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Photoionization cross sections for excited laser-cooled cesium atoms

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Losses from a cesium magneto-optical trap induced by an additional laser ionizing the atoms in the $6P_{3/2}$ state have been measured as a function of the intensity and frequency of the ionizing laser. The absolute cross section for ionization of the $6P_{3/2}$ state has been derived as a function of the wavelength of the ionizing laser. Our values are compared with previous ones and available theoretical predictions. A simple model describing the photoionization processes has been developed.

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The laser cooling of atoms has been established as a very powerful and flexible technique for preparing a dense and cold sample. The low temperature reached by laser cooling allows one to produce samples with interesting spectroscopic features. Even if alkali-metal, alkaline-earth, and rare-gas atoms have been laser cooled and novel atomic properties discovered in collisions and in nonlinear optics of cooled atoms, the impact of laser cooling on atomic spectroscopy has been somewhat limited. On the contrary, cooled atomic samples have provided a large amount of information on diatomic molecular systems, mainly through photoassociation spectroscopy. A main limitation imposed by laser-cooled samples on the precise determination of atomic parameters is the perturbation associated with the presence of the strong trapping lasers, so that usually measurements of atomic parameters must be performed by switching off the trapping lasers. In the present investigation direct use is made of the atomic excitation by the trap lasers.

Photoionization of cold atoms was introduced by Dinneen *et al.* to measure the photoionization cross section in a rubidium magneto-optical trap (MOT) loaded from an atomic beam [1]. More recently [2], the change in the loading rate for a rubidium MOT loaded from the background gas has been used to measure the photoionization cross section at a different photoionizing wavelength. We report here on measurements of the photoionization of laser-cooled cesium atoms at various wavelengths; specifically, accurate absolute data for the photoionization cross section of cesium atoms in the $6P_{3/2}$ state are presented. For these measurements the cesium occupation of the excited state is directly provided by the trapping lasers. The modifications of the MOT loading rate and of the steady-state number of trapped atoms allow us to measure the dependence of the cesium photoionization cross-section on the laser wavelength for absorption from the excited $6P_{3/2}$ state. We compare our cross-section values to those measured in previous investigations and to those derived in different theoretical analyses. Our work demonstrates the accuracy, flexibility, and simplicity achieved in cold-atom photoionization measurements. We have developed a model for the photoionization rate of cold atoms and discovered that collisions between cold atoms and ions play a role in the ion production. More reliable theoretical models are required to properly fit our cross-section data.

Our MOT, described in [3], was operated with σ^+,σ^- circularly polarized light, a magnetic field gradient of 0.09 T/m, and trap laser detuning $\delta_T=-3\Gamma$, where Γ is the excited-state spontaneous decay rate. High Rabi frequencies were used for both the trapping and repumping lasers, with total intensity $I_T=170$ mW/cm^2 for the trapping transition between the hyperfine $F=4$ lower level and the excited $F'=5$ level. A total laser intensity $I_R=30$ mW/cm^2 was used on the repumping transition. Different intensities of the six laser beams were chosen in order to balance the asymmetry in the magnetic-field gradient, so that a spherical distribution of the cold atoms was realized. The spatial distribution function, $g(r,N)$ with $g(0,N)=1$, was derived from charge-coupled-device (CCD) images of the MOT. The number N of trapped atoms, derived from the MOT fluorescence flux measured through a calibrated photodiode, was around 10^7, and the density of cold atoms was around 8×10^{10} cm^{-3}. MOT operation with a low atomic density, determined by the low-magnetic-field gradient, was chosen in order to decrease the role played by cold collisions in the MOT balance and to simplify the analysis of the photoionization data.

For the photoionization we used several lines from an argon-ion laser operating in single-line mode with 100-mW maximum power. In addition, radiation at 423 nm with power up to 10 mW from a frequency-doubled diode laser was also used. The photoionizing beam was mildly focused at the MOT center. Its beam waist, for instance 3.3 mm for the argon laser, was large compared to the MOT radius of 0.3 mm, and the intensity of the photoionizing radiation, uniform over the MOT volume, corresponded to the peak laser intensity within the Gaussian distribution. The occurrence of the photoionization process was measured through the modification of the fluorescence light emitted by the MOT. The CCD images confirmed that in our operating conditions the spatial distribution of the cold atoms in the MOT was not modified by the photoionization process.

