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Molecular Cesium Component in Multiphoton Ionization of a Cesium Atomic Beam by a Q-Switched Neodymium-Glass Laser at 1.06 μm

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In the interaction of a Q-switched Nd-glass laser radiation with cesium vapor, the molecular cesium component seems to play an important role in the multiphoton ionization process at low laser intensity.

In a recent paper,[1] we have reported experimental results concerning multiphoton ionization of cesium and potassium atoms by the radiation from a Q-switched neodymium-glass laser. The laser beam was focused with a plano-cylindrical lens, with laser power varying from 3×10^6 to 3×10^7 W for cesium.

The previously poor detection sensivity of ions (10^5 ions) has been improved by using a Bendix type 310M magnetic electrom multiplier (MEM). The study of the multiphoton ionization of cesium has been extended towards lower laser power (10^5 W). Figure 1 illustrates the experimental system used. The collection of the ions produced in the focal volume is achieved with the help of a transverse electric field of 200 V cm^{-1}. Ions resulting from multiphoton ionization of Cs atoms or Cs molecules have an initial kinetic energy that can be completely neglected[2] compared to the final kinetic energy gained in the 200 V cm^{-1} electric field. About 1% of the total number of ions created in the focal volume pass through a circular aperture (3 mm diam) in the collector plate of the Faraday cup and are then detected by the electron multiplier. The direct ion signal I_c and the multiplier output signal I_M are simultaneously recorded on the oscilloscope. Such a calibration prevents any possible variation in multiplier gain after a long operation with a cesium-covered dynode strip.

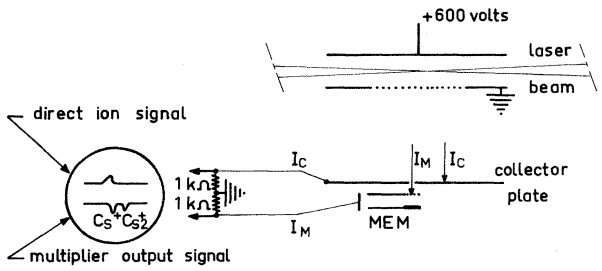

FIG. 1. Experimental apparatus.

Figure 2 shows the variation of the number of

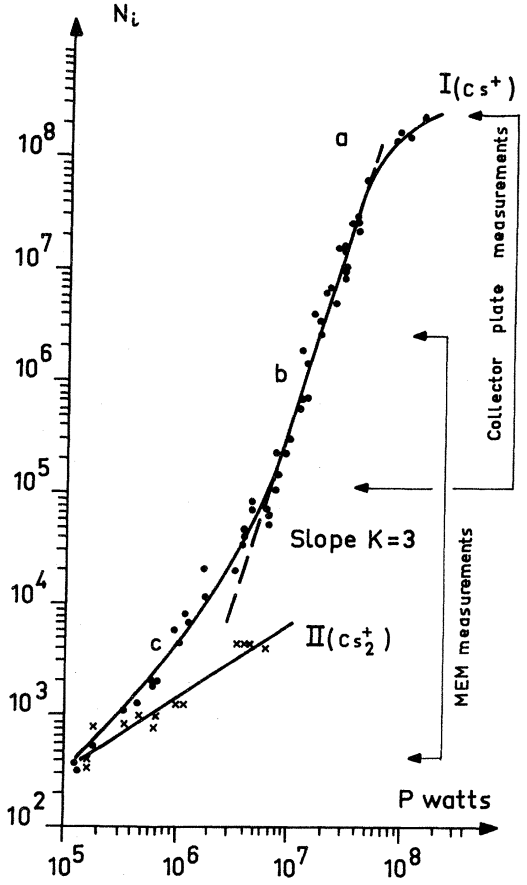

FIG. 2. Variation of the total number N_i of atomic (curve I) and molecular (curve II) cesium ions created as a function of laser power P in the focusing region.

cesium ions created as function of laser power. Curve I describes atomic cesium ions, and curve II concerns molecular cesium ions. For laser power less than 5×10^6 W, a second ionic signal appears in multiplier output current. The resulting time-of-flight mass analysis makes a convincing identification of this collected species as Cs_2^+

Concerning atomic cesium ions, curve I displays three different parts. (a) In this part, the slope of curve I diminishes gradually for laser power higher than 4×10^7 W, corresponding to total ionization of all the atoms present in the interaction volume between the focused laser beam and the beam of cesium atoms.[2] (b) Here the slope of the straight line is $K=3$ for laser power between 5×10^6 and 4×10^7 W. The photon energy we are dealing with is 1.17 eV. It would thus take four of these photons to ionize cesium atoms (the ionization energy is 3.89 eV), and the slope for Cs^+ should be $K=4$. But an examination of the scheme of the atomic levels of cesium shows that the difference between the energy of three quanta (28 339 cm^{-1}) and the energy of the $6f$ level (28 330 cm^{-1}) is smaller than the bandwidth (10 cm^{-1}) of the laser radiation. Thus the cesium atom seems to be ionized via the resonant $6f$ level as previously mentioned,[1] and a slope $K=3$ is observed, corresponding to a three-photon resonant excitation of the $6f$ level.

The triple-quantum ionization of cesium by ruby-laser radiation[3] does not exhibit such an ionization via a resonant intermediate level because no resonance can be found between the energy of an integer number of ruby photons and the energy of a cesium atomic level.

(c) In this section, the slope is roughly equal to 1 for laser power less than 5×10^6 W. In this part of the curve I, the collected atomic ions Cs^+ have been created essentially from a partial dissociation of Cs_2^+ by the laser radiation itself according to a one-photon process (dissociation energy of $Cs_2^+ \simeq 0.7$ eV). The multiphoton ionization of cesium atoms, as for higher laser power, becomes negligible at low laser power.

Some remarks should be made about curve II concerning the molecular cesium ions. The respective ionization energies of cesium atoms and molecules are 3.89 and 3.17 eV. This multiphoton ionization cross section of Cs_2 must be greater than the multiphoton ionization cross section of Cs for a given laser power.[4,5] Both the high ionization cross section for Cs_2 and very low molecular density lead to a total ionization of the molecules present at the focus of the lens even for the low laser power range where Cs_2^+ is detected. This saturation effect may explain the small slope of curve II. This curve II has not been drawn out for laser power higher than 5×10^6 W because the Cs_2^+ signal is hampered by the increase in simultaneous Cs^+ signal with laser power P as P^3.

Very few molecular cesium ions have been detected in the interaction of a laser radiation at 0.53 μm with a cesium beam in the same conditions as above. Moreover, the variation of the number of atomic cesium ions created as a function of laser power in focal volume displays a slope close to the value 2, below the saturation part, corresponding to a two-photon ionization process, even for low laser power. It is quite possible that the very small number of cesium molecules present in the focal volume is nearly completely dissociated by the laser radiation at 0.53 μm.

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