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Observation of Full Ponderomotive Shift for the Photodetachment Threshold in a Strong Laser Field

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Electron-energy-resolved measurements of the threshold shift of negative chlorine ions have been performed under the influence of a strong (4.5×10^{13} W/m^2) infrared laser field. The threshold shift is found to be in accordance with a full ponderomotive shift. Explanations are proposed for previously measured smaller-than-expected threshold shifts.

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In an electromagnetic field the average kinetic energy of a free electron is raised by $U_p = e^2E^2/4m\omega^2$ (E and ω are the strength and frequency of the light field and e and m are the charge and mass of the electron, respectively). Because of this so-called ponderomotive energy the minimum energy needed to detach an outer electron from an atom or ion is increased. Despite several years of research by various groups there is still a controversy concerning this light-induced energy shift. Trainham *et al.* [1] were the first to directly observe such a threshold shift in a negative ion. In their experiment the behavior of the detachment threshold under the influence of a strong infrared laser field was probed by means of ultraviolet light. The magnitude of the shift they found, however, was only 25% of the expected value. In a later experiment by Baruch, Gallagher, and Larson [2] a microwave field was used as the shift-inducing field. This time, in the presence of the microwave field, no increase in the UV-photon energy needed to reach the threshold was observed at all. Therefore it was concluded that no threshold shift had occurred. As a result of these experiments Bloomfield [3,4] performed calculations which showed that smaller threshold shifts could indeed be expected when the frequency of the shift-inducing field is very low. In an electromagnetic field with very low frequency, the excursion of the electron, forced by the field, would be so large compared to the size of the unperturbed orbit that the electron could become "lost" on its way, instead of becoming reattached in the next half cycle of the field. This effect was called "leakage detachment" below the shifted threshold. Another explanation which has been given for the observed effects, in the limit of very low frequencies, is that the field hardly changes on the time scale in which the detachment process takes place. In that case detachment takes place in an essentially constant field and, therefore, no threshold shift should be seen.

More claims concerning smaller-than-expected threshold shifts have been made but some of them have been attributed to experimental circumstances. Smith *et al.* [5] initially observed 30% of the expected shift of the minimum energy for two-photon detachment of H^-. Later, this group [6] measured a shift of 45% for the same physical process, then, however, attributing the difference to the uncertainty in the laser intensity.

Although the idea of leakage detachment and the notion of an essentially constant electric field seem to explain the experimental results obtained in the case of detachment in a microwave field very well, it should be noted that threshold measurements are complicated by the fact that at high light intensities absorption of excess photons in the detachment process can occur. In the work discussed so far *total* detachment yields were investigated, which means that the different detachment channels were not investigated separately. Nevertheless, claims of smaller-than-expected threshold shifts were based on the assumption that the main contribution to the total yield came from a process with a fixed number of photons; in particular, in the case of Ref. [2], from a one-photon process. Although this assumption can be valid in the case where the shift-inducing field has a high frequency, in the low-frequency limit absorption of (excess) photons from the shift-inducing field will certainly occur. In the latter case the average number of absorbed photons is accurately described by classical arguments [7], and given by $U_p/\hbar\omega$ (with $\hbar\omega$ the photon energy). This means that the total energy gained by absorption of photons from the shift-inducing field exactly equals the expected shift of the threshold, U_p. Therefore, the conclusion which should be drawn from the microwave experiments is not that no threshold shift has occurred, but that in the limit of very low frequencies the extra energy needed to detach the electron is supplied by the shift-inducing field instead of the probing field. One has to perform electron-energy-resolved measurements to directly observe the absorption of the low-frequency photons. In the case of detachment by a one-color radiation field with a low frequency ($\hbar\omega < U_p$) the situation would be similar. The ponderomotive shift makes detachment by the minimum number of photons energetically impossible, but the absorption of excess photons is still allowed. The result of the ponderomotive shift would be the disappearance of the low-energy peak(s) in the electron-energy spectrum. This effect of channel closure is well known from multiphoton experiments on atoms in the nonperturbative regime [8].

