Tuning the scattering length on the ground triplet state of Cs_2

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Two schemes for tuning the scattering length on the ground triplet state of Cs_2 are developed. The absolute value of the triplet scattering length of $^{133}Cs_2$ is determined from the experimental data [A. Fioretti, D. Comparat, C. Drag, C. Amiot, O. Dulieu, F. Masnou-Seeuws, and P. Pillet, Eur. Phys. J. D **5**, 389 (1999)], we demonstrate that the large scattering length can be made small and positive by coupling the $^3\Sigma_u^+(6S+6S)$ potential to the $^3\Pi_g$ state by strong off-resonant radiation. A weaker laser field coupling the $^3\Sigma_u^+(6S+6S)$ continuum to the lowest bound level of the excited $^3\Sigma_g^+(6S+6P)$ state also leads to a small positive scattering length. The scattering length of the ^{135}Cs isotope is found to be positive. The method used solves the Schrödinger equation for two electronic states coupled by an electromagnetic field with no approximations employed. The scattering length is determined from the calculated continuum wave functions at low energies. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1343080]

A positive scattering length is crucial to obtain Bose–Einstein condensation in a gas of ultracold atoms. The scattering length controls the long range repulsion between the atoms and therefore the stability of the condensation process. Manipulation of the scattering length adds new candidates to the list of atomic systems which form a condensate. Schemes to control the scattering length have been suggested based on manipulating the interatomic interactions by external fields. They include the use of a magnetic field to induce a Feshbach resonance,[1–7] the use of radio-frequency[8] and dc electric fields,[9] and off-resonant strong electromagnetic fields.[10–12] In the present study, two schemes for modifying the scattering length of the ground triplet state of Cs_2 using an electromagnetic field are proposed. We calculate the scattering length a_T of triplet $^{133}Cs_2$ and discuss the sensitivity of a_T to both the C_6 coefficient and the uncertainty of the experimental data. We develop two modification schemes to tune the scattering length to small positive values using a continuous wave laser field with experimentally feasible characteristics. The scattering length of ^{135}Cs is found to be positive and thus ^{135}Cs is a suitable candidate for Bose–Einstein condensation.

The best known potential for the ground triplet state of Cs_2 $^3\Sigma_u^+(6S+6S)$, is a combination of quantum chemistry calculations by Foucrault *et al.*[13] fitted at the distance ≈ 20 Bohr to the asymptotic behavior C_6/R^6. In the present calculation we used two different values of the coefficient C_6 calculated by Marinescu and Dalgarno[14] and by Derevianko *et al.*[15] This potential has 55 ± 2 bound vibrational levels. The uncertainty of the number of levels is due to the uncertainty of the potential curves at small distances. Other triplet potentials[16] lead to a different number of bound vibrational levels, 58 ± 2. The influence of the higher-order dispersion terms of the long-range potential on the results of the calculations was checked. Since no significant differences have been found, only the term C_6/R^6 was kept as representative of the asymptotic behavior. Using this potential all the bound state wave functions and the threshold scattering wave functions were calculated using the mapped Fourier grid method.[17–19] This approach enabled us to obtain an exact and fully-quantum time-independent solution of the Schrödinger equation without approximation. A grid of 557 points covering 20 000 bohr was used. Due to the exponential uniform convergence of the method the error in the phase of the wave functions was converged to an accuracy of $\sim 10^{-6}$. At the far end of the grid we used either fixed or absorbing boundary conditions. The threshold continuum wave functions were not affected by the choice of boundary conditions. The accuracy of the calculation is determined by the short-range part of the potential where the inaccuracy of the depth of the well is estimated to be about 40 cm^{-1}.[13] This inaccuracy leads to an asymptotic accumulated phase of more than $\pm 2\pi$ which makes the determination of the scattering length from *ab initio* calculations impossible.

