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Verification of the ponderomotive approximation for the ac Stark shift in Xe Rydberg levels

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We report direct measurements of the ac Stark shifts of Xe Rydberg levels in high-intensity laser fields (up to 6 GW/cm^2), demonstrating the expected near-ponderomotive behavior. These measurements provide extensive unambiguous evidence for the validity of perturbative calculations of the nonresonant ac Stark shift at these intensities.

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The ac Stark shift from intense laser fields has been the subject of many recent investigations. In particular, the ponderomotive shifting of ionization potentials and high-lying atomic or molecular levels has been widely discussed to explain photoelectron energy spectra in above-threshold ionization experiments, and to interpret negative-ion photodetachment threshold shifts [1]. The ponderomotive energy, $U_p = e^2 E_0^2/4m\omega^2$, is simply the classical mean kinetic energy imparted to a free electron by an oscillating radiation field $E = E_0 \cos\omega t$. Second-order perturbation theory predicts that in the high-frequency approximation (where the photon energy is large compared to the binding energy of a Rydberg level) the net shift Σ of the transition energy between the ground state and a Rydberg state should be $\Sigma = U_p + \Delta E$, where ΔE is the difference between the shifts due to the static polarizabilities of the neutral ground state and the ionic core [2]. If the perturbing field photon energy, while larger than the Rydberg binding energy, is small compared to the energy of any transitions out of the ground state, ΔE is a small fraction of U_p, and essentially ponderomotive ac Stark shifts are expected. The net ac Start shift of the Rydberg level is then $\Sigma \approx U_p = e^2 E_0^2/4m\omega^2 = e^2 I/2c\epsilon_0 m\omega^2$, a linear function of the perturbing field intensity I.

Previously reported direct and indirect measurements of the ac Stark shift under conditions appropriate for the application of the perturbative high-frequency approximation have been contradictory. For example, Liberman, Pinard, and Taleb [3] directly measured the ac Stark shift of the $22p\,^2P_{3/2}$ level in rubidium perturbed by 1064-nm radiation at intensities up to 12 MW/cm^2, obtaining values within about 10% of the expected ponderomotive shifts. Trainham *et al.* [4] measured the shift in the threshold for photodetachment from the Cl$^-$ ion perturbed by 1064-nm radiation up to 10^{10} W/cm^2. They found an increase in the photodetachment threshold of only about 25% of the expected near-ponderomotive value. Landen, Perry, and Campbell [5] measured the three-photon resonant, four-photon ionization profiles of krypton at intensities up to 3×10^{14} W/cm^2 in picosecond pulses near 300 nm, reporting ac Stark shifts of about 45% and 95% of the ponderomotive value for the $4p^5(^2P_{3/2})5d[1/2]_1$ and $4p^5(^2P_{3/2})4d[3/2]_1$ levels, respectively. Throughout this Communication, the standard Jl coupling notation is used, where the number in brackets indicates the K value resulting from coupling the core $(^2P_{3/2,1/2})$ total angular momentum with the orbital angular momentum of the excited electron, and the subscript indicates the total angular momentum of the level including the excited electron spin, $J = K \pm s$. Agostini *et al.* [6] measured the photoelectron energy spectrum for six-photon resonant, seven-photon ionization of xenon by subpicosecond pulses up to 3×10^{13} W/cm^2, inferring Stark shifts consistent with ponderomotive shifts for states in the $7p$ and $6f$ configurations. Normand *et al.* [7] directly measured the ac Stark shift in a large number of xenon Rydberg levels perturbed by 1064-nm radiation up to about 10^{10} W/cm^2, obtaining about 45% of the near-ponderomotive shift predicted by detailed multichannel quantum defect calculations [8]. Tang *et al.* [9] studied the photodetachment threshold in H$^-$ ions at intensities up to 1.2×10^{10} W/cm^2 and found threshold energy shifts about half the ponderomotive value, but attributed the discrepancy to variations in the intensity of the perturbing laser field. Very recently, Davidson *et al.* [10] observed a full ponderomotive shift for the photodetachment threshold of Cl$^-$ in perturbing fields up to 4.5×10^{13} W/cm^2. This partial list reflects the range of measured or inferred ac Stark shifts obtained in recent experiments under conditions expected to produce ponderomotive shifts.

This Rapid Communication presents the results of a precise set of direct measurements of ac Stark shifts in xenon Rydberg levels using two-color, three-photon resonance ionization spectroscopy. We report excellent agreement with both the simple ponderomotive predictions and with detailed second-order calculations of the expected ac Stark shifts.