The rate equation that governs the temporal evolution of the number $N(t)$ of trapped atoms in the MOT is [4]

$$\frac{dN}{dt}=L-\gamma N-\beta n_{max}f(N)N, \tag{1}$$

where L is the loading rate for collection of atoms from the background vapor, and γN is the trap loss rate for collisions with the background vapor. The last term, characterized by the rate coefficient β and the peak atomic density n_{max}, describes the loss due to collisions between trapped atoms. The factor $f(N)=\int g^2(r,N)d^3r/\int g(r,N)d^3r$ measures the reduction in the collision rate due to the spatial atomic distribution. For the operating conditions of our MOT the cold collision term is smaller than the term for the collisions with background vapor. If we denote by $R=\gamma+\beta n_{max}f(N)$ the total loss term of the MOT, the steady-state value for the number of atoms in the trap becomes $N_0=L/R$.

A laser with wavelength λ_P, excess energy ΔE above the ionization threshold, and intensity I_P, produces photoionization of the cesium atoms in the $6P_{3/2}$ state and modifies several terms in Eq. (1). As main modification of the MOT balance, the photoionization introduces an additional loss rate R_P for the N atoms contained in the MOT given by

$$R_P=p_e\frac{\sigma_P I_P\lambda_P}{hc} \tag{2}$$

where σ_P represents the photoionization cross section and p_e the excited-state fraction of cooled atoms in the MOT. We assume a photoionization rate proportional to the laser intensity, because in our conditions I_P is lower than the photoionization saturation intensity.

Another modification produced by the photoionization laser is associated with the loading rate L. This rate depends on the number of background atoms entering the trap region with an initial velocity lower than the capture velocity and captured by the trapping laser [5]. In the presence of the photoionization laser a fraction of these background atoms is ionized and is lost for the trap, reducing the loading rate. The time evolution for the density of ground and excited atoms contained in the background region, $n_{g,b}+n_{e,b}$, is given by

$$\frac{d(n_{g,b}+n_{e,b})}{dt}=-\frac{\sigma_P I_P\lambda_P}{hc}n_{e,b}=-R_P(n_{e,b}+n_{g,b}), \tag{3}$$

where the excited-state density $n_{e,b}$ was approximated as $p_e(n_{e,b}+n_{g,b})$, supposing the background excitation equal to that in the MOT. This background photoionization process acts during the capture time τ_c required for the atoms to reach the MOT. The time integration of Eq. (3) produces an exponential decay, $\exp(-R_P\tau_c)$, for $n_{g,b}(\tau_c)+n_{e,b}(\tau_c)$. Inserting this MOT loading source into the derivation of the loading rate given in Ref. [5], we obtain a reduced loading rate L_P,

$$L_P=Le^{-R_P\tau_c}. \tag{4}$$

Solving Eq. (1) with loading rate L_P when the trap laser is switched on at time $t=0$, in the density limited regime the time dependence of $N(t)$ becomes

$$N(t)=N_s[1-e^{-(R+R_P)t}], \tag{5}$$

with the steady state value N_s for the number of atoms given by

$$N_s=\frac{N_0}{1+R_P/R}e^{-R_P\tau_c}. \tag{6}$$

The photoionization laser could also modify the factor $f(N)$ for the spatial distribution of the MOT atoms. However, as stated above, no modification of the spatial distribution was observed in the experiment and it has not been included in our model. The light shift produced by the photoionizing laser on the optical transition driven by the trap laser could also modify the trap process, but for the intensity of our photoionizing laser that light shift can be neglected. In our experimental observations the irradiation of the MOT by the strong argon-ion line at 514.5 nm, below the photoionization threshold for the $6P_{3/2}$, did not modify the number of atoms or the MOT loading rate. Thus one-photon ionization from the excited state is the relevant process.

The photoionization produces cesium ions escaping from the MOT. The ion escape rate is larger than the ion production rate R_P because the MOT dimension is small and the created ions acquire a kinetic energy from the share, between ion and electron, of the excess energy ΔE [6]. In consequence, the fraction of ionized species is small, in our MOT less than one part in a thousand. For the cesium ions produced by the photoionization in the background, owing to the low background pressure, the mean-free path is large. However, recombination into molecular ions, or other atomic recombination processes leading to molecule formation, correspond to an effective loss of the background atoms. Using the only available data for the recombination rate of cesium ions, at room temperature [7], we derived that these recombination processes produce a negligible modification of the loading rate for the cold atoms.

