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s-wave photodetachment from S^- ions in a static electric field

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Photodetachment from negative S ions has been studied in external electric fields up to 220 V/cm for laser polarization parallel to the electric field direction. Fast neutrals produced by a pulsed laser beam crossed with a 4 keV S^- ion beam were detected to measure the relative cross sections. Increased ion currents, lower background signals, analog detection methods, and more precise wavelength calibration have produced higher quality data than previous direct cross-section ratio measurements on nonhydrogenic ions. The ratio of the electric field-on to electric field-off cross sections is found to be in excellent agreement with theoretical predictions, thus resolving a long-standing discrepancy with previous, less definitive experiments. As predicted, electron wave-function rescattering effects are very small in both the amplitude and the phase of the electric-field-induced oscillations.

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During the past two decades, electric field effects on photodetachment cross sections have been of great interest both theoretically and experimentally. Calculations began with simple models that treated the negative ion as an electron bound in a short-ranged potential and the detached electron as a free electron in the electric field and have proceeded to become more sophisticated [1–7]. In this picture the negative ion system provides essentially a one-dimensional electron wave-function interferometer. The two main effects of the electric field on the photodetachment cross section are below threshold tunneling through the now finite potential wall and amplitude oscillations due to interference above threshold. Early experimental investigations began in 1987 when Bryant *et al.* used a motional electric field to investigate the effects on the H^- photodetachment cross section [8]. Publication of this important work led Greene and Rouze [9] to explain oscillations previously observed on the Rb^- cross section [10,11] as being due to an electric field. Further experimental work on H^- [12,13] was compared to theory and found to be in qualitative agreement, however, quantitative comparisons were not made [14–20]. The present experiments make a direct, quantitative test of theory.

During the early 1990's, four experiments were carried out to investigate photodetachment from nonhydrogenic ions in electric fields. Photodetachment from S^- and Cl^- was performed in a standing-wave microwave cavity that exposed the ions to a range of electric fields that were constant on the time scale of the detachment [21,22]. Possible discrepancies between the Cl and S data led to photodetachment experiments in truly static electric fields [23,24]. To within the experimental uncertainty, *p*-wave photodetachment from Au^- was found to be in agreement with theory [24]. The periodicity of *s*-wave oscillations for S^- and Cl^- also appeared to agree with theory. However, the measurements showed a 20% reduction in the amplitude of the interference oscillations for the *s*-wave case, specifically for S^-, which was suggested to be due to rescattering effects [23]. More recently, tunneling and cross-section modulations due to stray electric fields have been observed in high-resolution near-threshold photodetachment spectra by Haugen's group at McMaster University. [25]

Electric field effects on photodetachment have been investigated using a different and complimentary approach in a beautiful experiment using a photodetachment microscope [26] to record the two-dimensional interference rings produced by *s*-wave photodetachment from O^- in static electric fields of 4.23 V/cm [27,28]. Blondel and co-workers conclude that photodetachment from O^- in an electric field of a few volts per centimeter, within 2 cm^{-1} of threshold, is free from rescattering effects. However, they note that the rescattering of the detached electron's wave function may be more important for a larger ion such as S^- [27].

For the relatively low (on the atomic scale) electric fields investigated here, most theoretical analyses reduce to similar results independent of the complexity of the model. The present experiment is directly compared with the results of Fabrikant [29], which specifically addressed photodetachment and rescattering effects for S^- at electric fields of the same magnitude as used here. The photodetachment cross section in an electric field of strength F is given by [19,29]

$$\sigma_F = H(E,F)\,\sigma_{F=0}, \tag{1}$$

where $H(E,F)$ is a modulation factor that oscillates with both F and the final-state electron energy E. For the present case of photodetachment from S^-, *s*-wave detachment is by far the dominant channel near threshold; therefore, the cross section in zero electric field is given by the Wigner law as $\sigma_{F=0} \propto E^{1/2}$. In the absence of rescattering effects, the modulation factor for *s*-wave detachment is given in atomic units by [19]