A binary scattering event is completely determined by the interatomic potential and the boundary conditions. Since the scattering length is determined by the conditions of vanishing asymptotic kinetic energy it is exclusively determined by the potential. For heavy colliders such as Cs_2 the scattering length is an extremely sensitive function of the potential. Its value is related to the accumulated phase ϕ of the wave function from the inner turning point at zero energy to the infinity. The phase ϕ for the zero energy is estimated semiclassically as $\int_{R_i}^{\infty}\sqrt{-2\mu U(R)}\,dR$ where R_i is the inner turning point of the potential, $U(R)$ is the potential, and μ is the reduced mass of $^{133}Cs_2$. For the potential used for Cs_2, ϕ is 54.6π. From the Bohr–Sommerfeld quantization condition the number of bound levels is given as 54. The number of levels obtained by a direct solution of the Schrödinger equation is the same. Variations of $\pm\pi/2$ change the scattering length from negative to positive. These facts pose a computational challenge for *ab initio* determination of the scatter-

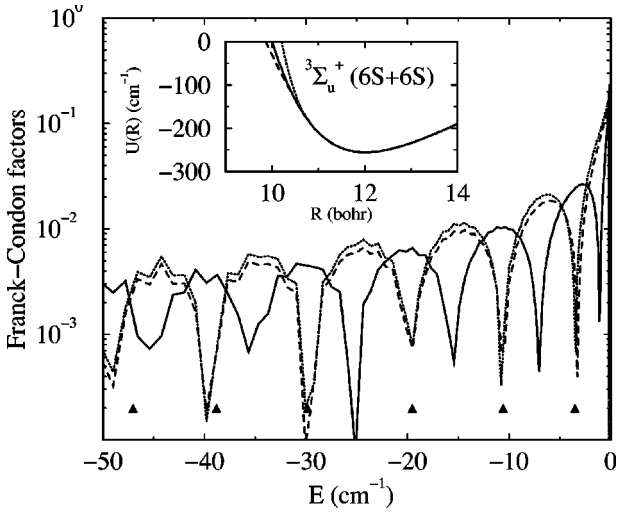

FIG. 1. The variation of the Franck–Condon factors for the transition between the $^3\Sigma_u^+(6S+6S)$ and the $0_g^-(6S+6P_{3/2})$ states for the original (full line) and modified potentials (dotted and dashed lines) with the detuning E. Triangles indicate position of the experimental spectral nodes. Inset shows also the original (full line) potential and two modified potentials (dotted and dashed lines) which both reproduce positions of nodes observed experimentally.

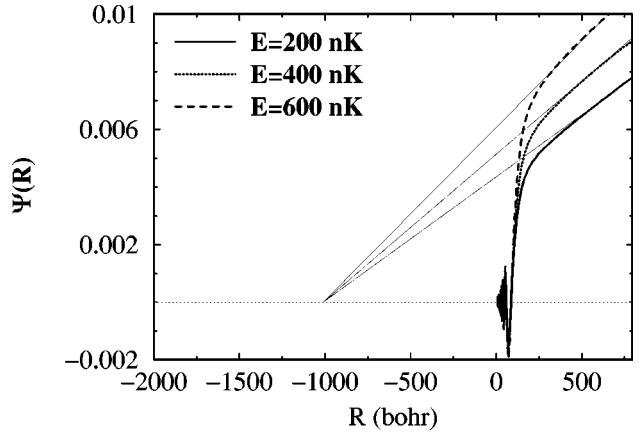

FIG. 2. The determination of the $^3\Sigma_u^+$ scattering length from the continuum wave functions slightly above the dissociation limit. The wave functions are calculated using the potential adjusted to reproduce correctly the oscillations in the photoassociation spectrum.

ing length but they also offer an opportunity for experimental manipulation.

To overcome this problem, an independent method of determining the phase of the wave function is needed. The photoassociation spectrum from the triplet to the $0_g^-(6S+6P_{3/2})$ state obtained by Fioretti *et al.*[20,21] may be used to obtain the phase and to adjust the ground state potential. The spectrum shows oscillating behavior which reflects the variation of the Franck–Condon factors of the transition between the $^3\Sigma_u^+(6S+6S)$ and rovibrational levels of the $0_g^-(6S+6P_{3/2})$ state. The oscillations in the spectrum reflect the nodal structure of the continuum wave functions in the energy range of (200 μK).[21] Using the mapped Fourier grid method,[17–19] the Franck–Condon factors between the vibrational wave functions of the $0_g^-(6S+6P_{3/2})$ state and the continuum wave functions of the $^3\Sigma_u^+(6S+6S)$ state were calculated. These factors give relative intensities of the spectral lines of the $0_g^-(6S+6P_{3/2})$ spectrum. The excited potential curve $0_g^-(6S+6P_{3/2})$ for short distances was obtained by a diagonalization of the $^3\Sigma_g^+(6S+6P)$ and $^3\Pi_g^+(6S+6P)$ curves of the $0_g^-(6S+6P)$ symmetry while including the spin–orbit interaction ($V_{so}(Cs)=554.1$ cm^{-1}). In the asymptotic region the potential is fitted to the Rydberg–Klein–Rees potential obtained from experiment.[21]