Because our time constant $1/(R+R_P)$ was a fraction of a second, the MOT loading of Eq. (5) could be directly monitored on the fluorescence emission from the trap. From the change in the loading rate produced by the ionization loss, R_P was easily measured. Even if not properly describing the very initial growth, when the MOT was not yet in the density-limited regime, the exponential dependence of Eq. (5) fitted most parts of the temporal increase in the fluorescence, as shown in Fig. 1. The measured values of the loss rate $R+R_P$ are plotted in Fig. 2(a) versus the intensity I_P of the photoionization laser, at fixed values of the trapping laser intensity and frequency. The data are well fitted through a linear dependence with slope $p_e\sigma_P\lambda_P/hc$. The photoionization rate R_P was also derived from the steady-state value for the number of trapped atoms. The dependence of N_s/N_0 vs the laser intensity I_P, shown in Fig. 2(b), fitted by Eq. (6), allowed us to derive R_P and τ_c. We have used both the MOT loading rate and the steady-state value for the number of trapped atom to derive R_P. The derivation of σ_P from R_P required a measure of the laser intensity I_P and of the fraction p_e for the excited trapped atoms in the trap. The fraction p_e depends on the total Rabi frequency $\Omega_{R,T}$ and detuning δ_T of the trapping beams through the following expression introduced in [8,9]:

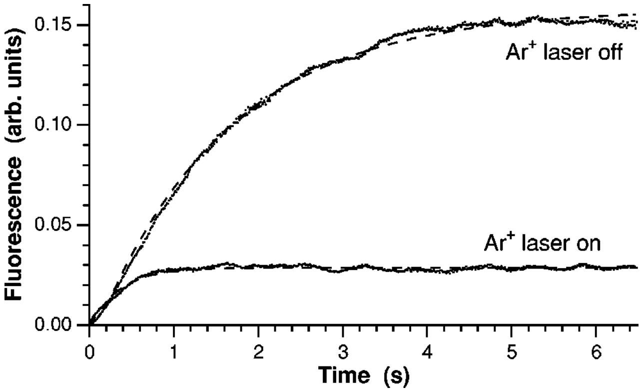

FIG. 1. Time evolution of MOT loading in the absence and in the presence of a photoionizing laser at wavelength λ_P=497 nm with intensity I_P=330 mW/cm^2 (continuous lines) and exponential fits by Eq. (5) (dashed lines).

$$p_e = \frac{1}{2}\,\frac{c^2\Omega_{R,T}^2/2}{\delta_T^2+\Gamma^2/4+c^2\Omega_{R,T}^2/2}. \qquad (7)$$

At low $\Omega_{R,T}$, where the Zeeman sublevels of the ground state are equally populated, c^2 is equal to the average of the Clebsh-Gordan coefficients over the Zeeman sublevels; at large $\Omega_{R,T}$, as for our MOT, a preferential population of the high-m_F stretched states produces a value c^2=0.73 [10].

The cross sections σ_P derived from the measured R_P rates, using either the loading rates or the steady-state values of the number of atoms, are shown in Fig. 3. Except for the highest ΔE point, where a low intensity was provided by the frequency-doubled diode, there is good agreement, within error bars, between the cross sections derived in the two different ways. Such an agreement confirms the validity of our simple model. Figure 3 shows also the values measured by previous authors and some theoretical predictions. The agreement with the previous measurement of Nygaard *et al.* [11], obtained from the photoionization of a cesium atomic beam with a discharge lamp, is only fair. Those cross sections are consistently about 30% above our results. We believe that the discrepancy is due to a systematic error in those older results, since they were normalized for a specific wavelength (λ_p=436 nm) to the semiempirical theoretical result of Weisheit [12], also shown in Fig. 3. In the energy range both experiments have in common, the ΔE dependence of our cross section and that of Nygaard *et al.* [11] are quite similar. There appears to be some problem, however, with the three higher-energy points of Ref. [11]. Three other theoretical cross sections are also shown in Fig. 3: another semiempirical result due to Norcross [13], an *ab initio* Hartree-Fock result of Msezane [14], and an *ab initio* Hartree-Slater result of Lahiri and Manson [15]. In principle, the Hartree-Fock result should be the most accurate, but it is clear from Fig. 3 that the Hartree-Slater result gives the best overall agreement with the present experimental cross section. Actually, the Hartree-Fock result shown in Fig. 3 is an average of the length and velocity cross sections that differ from each other by about 15%. We have also verified that the Hartree-Slater calculation gives a threshold energy that is slightly closer to experiment than the Hartree-Fock threshold. In any case we believe that the somewhat better agreement of our experimental results with the simpler *ab initio* Hartree-Slater calculation is probably accidental. However, the general agreement between the nonrelativistic calculations and the measurements for the $6P_{3/2}$ state is strong evidence that the photoionization cross sections of these excited states are virtually independent of J.

