Resonant Multiphoton Ionization of a Cesium Atomic Beam by a Tunable-Wavelength Q-Switched Neodymium-Glass Laser

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The narrow spectral bandwith (0.4 Å) of a Nd-glass laser was tuned over a range of 40 Å to populate selectively the $6f$ level of cesium atoms. The resulting variation of the multiphoton ionization probability was investigated as a function of the laser wavelength.

In a previous paper[1] we reported experimental results concerning multiphoton ionization of Cs atoms subjected to radiation from a Q-switched Nd-glass laser. The difference between the order of nonlinearity measured, $k_{\text{expt}} = 3$, and the theoretical value $k_0 = 4$ was attributed to the contribution of a resonant multiphoton excitation of the $6f$ atomic level, the difference between the $6f$-level energy (28 329.7 cm^{-1}) and the energy of three quanta (28 339 cm^{-1}) being smaller than the spectral band width (10 cm^{-1}) of the laser radiation previously used. Both the possibility of accurately tuning the laser wavelength and the better monochromaticity of the radiation now allow us to investigate the respective roles of the direct multiphoton ionization and the ionization via an intermediate excited state.

In the multiphoton-ionization experiment to be described, interaction takes place between the tunable-wavelength laser beam (10 560 Å $< \lambda_L$ $<$ 10 604 Å) and a Cs atomic beam. When there is no correspondence between the energy of an integer number of photons and the level energies of the Cs spectrum, the atom may be ionized only by direct absorption of k_0 photons, where k_0 is the next integer greater than the ionization energy divided by the photon energy; $k_0 = 4$ for the Cs atom. The ionization probability is proportional to the laser intensity raised to the power k_0, where k_0 is the theoretical value of the order of nonlinearity of the interaction. On the other hand, if the laser wavelength is adjusted so that the energy of three photons exactly corresponds to the energy of the $6f$ level, most of the ions are produced by a two-step ionization process: a three-photon excitation of the $6f$ level with the probability $W_{0\text{-}1}$, followed by a one-photon ionization of the excited $6f$ state with the probability $W_{1\text{-}2}$. In such a process $W_{1\text{-}2} \gg W_{0\text{-}1}$ and $W_{\text{ioniz}} \simeq W_{0\text{-}1}$. The ionization probability is now proportional to the laser intensity raised to the power 3, and its value is much greater than in the case of direct multiphoton ionization which is a fourth-order process. However, the lifetime of the excited state may decrease as a consequence of the high transition probability from this excited level to the continuum under the influence of the laser field. In these circumstances, an "instant" ionization of the excited state can take place, and a two-step ionization process considered as two successive processes is misleading.

The experimental apparatus is shown in Fig. 1. The Nd-glass laser consists of an oscillator Q switched by a rotating prism and one amplifier. It can deliver 6 J in 30 nsec. A birefringent filter is inserted in the oscillator cavity to select a spectral band in the fluorescence spectrum. Each element of the filter consists of a calcite plate, cut parallel to the optical axis, between two linearly polarizing plates.[3-6] Both polarizers are so oriented that their direction of polarization is 45° to the principal axes of the crystal. When the faces of the plate are normal to the laser axis, the transmission function of the filter is given by $I = \cos^2(\pi e\,\Delta n/\lambda)$, where Δn is the difference between the extraordinary and ordinary refractive indices, e is the thickness of the plate, and λ is the wavelength. Tilting the birefringent plate around the x_{hor} and y_{vert} axes (z being the laser axis) results simultaneously in a shift of the interference pattern and a reduction of the interval $\Delta\lambda$ between two successive maxima. The laser line is narrowed by the multiple pass-

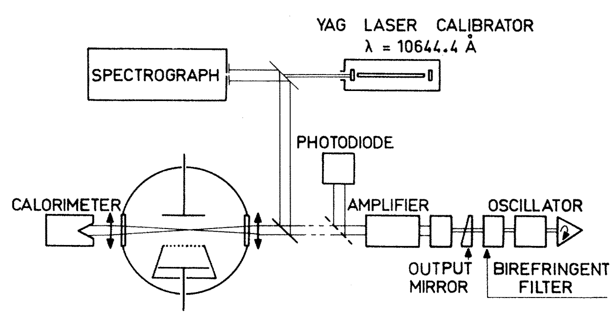

FIG. 1. Schematic diagram of the experimental apparatus.

