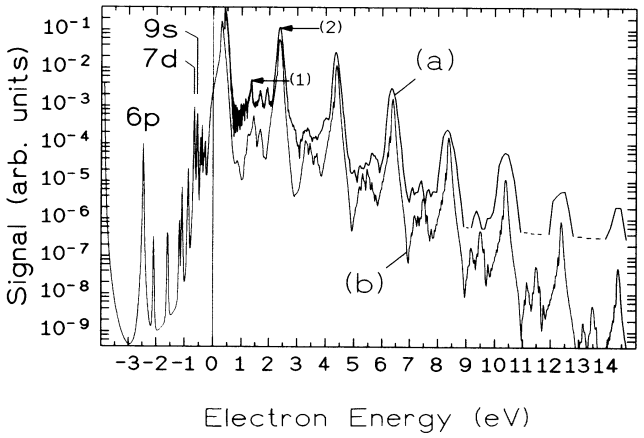

FIG. 1. Curve *a*: Experimental electron energy spectrum measured after multiphoton ionization of Cs with laser pulses of about 90 fs duration and a peak intensity of about 10^{14} W/cm^2 at a wavelength of 621 nm. Curve *b*: TD calculation at an intensity of 5×10^{12} W/cm^2. The pulse shape is trapezoidal with 6 optical cycles turning the field on or off and 30 cycles at maximum. The peaks below threshold correspond to population retained in excited states. Peaks (1) and (2) correspond to the $6s$-$6p$ ac Stark doublet. Curves *a* and *b* are separated slightly in the vertical direction for visual convenience.

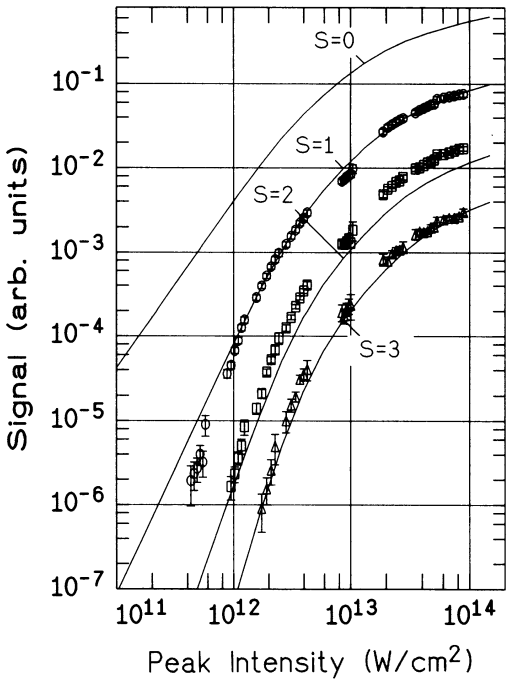

FIG. 2. Intensity dependence of the first three ATI peaks measured in the direction of the polarization at 635 nm with 70 fs pulse duration. ○, $S=1$; □, $S=2$; △, $S=3$. The solid lines are the result of the PT model including the temporal and spatial pulse shape. The measured peak intensity has to be corrected only by a factor of 0.8. All data are fitted in the vertical direction by a global factor, because we have no absolute measurements of the photoelectron signal.

one observed in earlier experiments [4], where highly excited states shift through multiphoton resonance by an amount practically equal to the ponderomotive shift. The shift of the ground state, on the other hand, affects the position of the high peak of the splitting (see Fig. 1, curve *b*), which appears as the dominant peak in the data.

Figure 2 shows the intensity dependence of the ATI signal from $S=1$ to $S=3$. The intensity is changed by turning the polarization by a quarter-wave plate in front of a thin Rochon polarizing prism. As mentioned above, the laser pulse energy is monitored constantly during a measurement. Thus in one single run, one can obtain electron spectra for six diferent laser energies which correspond to six neighboring data points in the diagrams. Pulse duration and focal diameter of the laser beam are obviously not influenced by the energy fluctuations of the laser system. Otherwise the different groups of data points would not fit precisely to each other.

For comparison with theory we use a model based on the pertubative ionization rate for $(N+S)$-photon ionization: $dP/dt=\sigma_{(N+S)}I^{N+S}$, where S is the number of the ATI peak and the generalized cross sections $\sigma_{(2)}=6\times10^{-49}$ cm^{4}s, $\sigma_{(3)}=7.33\times10^{-81}$ cm^{6}s^2, $\sigma_{(4)}=6.15\times10^{-113}$ cm^{8}s^3 have been obtained through the Green's-function technique [14]. To fit experiment and theory for $S=3$ we use $\sigma_{(5)}=4.6\times10^{-145}$ cm^{10}s^4, which could not be obtained reliably through our Green's-function calculation. The temporal shape of the laser pulse was measured by a standard autocorrelation technique and the spatial shape by a 2 μm pinhole mounted on a stepper. We assume a hyperbolic secant for the temporal and a Gaussian for the spatial pulse shape. The pulse parameters are fitted to the experimentally determined shape with reasonable agreement. In order to take into account pulse shape and saturation one has to integrate over time and space.

