Photoassociative Spectroscopy as a Self-Sufficient Tool for the Determination of the Cs Triplet Scattering Length

C. Drag,[1] B. Laburthe Tolra,[1] B. T'Jampens,[1] D. Comparat,[1] M. Allegrini,[1,2] A. Crubellier,[1] and P. Pillet[1]
[1]*Laboratoire Aimé Cotton, CNRS II, Bâtiment 505, Campus d'Orsay, 91405 Orsay Cedex, France*
[2]*INFM, Unitá di Pisa-Universitá, Piazza Torricelli, 2 56122 Pisa, Italy*
and Dipartimento di Fisica MTFA, Universitá di Messina, 98166 Messina, Italy
(Received 23 December 1999)

In photoassociation spectroscopy, the line intensities of a given vibrational progression exhibit zero-signal modulation reflecting the node structure of the s-wave ground state wave function of two free colliding atoms. This leads to the determination of the scattering length. We performed photoassociation of cold Cs atoms polarized in the Zeeman sublevel $f = 4$, $m_f = 4$. We analyzed the intensities of the lines associated with the Cs_2 0_g^- state dissociating to the $6s_{1/2} + 6p_{3/2}$ asymptote. This yields a value of the Cs triplet state scattering length, $a_T = -530a_0$, while consistency requirements impose a value of the multipole ground state molecular coefficient, $C_6 = 6510$ a.u.

PACS numbers: 32.80.Pj, 33.20.–t, 34.20.–b

Collisional properties of ultracold atoms are of crucial importance for Bose-Einstein condensation. This phenomenon has presently been demonstrated for rubidium, sodium, lithium, and recently hydrogen. The scattering length is a key parameter for the production and the stability of a condensate; in particular, in a magnetic trap, a negative value of the scattering length strongly limits the number of atoms inside the condensate [1]. The value of the scattering length is also critical in the interpretation of the collisional frequency shift. Regarding the present development of high-precision atomic clocks using cold atoms, the knowledge of scattering lengths is therefore very important in metrology. The scattering length of cesium has thus been the object of a substantial research effort [2–6].

Molecular photoassociative (PA) spectroscopy of alkali dimers allows an accurate determination of the long-range part of molecular potential curves correlated to the $6s + 6p$ asymptotes, through the precise measurement of the ro-vibrational energy levels of the different molecular states [7]. In a previous article [8], we reported the PA spectroscopy of the Cs_2 0_g^- long-range molecular state dissociating to the $6s_{1/2} + 6p_{3/2}$ asymptote. Photoassociative excitation of the Cs_2 0_g^- long-range molecular state produces, by spontaneous decay, translationally cold Cs_2 molecules in the lowest triplet ground state $a^3\Sigma_u^+$ [9]. The spectroscopic data of the 0_g^- series have been analyzed in terms of Rydberg-Klein-Rees (RKR) and near dissociation expansion (NDE) procedures [8]. In that paper, we already noticed a marked modulation in the intensities of the spectral lines that we attributed to the PA transition between the initial state consisting in two free cold atoms and the final state which is a ro-vibrational level of the 0_g^- state. As discussed in several references [7,10–12], the observed modulation gives access to knowledge of the nodes of the initial wave function of the two colliding atoms and can then lead to the determination of the scattering length of the initial channel. There exists indeed a one-to-one relation between the position of the nodes and the value of the scattering length [13], which involves only the asymptotic part of the ground state potential. It was, however, difficult to fully analyze the data published in Ref. [8] because they correspond to a degenerate dissociation limit and thus to several, *a priori* distinct, scattering lengths.

In order to determine the Cs triplet scattering length, we have now performed a new experiment consisting in molecular PA of cold atoms polarized in the ground state Zeeman sublevel, $f = 4$, $m_f = 4$. The colliding atoms are in a two-atom state with a total angular momentum $F_t = 8$, with projection on the quantization axis $M_F = 8$; this state is correlated only with the molecular Cs triplet ground state, leading to a true one-channel problem. An accurate value of the C_6 parameter is needed for the determination of the scattering length, especially in the case of cesium where the absolute value of the scattering length is known to be large, so that it is very difficult to fix its sign [9]. In this Letter, by carefully analyzing the intensity minima of the PA spectrum, we determine not only the triplet scattering length a_T but also the value of the ground state molecular C_6 parameter. As experimental determination of both parameters is in general difficult, the method described here can be very helpful to define the criteria for Bose-Einstein condensation experiments, for cesium and other elements as well.

