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Longer Wavelengths Require Lower Intensity in Multiphoton Detachment of Negative Ions

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Negative chlorine ions, held in a Penning ion trap, are irradiated by intense (up to 2×10^{16} W/m^2) Raman-shifted light of a Nd:YAlG laser with a wavelength of 1908 nm and a pulse duration of 30 ps. Electron energy spectra are obtained showing up to 11-photon absorption. In the case of circularly polarized light suppression of the low-energy channels is observed. A saturation intensity of $(8.5\pm2.5)\times10^{15}$ W/m^2 is found for multiphoton detachment of Cl^- by 1908-nm light. This result is counterintuitive because it is lower than the saturation intensity for multiphoton detachment at 1064 nm.

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With the recent breakthrough in electron spectroscopy of negative ions in strong laser fields [1-3] there has been a sudden extension of the widely studied subject of atoms in strong fields. The next step is to study this interaction in the nonperturbative regime. Just as with atoms, this study is hindered by the fact that even at relatively low intensities the process rate is so high that depletion of the original target occurs before the maximum light intensity is reached. The standard solution for this problem is to increase the order of nonlinearity of the process by increasing the minimum number of photons that are required to drive the process. In the case of atoms this consideration leads to a choice of atoms with high ionization potentials compared to typical photon energies, notably the rare gases. In the case of negative ions this is no option since in the whole periodic table there are no binding energies found larger than about 3.6 eV. Up to now saturation intensities have been measured for detachment processes with nonlinearities up to four [4]. In the present experiment we therefore reduced the photon energy.

With a wavelength of 1908 nm for the optical pulse and a detachment energy of 3.6 eV in the case of the negative chlorine ion, there are six photons required to detach the most loosely bound electron. Up to now there were no high-intensity experiments reported at this wavelength, either with atoms or with ions. Apart from the obvious reasons of technological limitations, the choice of this particular wavelength is given by the trade-off between using a long wavelength for the above-mentioned reason and still using a photon energy large enough to have a resolvable structure in the electron spectrum.

It is not only the details in the electron energy spectra that deserve our attention, but also the energy-integrated process yield. As a result of the summation over various detachment channels, these yields tend to be less informative than the electron spectra, but, in this particular case, they are worth a more detailed inspection. Calculations of Crance [5] indicate that the saturation of the process yield due to depletion of the ground state is decreasing for an increasing wavelength of the detaching pulse. This result is very counterintuitive since it means that the process has a higher rate when more photons are involved (with the same initial state, final state, and light intensity). Evidently, even if this prediction is true, it can only be so in a restricted range of numbers of absorbed photons, as was already pointed out by Crance. This raises another interesting question, namely, whether the surprising behavior of the saturation intensity is typical for negative ions or whether this is more generally true for the investigated range of photon energies and electron binding energies.

In the experiment negative chlorine ions are held in a Penning ion trap [6] which has been integrated in a magnetic-bottle electron spectrometer [7]. This unique combination has been described previously [2]. About 1000 ions are confined in a cloud of about 1 mm^3. The background pressure is 1×10^{-6} mbars. The produced photoelectrons are energy analyzed on the basis of their time of flight over a 50-cm-long flight path. As a light source for irradiating the Cl^- ions we used a Raman-shifted output of a Nd-doped yttrium-aluminum-garnet (Nd:YAlG) laser. The actively and passively mode-locked Nd:YAlG laser produces 40 mJ of energy in 35 ps at a wavelength of 1064 nm and a repetition rate of 10 Hz. This light is focused with a telescope of 2-m effective focal length in the center of a 2-m-long Raman cell filled with hydrogen gas under a pressure of 8 bars. The windows of the Raman cell are made of 0.5-in.-thick suprasil. The various wavelengths are separated by means of a fused-silica Pellin-Broca prism. In this way we obtained 4 mJ in the first Stokes line at a wavelength of 1908 nm and excellent beam quality. The 1908-nm beam showed hardly any divergence after being transmitted over several meters. We measured the transmittance of the beam through various diaphragms, which showed a near-Gaussian profile, indicating a TEM_{00} mode. A lens of 15-cm focal length is used to focus the 1908-nm light into