The inaccuracy of the potential leads to a shift of the position of the nodes of the calculated spectrum. Figure (1) shows the calculated intensities (full line) and positions of nodes observed in the experiment. The calculated spectrum is almost completely out of phase with the experiment. By slightly modifying the inner part of the potential within the estimated range of accuracy, the experimental observations can be reproduced. The variation in the potential can lead either to addition or subtraction from the phase of the asymptotic part of the wave function. In both cases, very good agreement with the experimental nodal structure as illustrated in Fig. 1 was obtained. With these adjustments the number of bound levels is either 54 or 55.

In order to see the sensitivity to the inaccuracy of the C_6 coefficient we used two different values of C_6 calculated by Marinescu and Dalgarno[14] (-6331 a.u.) and by Derevianko *et al.*[15] (-6899 a.u.). As a result, the inner part of the fitted potentials are slightly different. Figure 1 shows the fits for $C_6=-6899$ a.u.

Using the adjusted potential, the scattering length is recalculated (Fig. 2) leading to $a_T=-350$ bohr for $C_6=-6331$ and $a_T=-950$ bohr for $C_6=-6899$. For $C_6=-6899$ we made additional calculations in order to see if the inaccuracies might result in a positive scattering length. First the uncertainty of the position of minima in the experimental spectrum was considered. The fitted potential is changed in such a manner that the calculated minimum prior to the last minimum (the last node is not very pronounced) shifts by 0.2 cm^{-1} in either direction. With this condition, the last node shifts by 0.1 cm^{-1}. The scattering length becomes $a_T=-1040$ bohr when the nodes shift in the direction of increasing the binding energy and becomes $a_T=-850$ bohr for the opposite direction.

Also, the sensitivity of the determination of a_T to the number of bound levels was verified by calculating a_T for the potentials giving 54 and 55 bound levels (dotted and dashed lines in the inset of Fig. 1, respectively). Once the two potentials are fitted to the same nodal positions, the difference in a_T for the two potentials is smaller at least by a factor 10 than the difference due to the uncertainty of the positions of minima. This result is easily explained in a spirit of quantum defect theory: if the long-range behavior of the potential and positions of last nodes of the last bound wave function are fixed, the short range dynamics (in particular, the total number of nodes of the wave function at small distances) does not influence the scattering properties.

The value of a_T obtained is in good agreement with the previous experimental and theoretical results. Arndt *et al.*[22] and Leo *et al.*[23] got the absolute value >260 bohr and >600 bohr, respectively. Kokkelmans *et al.*[16] published the value -315 to -380 bohr, and Legere *et al.*,[24] ≈ -400 bohr. Re-

TABLE I. The scattering length determination.

	C_6	a_T
Present work	-6331	≈ -350
Ref. 25	-6510	-370 to -825
Present work	-6899	≈ -950
Present work	-7050	the transition

cently, Drag *et al.*[25] analyzed the experiment. The use of polarized atoms allowed to exclude completely the inaccuracy due to the uncertainty of node position, but the greater uncertainty due to the C_6 coefficient was not included. The value of $C_6=-6510$ was determined and used in their calculations.[26] In addition, we determined the critical C_6 coefficient for which the scattering length becomes infinite: for $C_6=-7000$ it is still negative, but $C_6=-7050$ equals to the transition value. For $C_6=-7100$ the scattering length becomes positive. We emphasize the comparison of the presented results with those of Drag *et al.*,[25] Table I, since both approaches analyze the same set of experimental data by different methods.

The absolute value of the scattering length is so large that small variations of the potential can lead to a change of the sign of a_T. For example, a low-intensity laser field coupling the ground molecular state with one of excited states at large distances can modify the scattering length. This possibility was discussed by Kagan *et al.*[12] This sensitivity of the sign of a_T to small perturbations has crucial importance for behavior at ultracold temperatures. Below we discuss how a_T can be made small and positive.