The essential idea of our analysis is depicted in Fig. 1, which shows schematically the potential curves for the 0_g^- state and for two ground state atoms. As already mentioned, the 0_g^- potential is known from previous measurements of the ro-vibrational structure. Figure 1 also shows the wave function for a particular v value, with its outer lobe at R_M. If the ground state wave function has an antinode at R_M, there is a maximum in the PA signal, while if there is a node at R_M, the PA signal vanishes. Whether there is a node or antinode at R_M depends on the scattering length but also on the asymptotic part of the potential

0031-9007/00/85(7)/1408(4)$15.00

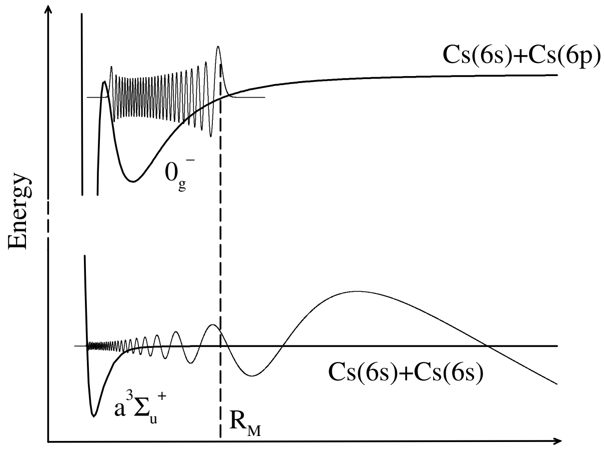

FIG. 1. Potential curves and wave functions.

TABLE I. Theoretical values of the multipole expansion coefficients for the $6s + 6s$ asymptote of the Cs_2 molecule, in atomic units.

Source	10^{-3} C_6	10^{-5} C_8	10^{-7} C_{10}
[17]	6.331	9.630	15.20
[18]	6.408	9.091	13.03
[19]	6.652	9.546	13.58
[20]	6.851 ± 74		
[21]	6.855	9.025	13.01
Present	6.510 ± 70		

[13], i.e., on the C_6, C_8, and C_{10} coefficients. As no experimental value of these coefficients is available, one has to choose among a number of theoretical values, with a general dispersion of about 10% (see Table I). The dependence on C_8 and C_{10} is weak, so that values calculated to 10% accuracy are sufficient. However this is not true for C_6, and consequently all previous determinations of scattering lengths of cesium have relied upon involved calculations of C_6. We will show here that we can determine simultaneously a_T and C_6 by simply using the positions of several zeroes.

The principle of the PA experiment with cold Cs atoms and the details of the experimental setup have been described in Ref. [8,9,14]. The novelty here consists of the use of polarized atoms. The cold Cs atoms are first produced in a vapor-loaded magneto-optical trap (MOT) and they are polarized using the following temporal sequence. First, at time $t = 0$, the MOT cooling laser and the magnetic field gradient are switched off for 8 ms and 4 ms, respectively, and a 6 G magnetic field, oriented along the PA laser propagation direction, is applied simultaneously. At $t = 1$ ms, which is about the rise time of the magnetic field, we apply a pulsed laser (pulse duration 3 ms, pulse intensity ~ 20 μW/cm^2), propagating along the same axis as the PA laser with a σ^+ polarization and tuned to the $6s_{1/2}, f = 4 \rightarrow 6p_{3/2}, f' = 4$ atomic transition. We then apply, after 500 μs and for a time of 2.5 ms, the PA laser which is a Ti:Sapphire laser (Coherent 899 ring laser) pumped by an Argon ion laser. At this time, the measured atomic polarization in the "dark" Zeeman sublevel $f = 4$, $m_f = 4$ is larger than 90%. The symmetry of the wave function thus prohibits odd values of ℓ and the s wave yields the main contribution to the line intensities. This can be checked on the 0_g^- spectrum, as shown in Fig. 2. The absence of a signal for $J = 4$ in both cases indicates that the amount of d wave in our experiment is almost zero, as expected at the measured temperature of 130 μK [15]. In the following, we will thus neglect partial waves with $\ell \neq 0$. The photoassociative reaction reads

$$\mathrm{Cs}(6s_{1/2}, f = 4, m_f = 4) + \mathrm{Cs}(6s_{1/2}, f = 4, m_f = 4) + h\nu_L \rightarrow \mathrm{Cs}_2[0_g^-(6s_{1/2} + 6p_{3/2}; v, J)].$$

That PA has occurred is detected through photoionization of the cold ground state molecules to ${Cs_2}^+$ ions by using a pulsed dye laser (wavelength $\lambda_I \sim 716$ nm, pulse duration 7 ns, pulse energy ~ 1 mJ) pumped by the second harmonic of a Nd-YAG laser, running at a 10 Hz repetition rate. The ions are expelled from the interaction region to a field-free zone providing a time-of-flight mass spectrometer which separates ${Cs_2}^+$ from Cs^+ ions. The ions are then detected by a pair of microchannel plates.

