Photodetachment Threshold Shift in a Strong Laser Field

R. Trainham, G. D. Fletcher,[a] N. B. Mansour,[b] and D. J. Larson
Jesse W. Beams Laboratory of Physics, Department of Physics, University of Virginia, Charlottesville, Virginia 22901
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A shift in a continuum threshold due to the presence of a strong laser field has been directly observed. An increase in the photodetachment threshold of Cl^- was observed in the presence of 1064-nm-wavelength radiation.

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Light shifts in photoabsorption processes play an important role in many circumstances and have been the subject of numerous investigations. The ac Stark effect was first studied in microwave spectroscopy of molecules many years ago,[1] and has since been investigated in optical transitions.[2] Start shifts caused by blackbody radiation have been measured as well.[3] In addition, the effect of an oscillating electromagnetic field on a free electron, giving rise to the field gradient or ponderomotive force, has been discussed[4] and observed.[5] Threshold shifts due to the ponderomotive potential have been invoked to explain the disappearance of the lowest-energy photoelectron peaks in multiphoton ionization of rare gases.[6-10] Models which do not include threshold shifts have also been proposed to explain the observed electron energy distributions. Despite the interest in the behavior of atomic systems in strong laser fields and the likely importance of threshold shifts in many situations, previous experiments have provided little explicit evidence for threshold shifts due to strong oscillating fields. Implicit evidence comes from several experiments on multiphoton ionization and from the experiment of Liberman, Pinard, and Taleb, who measured the shift between the ground state and a Rydberg state of the rubidium atom.[11] More evidence comes from the recent multiphoton photoionization experiments of Freeman *et al.* with picosecond pulses.[12] In this Letter, we discuss the contributions to a continuum threshold shift and present a direct observation of such a shift due to a strong laser field. An increase in the photodetachment threshold of Cl^- was observed in the presence of 1064-nm radiation from a pulsed neodymium-doped yttrium aluminum garnet (YAlG) laser.

There are good reasons for the use of negative ions in the study of the effects of strong laser fields. The negative ion differs from the neutral in its lack of bound excited states. Such states can provide difficulty in experimental determinations of the location of photoionization thresholds because they allow excitation below threshold and subsequent ionization due, for example, to collisions or stray electric fields. These states can also make theoretical calculations more difficult.[13,14] A first-order approximation to a negative ion is an electron bound in a zero-range potential, a convenient model for the consideration of the addition of strong laser fields.[9,15] Thus the negative ion is an attractive atomic system in which to measure and understand the effects of strong optical fields.

The Penning ion-trap apparatus used in these experiments has been described previously.[16] Cl^- ions were produced in the trap by dissociative attachment of low-energy electrons to CCl_4 gas which was leaked into the vacuum system at a pressure of less than 10^{-8} Torr. Photodetachment was observed by measurement of the number of trapped ions before and after 343-nm ultraviolet light pulses were sent through the trap. The 343-nm pulses were produced by frequency-doubling pulses of 686-nm wavelength from a Nd:YAlG-pumped tunable dye laser. The data consisted of the ratios of the ion signals before and after photodetachment. To observe the effect of a strong laser field on the detachment cross section, 1064-nm pulses of high intensity from the Nd:YAlG laser were also sent through the trap and overlapped in space and time with the ultraviolet light. The infrared pulses were larger in both spatial and temporal extent than the ultraviolet pulses. Thus the ultraviolet light was used to probe the detachment cross section which was modified by the intense infrared light.

Two types of data sets were taken. The first consisted of the measured ion depletion ratios as a function of ultraviolet (uv) frequency in the vicinity of the photodetachment threshold. All of the data unambiguously show a reduction in the detachment rate near threshold due to the infrared (ir) light. This is interpreted to be an intensity-dependent shift in the threshold position, with broadening due to the spatial and temporal variations of the ir intensity and, hence, the threshold shift. Data with and without the ir light were fitted separately by a function which assumed Wigner-law s-wave detachment and included a broadening factor to take into account laser, motional, and other linewidths. Laser linewidths near 1 cm^{-1} were responsible for most of the broadening observed in the absence of the infrared pulses. The linewidth was large enough so that the structure in the cross section due to the magnetic field of the trap did not need to be considered.[17] The fits resulted in a value for the photodetachment threshold with a typical uncertainty of less than 0.1 cm^{-1}. An example of this type of

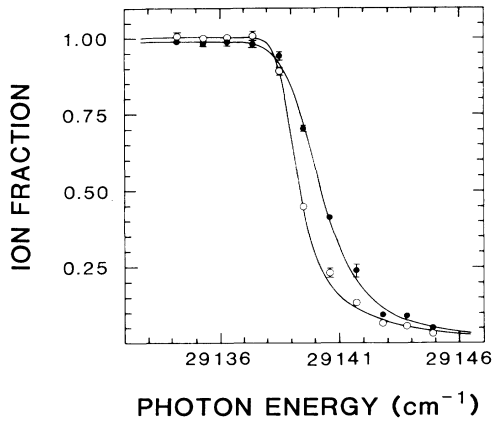

FIG. 1. Photodetachment thresholds of Cl^- showing the effects of an intense 1064-nm infrared pulse. The fraction of ions surviving illumination by ultraviolet light with a frequency near the photodetachment threshold is plotted as a function of that frequency. The open circles are points taken without the infrared present and the filled circles are points taken with the infrared present. The solid lines show the results of least-squares fits by the Wigner threshold law convolved with a Gaussian width function. Both the width and the threshold energy are increased for the data taken with the infrared light present.