$$H(E,F) = (\pi/\eta^{1/2})[(\mathrm{Ai}')^2 + \eta\,\mathrm{Ai}^2], \tag{2}$$

where $\eta = (2^{1/3}E)/F^{2/3}$, Ai is the Airy function, and the argument of both Ai and its derivative Ai' is $-\eta$. Fabrikant [29] explicitly evaluates the changes in the modulation factor H due to final-state electron-atom interactions for S^- in an electric field of 1 kV/cm based on scattering lengths. The calculations indicate that rescattering causes a relative change of H of at most $\sim$1.6% at zero energy, and that the deviation rapidly decreases to less than 1% as the electron energy increases above 2 cm^{-1}. This relative deviation is far too small to account for the reduction of the oscillation amplitude observed by Gibson *et al.* [23], thus leaving a significant discrepancy between the best previous theoretical and experimental results. The present experiments were designed to produce higher quality cross section measurements to further investigate this long-standing discrepancy.

These measurements were made using a crossed laser and ion-beam apparatus. Negative ions were formed from CS_2 support gas in a Colutron ion-source Model 100-*Q* and accelerated to 4 keV. After focusing for collimation, the ions were mass analyzed to select only isotope ^{32}S using a 45° bending magnet. Approximately 1 m away in the detection chamber, the circular ion beam was apertured to approximately 3 mm in diameter and a 1 cm horizontal ion-beam deflection was employed immediately before the interaction region in order to remove neutrals produced by background gas stripping. Background gas pressures were typically in the 1×10^{-7} Torr range. About 18 nA of mass analyzed S^- was typical for data collection.

In the center of the interaction region, a laser beam intersected the ion beam at an angle of $90°\pm2°$. Following the interaction region, remaining negative ions were deflected into a Faraday cup, while neutral atoms continued on undeflected. Direct detection of neutral atoms was accomplished by using an ETP AF150 electron multiplier detector and standard time-of-flight techniques. The best signal-to-noise ratio was obtained running the detector in analog mode and then using an SRS SR440 amplifier to input the signal into a LeCroy LC534AM 1 GHz oscilloscope operating at 500 megasamples per second. The scope was operated as a gated integrator and boxcar averager, with the photodetachment signal determined by subtracting the average background voltage from the integrated amplifier output voltage during the time window for arrival of photodetached neutrals. The LABVIEW programming language was used to interface the computer to the scope, the laser power meter and the picoammeter monitoring ion-beam current.

The interaction region was carefully constructed to have a uniform electric field throughout the entire region that ions interact with the laser. The interaction region was defined by two parallel stainless steel plates with apertures in the center in order to pass the beam normal to their surfaces. A positive voltage was applied to the first field plate and the second field plate was biased to an equal negative voltage such that the center of the interaction region was at ground potential. A series of grounded plates prevented the electric field from leaking out in any direction and provided more uniformity to the field in the interaction region. Models of the interaction region using the SIMION program showed the uniformity of the electric field to be better than 2% and the strength of the field was determined to be the applied voltage divided by the distance between the plates (1.016 cm) multiplied by 0.9385.

The two-part laser system was composed of a Coherent Infinity 40-100 Nd:YAG (yttrium aluminum garnet) pulsed pump laser operating at 50 Hz and a Lambda Physik ScanMate OPPO tunable laser. The ScanMate uses the tripled Nd:YAG fundamental to pump both a grating tuned seed dye laser and a nonlinear optical crystal. For every 355 nm pump photon mixing with the seed beam in the crystal, the output consists of a visible photon of the seed wavelength and a complementary ir photon such that the two output photon energies sum to the input photon energy. The desired output beam is separated from the second output using a combination of optical alignment and glass absorption filters. The laser light was sent into the interaction region linearly polarized parallel to the applied electric field. Laser diagnostics were performed using a Burleigh WA-4500 pulsed wavemeter. A wavelength calibration accuracy of 0.003 nm was achieved by continuously monitoring 3% of the main laser beam and the measured laser linewidth was 0.1 cm^{-1}. The pulsed wavemeter also provided a method for ensuring that the mixing crystal was properly tuned for maximum seeding. This diagnostic was crucial for ensuring that no unseeded, broadband light was propagated into the interaction region with the narrow linewidth, seeded laser light. The laser was coupled into the vacuum system using two wedged brewster angled windows to minimize reflections and eliminate etalon effects. Typical average laser power into the interaction region was 30–60 mW which put on the order of 1 mJ per pulse [3 ns FWHM (full width at half maximum)] into a 3 mm by 2 mm laser spot.