Figure 3 shows the intensities of the spectral lines of the ro-vibrational levels with $J = 2$ versus the frequency detuning of the PA laser. Nine clear minima appear in the modulation of the intensity. The intensities of the spectral lines result from several factors corresponding to the various steps leading from the atom pairs to the detected ${Cs_2}^+$ ions. Zero values corresponding to given v values can, however, only be due to PA, more precisely to zeroes of the probability of the transition between the initial state, corresponding to two free cold atoms, and the final bound state of the excited potential.

To analyze the data, we use a procedure based on the calculation of the electronic dipole moment between initial and final radial wave functions of the PA process. The R dependence of the dipole moment, $\mu(R)$, is taken into account and is obtained from the relevant adiabatic asymptotic potential curves related either to the $6s + 6s$ or to the $6s + 6p$ asymptotes, calculated by including fine and hyperfine interactions, as in Ref. [16]. For the final state, we calculate the vibrational wave functions of the 0_g^- potential determined by the RKR and NDE approaches [8]. For the initial state, we use integration from infinite R in the asymptotic potential, with a boundary condition fixed by $l = a_T/\alpha$, where $\alpha = (mC_6/\hbar^2)^{1/4}$ is a scaling factor (m is the Cs_2 mass), or, equivalently, by $\theta = \tan^{-1}(l)$. In this asymptotic potential, we have included the C_6, C_8,

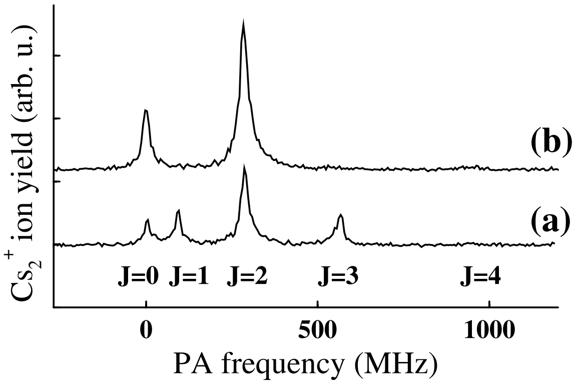

FIG. 2. Rotational structure of the $v = 6$ line of the 0_g^- series of the photoassociation spectrum, obtained with either nonpolarized (a) or polarized (b) atoms.

and C_{10} terms and the exchange interaction. As we are concerned only by the $F_t = 8, M_t = 8$ asymptote, the hyperfine structure acts merely as an additional constant. We have also checked that the retardation effects are negligible for both $s + s$ and $s + p$ asymptotes. To avoid problems connected with absolute intensity measurements and calculations, we focused our analysis on quasizero signals. For each minimum, we determined by interpolation the detuning $\Delta_i^{\text{exp}} \pm \sigma_i$ which would correspond to zero intensity. We then chose a given asymptotic potential. For a few v values around each minimum, we calculated the dipole moment integral for different values of a_T. We found by interpolation the detuning Δ_i^{theor} which would correspond to a zero-dipole-moment integral and we adjusted the value of a_T to make it match the corresponding experimental value. A first trial was made using in turn the various theoretical values found in the literature for the multipole expansion parameters [17–21] and for the exchange terms [22,23]. The values of the scattering length that were obtained for the various sets of parameters and also, in general, for the various minima were all different and incompatible with the experimental error bars. The value of the C_6 parameter, in particular, is of great importance and the differences of about 10% that are observed among the theoretical values (see Table I) are sufficient to change the sign of a_T. The dependence of a_T on C_6 is, however, different for the various minima (see Fig. 4), and we can choose the C_6 value to ensure compatibility between the results corresponding to each minimum. More precisely, we calculated the Δ_i^{theor} values for various pairs (a_T, C_6) and we found the minimum value of $\chi^2 = \sum_i [(\Delta_i^{\text{theor}} - \Delta_i^{\text{exp}})/\sigma_i]^2$. We obtained in this way $\theta = 110.7°$ and $C_6 = 6510$ a.u. Error bars coming from the uncertainties σ_i on the positions of the zeros of intensity are estimated to 1.5° for θ and 12 a.u. for C_6. However, the compatibility between the various minima is better characterized by a doubling of the minimum χ^2 value, which yields an error bar of 7.5° for θ and of 70 a.u. for C_6. The value obtained for the triplet scattering length is thus $-530a_0$, with limiting values $-825a_0$ and $-370a_0$, and the value for C_6 is 6510 ± 70 a.u.