In the high-frequency limit, where the detachment pro-

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cess is dominated by the lowest-order channel, the extra energy needed for the forced oscillation of the electron has to be supplied by the UV photon. In the experiments by Trainham *et al.* [1] the energy of the infrared photons is much larger than the threshold shift and, therefore, one expects the experiments to be an example of the high-frequency case. Nevertheless, a deviation was found from the expected value of the threshold shift.

In order to shed some light on the questions raised above, we have repeated the experiments by Trainham *et al.* [9], but this time in an electron-energy-resolved measurement. In this way we could separate the different detachment channels. In addition, we used a seeded Nd-doped yttrium-aluminum-garnet (Nd:YAlG) laser to minimize experimental uncertainties.

In the experiment negative chlorine ions are held in a Penning ion trap [10] which has been integrated in a magnetic-bottle electron spectrometer [11]. Since this setup has been described in detail elsewhere [12], we only give a brief discussion. About a few thousand ions are confined in a cloud of about 1 mm^3. The background pressure is 1×10^{-6} mbar. The produced photoelectrons are energy analyzed on the basis of their time of flight over a 50-cm long flight path. To measure the position of the threshold for detachment from the negative ions, a home built tunable dye laser was used which was pumped by the frequency-doubled output of the seeded Nd:YAlG laser. The Nd:YAlG laser produced pulses with a wavelength of 1064 nm at a repetition rate of 30 Hz. The energy of the infrared pulses was around 400 mJ and the energy of the frequency-doubled pulses around 150 mJ. The dye laser consisted of a Hänsch-type oscillator and six amplifiers, using Piridin 1 dissolved in methanol as a gain medium. The output of the dye laser was frequency doubled in a KDP crystal to obtain 342-nm light with pulse energies up to 1.3 mJ and a bandwidth of 1 cm^{-1}. The absolute wavelength of the UV light was measured with a monochromator to an accuracy of 0.025 nm. The size of the wavelength steps, made by turning the grating of the dye laser by means of a stepping motor, was measured with etalons and determined to be 0.53(5) cm^{-1}. The UV light was focused in the center of the Penning trap with a lens of 25-cm focal length. Using a dichroic mirror the IR light was focused in the trap from the same side with a lens of 50-cm focal length. Spatial overlap of the foci was found by reflecting the light before entering the spectrometer, placing a pinhole at the position of the UV focus, and optimizing the transmittance of the IR light through the pinhole. The size of the foci was determined by measuring the transmittance of the beams through various pinholes as well as by scanning a 35-μm pinhole through the focus in the case of the IR light and a 12.5-μm pinhole in the case of the UV light. The profiles of the foci are found to be nearly Gaussian. Pulse duration and pulse overlap were measured with a fast photodiode and a 9450 LeCroy digital oscilloscope. The response of the detection system to a 35-ps laser pulse was a pulse with a width of 1.3 ns. The temporal overlap of the 343-nm and 1064-nm pulses was accurate to within 0.3 ns. The infrared pulse energy has been measured with a Scientech AC2501 volume absorber. The shot-to-shot fluctuations were less than 2%. For the energy measurement of the UV power the volume absorber was calibrated with a surface absorber. The energy of the UV laser is measured on a shot-to-shot basis and the electron spectra are binned accordingly. The energy fluctuations in one bin are about 8%.

The main experimental results are given in Fig. 1. It shows a threshold scan taken with the following experimental parameters: pulse energy UV and IR 14(2) μJ and 14.8(5) mJ, respectively; pulse duration UV and IR 3.4(2) ns and 12.9(5) ns, respectively; focus diameter UV and IR 36(4) μm and 146(5) μm, respectively. The maximum intensities of the UV and IR pulses are 2.6×10^{12} W/m^2 and 4.5×10^{13} W/m^2, respectively. The electron signal shown in Fig. 1 is energy resolved and is due to the absorption of only one UV photon. Calculations, based on the Wigner threshold law [13], have been performed and are shown in the same figure. In the calculations an integration is made over space and time. A Gaussian spatial profile is assumed and the temporal profiles of the UV and IR laser pulses are taken from measured data. A constant intensity along the beam axis is assumed, since the Rayleigh range of the laser focus is much larger than the size of the ion cloud. The theoretical calculations by Robinson and Geltman [14], corrected by the experimental values obtained by Mandl [15], are used for the cross section for one-photon detachment as a function of the photon energy. At an energy of 1 meV above the threshold σ_1 is 1.17×10^{-4} $W^{-1}m^2s^{-1}$. Apart from the ponderomotive shift, which is assumed to be $(1.06\times10^{-17}\ eV)I_{1064}$, where I_{1064} is the intensity of