Cross-section ratios were measured at each wavelength by collecting data for 400 laser pulses with the electric field-on and then with the field-off, normalizing each signal to the respective laser power and ion current, and then dividing the two normalized signals. This direct ratio method is the most precise way to measure the effects of the electric field since the field-on and field-off measurements are taken successively over a short time period of $\sim$20 sec, thus minimizing uncertainties due to changes in the laser and ion beams, fluctuations in the beam overlap, and changes in the detector efficiency. Cross-section ratios presented here are the average of 19 scans across the photon energy range. The experimental system was dynamically tested by measuring the ratio of field-on to field-off cross section far above threshold, where the interference oscillations should damp out. The modulation factor H was measured to be 1.00 ± 0.01 at 44 cm^{-1} above the zero-field threshold in electric fields up to 400 V/cm, in agreement with expectations. As a further test of the system, the zero-field threshold for the lowest energy transition ($^2P_{1/2}\rightarrow{}^3P_2$) was determined by a Wigner law fit to the field-off cross-section data; the measured threshold of 16 269.34(30) cm^{-1} agrees with the more accurate accepted value of 16 269.426(13) cm^{-1} [30].

For illustrative purposes, the full photodetachment cross section in an electric field can be displayed by matching direct measurements of the field-on cross section below the zero-field threshold together with the above threshold cross sections obtained by multiplying the measured H ratios by the Wigner law, Eq. (1) (Fig. 1). The data show both the interference oscillations and the tunneling below the zero-field threshold in complete agreement with the theoretical calculations of Fabrikant [29].

The measured modulation factor H as a function of photon energy for S^- at an applied electric field of 220.5(4.0) V/cm is shown in Fig. 2, together with a fit of the theoretical formula Eq. (2), in which the field strength was taken as the

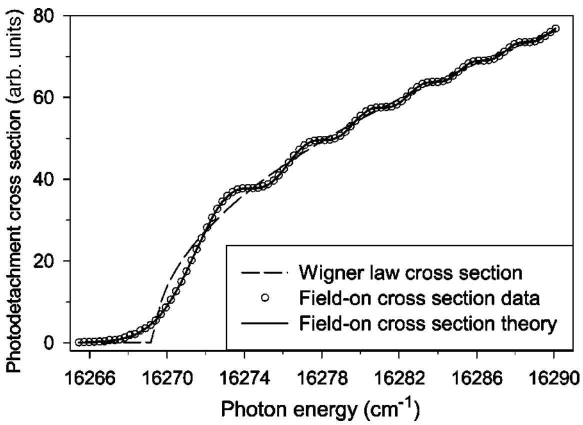

FIG. 1. Photodetachment cross section in a static electric field. The circles show the data scaled to the Wigner law above the zero-field threshold and scaled to the cross-section magnitude measurements below threshold. The dashed line is the Wigner law field-off cross section scaled to the data far above threshold and the solid line is the theoretical curve in an electric field [29].

only adjustable parameter. The data is in excellent agreement with theory without the inclusion of rescattering for the amplitude, phase, and periodicity of the interference oscillations. The value of the electric field from the least-squares fit to the data is 220.9(3.0) V/cm, which is within 0.2% of the applied electric field. More importantly, the amplitude of the measured oscillations matches very closely the theoretical curve, with an overall deviation of less than 0.3% over the entire energy range. The results confirm Fabrikant's theoretical conclusion [29] that rescattering effects are very small for photodetachment from S^- in electric fields up to several hundred volts per centimeter, causing less than a 1% change to the amplitude of the first interference minimum at only 0.7 cm^{-1} above the zero-field threshold. The previous discrepancy between earlier experiments [23,24] and theory must now be explained.