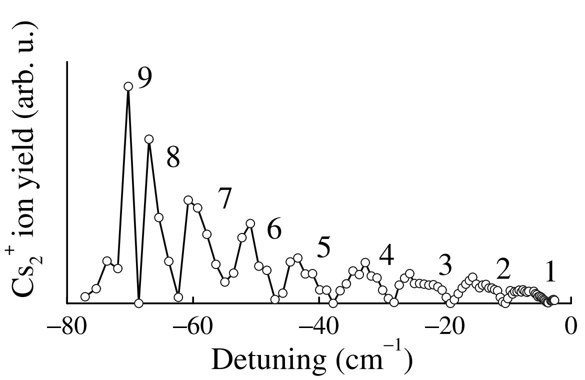

FIG. 3. Relative maximum intensities of the $J = 2$ rovibrational spectral lines versus the frequency detuning of the PA laser. The origin is taken at the atomic transition $6s_{1/2}, f = 4 \rightarrow 6p_{3/2}, f' = 5$.

In the final result we have taken the C_8 and C_{10} values from Ref. [19], whose C_6 value is closest to our result. A change of 15% of the C_8 and C_{10} values does not change significantly the result; it also does not yield a smaller χ^2, so that we cannot derive a value for these parameters from our data. We have used in turn the exchange terms found in Refs. [23] and [22], which differ by roughly a factor of two. The latter is preferable because the corresponding value of χ^2 is twice as small, whereas the results for a_T and C_6 are almost identical. Our calculations concern the $E = 0$ wave function only. In fact, a whole range of continuum states with energies of the order of $k_B T$ are initially populated. Their influence on the intensity minima can be estimated by using an extrapolation from the position

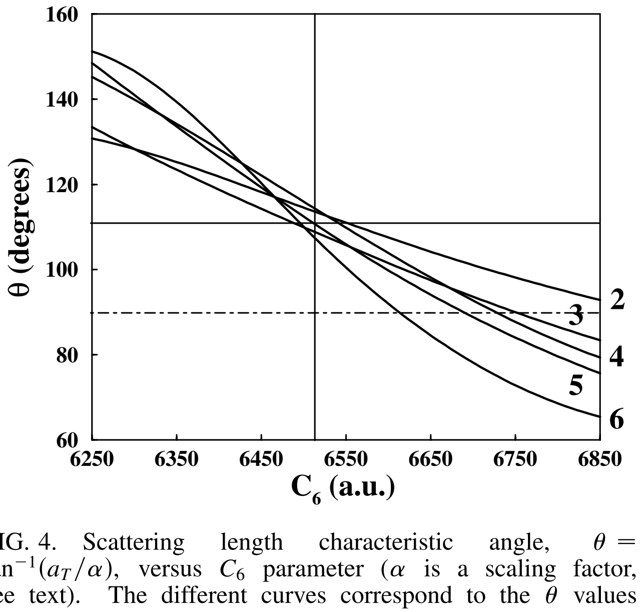

FIG. 4. Scattering length characteristic angle, $\theta = \tan^{-1}(a_T/\alpha)$, versus C_6 parameter (α is a scaling factor, see text). The different curves correspond to the θ values obtained for second to sixth intensity minima. For θ values superior to 90° (dot-dashed line), the scattering length a_T is negative.

of the last node to determine the variation with E of the phase shift of the continuum s-wave wave functions: the variation of the position of the minimum is found to be small, even for the last node (less than 1% for a temperature of 200 μK). The final result is based on the second to the sixth minima only. The first one was excluded because it was not well determined experimentally. We have also excluded the minima numbered 7, 8, and 9. In fact, the number of v values between successive zeroes decreases rapidly as the detuning increases. For the last minima, the determination of the position of zero intensity is thus much less precise. Furthermore, this determination involves neighbor intensities which are rather high, so that dynamical saturation effects are suspected.