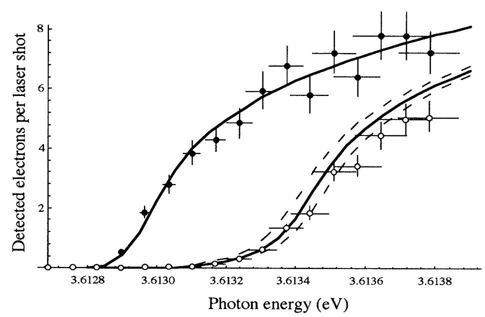

FIG. 1. Shift of the threshold for electron detachment from Cl^- under the influence of a strong infrared laser pulse. The filled circles show the electron signal after absorption of one UV photon only, as a function of the UV-photon energy without the infrared field. The open circles show the electron signal with the infrared field turned on. The solid lines show the results from calculations based on the Wigner threshold law, assuming a full ponderomotive threshold shift. The dashed lines denote the error in the calculated curve due to the uncertainty in the intensity of the infrared field.

the infrared laser field in W/m^2, there are two other, much smaller contributions to the total threshold shift which have been taken into account in the calculations. These are the ac Stark shift of the ion ground state and the neutral atom ground state. The static polarizability of the chlorine atom is 2.18×10^{-30} m^3 [16]. Since the energy of the infrared photons is far from the energies needed to excite the chorine atom, it is reasonable to use the static polarizability as an estimate for the shift at 1064 nm. This shift is $(-3 \times 10^{-19}\ \text{eV}) I_{1064}$. The theoretical values calculated by Kutzner, Felton, and Winn [17] have been used for the frequency-dependent polarizabilities of the negative chlorine ion. At 1064 nm their value is 5×10^{-30} m^3, which results in a shift of $(7 \times 10^{-19}\ \text{eV}) I_{1064}$. The calculation for the UV-only signal has been fitted to the experimental data points by adjusting the ion density, the position of the threshold, and an additional spectral broadening of the threshold. This broadening is due to a nonzero bandwidth of the laser and Doppler broadening, as a result of the velocity of the ions in the trap. On the basis of the fit the broadening is found to be about 1.4 cm^{-1}, somewhat larger than the expected 1.1 cm^{-1} on the basis of the laser linewidth and the Doppler width (estimated to be about 0.1 cm^{-1}). The same values have been used for the calculation with the IR field on. The uncertainty in this latter calculation is determined by the uncertainty in the infrared intensity, which is about 8%.

As can be concluded from Fig. 1, the experimental data are in good agreement with the calculations. It should be stressed that the small tail in the scan with the infrared field turned on is due to a tail in the temporal profile of the UV laser pulses and is not due to the leakage detachment, which has been discussed in the introduction. The experimental data are therefore consistent with a full ponderomotive shift of the threshold.

Trainham *et al.* used for their experiments the same negative ion and laser frequencies as have been used in this experiment, but nevertheless found a deviation from the expected threshold shift. We can think of a number of reasons why Trainham *et al.* observed a smaller-than-expected threshold shift.

(1) To find a threshold shift which corresponds with the maximum infrared intensity that is reached, the infrared laser pulse should vary only slightly in temporal and spatial dimensions when and where the ultraviolet laser is on. This means that the pulse duration and the focus size of the UV light have to be much smaller than the pulse duration and the focus size of the IR light. If the UV intensity is too high, strong saturation of the detachment process occurs. As a result, the production is strongly enhanced in the wings of the temporal and spatial profile of the UV laser pulses. This effect gives rise to a much larger effective production volume than one would normally predict and in this case a smaller threshold shift is found. To investigate the influence of the ratio between the focal size and pulse duration of the UV and IR laser pulses, we have performed numerical calculations in which this ratio is varied. In the calculations we used the accurate threshold energy of 3.61269(6) eV from Ref. [18]. The deviation of the unshifted threshold energy found in Fig. 1 is due to the uncertainty in the absolute wavelength calibration. The intensities of the UV and IR fields used in the calculations are 1.6×10^{12} W/m^2 and 5×10^{13} W/m^2, respectively, and strong saturation already occurs at UV-photon energies, which are only slightly larger than the threshold energy. In Fig. 2, which shows the results of the calculations, it can be seen that even if the UV pulse has half the focal size and half the pulse duration of the IR pulse, the "apparent" threshold shift is almost half the "expected" shift. Experimental data taken with a higher UV intensity and a larger focus than presented in Fig. 1 are in agreement with the calculations, which means that saturation effects are a likely candidate to explain the previously measured smaller-than-expected threshold shifts by Trainham *et al.*.

