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First Experimental Evidence of Destructive Interference Effects in Two-Photon Ionization of Cs Atoms

J. Morellec, D. Normand, G. Mainfray, and C. Manus

Centre d'Etudes Nucléaires de Saclay, Service de Physique Atomique, F-91190 Gif-sur-Yvette, France

(Received 28 February 1980)

This Letter presents the first experimental evidence of destructive interference effects in multiphoton ionization. The experiment involves a Cs atomic beam crossed with a linearly polarized dye-laser beam whose intensity reaches 20 GW/cm^2 in the 460–540-nm wavelength range. As predicted by second-order perturbation theory, the generalized ionization cross section exhibits a deep minimum which is found to equal $(9 \pm 5)\times 10^{-52}$ cm^4 s at 480 nm. Discrepancies with previous experiments are tentatively explained.

PACS numbers: 32.80.Kf, 32.80.Fb, 32.30.Jc

The two-photon ionization of Cs atoms in the 460–540-nm wavelength range has been the subject of several quantitative calculations based on second-order perturbation theory.[1-3] Apart from small discrepancies mainly due to the choice of atomic wave functions, these calculations agree in predicting a deep minimum in the ionization cross section arising from cancellation of $6P$ and $7P$ intermediate-state contributions. However, the first experiment carried out by Granneman and Van der Wiel[4] (which we shall refer to as the GVW experiment) did not exhibit any minimum and presented experimental cross-section values up to 4 orders of magnitude higher than theoretical predictions. Attempts were then made to modify the theoretical predictions in the hope of obtaining better agreement with the GVW experiment. First, the introduction of time-dependent effects[5] in the previous theoretical work led to a partial disappearance of the deep minimum. Second, another model taking into account laser-bandwidth effects[6] even succeeded in fitting the experimental results of GVW. In fact, the cause of the discrepancy most frequently evoked in the literature was a possible contribution of a residual molecular component in the atomic beam. This phenomenon is highly probable as the absorption band for Cs_2 dimers presented a maximum around 480 nm where the ionization cross section for atoms is expected to be minimum.[7] However, in the GVW experiment it is assumed that the molecular contribution is minimized to at most 15% of the detected signals. Even when GVW repeated their previous experiment,[8] they confirmed their previous results.

Recently, an absolute measurement[9] of the two-photon ionization cross section of Cs atoms by the second harmonic of Nd^{3+} glass laser radiation gave an experimental value in good agreement with the calculated values of Refs. 1–3. In addition, this experiment demonstrated the importance of the residual molecular component in the atomic beam, even after reduction by thermal dissociation in a superheater. These undissociated dimers introduce a molecular background in the ionic signal which seems incompatible with a low-laser-intensity experiment with use of high-detectivity counting methods as reported with GVW. Therefore, we present a new experiment on the subject, carried out at laser intensities of around 10^{10} W/cm^2, whereas GVW worked in the 10^4–10^5-W/cm^2 range. We thus taken advantage of the weak dependence of the saturated molecular process on the laser intensity I, and of the I^2 dependence of the atomic process, so that the atomic signal emerges clearly above the molecular background.

For this purpose, we have constructed a three-stage oscillator-amplifier dye laser transversely pumped by the second harmonic of a single-mode ruby laser ($\lambda = 347.1$ nm). The oscillator cavity is tuned by means of a beam-expander–diffraction-grating system reducing the laser linewidth to 2×10^{-2} nm. An energy of 20 mJ per pulse in 10 ns can be obtained throughout the 460–540-nm wavelength tunability range with C481F and C500F coumarin dyes. A spectral filter inserted between the two amplifiers avoids a background of broadband amplified spontaneous emission. The laser radiation is focused into the atomic beam by a planospherical lens, the effective laser beam section being equal to 10^{-4} cm^2. The laser energy and interaction volume characteristics are measured in the same way as in Ref. 9. Compared with Ref. 9, the only modification in the experimental setup concerns the superheater: It is now separated from the boiler by an independently heated pipe in which molecules are expected to undergo many dissociating wall collisions. We have measured an improvement of about a factor of 2 in the dissociation efficiency due to this modification.

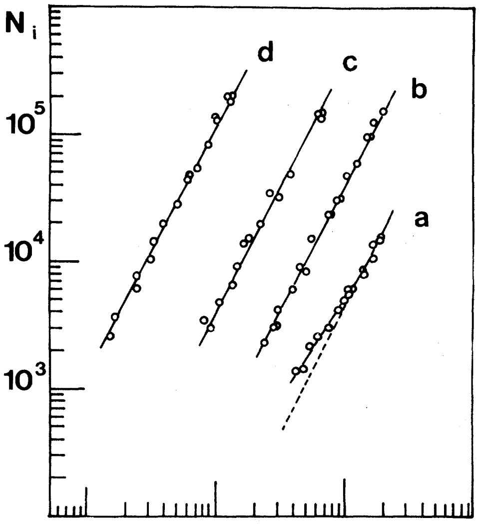

FIG. 1. Ionic signal (absolute values) as a function of laser intensity for four typical wavelengths: curve a, $\lambda = 480$ nm; curve b, $\lambda = 490$ nm; curve c, $\lambda = 540$ nm; and curve d, $\lambda = 460$ nm.