To conclude, we have simultaneously determined the triplet scattering length of cesium, a_T, and the molecular asymptotic parameter of the van der Waals interaction C_6. The value of a_T agrees with the values given in Table II, obtained from various experiments [2–6]. The value of C_6 differs significantly from the value 6851 ± 74 a.u., obtained very recently through precise many-body calculations [20] which have been found in good agreement with experimental values in rubidium. We however emphasize here that our value depends very little on the chosen parameters (C_8, C_{10}, and exchange term), so that it comes almost entirely from the experiment. Our basic assumption is that the PA rate near the intensity minima is proportional to the squared dipole matrix element, as it is found in a simple perturbation treatment of photoassociation, like in Ref. [12]. Our approach, based on the analysis of the PA spectrum of the 0_g^- state obtained with polarized cold atoms, is almost completely self-sufficient. The determination of the scattering length requires first knowledge of the 0_g^- potential curve: we deduce it from previous spectroscopic study of the 0_g^- series of the PA spectrum. It also requires knowledge of the asymptotic part of the ground state: the most important parameter, C_6, is fully determined through self-consistency requirement of the analysis. The indisputable discrepancy between our value and the one calculated in the many-body approach clearly questions the validity of theory, either in the many-body calculations or in our photoassociation model. To go further, some new points could be introduced, such as dynamical saturation effects or channel coupling: experimental and theoretical studies on these points are both in progress. In any case, PA spectroscopy clearly appears as a wonderful tool for accurate determination of collisional parameters. It will certainly also become very useful for Feshbach resonance studies, already started for cesium [24].

TABLE II. Determination of the triplet ground state scattering length of cesium, in Bohr radius.

Source	Triplet scattering length
[2]	$-1100 < a_T < -200$
[3]	$\lvert a_T \rvert > 260$
[4]	$-380 < a_T < -315$
[5]	$\lvert a_T \rvert > 600$
[6]	$a_T \approx -400$
Present	$-825 < a_T < -370$

The authors gratefully thank C. Amiot, C. Dion, O. Dulieu, A. Fioretti, T.F. Gallagher, S. Guibal, F. Masnou-Seeuws, and C. Williams for many valuable discussions. Laboratoire Aimé Cotton is associated with Université Paris-Sud. M. A. was an invited Professor by Université Paris-Sud.

[1] C.C. Bradley *et al.,* Phys. Rev. Lett. **75**, 1687 (1995).
[2] B. Verhaar *et al.,* Phys. Rev. A **48**, R3429 (1993).
[3] M. Arndt *et al.,* Phys. Rev. Lett. **79**, 625 (1997).
[4] S.J.J.M.F. Kokkelmans *et al.,* Phys. Rev. Lett. **81**, 951 (1998).
[5] P.J. Leo *et al.,* Phys. Rev. Lett. **81**, 1389 (1998).
[6] R. Legere and K. Gibble, Phys. Rev. Lett. **81**, 5780 (1998).
[7] H.R. Thorsheim *et al.,* Phys. Rev. Lett. **58**, 2420 (1987).
[8] A. Fioretti *et al.,* Euro. Phys. J. D **5**, 389 (1999).
[9] A. Fioretti *et al.,* Phys. Rev. Lett. **80**, 4402 (1998).
[10] R. Côté *et al.,* Phys. Rev. Lett. **74**, 3581 (1995).
[11] P.S. Julienne, J. Res. Natl. Inst. Stand. Technol. **101**, 487 (1996).
[12] P. Pillet *et al.,* J. Phys. B **30**, 2801 (1997).
[13] A. Crubellier *et al.,* Euro. Phys. J. D **6**, 211 (1999).
[14] D. Comparat *et al.,* J. Mol. Spectrosc. **195**, 229 (1999).
[15] A. Fioretti *et al.,* Phys. Rev. Lett. **82**, 1839 (1999).
[16] D. Comparat *et al.,* Euro. Phys. J. D (to be published).
[17] M. Marinescu *et al.,* Phys. Rev. A **49**, 982 (1994).
[18] S.H. Patil and K.T. Tang, Chem. Phys. Lett. **301**, 64 (1999).
[19] S.H. Patil and K.T. Tang, J. Chem. Phys. **106**, 2298 (1997).
[20] A. Derevianko *et al.,* Phys. Rev. Lett. **82**, 3589 (1999).
[21] F. Maeder and W. Kutzelnigg, Chem. Phys. **42**, 95 (1979).
[22] G. Hadinger *et al.,* J. Mol. Spectrosc. **175**, 441 (1996).
[23] M. Marinescu and A. Dalgarno, Z. Phys. D **36**, 239 (1996).
[24] V. Vuletić *et al.,* Phys. Rev. Lett. **82**, 1406 (1999).