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the center of the trap. The diameter of the focused beam at half maximum is determined by measuring the transmittance through a 30-μm pinhole, scanned over the laser focus. The diameter is measured to be 50 μm, close to diffraction limited and with a near-Gaussian shape. The intensity autocorrelation function of the second Stokes line was measured in a lithium-niobate crystal. The result was approximately Gaussian, with FWHM of 30 ± 3 ps, slightly shorter than the pulse duration of the 1064-nm light. Since the output intensity is a very large exponential of some other quantity (namely, the Raman gain) that evolves through a maximum, a Gaussian shape does not seem unreasonable. The absolute pulse energy was measured by means of a thermal joulemeter. Pulse-to-pulse variations in the energy during the measurements were monitored by means of a pyroelectric detector. From all these parameters the maximum peak intensity used is calculated to be 1.7×10^{16} W/m^2.

The chlorine atom has an electron affinity of 3.613 eV [8] and can be obtained after detachment either in the $^2P_{3/2}$ state or in the $^2P_{1/2}$, state, 109 meV higher in energy [9]. Figure 1(a) shows the energy spectrum of the detached electrons at a laser intensity of 1.3×10^{16} W/m^2 when linearly polarized light is used with a wavelength of 1908 nm. The light is polarized in the direction of the magnetic field of the spectrometer. Up to 11-photon absorption is visible. Ignoring shifts due to the finite pulse duration and focus dimensions, we expect electron energies of $(0.29+n\times0.65)$ eV and $(0.18+n\times0.65)$ eV. The energy peaks have a width of about 300 meV in which both ^2P states coalesce. The broadening of the peaks can be explained by the following effects. At a wavelength of 1908 nm the ac Stark shift of the threshold is 340 meV/(10^{16} W/m^2). We are in the intermediate pulse regime where the detached electrons travel through only part of the laser focus, while the intensity drops to zero again. We therefore find shifts of the electron energies towards lower as well as higher energies, depending on the moment of detachment. If the electron is detached before the light intensity has reached its highest point, the electrons can "surf" out of the laser focus and gain energy [1,10]. Comparison of spectra measured at various intensities higher than 1.3×10^{16} W/m^2 shows that the peaks of subsequent detachment channels completely overlap but that the envelope of the spectrum hardly changes. The spectrum in Fig. 1(a) is therefore taken at a peak intensity above the saturation value.

Figure 2 shows the averaged number of detected electrons per laser shot at 1064 and 1908 nm as a function of light intensity on a double-logarithmic scale. The light intensity was varied by means of neutral-density filters. Numerical calculations are fitted to the experimental data points. We calculated the total production of photoelectrons as a function of light intensity, assuming a Gaussian temporal and radial distribution of the light intensity, which is reasonable taking into account the experimental checks discussed above. We assumed a con-

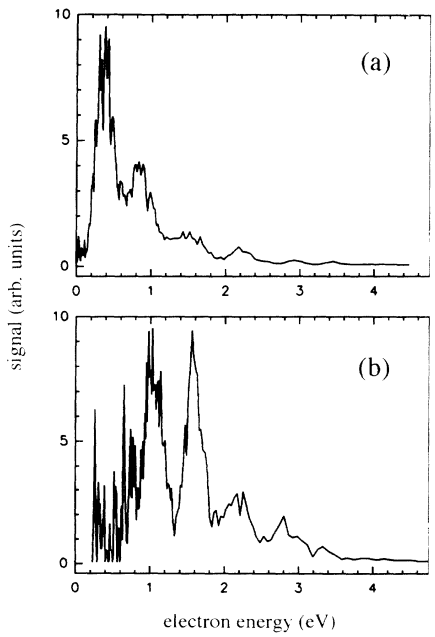

FIG. 1. Electron energy spectra measured after multiphoton detachment of Cl^-. Wavelength of 1908 nm (0.650 eV), pulse duration of 30 ps, and peak intensity of 1.3×10^{16} W/m^2: (a) linearly polarized and (b) circularly polarized.