Our experimental results, obtained with linearly polarized light, are displayed in Fig. 1 on log-log coordinates. We have plotted there the number of ions created by each laser pulse as a function of the laser intensity for a set of characteristic laser wavelengths in the 460–540-nm range. We have checked that the effective laser section S_0 is constant within 10% throughout this wavelength range. The ionic signal is found to depend quadratically on the laser intensity as expected for a two-photon process. The I^2 dependence is demonstrated by the slope 2 of the curves b–d and the upper part of curve a. The deviation from slope 2 of the lower part of curve a is due to a molecular contribution to the ion measurements which has already been observed in similar experiments[9, 10] and will be seen in detail in Fig. 2 of the present paper. It is then possible to derive the generalized cross section σ_2 from the set of curves of Fig. 1 using Eq. (2) of Ref. 9 in

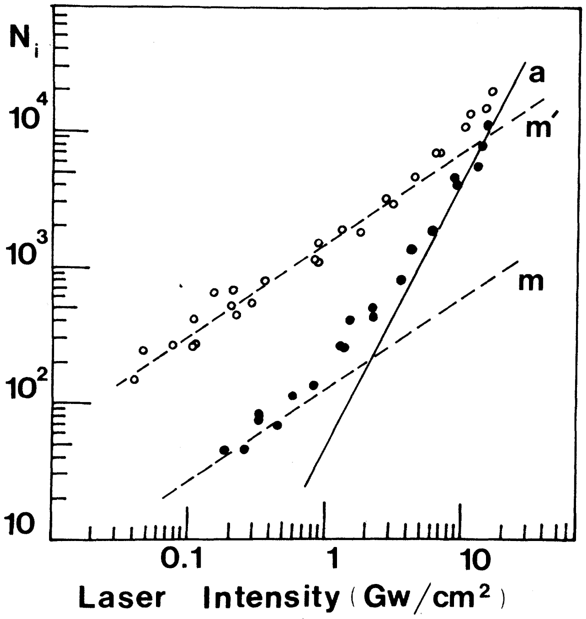

FIG. 2. Ionic signal (absolute values) as a function of laser intensity for $\lambda = 480$ nm. Experimental points: superheater in (solid circles) and out of (open circles) operation. Curve a represents the pure atomic contribution, with curves m and m' representing the molecular background with and without superheating, respectively.

which we introduce the second-order normalized correlation function $g^{(2)}$ to take into account the multimode character of our dye-laser radiation,

$$\sigma_2 = \frac{N_i}{g^{(2)} n_0 V_2 t_2 I^2},$$

where n_0 is the neutral density = 3.6×10^9 atoms/cm^3, V_2 and t_2 are, respectively, the second-order interaction volume and interaction time. Since the laser line contains at least 50 axial modes of the oscillator cavity, the second-order normalized correlation function $g^{(2)}$ is equal to 2 (see Ref. 11).

In Fig. 3, the generalized cross section σ_2 is plotted as a function of photon energy and compared to the results of GVW.[4,5] In contrast with the GVW results, our experimental curve exhibits very rapid variations—indeed σ_2 decreases by two orders of magnitude when the photon energy varies from 2.3 to 2.58 eV and then increases rapidly by more than five orders of magnitude when approaching the resonance of the $7P_{1/2}$ state.

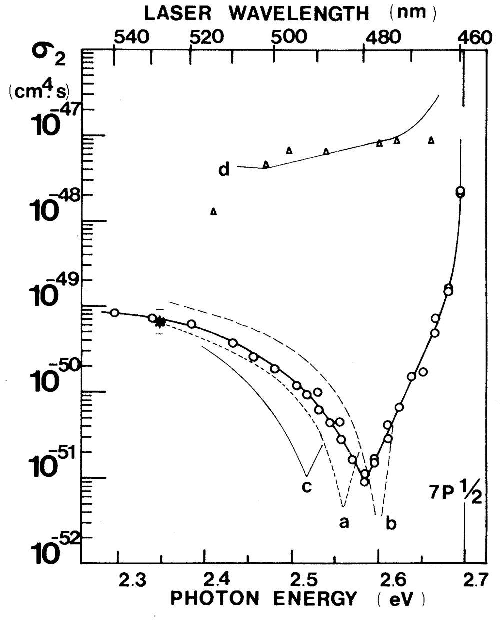

FIG. 3. Two-photon ionization cross section of Cs atoms as a function of photon energy and laser wavelength. Triangles, experimental points of GVW (1975) renormalized by Theodosiou and Armstrong (Ref. 5) according to new photoionization cross sections by Cook *et al.* (Ref. 14); circles, this experiment; asterisk, experimental measurement of Ref. 9. Theoretical calculations: curve *a* Bebb (Ref. 1); curve *b*, Teague *et al.* (Ref. 2); curve *c*, Rachman, Laplanche, and Jaouen (Ref. 3); and curve *d*, Armstrong and Eberly (Ref. 6).