The result is plotted as solid lines in Fig. 2. For small intensities, the lines follow the simple power law since the main contribution always comes from the center of the pulse. Above 4×10^{12} W/cm^2 the influence of saturation can be seen. The slope of the curves decreases and the slight increase in ionization is mainly due to the fact that the interaction region becomes larger. The experimental results in the saturation region are described well by the theoretical model. The slight discrepancy in the slopes for $S=1$ and $S=2$ below saturation is too small to serve as a criterion for the breakdown of PT. The agreement between theoretical and measured peak intensity, on the other hand, is quite good (see caption of Fig. 2).

The angular distributions of the emitted electrons are shown in Fig. 3 for the first and the second ATI peaks. The distributions are slightly more peaked at zero for the second ATI peak, as expected. Qualitatively this holds also for higher S values. The agreement between experiment and theory is good, especially for the first ATI peak, but there is a significant difference from the data by

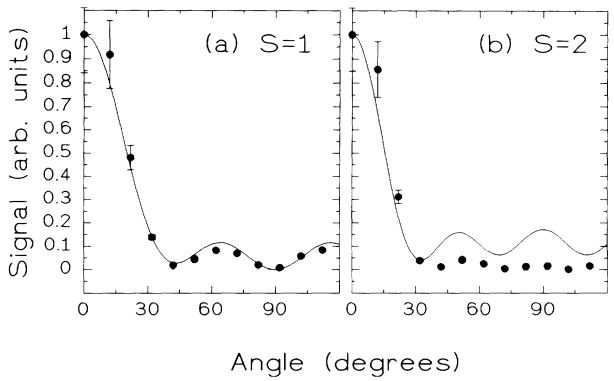

FIG. 3. Photoelectron angular distribution for (a) the first and (b) the second ATI peak at 623 nm with 80 fs pulse duration. The solid line represents the PT calculation. The curves are adjusted in the horizontal axis by 2° in order to eliminate the zero-point calibration error and normalized to 1 in the vertical axis.

Dodhy, Compton, and Stockdale [8] obtained with longer pulses, which may explain the difference.

In contrast to other ATI experiments [4] we observe that the positions of the ATI peaks shift to higher energies with rising laser intensity. The reason for this is the ac Stark shift of the ground state which had an opposite sign and much smaller size in previous experiments. The shift of the ATI peaks (in atomic units) in first nonvanishing order is given by

$$\Delta E_g-\Delta E_{\text{thres}}=\frac{I}{4}\left[-\alpha_g(\omega)-\frac{1}{\omega^2}\right], \tag{1}$$

where ΔE_{thres} is the shift of the ionization limit which is equal to the ponderomotive shift and ΔE_g is the shift of the ground state determined by its polarizability α_g which in our case is negative. The shift of the ionization threshold by 0.38 eV at $I=10^{13}$ W/cm^2 is overcompensated by the ground-state shift amounting to 0.90 eV. If we include the temporal saturation ($I_{\text{sat}}=4.2\times10^{12}$ W/cm^2) by calculating the position of the maxima of the first two ATI peaks, we obtain the solid lines in Fig. 4. The shifts in the low-intensity region are in good agreement with (1). At higher intensities the shifts become constant, since we reach saturation. The mismatch between the theoretical and experimental saturated values (Fig. 4) can be explained if we assume that our theoretical saturation intensity is too large by about a factor of 2.