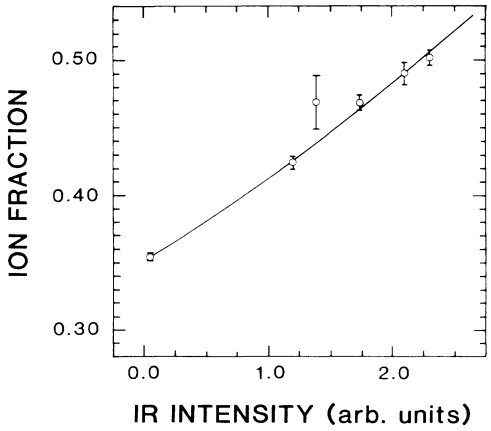

FIG. 2. Fraction of Cl^- ions surviving as a function of infrared pulse intensity for a fixed ultraviolet frequency. For these data the frequency of the ultraviolet light was set about 1 cm^{-1} above threshold and the infrared pulse energy was varied. The data are fitted by the threshold function shown in Fig. 1 with a threshold position which is assumed to increase linearly with infrared intensity.

data set is shown, together with the results of the fits, in Fig. 1. The peak ir intensity was near 10^{10} W/cm^2 for those data. The fits give a threshold shift of 0.96(13) cm^{-1} and an increase in broadening of 0.66(18) cm^{-1}. The average shift for data of this type is 1.7 cm^{-1} at 10^{10} W/cm^2.

The second type of data set was taken at a fixed uv frequency. The frequency of the uv light was selected to be just above threshold in the absence of the infrared pulse. The ion depletion fraction was measured as a function of the intensity of the ir pulse. An example of this type of data set is shown in Fig. 2. The data show an increase in ion number as the ir intensity is increased, i.e., a decrease in the detachment rate with increasing ir intensity. The data are fitted by a broadened Wigner-law s-wave threshold with a shift of the threshold position linear in ir intensity. Some of the data of this type show a reduction in slope and even a decrease in ion number at the higher intensities. Possible explanations for this effect include two-color two-photon detachment, loss of ions due to the ir beam's grazing a trap electrode, and misalignment of the ir beam at the higher intensities. In any event, all five of the data sets of the type shown in Fig. 2 exhibit similar initial slopes consistent with threshold shifts in the range of 2 to 3 cm^{-1} at an intensity of 10^{10} W/cm^2. Thus both types of data are consistent with the assumption of a threshold shift linear in ir intensity with a value near 2 cm^{-1} at an intensity of 10^{10} W/cm^2.

The major experimental uncertainty in the numerical value for the threshold shift is due to uncertainty in the ir pulse intensity at the position of the uv pulse. The ir intensity was obtained from measurements of the average power, the pulse length, and the size of the focused spot. The average power was measured with a thermal power meter, and the total power available in the ir beam was typically about 1 W at a repetition rate of 20 Hz. The pulse length was measured with a fast photodiode. For the data of Fig. 1, the size of the focused spot was measured to be approximately 110 μm, much larger than the 50 μm measured for the uv photodetaching beam. The overlap of the two pulses in the trap was adjusted by our setting the uv frequency to be above, but near, threshold and observing the change in the ion signal as the position of the ir beam was changed. The uncertainty in the ir intensity comes from the uncertainties in pulse energy, length, and area, and possibly from the issue of pulse overlap. In addition, there are questions associated with the spatial and temporal structure in both pulses. The laser output has temporal fluctuations within the pulse, leading to large fluctuations in the threshold shift during the pulse. Even though the shift should be linear in intensity, it may not be reasonable to expect an observed shift corresponding to the average intensity. For example, the observed detachment could be largely occurring during the times in the pulse when the ir intensity is low. If we do not attempt to account for the spatial and temporal substructure within the pulses and use the measured values for the pulse parameters together with the observed shifts, we obtain an average value from all the data for the threshold shift of 2 cm^{-1} at 10^{10} W/cm^2.