Fourteen levels in the $5p^5np$ $(n=10-15)$ and $5p^57f$ configurations of Xe were studied. Xe atoms were subject to intense (up to 6 GW/cm^2) monomode infrared (ir) radiation from a pulsed neodymium-doped yttrium aluminum garnet laser (1064 nm). Simultaneously, the transition energy between the ground state and a perturbed Rydberg state was probed by scanning a tunable

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laser through the two-photon ultraviolet (uv) resonance ($\sim$205–210 nm). The ir radiation shifted the energy levels and ionized the Rydberg atoms, and the ionization signal as a function of uv laser frequency was recorded with a simple time-of-flight mass spectrometer. The uv and ir lasers were spatially and temporally overlapped and focused into a chamber containing $\sim 10^{-4}$ Pa of Xe. Both lasers were linearly polarized with parallel polarizations. Great care was taken to assure that the atoms excited by the uv laser were subject to a well-characterized ir perturbing field. Before overlapping with the uv laser, the ir laser was spatially filtered and its divergence adjusted so that in the interaction region around the uv focal plane the ir laser spatial profile was near-Gaussian and large compared to the uv laser beam diameter. An aperture in the ion collection electrodes—3 mm along the laser axes—limited detection of ions to a region where both the uv and ir spatial profiles were nearly constant. The uv confocal parameter was about 10 mm and the ir beam diameter varied by less that 4% over $\pm$10 mm around the uv focus. The uv beam waist was about 70 μm [full width at half maximum (FWHM)], while the ir beam diameter at the uv focal plane was systematically varied from about 160 to 210 μm. A video camera with a charge-coupled-device detector sensitive to both the uv and ir radiation was used to measure the laser spatial profiles and assure optimal spatial alignment of the lasers at the uv beam waist (see Fig. 1). Fast photodiodes were used to verify that the uv and ir laser peak intensities were temporally coincident—the uv pulse length was about 3 ns (FWHM) compared to the $\sim$9 ns (FWHM) ir pulse length. For each Rydberg level selected, at least ten separate uv laser scans were recorded for each of at least six different ir laser intensities. The ir laser power was continuously monitored with a calibrated volume absorber and power meter. Figure 2 shows typical resonance ionization profiles as a function of uv laser detuning from the unperturbed two-photon resonance, as well as predicted profiles as discussed below.

Accurate measurement of the ac Stark shifts from such resonance ionization profiles depends critically on the experimental conditions. The ac Stark shift is only one factor determining the energy shifts of the peaks of the ionization profiles—other atomic parameters and experimental conditions influence the peak energy shifts as well as the widths and shapes of the ionization profiles. L'Huillier *et al.* [8] developed a detailed theory of the two-color, three-photon resonance ionization process for the Rydberg levels of Xe, including estimation of the atomic parameters necessary to calculate the ionization profiles for the different levels in this study, such as the uv two-photon Rabi rates and the ir photoionization rates. The model implies that if the spatiotemporal profile of the (perturbing) ir laser is sufficiently large compared to the (probe) uv laser, and if the uv (two-photon) excitation is not saturated—these conditions assuring that the two-photon excitation occurs in a region of near-constant ir laser intensity—then the ac Stark shift should be closely approximated by the observed energy shifts of the peak of the ionization profiles at the intensities used in this experiment. Under these conditions, the ionization profile peak energy shifts become relatively insensitive to the two-photon Rabi rates and photoionization rates, which are difficult to accurately measure or calculate.

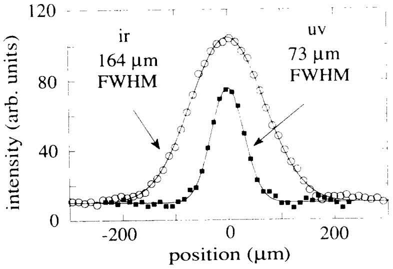

FIG. 1. Cross-sectional profiles of the ir and uv laser intensities at the uv focal plane. The open and filled symbols show the measured relative intensities and the solid lines the least-squares Gaussian fits.

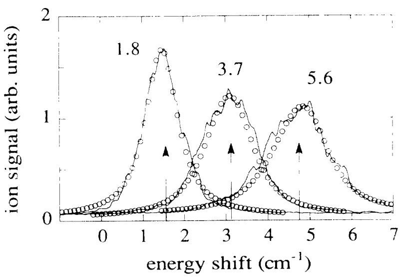

FIG. 2. Sample resonance ionization profiles (solid lines) for the $5p^5(2P_{3/2})10p[1/2]_0$ level of Xe perturbed by varying intensities of ir (1064 nm) radiation. The numbers by each curve indicate the ir intensity in GW/cm^2. Each profile is the average of three consecutive scans. Also shown are the corresponding predicted profiles (dotted) using the model described in the text and assuming a ponderomotive ac Stark shift. The arrows indicate the expected ponderomotive shift at each ir intensity.