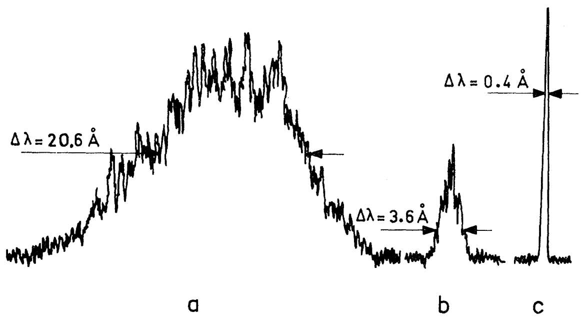

FIG. 2. Bandwidth of the laser emission. (a) Without birefringent filter in the laser cavity; (b) with one calcite plate of thickness 1 mm; (c) with two calcite plates of thickness 1 and 4 mm.

es through the filter when inserted in the cavity. The first birefringent-plate thickness is chosen so that $\Delta\lambda$ is large enough to obtain only one transmission maximum of the fluorescence spectrum. The second birefringent plate, several times thicker than the first one, reduces the spectral bandwidth of the laser radiation. Figure 2 represents the laser bandwidth with (a) no birefringent filter, (b) a one-element filter, and (c) a two-element filter. It is possible to obtain better monochromaticity by inserting a third element in the cavity, but temperature control and regulation have to be improved because of the large temperature dependence of the birefringence. The coherence effects of the radiation do not seem to depend on the laser tuning. The spectral linewidth of the laser radiation has been verified to be constant when the laser wavelength is tuned. The laser wavelength is measured by a diffraction-grating spectrograph and recorded on spectrographic plates simultaneously with an yttrium aluminum garnet (YAlG) laser beam whose reproductibility has been established and the wavelength calibrated with an iron arc: $\lambda_{\text{YAlG}} = 10\,644.4 \pm 0.4$ Å. Values of the Nd-glass laser wavelength are determined with a relative error of ± 0.1 Å. The uncertainty of the absolute value of the laser wavelength is ± 0.4 Å. The nonlinear interaction takes place between the laser beam focused by the same cylindrical lens as that employed in previous experiments[1] and a Cs atomic beam of rectangular cross section whose neutral density, measured by a surface ionization detector, is found to be 8×10^9 cm^{-3}. Ions formed from the interaction are accelerated by a transverse electric field of 750 V cm^{-1}, separated in mass from other possible ionic species by a time-of-flight spectrometer, and then detected. The

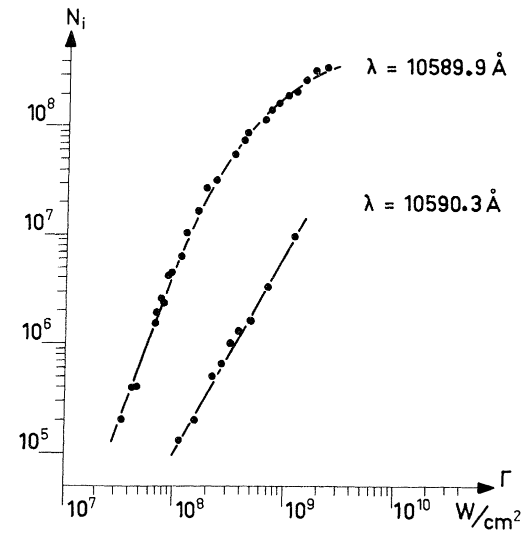

FIG. 3. Variation of the number N_i of Cs ions formed as a function of laser intensity Γ for two different laser wavelengths corresponding to a detuning from resonance of 0.4 Å. k_{expt} values are derived from the linear portion of curves. $k_{\text{expt}} = 2.9$ at $\lambda = 10\,589.9$ Å, and $k_{\text{expt}} = 1.8$ at $\lambda = 10\,590.3$ Å.

energy is measured by a TRG calorimeter, the time distribution of the laser intensity by a photodiode, and the spatial distribution in the interaction volume by a photometric method described in a previous paper.[7]