(2) The Nd:YAlG laser that has been used in Ref. [1] was unseeded, which means that the nanosecond laser pulses must have shown large fluctuations on a picosecond time scale. Since a laser dye (Piridin 1) has been used with a very fast response time to the pump pulse, it is to be expected that the dye-laser output also showed a similar temporal substructure. It is therefore very well possible that, while the UV and IR pulses were overlapping on a nanosecond time scale, fluctuations occurred in the overlap on a picosecond time scale, resulting in a broadening of the threshold shift. Note that every broadening mechanism of the threshold shift in combination with strong saturation immediately results in a smaller-than-expected value of the threshold shift.

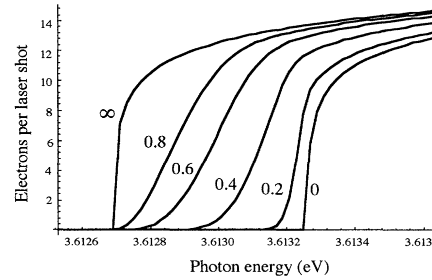

FIG. 2. Calculated electron signal when the outer electron is detached from Cl^- by means of a UV pulse under the influence of a strong IR pulse. The label of the curves indicates the values for both the ratio between the focal sizes and the ratio between the pulse durations of the UV pulses and the IR pulses, as used in the calculations. The intensities of the UV and IR fields are 1.6×10^{12} W/m^2 and 5×10^{13} W/m^2, respectively. Although the IR intensity stays the same, the apparent threshold shift becomes smaller when the duration and the focal size of the UV pulses are increased. A ratio of zero gives a threshold shift which corresponds to the maximum IR intensity and a ratio of infinity gives the unperturbed threshold.

(3) In the experiment the spatial overlap was optimized by minimizing the detachment rate of the negative ions when the UV-photon energy was just above the threshold for detachment. Since in the experiment by Trainham *et al.* ion yields were measured, no distinction could be made between detachment by means of one UV photon and detachment by means of one UV photon and one IR photon. Finding the spatial overlap is therefore complicated by the effect that minimizing the detachment rate by means of one UV photon also means optimizing the rate of detachment by means of one UV photon and one IR photon [19].

It has been predicted that in the vicinity of the threshold for N-photon absorption cusp structures can occur in the cross sections for the higher-order $(N+n)$ processes [20]. These so-called Wigner cusps could result, for example, in a sharp rise in the cross section for the absorption by 1 UV$+n$ IR photons, in the vicinity of the threshold for absorption by one UV photon only. The occurrence of the higher-order processes has been observed by Trainham *et al.* (as mentioned in Ref. [21]), but the influence on the threshold shift could not be determined. In our experiment the various detachment channels are separable, which means that the behavior of the cross section of the higher-order processes can be investigated directly. In our experiment we observed, next to the one-photon process, only the absorption of one additional IR photon. Although the electron production due to the absorption of one UV photon and one IR photon is certainly not negligible when the UV intensity is high, it remains constant within the accuracy of our experiment. This result is in agreement with previous measurements of the cross section for two-photon absorption by Cl^- near the threshold for one-photon absorption [22]. We therefore do not find evidence for Wigner cusps as an explanation for the smaller-than-expected threshold shifts.