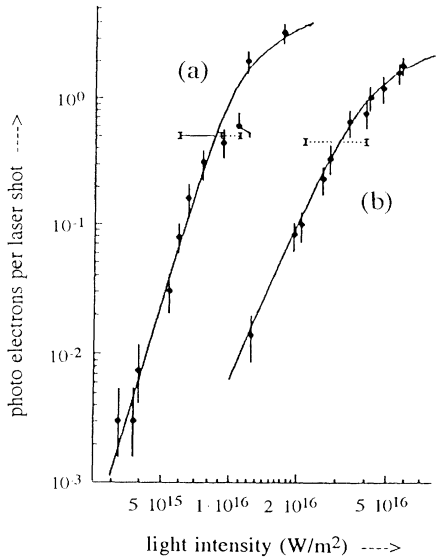

FIG. 2. Detected electrons per laser shot as a function of light intensity, when Cl^- is detached with linearly polarized light, with a wavelength of (a) 1908-nm light and a pulse duration of 30 ps and (b) 1064-nm light and a pulse duration of 35 ps. The experimental data are fitted with numerical calculations. Fitting parameters are the cross section and ion density. The horizontal error bars apply to the curves as a whole. The solid error bar designates the error based on ponderomotive arguments; the dashed one, that based on the measurement of focal parameters and pulse energy.

stant intensity distribution along the beam axis. Furthermore, we only used one single intensity-independent cross section for the lowest detachment channel, so higher-order processes and channel closure are neglected (nevertheless, in the case of atoms it is normally found that the ionization probability follows the same order of nonlinearity as the order of the lowest ionization channel, even in cases where there is strong above-threshold ionization [11]). Fitting parameters are the cross section and ion density, which both only affect the position of the calculated curve in the figure in the x direction and y direction, respectively, but do not alter the form of the curve. On the basis of the data given in Fig. 2, we obtain the cross section σ_6 for 1908 nm (in $\mathrm{sr}^{-1}\mathrm{W}^{-6}\mathrm{m}^{12}$):

$$\log_{10}\sigma_6 = -85.05 \pm {}^{0.90}_{0.68}.$$

On the basis of this value of the cross section and a pulse duration of 30 ± 3 ps we derive $I_S = (8.5 \pm 2.5) \times 10^{15}$ $\mathrm{W/m^2}$. The uncertainty in the assumption about the temporal and spatial profile being Gaussian is included in the error limits. A much more accurate upper limit for the saturation intensity can be deduced from the electron energy spectrum at saturation, shown in Fig. 1(a). As a result of the ac Stark shift of the threshold, closure of the six-photon detachment channel occurs at 8.4×10^{15} $\mathrm{W/m^2}$. More than half of the electron counts are in this channel, though, and the detachment events producing those electrons [as well as those producing possibly associated excess-photon detachment (EPD) peaks] must have occurred below the channel-closure intensity. In fact, an amount of EPD equal to the amount observed is likely to be associated with the production of the six-photon peak. If we assume that the increase in detachment potential is completely dominated by the ponderomotive shift (which is very likely at these low frequencies), a much more accurate upper limit can be obtained. Even if none of the excess-photon signal is produced before channel closure, at the closure intensity apparently only 50% detaches and a $2^{1/6}$ times larger intensity would be needed to saturate detachment. Therefore the saturation intensity cannot be much larger than 9×10^{15} $\mathrm{W/m^2}$. Note that this result is independent from any knowledge about the actual peak intensity and temporal profile of the laser pulse.

To compare saturation intensities, obtained at different pulse durations, we can introduce a "standard pulse duration." For this purpose, Crance uses the time $\tau = h/A$ [5], where A is the detachment energy of the ion. In the case of the chlorine ion this time is 1.14 fs. Normalized in this way, we obtain a saturation intensity $I_{S,N}$ of $(4.6 \pm 1.4) \times 10^{16}$ $\mathrm{W/m^2}$.

Along the same lines we obtain the cross section σ_4 for 1064 nm (in $\mathrm{sr}^{-1}\mathrm{W}^{-4}\mathrm{m}^{8}$):

$$\log_{10}\sigma_4 = -55.51 \pm {}^{0.60}_{0.44}.$$

At a pulse duration of 35 ps, $I_S = (3.1 \pm 0.9) \times 10^{16}$ $\mathrm{W/m^2}$. Using $\tau = 1.14$ fs we obtain $I_{S,N} = (41 \pm 12) \times 10^{16}$ $\mathrm{W/m^2}$. The latter result can be compared to the value obtained by Blondel and Trainham [4], $I_{S,N} = (32 \pm 5) \times 10^{16}$ $\mathrm{W/m^2}$, and shows good agreement.

If one compares the saturation intensities at 1064 and 1908 nm, one finds the value at 1908 nm to be more than a factor of 2 lower than that at 1064 nm. This is a remarkable result since detachment with 1064-nm light is a process with a lower order of nonlinearity than detachment with 1908 nm.