The experiment consists of measuring the number of ions N_i formed as a function of the laser intensity. For each value of the laser wavelength, the experimental value of the order of nonlinearity of the interaction k_{expt} is determined by the slope of the linear portion of the curves $N_i = f(\Gamma)$ in log-log coordinates, such as in Fig. 3. The multiphoton ionization probability is

$$W = N_i / n_0 V_k \tau_k,$$

where n_0 is the neutral density of cesium atoms, V_k is the effective interaction volume for a kth-order process, and τ_k is the effective interaction time for a kth-order process. For this form of W, V_k and τ_k are modified to take into account the variation of k_{expt} as a function of the laser wavelength. It should be pointed out that this procedure does not significantly change the law of variation of W as a function of the laser wavelength compared to the case of $k = 4$ throughout. Experimental results are summarized in Fig. 4 by expressing the variation of K_{expt} as a function of the laser wavelength, and in Fig. 5 representing the variation of the multiphoton ionization

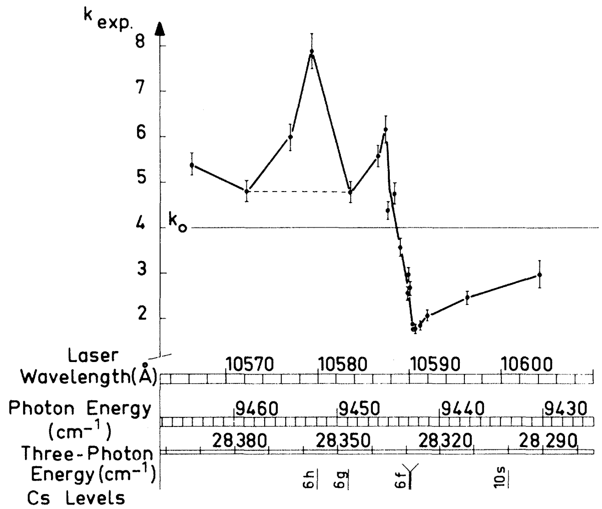

FIG. 4. Variation of the slope k as a function of the laser wavelength. The dashed line disregards the additional process which takes place at approximately 10 579 Å.

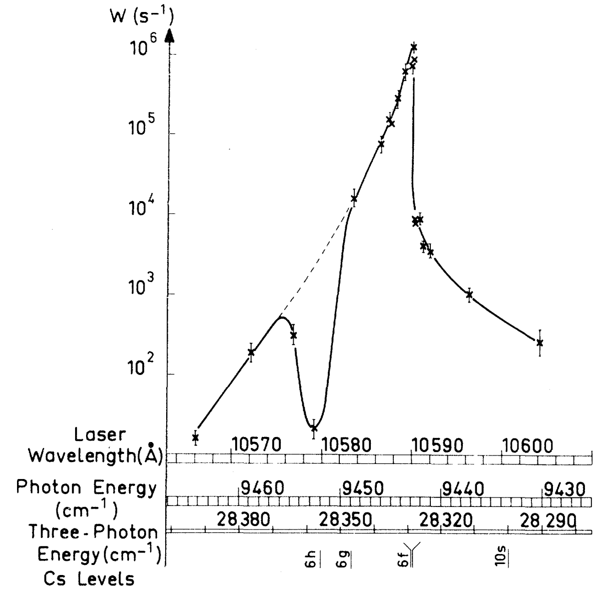

FIG. 5. Variation of the multiphoton ionization probability W as a function of the laser wavelength, for a given laser intensity $\Gamma = 1.4\times10^8$ W/cm^2. The abscissa axis also displays the photon energy, the corresponding three-photon energy, and the positions of the atomic levels. The dashed line disregards the additional process which takes place at approximately 10 579 Å.

probability W as a function of the laser wavelength for a given laser intensity $\Gamma = 1.4\times10^8$ W/cm^2. These two curves express the same resonant phenomenon; Fig. 4 displays the dispersion-like curve of the resonance, and Fig. 5 gives the absorption curve of the resonance. We find classical resonance curves, except for an additional process appearing at approximately 10 579 Å, the origin of which is not well known at the present time. However, it should be pointed out that this process takes place close to the $6h$ level. Considering these curves, one can make several observations. First, the curve $W = f(\lambda)$ is marked by a strong asymmetry. The ionization probability slowly increases when the laser wavelength is tuned toward the wavelength of resonance (λ_R) from the shorter-wavelength side, and sharply decreases for a small detuning of the laser wavelength toward longer wavelengths when $\lambda_L = \lambda_R$. The sharp variation of the ionization probability is illustrated by the two curves of Fig. 3, where the number of ions N_i is expressed as a function of the laser intensity for two values of λ_L separated by only 0.4 Å. It appears that an increase of wavelength of 0.4 Å induces a decrease of N_i of 1 to 2 orders of magnitude for a constant laser intensity. If we take into account the laser linewidth, which is 0.4 Å, such a decrease in the number of ions looks like a discontinuity.

