Competition Between Photoionization and Two-Photon Raman Coupling

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In transitions between bound states, multiphoton processes become important at higher laser intensities when the Rabi-frequency is comparable to or larger than the width of the transition line. This has been shown, e.g., in the study of resonance fluorescence of a two-level atom [1]. Bound-free transitions, however, cannot be saturated owing to the large width of the continuum. Consequently, multiphoton processes should not be important. The only effects expected at higher laser intensities are depletion of the bound state population and possibly continuum-continuum transitions, the latter resulting in multiple peaks in the photo-electron energy spectrum [2].

The experiment described in the following focusses on the intensity dependence of the photoionization process leading to the first, lowest energy electron peak, which can be reached by one-photon absorption. According to the simple picture discussed above, this bound-free transition should not show any intensity dependence apart from depletion of the bound state. In contrast to this expectation we demonstrate that a third-order process involving Raman coupling to a nearby nearly degenerate state may effectively compete with the one-photon absorption process.

In the experiment sodium atoms of a thermal beam have been excited via the $3^2P_{1/2}$ to the $n^2D_{3/2}$ state, using pulsed dye lasers pumped by the second and third harmonic of a Nd:YAG laser. The fundamental beam of the Nd:YAG laser at λ=1.06μ was optically delayed and also directed onto the atomic beam. The delay was adjusted such that the 1.06μ laser pulse interacted with the atoms only after the exciting dye laser pulses had left the interaction region. With this scheme we studied one-photon ionization out of the excited $n^2D_{3/2}$ state. From measurements of total ionization signal as a function of laser intensity, a linear dependence of the signal on the intensity was found at low laser intensities. At higher intensities ($\sim 10^8$ W/cm^2 for n=8) the signal leveled off. This is consistent with the bound-state depletion mentioned above. However, the total ionization signal is not very sensitive to the characteristics of the intermediate state. A more stringent test is provided by measurement of the angular distribution of the photoelectrons [2] by simultaneously rotating the linear polarizations of all nearly collinearly propagating lasers, and by detecting electrons emitted at right angles to the laser beams. Based on the simple argument made above, this distribution should not change at higher intensity of the Nd:YAG laser. The only effect of increasing the laser intensity should be that more and more atoms are ionized during the early part of the laser

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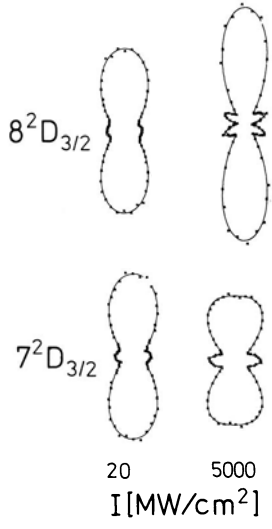
pulse. The kinetic energy of the photoelectron was measured using the time-of-flight technique, proving that no continuum-continuum transitions were involved in this experiment.

The Figure shows polar diagrams of angular distributions of photoelectrons measured for two different Nd:YAG laser intensities and for two different intermediate states, $7^2D_{3/2}$ and $8^2D_{3/2}$. The data are represented by the points and the solid lines result from a least squares fit. At the lower laser intensity, the observed angular distribution has the shape predicted by lowest order perturbation theory, which is the same for n=7 and 8. At the high laser intensity, however, the distributions change drastically, and, what is more, they then differ drastically for n=7 and 8. We explain this effect by two-photon Raman coupling from the $n^2D_{3/2}$ to the $n^2D_{5/2}$ state [3]. Similar Raman coupling between nearly degenerate states has been taken into account in the theoretical description of, for example, quantum beats in photoionization [4,5]. The experiment discussed here has the advantage to isolate the effects of Raman coupling.

For the $8^2D_{3/2}$ state the effect was found to be especially strong owing to the near resonance with the 4p state. The difference in shape at high intensity for n=7 and 8 is due to the fact that $E(n^2D_{3/2}) - h\nu$ is slightly below (above) E(4p) for the n=7 (n=8) state, so the sign of the coherent admixture of the $n^2D_{5/2}$ state is different in the two cases.

The photoelectron angular distribution can be described by a linear combination of $|Y_{30}|^2$ and $|Y_{31}|^2$ [2]. The Raman coupling leads to a mixing of the two fine structure states $|J=3/2,M=1/2\rangle$ and $|J=5/2,M=1/2\rangle$. If the Raman coupling term has a negative sign, as in the case of the 8d-state, this results in an enhancement of the $|Y_{30}|^2$ contribution to the angular distribution at higher laser intensities as shown in the Figure. Similarly, the positive sign of the Raman term in the case of the 7d state enhances the $|Y_{31}|^2$ contribution. The measurement of the electron angular distributions at different laser intensities together with a more detailed theoretical analysis allows one to study the dynamics of the competition between the one-photon and the three-photon process.



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