The minimum value of σ_2 is $(9 \pm 5) \times 10^{-52}$ cm^4 s.

The lines *a–c* represent the position and the depth of the σ_2 valley according to the predictions of Refs. 1–3, while curve *d* refers to the calculation of Armstrong and Eberly.[6] It is obvious from the figure that this experiment confirms the results of second-order perturbation theory even if the different calculational methods and radial dipole matrix elements used in the theory lead to small differences in the position of the minimum.

The question then arises to determine whether the depth of the σ_2 valley is a unique characteristic of the ionization of Cs atoms or whether it is also relevant to Cs_2 dimers. To answer this question, we measured the ionic signal obtained at the wavelength minimum—$\lambda = 480$ nm—when the superheater is successively put into (solid circles) and out of (open circles) operation (Fig. 2). In the first case, the quadratic dependence of N_i on I is only observed between 10 and 20 GW/cm^2; between 0.3 and 10 GW/cm^2, a parasitic signal due to Cs_2 dimers is superimposed on the atomic one. By subtracting the expected quadratic atomic curve (*a*) from the experimental points, we obtain the molecular background (curve *m*) which is completely saturated at this intensity level. By switching off the superheater, we obtain the open circles (curve *m*′) where the molecular signal is now predominant. It can then be directly deduced from curves *m* and *m*′ that the superheater reduces the molecular component in the beam by more than one order of magnitude. In summary, we can say that when the superheating system is in operation, the molecular component in the Cs beam is kept low enough so that, in the 10–20-GW/cm^2 laser intensity range, the atomic signal predominates over the molecular signal. The depth of the σ_2 valley is therefore characteristic of only atoms. It is to be noted that the slopes of curves *m* and *m*′ are about 0.7 and not $\frac{3}{2}$ as predicted for a saturated process when spherical focusing is used.[12] This slope of 0.7 arises from limiting of the Cs atomic beam to the interaction volume, preventing it from extending along the laser axis.

Would it then be possible to explain the results of GVW? In fact, the GVW experimental conditions were very different from ours since the laser intensities used were five orders of magnitude lower. The atomic signal which varies as I^2 was consequently reduced by a factor of 10^{10} in the GVW experiment. In our opinion, they were therefore unable to detect any atomic signal, but probably saw only the molecular background which we found to be predominant over the atomic signal as soon as soon as the laser intensity decreases below 1 GW/cm^2.

In conclusion we would like to emphasize that the validity of second-order perturbation theory is well verified for the two-photon ionization of cesium in the 460–540-nm wavelength range. The deep minimum in ionization cross section predicted by theory is clearly demonstrated in the present experiment and absolute values of σ_2 are found in satisfactory agreement with theoretical results. Finally, as far as the position and the depth of the σ_2 valley are concerned, our experimental measurements are sufficiently accurate to be useful to test the accuracy of the different calculational models proposed in the literature.[2]

Moreover, it is obvious from Fig. 3 that the "laser-bandwidth effects" predicted in Ref. 6 are not observed in this experiment, although our laser line is five times wider than the line of the argon-ion laser used by GVW. Finally, with regard to the "time-dependent effects" of Ref. 5, we point out that under our experimental conditions the sudden approximation[13] does not apply since the laser-pulse rise time is orders of magnitude larger than the Rabi oscillations period $2\pi/\delta$, where δ is the detuning from the resonance $7P_{1/2}$ state.

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Direct Inversion of Rotationally Inelastic Cross Sections: Determination of the Anisotropic Ne-D$_2$ Potential

R. B. Gerber and V. Buch
Department of Physical Chemistry, The Hebrew University of Jerusalem, Jerusalem, Israel

and

U. Buck, G. Maneke, and J. Schleusener
Max-Planck Institut für Strömungsforschung, 3400 Göttingen, Federal Republic of Germany

(Received 14 January 1980)

The anisotropic interaction potential for Ne-D$_2$ is determined by a direct inversion method from the measured rotationally inelastic $j=0 \rightarrow j=2$ and elastic $j=0 \rightarrow j=0$ differential cross sections. The values obtained yield the repulsive part of the interaction in the range 2.43 to 2.77 Å. This is the first inversion of inelastic scattering data. The method is restricted to weak inelastic systems. It is computationally simple, and involves only errors of ~ 5% in the inverted potential.

PACS numbers: 34.20.-b, 34.50.Ez

The purpose of this Letter is to present a first direct inversion of measured inelastic molecular cross sections, to yield the underlying intermolecular potential energy surfaces. For purely elastic atom-atom scattering systems, direct inversion methods have been available for some time,[1,2] and have proved successful in avoiding the difficulties of trial-and-error fitting. The motivation to develop such methods for inelastic scattering stems from recent progress in molecu-