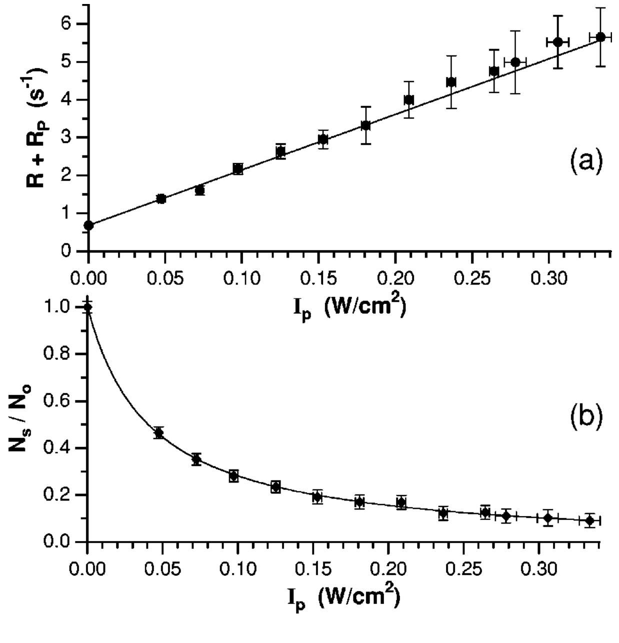

FIG. 2. In (a) loss rate $R+R_P$ and in (b) reduced steady state N_s/N_0 vs intensity I_P of the laser at λ_P=497 nm, and MOT parameters $\delta_T=-3\Gamma$, I_T=170 mW/cm^2. Using the p_e=0.33 value, from the fit we derived σ_P=17.2±1.6 MB from the loss-rate data and σ_P=19.2±1.7 MB from the data for the number of atoms. A capture time τ_c=34±5 ms was derived from the dependence of N_s/N_0 on I_P.

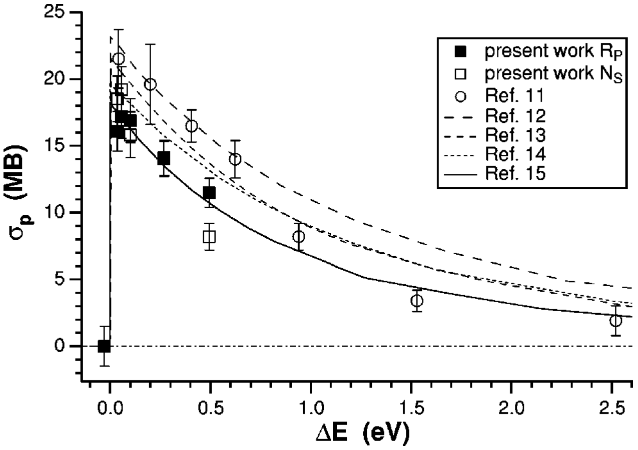

FIG. 3. Experimental data and theoretical predictions for the photoionization cross section σ_P vs the excess energy ΔE of the photoionizing laser. The filled squares represent our data derived from the MOT loss rate, while the open square data are derived from the number of atoms in the MOT. The circles represent cross sections measured by Nygaard *et al.* [11], and the various curves are theoretical results detailed in the text.

The values of capture times τ_c derived from our analysis of N_s for different laser wavelengths were typically in the 16–35-ms range. In effect, in Ref. [16] τ_c was measured between 20 and 50 ms for cesium atoms in trap conditions similar to ours, and was estimated at 1.7 ms, using the Doppler theory for the atomic motion. Our τ_c values derived from the photoionization at different laser wavelengths were consistent, except for the photoionization data at 501 nm, close to the ionization threshold, where a τ_c of 300 ms was derived. Such an inconsistency remained even when we modified our model to include the MOT fluorescence originated from cesium atoms during their capture process and from background cesium atoms excited by the trapping laser and photoionized. We have concluded that some additional weak processes, neglected in our analysis, influence the ionization balance. The ΔE dependence of τ_c suggests that ion recombination, a strongly energy-dependent process, plays an important role in the ion balance, either in the MOT or in the background. Further investigations of the MOT photoionization process for different geometries of the trap and photoionization lasers could be used to measure the ion collision cross sections. Those collisions determine the efficiency of the ion production in the present experiment as well in photoassociation investigations.

In conclusion, in the present cold-atom investigation atomic parameters have been measured with high accuracy, making proper use of the trap laser excitation. Our photoionization measurements demonstrated the demand for a detailed theoretical investigation of the overall atomic potentials. Previously very precise ground-state interatomic potentials were required to analyze ground-state collisions. Moreover, the interpretation of excited-state cold collisions required precise interatomic potentials for the excited state. Our study demonstrates also that recombination processes involving cold ions and atoms, and more generally ion-atom cold collisions, are an important issue deserving future experimental and theoretical investigations.

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