In summary, we have observed up to 7 ATI peaks in Cs with substructure due to Rabi oscillations, which are in general agreement with TD calculations despite the lack of spatial integration. Intensity and angular dependence of the lower-order peaks can be described by a simple model based on perturbation theory including the temporal and spatial pulse shape. According to the model and the observed shifts, the maximum intensity to which

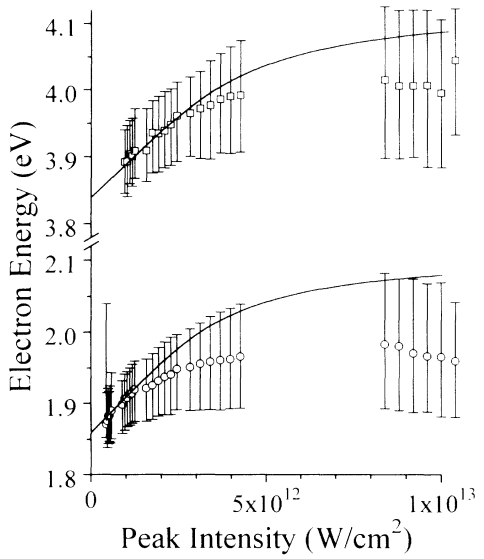

FIG. 4. Shift of the first and second ATI peak vs laser peak intensity. The dots show the mean energy, and the error bars indicate the width of the peaks (i.e., the region that contains 68% of all counts). The solid lines indicate the shift expected from PT including saturation. The pulse duration is 70 fs. As in Fig. 2 the measured peak intensity is corrected by a factor of 0.8.

the electrons are really exposed is limited by saturation to about 4.2×10^{12} W/cm^2. The observed intensity-dependent blueshift in the electron spectrum is due to the fact that the energy difference from the ground state to the first excited state of Cs at zero field is lower than the photon energy. The further experimental delineation of the peak substructures (especially in other alkalis on which we expect to report soon) as well as of the possible effect of double electron excitations (given that the last two ATI peaks are within the energy range of doubly excited states) represent exciting avenues of future investigations. Large-scale further computations to quantify perturbative and nonperturbative aspects are also much desirable.

The authors want to thank Professor W. Christian, Professor S. Dinev, and Professor K. Rzaczewski for helpful discussions. The work was supported in part by NSF under Grant No. PHY-9013434 and DOE under Grant No. DE-FG03-87ER60504.

[1] P. Agostini, F. Fabre, G. Mainfray, G. Petite, and N. K. Rahman, Phys. Rev. Lett. **42**, 1127 (1979).
[2] P. Kruit, J. Kimman, H. G. Muller, and M. J. van der Wiel, Phys. Rev. A **28**, 249 (1983).
[3] T. J. McIlrath, P. H. Bucksbaum, R. R. Freeman, and M. Bashkansky, Phys. Rev. A **35**, 4611 (1987).
[4] R. R. Freeman, P. H. Bucksbaum, H. Milchberg, S. Darack, D. Schumacher, and M. E. Geusic, Phys. Rev. Lett. **59**, 1092 (1987).
[5] H. G. Muller, H. B. van Linden van den Heuvell, P. Agostini, A. Antonetti, M. Franco, and A. Migus, Phys. Rev. Lett. **60**, 565 (1988); P. Agostini, P. Breger, A. L'Huillier, H. G. Muller, G. Petite, A. Antonetti, and A. Migus, Phys. Rev. Lett. **63**, 2208 (1989).
[6] H. Rottke, B. Wolff, M. Brickwedde, D. Feldmann, and K. H. Welge, Phys. Rev. Lett. **64**, 404 (1990); M. Dörr, R. M. Potvliege, and R. Shakeshaft, Phys. Rev. Lett. **64**, 2003 (1990).
[7] G. Petite, F. Fabre, P. Agostini, M. Crance, and M. Aymar, Phys. Rev. A **29**, 2677 (1984).
[8] Adila Dodhy, R. N. Compton, and J. A. D. Stockdale, Phys. Rev. Lett. **54**, 422 (1985); Phys. Rev. A **33**, 2167 (1986).
[9] S. L. Chin, Claude Rolland, P. B. Corkum, and Paul Kelly, Phys. Rev. Lett. **61**, 153 (1988).
[10] J. A. Valdmanis, R. L. Fork, and J. P. Gordon, Opt. Lett. **10**, 131 (1985).
[11] C. Hirlimann, O. Seddiki, J.-F. Morhange, R. Mounet, and A. Goddi, Opt. Commun. **59**, 52 (1986); R. L. Fork, H. Avramopoulos, H. L. Fragnito, P. C. Becker, K. L. Schehrer, and C. Hirlimann, Opt. Lett. **14**, 1068 (1989).
[12] K. C. Kulander, Phys. Rev. A **35**, 445 (1987).
[13] Huale Xu, X. Tang, and P. Lambropoulos, Phys. Rev. A **46**, R2225 (1992).
[14] Mark Edwards, X. Tang, P. Lambropoulos, and Robin Shakeshaft, Phys. Rev. A **33**, 4444 (1986); Mark Edwards, X. Tang, and Robin Shakeshaft, Phys. Rev. A **35**, 3758 (1987).