As previously mentioned, there are theoretical calculations by Crance [5] that show decreasing saturation intensities when the number of photons involved in the detachment process increases. In these calculations it is found that for the negative fluorine ion the variation of $I_{S,N}(n)$ fits with the relation $I_{S,N}(n) = 10^{18.72}/n^{2.19}$ $\mathrm{W/m^2}$ [for $n = 4$–30, and $I_{S,N}(n)$ calculated for $\tau = h/A$].

There are two reasons which make it difficult to compare experimental results with this theoretical relation. First, especially when many photons are involved, n cannot be seen as a fixed number due to the effect of excess-photon absorption. Second, to compare saturation intensities for a different number of absorbed photons one would prefer to obtain detached electrons with the same energy. This is experimentally not actually possible when many photons are involved. The ac Stark shift of the threshold leads to different electron energies depending on time and place in the focus at the moment of detachment. Still, the decrease of saturation intensity with increasing number of absorbed photons is clearly visible. One might argue that the difference in saturation intensity is due to the energy difference of the photoelectrons, as indicated by the Wigner threshold law [12], $\sigma \sim \varepsilon^{(l+1/2)}$, in which ε is the kinetic energy and l is the angular momentum of the photoelectrons. We would like to point out that the Wigner law tends to suppress the 1064-nm rates compared to the rates at 1908 nm, and therefore makes the observed effect even more dramatic. The explanation given by Crance is that enhancement of the detachment rate occurs at lower frequencies because there the quiver velocity of the liberated electron equals the velocity of the electron in the initial state at a lower intensity.

The unexpected behavior of the saturation intensity prevented us from penetrating as deep in the nonperturbative regime as we would have liked. To boost the intensity even further we resorted to a second well-known approach, namely, the use of circularly polarized light. We produced intense, circularly polarized 1908-nm light by sending the linearly polarized light through a Soleil-Babinet compensator.

Figure 1(b) shows an energy spectrum where circularly polarized light is used at an intensity of 1.3×10^{16} $\mathrm{W/m^2}$. This spectrum shows a strong suppression of the lower detachment channels. When circularly polarized light is used in a multiphoton process like multiphoton detachment the electrons gain a high angular momentum. This results in a poor overlap between initial and final state

and therefore a reduction of the cross sections for the various detachment channels. Nevertheless, an increment of the number of absorbed photons also means an increase of the kinetic energy of the electron, which makes it easier to overcome the centrifugal barrier [13]. The combination of these two effects gives rise to an optimum, which in this case lies around the absorption of seven or eight photons.

In conclusion, we have performed measurements on the photodetachment of negative ions in the nonperturbative regime. We find that the saturation intensity decreases when more photons are needed to reach threshold. Despite the substantial amount of experimental data for atoms in strong fields, this behavior was never before found for atoms.

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[1] C. Blondel, M. Crance, C. Delsart, and A. Giraud, J. Phys. B **24**, 3575 (1991).
[2] M. D. Davidson, H. G. Muller, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **67**, 1712 (1991).
[3] H. Stapelfeldt, P. Balling, C. Brink, and H. K. Haugen, Phys. Rev. Lett. **67**, 1731 (1991).
[4] C. Blondel and R. Trainham, J. Opt. Soc. Am. B **6**, 1774 (1989).
[5] M. Crance, J. Phys. B **21**, 3559 (1988).
[6] 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.
[7] P. Kruit and F. H. Read, J. Phys. E **16**, 313 (1983).
[8] R. Trainham, G. D. Fletcher, and D. J. Larson, J. Phys. B **20**, L777 (1987).
[9] L. J. Radziemski, Jr., and V. Kaufman, J. Opt. Soc. Am. **59**, 424 (1969).
[10] P. H. Bucksbaum, M. Bashkansky, and T. J. McIlrath, Phys. Rev. Lett. **58**, 349 (1987).
[11] P. Kruit, J. Kimman, H. G. Muller, and M. J. Van der Wiel, Phys. Rev. A **28**, 248 (1982).
[12] E. P. Wigner, Phys. Rev. **73**, 1002 (1948).
[13] P. H. Bucksbaum, M. Bashkansky, R. R. Freeman, T. J. McIlrath, and L. DiMauro, Phys. Rev. Lett. **56**, 2590 (1986).