The sensitivity of the asymptotic phase to the inner part of the potential is the key to modifying the scattering length. The first proposed scheme employs a continuous wave (CW) laser field to couple the inner part of the $^3\Sigma_u^+(6S+6S)$ potential to the $^3\Pi_g(6S+6P)$ electronic state. The bound and continuum levels are recalculated for the coupled two-surface potentials. The rotating wave approximation for the coupling by a field is used; the $^3\Pi_g(6S+6P)$ potential is from Ref. 27.

In order to change the scattering length sufficiently, the adiabatic transition probability, $P(^3\Sigma_u^+ \rightarrow {}^3\Pi_g)$, for levels with energy close to the $6S+6S$ threshold, should be non-negligible. For relatively small laser intensity, this condition is fulfilled only if the potentials $^3\Sigma_u^+(6S+6S)$ and $^3\Pi_g(6S+6P)$ cross at an energy close to the $6S+6S$ dissociation limit. The insert in Fig. 3 shows the total two-channel potential in the adiabatic representation. The $^3\Pi_g(6S+6P)$ potential is shifted down by the energy, $E_f=\hbar\omega$, determined by the frequency, ω, of the field. When the intensity, I, of the laser field is low, the region of a pseudocrossing is very narrow. Figure 3 shows two wave functions obtained for the two different intensities for the potentials shown at the inset. With increasing field intensity, the value of the scattering length gradually moves from negative to positive. A change of sign is obtained for the intensity of 300 kW/cm^2.

The second scheme is based on a resonant coupling of the continuum wave functions of the $^3\Sigma_u^+(6S+6S)$ state to one of the vibrational levels of the $^3\Sigma_g^+(6S+6P)$ electronic excited state.[27] Such a Feshbach resonance results in modification of the triplet state scattering length. The sign and value of the scattering length depends on the position of such a level in respect to the $6S+6S$ threshold.

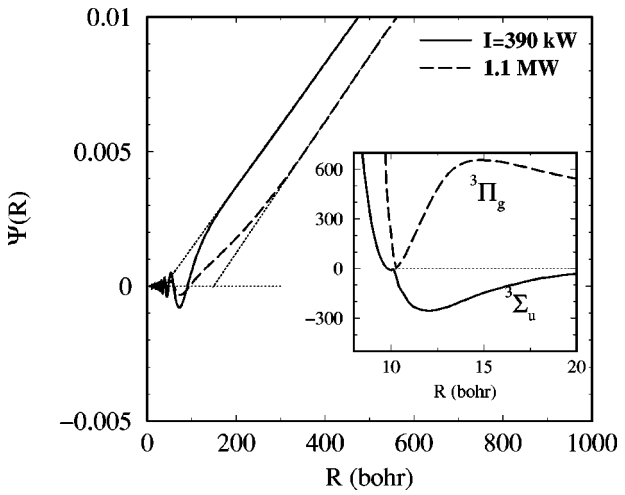

FIG. 3. The scheme of the modification of the inner part of the $^3\Sigma_u^+(6S+6S)$ potential (the inset). The two coupled potentials (in cm^{-1}) are shown in the adiabatic representation using the rotating wave approximation. Wave functions with energy $E=0.4$ μK, calculated for two different intensities of the laser field. Scattering lengths are 30 bohr for a intensity $I=390$ kW and 150 bohr for $I=1.1$ MW. The energy shift $E_f=\hbar\omega$ is the same for both cases, $E_f=11408$ cm^{-1} by 324 cm^{-1} red-detuned to the $6S\rightarrow 6P_{3/2}$ transition.

Employing the mapped Fourier grid method, the low energy continuum wave functions are calculated for two coupled potentials for different intensities of the laser field. The Feshbach resonance is induced by the lowest vibrational level of the excited state negatively detuned from the ground state dissociation threshold by the energy $\Delta=90$ MHz. A gradual increase of the value of the scattering length is observed reaching high positive values. Since the position (and width) of the Feshbach resonance depends strongly on the coupling, the scattering length can be changed employing a much smaller field intensity compared to the off-resonant laser manipulation (Fig. 3). The scattering length is modified from -350 to 300 bohr using a field intensity of only 2.5 kW/cm^2. The projected population in the excited state is less than 10^{-3}.