In conclusion, it has been shown that the threshold shift due to a strong (4.5×10^{13} W/m^2) infrared laser field of high frequency is in accordance with a full ponderomotive shift. We have given possible explanations for previously measured smaller-than-expected threshold shifts in similar circumstances. Presumably temporal substructure of the laser pulses and a high UV intensity are responsible for the observed effects. Furthermore, we have argued that results obtained in experiments performed in the low-frequency limit are also consistent with the assumption of a full ponderomotive shift, but in that case the extra energy needed to detach the electron is supplied by the shift-inducing field. We would like to bring to attention that similar results have been obtained in the case of threshold shifts in neutral atoms. The most recent measurements also indicate a full ponderomotive shift [23] in contrast to earlier reports [24].

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[1] R. Trainham, G. D. Fletcher, N. B. Mansour, and D. J. Larson, Phys. Rev. Lett. **59**, 2291 (1987).

[2] M. C. Baruch, T. F. Gallagher, and D. J. Larson, Phys. Rev. Lett. **65**, 1336 (1990).

[3] L. A. Bloomfield, Phys. Rev. Lett. **63**, 1578 (1989).

[4] L. A. Bloomfield, J. Opt. Soc. Am. B **7**, 472 (1990).

[5] W. W. Smith, C. Y. Tang, C. R. Quick, H. C. Bryant, P. G. Harris, A. H. Mohagheghi, J. B. Donahue, R. A. Reeder, H. Sharifian, J. E. Stewart, H. Toutounchi, S. Cohen, T. C. Altman, and D. C. Risolve, J. Opt. Soc. Am. B **8**, 17 (1991).

[6] C. Y. Tang, H. C. Bryant, P. G. Harris, A. H. Mohagheghi, R. A. Reeder, H. Sharifian, H. Tootoonchi, C. R. Quick, J. B. Donahue, S. Cohen, and W. W. Smith, Phys. Rev. Lett. **66**, 3124 (1991).

[7] H. B. van Linden van den Heuvell and H. G. Muller, in *Multiphoton Processes,* edited by S. J. Smith and P. L. Knight, Cambridge Studies in Modern Optics Vol. 8 (Cambridge Univ. Press, Cambridge, 1988), p. 25.

[8] H. G. Muller, A. Tip, and M. J. van der Wiel, J. Phys. B **16**, L679 (1983).

[9] Also the authors of the original publication have repeated their experiment [1] and have found results that are in agreement with the present work [D. J. Larson (private communication)].

[10] H. Dehmelt, in *Advances in Atomic and Molecular Physics,* edited by D. R. Bates and I. Estermann (Academic, New York, 1967), Vol. 3; *ibid.* (1969), Vol. 5.

[11] P. Kruit and F. H. Read, J. Phys. E **16**, 313 (1983).

[12] M. D. Davidson, H. G. Muller, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **67**, 1712 (1991).

[13] E. P. Wigner, Phys. Rev. **73**, 1002 (1948).

[14] E. J. Robinson and S. Geltman, Phys. Rev. **153**, 4 (1967).

[15] A. Mandl, Phys. Rev. A **14**, 345 (1976).

[16] E.-A. Reinsch and W. Meyer, Phys. Rev. A **14**, 915 (1976).

[17] M. Kutzner, M. Felton, and D. Winn, Phys. Rev. A **45**, 7761 (1992).

[18] R. Trainham, G. D. Fletcher, and D. J. Larson, J. Phys. B **20**, L777 (1987).

[19] M. Crance (private communication).

[20] F. H. M. Faisal and P. Scanzano, Phys. Rev. Lett. **68**, 2909 (1992).

[21] D. J. Larson, Bull. Am. Phys. Soc. **34**, 1209 (1989).

[22] M. D. Davidson, H. van der Hart, D. W. Schumacher, P. H. Bucksbaum, H. G. Muller, and H. B. van Linden van den Heuvell, in "Super-intense Laser Atom Physics," edited by B. Piraux (Plenum, New York, to be published).

[23] R. T. O'Brian, J.-B. Kim, G. Lan, T. J. MacIlrath, and T. B. Lucatoro (to be published).

[24] D. Normand, L.-A. Lompré, A. L'Huillier, J. Morellec, M. Ferray, J. Lavancier, G. Mainfray, and C. Manus, J. Opt. Soc. Am. B **6**, 1513 (1989).