The second observation concerns the good agreement between the energy of three photons of the laser corresponding to the maximum ionization probability, and the energy of the $6f$ resonant level of the Cs atomic spectrum ($\lambda_{6f} = 10\,589$ Å, $\lambda_{\max W} = 10\,589.9 \pm 0.4$ Å). It may be assumed from this statement that, at this value λ_R, the Stark shift of the $6f$ resonant level under the influence of the laser electric field is smaller than 0.4 Å. A similar observation has been reported for a resonant twelve-photon excitation of the $11p_{3/2}$ level of the neon atom under the influence of cooled ruby-laser radiation.[8] Such a result contrasts with other experiments reporting on multiphoton ionization through resonance where resonant levels were assumed to be shifted over tenths of angstroms.[9,10] If the Stark shift of the $6f$ level is unimportant at the exact resonant wavelength, it may be significant outside the resonance, and it suggests an interpretation of the observed asymmetry of the data in Fig. 5. On the short-wavelength side of the resonance, the sign of the Stark shift may improve the near resonance. Beyond the resonant wavelength, on the long-wavelength side, the $6f$ level may be repelled drastically by the Stark level shift, cutting sharply into the resonance enhancement of the multiphoton ionization probability.

The third point is relative to the variation of

k_{expt} as a function of the laser wavelength, illustrated in Fig. 4. Large discrepancies (Δk) are observed between the value $k_0 = 4$ and k_{expt} ($k_{expt\ max} = 8$, $k_{expt\ min} = 1.8$). The maximum value $k = 8$ is correlated to the significant decrease of the probability observed at $\lambda = 10\,579$ Å in Fig. 5. In the vicinity of a resonance, the observed slope k no longer corresponds to the nonlinear order of interaction between the laser radiation and the atoms. The dip observed near 10 579 Å could be interpreted as an interference between the nearly resonant intermediate state and the nonresonant background. Such dips arise from cancelation effects of the contributions from the energy levels above and below the photon energy.[11] However, recent theoretical calculations carried out by Gontier and Trahin[12] concerning the resonant multiphoton ionization of hydrogen atoms lead to the conclusion that even a small shift of the level can explain large positive and negative values of Δk. This process can also induce a second resonance or antiresonance far from the first one. This process may be correlated to the decrease of multiphoton ionization probability W observed at 10 579 Å. It should be pointed out that this decrease of probability is clearer for a small number of ions formed. Inhomogeneities of the laser intensity in the interaction volume may be responsible for the total ionization of the atoms in some parts of the interaction volume although the total number N_i of ions formed is less than the saturation limit observed in the upper part of the curves $N_i = f(\Gamma)$ in Fig. 3. A possible small shift of the resonant $6f$ level may take place selectively in the regions of the interaction volume where the laser intensity is higher. In the near future, another set of experiments will be carried out without focusing of the laser beam. In these conditions of large interaction volume, small ion and excited-atom densities, and weak laser intensity, better sharpness of resonance is expected, allowing an easier and more accurate interpretation.

The experimental study of multiphoton ionization of Cs atoms through resonance of the $6f$ atomic level by interaction between a tunable-wavelength Nd-glass laser radiation of narrow linewidth (0.4 Å) and Cs atomic beam has shown that the resonant level does not undergo a visible shift at the exact resonant wavelength under the influence of the laser electric field. However, outside the resonance, a Stark shift may take place, and this shift would explain the strong asymmetry in the law of variation of the multiphoton ionization probability as a function of the laser wavelength, in the resonance region. More experiments are needed to understand the meaning of the decrease of the multiphoton ionization probability W at approximately 10 579 Å and the correlated increase of the slope k reaching the value $k = 8$.

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