The ponderomotive energy of a free charge is the aver-

age energy associated with the oscillatory motion of the charge in a rapidly oscillating field. The ponderomotive energy for a free particle comes entirely from the square of the vector potential, **A**, when $\mathbf{p}\cdot\mathbf{A}$ and A^2 are used to calculate energies to second order in the applied field. Since A^2 is independent of the relative atomic coordinates in the dipole approximation, and therefore this term contributes equally to the electron in the ground state and in the continuum, one might be tempted to conclude that there is no ponderomotive shift of the threshold. This conclusion is wrong if the ground-state orbital frequency is large compared with the frequency of the field's oscillation. In this case of a "low-frequency" field, the bound electron has a much reduced energy of oscillation. Algebraically, this result appears as a near cancellation of the A^2 term by the $\mathbf{p}\cdot\mathbf{A}$ term taken in second order.[18] The sum of these two terms is simply a term proportional to the ordinary polarizability of the ground state, i.e., it is the same as the result obtained by taking $\mathbf{F}\cdot\mathbf{r}$ in second order, where **F** is the oscillating electric field.[19] Such a term causes a decrease in the ground-state energy in the presence of the field and thus adds to the shift contributed by the ponderomotive energy of the free electron.

A simple conservation-of-energy argument serves to identify the various contributions to the total threshold shift.[10] This argument applies when the pulse lengths are long compared with the time for motion of the electrons out of the field. Consider the negative ion outside a region of oscillating electric field, **F**, whose frequency is low compared with the ground-state orbital frequencies for both the ion and the atom. The total energy required to move the ion into the field, separate the electron from the atom, move the electron and atom out of the field, and recombine them must be equal to zero. To move the ion into the field requires the supply of an energy equal to the ponderomotive potential for the ion, U_p(ion), minus the energy due to the ac Stark shift of the ion ground state, ΔS(ion). The ponderomotive force pushes the ion out of the field while the negative ground-state Stark shift pulls the ion into the field. To separate the electron from the atom requires energy equal to the electron affinity in the presence of the oscillating field, $A_e(F)$. The electron is pushed out of the field, with a net increase in energy of U_p(electron), and the atom must be pulled from the field due to its negative ground-state Stark shift, ΔS(atom). Finally, the recombination of the electron and atom results in a net energy equal to the electron affinity, $A_e(0)$. The net work done is

$$U_p(\text{ion})-\Delta S(\text{ion})+A_e(F)-U_p(\text{electron})+\Delta S(\text{atom})-A_e(0)=0,$$

where all the quantities are taken to be positive. Rearrangement of terms gives the threshold shift due to the oscillating field,

$$A_e(F)-A_e(0)=U_p(\text{electron})-U_p(\text{ion})+\Delta S(\text{ion})-\Delta S(\text{atom}).$$

The ponderomotive energy for a charge q of mass M in a linearly polarized field of strength F and frequency ω is $U_p=q^2F^2/4M\omega^2$. Since the ion has a much larger mass than the electron, U_p(ion) can be neglected in comparison with U_p(electron), leaving a threshold shift equal to the ponderomotive energy of the electron plus the difference between the ac Stark shifts of the ion and atom ground states. This is the same as the result of Ref. 11 for the shift between the ground state and a Rydberg state of the rubidium atom, except that Ref. 11 does not explicitly include the Stark shift of the core, which corresponds to ΔS (atom) in the present case.

The ponderomotive energy of the electron has a value of 1 eV at 10^{13} W/cm^2 of 1064-nm light, or 8 cm^{-1} at 10^{10} W/cm^2. The static polarizability of the Cl atom is 2.18×10^{-24} cm^3.[20] Since the frequency of the ir light is low compared with the excitation frequencies for the Cl atom, use of the static polarizability should provide a reasonable estimate of the shift expected in the presence of the ir pulse. At 10^{10} W/cm^2, this polarizability corresponds to a shift of only 0.2 cm^{-1}. Dalgarno[21] has estimated the static polarizability of Cl^- to be approximately 3×10^{-24} cm^3. The polarizability of the negative ion is less than that of a neutral atom with comparable ionization energy since the oscillator strength is all above threshold. Again, since the ir frequency is small compared with the frequency of the detachment threshold, the static polarizability should provide a reasonable estimate for the negative ion, resulting in a shift of 0.3 cm^{-1} at 10^{10} W/cm^2. Combining these terms, we estimate that the threshold shift should be slightly more than 8 cm^{-1} at 10^{10} W/cm^2. This is several times the size of the observed shift. While some possible experimental problems, such as overlap of the ir and uv pulses, would result in measured values smaller than the real shift, it is very unlikely that the difference could be as large as the apparent discrepancy between the measured and predicted shifts. It seems quite clear, however, from the conservation-of-energy argument and from the experiment of Liberman, Pinard, and Taleb[11] extrapolated to the continuum limit, that the threshold shift should be greater than the ponderomotive energy of the free electron, at least in an experiment with continuous optical fields.

In summary, the experiment demonstrates the existence of a threshold shift in the presence of a strong laser field. The applied field causes an increase in the detachment energy, as expected, but the magnitude of the observed shift is less than that expected, possibly because of experimental circumstances such as the use of laser pulses with spatial and temporal substructure. Fu-

ture experiments with well controlled and more intense laser pulses should provide significant new data.

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[a]Present address: Lawrence Livermore National Laboratory, L-401 P.O. Box 808, Livermore, CA 94550.

[b]Present address: Physics Division, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439.

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