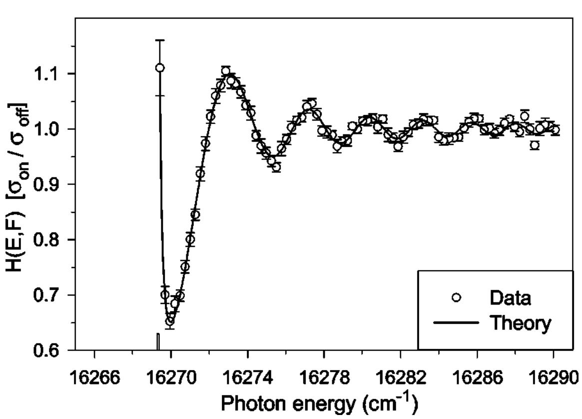

FIG. 2. The *H* factor is the ratio of the cross section measured in the electric field to the cross section measured with no field. The circles denote directly measured cross-section ratios with statistical uncertainties and the solid line is the theoretical fit to the data, Eq. 2. The vertical rectangle on the *x* axis shows the location of the zero-field threshold and its width denotes the relative experimental uncertainty.

One potential explanation for the reduced oscillation amplitude in previous experiments is the possible nonuniformity of the electric field. The simulations associated with this study show how critical it is to model the electric field uniformity during the design of the interaction region and how easily the field can vary across the size of the laser spot. Ions exposed to a range of different electric fields would be subject to varying phases for their oscillations and summing of these oscillation packets results in an electric-field-averaged *H* factor with reduced amplitude, just as was previously observed [23].

A second potential explanation for the reduced oscillation amplitude in previous experiments is nonlinearity of the complete detection system due to saturation of the pulse counting detector or saturation of the photodetachment process. Even if careful checks to the linearity were performed in a steady-state fashion, neutral bunching caused by laser detachment in the field region could account for much of the reduced oscillation amplitude. Due to the static electric field, neutral atoms are produced from negative ions at a range of electric potentials, thereby changing their kinetic energies. S atoms produced upstream of the center of the laser pulse have greater kinetic energy than those produced downstream of the center thus reducing the total arrival time window at the detector. In the present experiment, pulse compression reduced the main signal arrival window by 1/3 to 1/2 relative to the field-off case; higher time-resolution analog data (2 ns) have been critical in documenting this effect. The pulse counting detection method used in the previous experiments [23], with 50 times lower time resolution, was unable to distinguish these neutral bunching effects, which may have affected the measured oscillation amplitudes. Thus, linearity checks may have been required over a greater dynamic range.

In summary, the results of this study fully support theoretical predictions for the electric field effects on the photodetachment cross sections of negative ions. Both the phase and the amplitude of the oscillations above threshold are in excellent agreement with the theory. Since near threshold rescattering effects scale as $F^{1/3}$ [29], possible rescattering effects due to the 220 V/cm fields in this study should be at least four times larger than those due to the 4 V/cm field used by Blondel *et al.* for O^- [27], lending support to their conclusion that rescattering effects are not significant in their measurement. We carefully performed fits to the data in order to compare theory and the directly measured field-on to field-off cross-section ratios. For the present case of the laser polarization parallel to the electric field, the agreement between the theory without the inclusion of rescattering and the data is so good that the results of these fits are essentially indistinguishable from direct calculations of the expected effects using our initial zero-field threshold and electric field measurements.

Future studies are expected to include checks for electric field effects on photodetachment from heavier negative ions such as I^-, for which the larger scattering lengths may cause greater rescattering, and observations of electric field effects on resonant structures in photodetachment cross sections. In particular, polarization-dependent, high-resolution studies very near threshold could probe for possible rescattering effects due to higher electric fields, since even though the main *s*-wave electric field effects are not polarization dependent, the rescattering effects are predicted to be [29].

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