In this experiment, the absence of saturation of the two-photon excitation was verified by measuring the total number of ions produced by varying uv laser intensities at constant ir intensity. Figure 3 displays the results of one such investigation, demonstrating the quadratic (unsaturated) dependence of the ionization signal on the uv laser intensity. Most of the ac Stark shift measurements

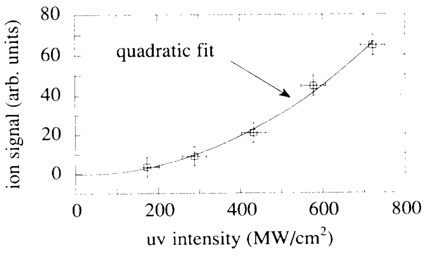

FIG. 3. The total ionization signal, at a fixed ir intensity of $\sim$5 GW/cm^2, as the uv laser intensity is varied for the $5p^5(2P_{3/2})11p[1/2]_0$ level of Xe. The quadratic dependence of the ionization signal on the uv laser intensity corroborates that the two-photon transition is not saturated at the intensities shown here.

reported here were made with uv laser intensities ~500 MW/cm^2, well within the unsaturated regime. Thus we expect in this experiment, with a uniform perturbing field and at uv laser intensities below saturation, that the energy shifts of the peaks of the ionization profiles should accurately reproduce the actual ac Stark shifts.

For Xe Rydberg levels subject to 1064-nm monomode radiation, the ac Stark shift is predicted to be the sum of the ponderomotive shift, $U_p = 0.85$ cm^{-1}/(GW/cm^2), and the net shift of the ground state vs ionic core, $\Delta E \cong 0.01$ cm^{-1}/(GW/cm^2) [8,11], for a net ac Stark shift of 0.86 cm^{-1}/(GW/cm^2)—that is, a shift of 0.86 cm^{-1} at a 1064-nm laser intensity of 10^9 W/cm^2. We will refer to this net predicted shift simply as the ponderomotive shift—the ~1% difference from the value of U_p is negligible compared to the ~15% uncertainty in the measurements reported here.

For over 1000 separate scans among the 14 levels, the mean measured ac Stark shift—inferred from the ionization peak energy shifts—was 0.87± 0.13 cm^{-1}/(GW/cm^2), in excellent agreement with the ponderomotive shift. The ~15% uncertainty is dominated by the uncertainty in the ir laser intensity measurement (~10%). The statistical (1σ) error in the separate determinations was about 7%. There was no detectable variation in measured Stark shifts among the different levels (see Fig. 4). As systematic checks, some data were recorded with Xe pressures varying between ~10^{-5} to 10^{-3} Pa, with the ir laser polarization rotated by up to 90° relative to the uv polarization, or with the uv laser intensity varied by a factor of ~10 for a give ir intensity. No appreciable variation in the measured ac Stark shift was detected in these studies.

To further test the agreement between ponderomotive theory and the observed energy shifts, we compared the model of L'Huillier *et al.* to the observed ionization profiles. Figure 2 demonstrates the good agreement between the model and the observed profiles typical in this experiment. In the calculations, a ponderomotive ac Stark shift was assumed and published multichannel quantum defect theory estimates of the two-photon Rabi rates and ir photoionization rates were used [8,12]. The good correspondence between the predicted and observed ionization profile shapes, relative peak heights, and peak energy shifts corroborates the simple peak energy shift measurement of the ac Stark shift in this experiment.

As noted above, the experimental conditions were adjusted to assure that the ionization profiles were relatively insensitive to the choice of two-photon Rabi rates and ir photoionization rates. Varying these parameters by a factor of 5 from the published values produced only minor changes in the model profiles. The principal effect of varying the ir photoionization rate is to change the width of the predicted ionization profiles without significantly altering the peak energy shifts or shapes. Varying the uv two-photon Rabi rate has the same effect as changing the uv laser intensity, since these two parameters enter into the calculation as the product of the Rabi rate and laser intensity. As the uv intensity (or Rabi rate) is increased, only minor changes in the model ionization profiles are seen until the intensity becomes sufficient to saturate the two-photon transition. Then the ionization profiles broaden asymmetrically as the two-photon excitation occurs in the spatiotemporal wings of the uv laser intensity, thus sampling a larger range of ir laser intensities. Figure 5 demonstrates the onset of saturation of the two-photon transition and the attendant suppression of the peak energy shift, which could lead to incorrect inference of the ac Stark shift if only the peak energy shifts were considered. Such asymmetric profiles were not seen in this experiment, and, as noted above, direct investigation of the uv intensity dependence showed the two-photon transitions to be unsaturated.

Probably the greatest technical challenges in direct measurement of Stark shifts using schemes similar to this experiment are the accurate characterization of the laser intensities and the optimal alignment of the two lasers. Depending on the relative sizes of the two laser beams in the interaction region, small misalignments of the beams result in reduction of the apparent ac Stark shift as the two-photon excitation occurs in a region of lower ir laser intensity. Such misalignments may produce good linear dependence of the ionization profile peak energy shifts on the perturbing laser intensity, although the apparent ac Stark shift may be significantly reduced from the ponderomotive value. Somewhat surprisingly, these misalignments need not be evidenced by obvious distortion of the ionization profiles. In Fig. 6, ionization

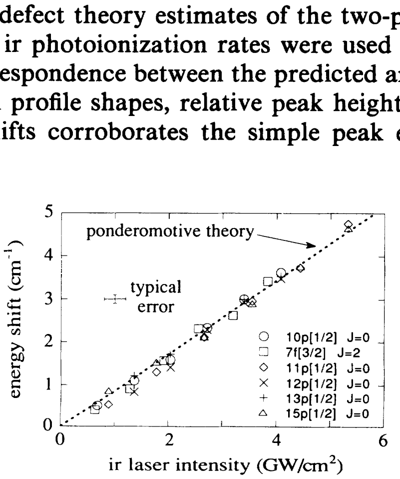

FIG. 4. Sample ac Stark shift data for six Rydberg levels in Xe. Each datum point is the average of three consecutive scans at a given ir laser intensity. The dotted line shows the ponderomotive shift.

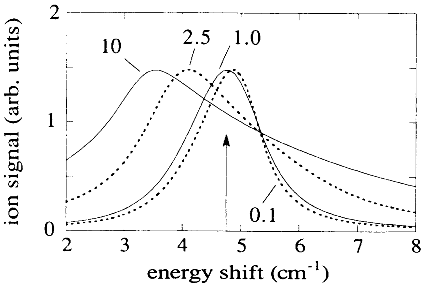

FIG. 5. Predicted ionization profiles assuming a ponderomotive ac Stark shift at fixed ir intensity (5.6 GW/cm^2) as the uv laser intensity varies, showing the effects of saturation of the two-photon transition. The number labels indicate the relative uv intensity for each curve (arbitrary units).

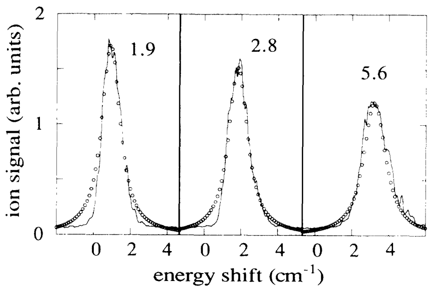

FIG. 6. Experimental resonance ionization profiles (solid lines) for the $5p^5(^2P_{3/2})10p[1/2]_0$ level of Xe with the uv and ir lasers intentionally misaligned to give an apparent Stark shift of 0.29 times the ponderomotive shift. The solid lines show the predicted profiles assuming (incorrectly) optimal alignment of the uv and ir lasers but an intrinsic ac Stark shift of 0.29 times the ponderomotive shift. The ir intensity (GW/cm^2) is shown by each curve.

profiles recorded with the uv and ir lasers intentionally misaligned are compared with predicted profiles. For this particular misalignment, the apparent ac Stark shift was ~ 0.25 cm^{-1}/(GW/cm^2), about 29% of the ponderomotive value. In the model, the alignment of the uv and ir lasers was considered optimal (contrary to the actual alignment), but instead of a ponderomotive Stark shift the reduced apparent value (29% of ponderomotive) was used. The agreement between the experimental profiles and the purposely miscalculated profiles demonstrates that the shapes of the ionization profiles are unreliable indicators of possible misalignments of the lasers—misalignment sufficient to cause a drastically lowered ac Stark shift can still generate rather "normal" appearing ionization profiles. The amount of misalignment that can be tolerated before the ionization profiles become obviously pathological depends partly on the relative sizes of the laser beams. (For any single-peaked spatial intensity distribution of the ir laser, misalignment will result in a reduced apparent ac Stark shift.) The difficulty in accurately characterizing the laser spatiotemporal profiles and in assuring optimal overlap of the lasers may explain some of the previously reported below-ponderomotive measurements of ac Stark shifts. Davidson *et al.* [10] discuss additional possible sources of error, including saturation effects due to rapid temporal fluctuations in laser beam intensity profiles using multimode (unseeded) lasers. The measurement of ponderomotive ac Stark shifts reported here should restore confidence in the simple ponderomotive picture for many atom-laser interactions in moderately strong fields (up to 10^{10} W/cm^2).

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