The present method of solution of the Schödinger equation on the electronic states coupled by a field does not rely on any approximate treatment like perturbation theory and is exact within a given physical model, in contrast to previous approaches by Fedichev *et al.*[10] and Bohn.[11]

It is advantageous to tune the light to one of lowest vibrational levels of the $6S+6P$ manifold which has non-negligible dipole moment for light-induced transition. In this way, the coupling with other vibrational levels of other molecular potentials and a two-photon excitation process to the $(6P+6P)$ manifold can be eliminated. The coupling is also chosen to occur in the short-range region where only a very small fraction of the atomic density resides. This will further reduce loss processes.

The proposed scattering length manipulation techniques

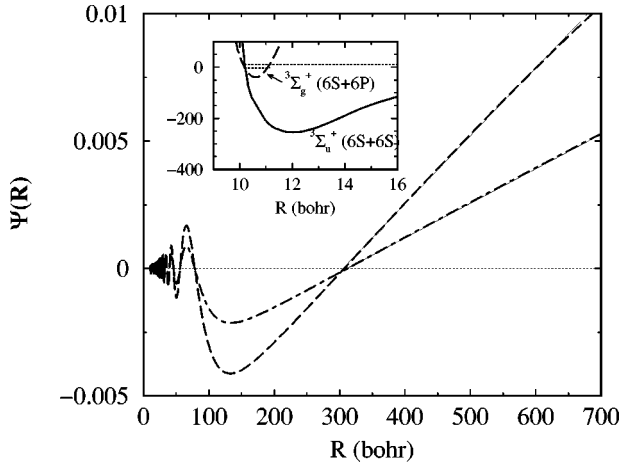

FIG. 4. The continuum wave functions with the scattering length of 300 bohr obtained by a resonant coupling of a vibrational level in the $^3\Sigma_g^+(6S+6P)$ state to the $^3\Sigma_u^+(6S+6S)$ potential. The field intensity was 2.5 kW/cm^2. The inset shows the position of the Feshbach resonance on the two dressed potentials (energy of the potentials in cm^{-1}). The energy shift E_f is 8545 cm^{-1} by 3187 cm^{-1} red-detuned to the $6S\rightarrow 6P_{3/2}$ transition.

employ light-induced coupling of the short-range part of the molecular electronic potential. Therefore, the variation of the dipole moment due to the molecular orientation with respect to the field vector and consequent variation of the coupling strength is to be considered. To solve this problem, using three perpendicular laser beams with orthogonal polarizations for irradiation of the cold cesium sample, which prevents any interference effect, is suggested. In this setup, the maximal variation of the dipole moment projection on the field is by $\approx 15\%$. We studied the robustness of the positive sign of the scattering length by computing its value for both proposed schemes using a field intensity that varies by several orders-of-magnitude. Both schemes are sufficiently robust to keep the interatomic interaction repulsive and, hence, to allow Bose–Einstein condensation.

Using the adjusted potential, the scattering properties of the isotopes ^{135}Cs and ^{137}Cs (half-life's are 2.3×10^6 and 30.2 years, respectively) were calculated. The scattering length of $a_T = 165$ bohr was obtained for ^{135}Cs. This result is in good agreement with the value 138 bohr published in Ref. 16. Therefore, the ^{135}Cs isotope is a good candidate for Bose–Einstein condensation. For the ^{137}Cs, almost zero scattering length was found, suggesting that this radioactive isotope can behave like a noninteracting Bose gas. In both cases, the value of the scattering length with respect to the number of bound levels of the original fitted potential was tested. No significant difference, specifically, ≈ 7 bohr for ^{135}Cs (55 and 56 bound levels) and ≈ 15 bohr for the ^{137}Cs isotope (56 and 57 bound levels), was found.

The spontaneous emission broadening of the Feshbach resonance discussed in Refs. 10,11 is eliminated in the present scheme in two different ways. First, the population transfer at the short-range part of the potential where the resonance is induced is very low due to the small amplitude of the ground state continuum wave function. That results in a small value of the transition dipole moment between the two electronic states. Since its order-of-magnitude is 10^{-4}, the resulting leading term in the spontaneous emission rate $|\langle \psi_g|\mu|\psi_e\rangle|^2$ is approximately 10^{-8} extending the spontaneous emission time scale to values beyond the microsecond range. That reduces the spontaneous emission broadening of the resonance width and it eliminates the related leakage mechanism competing with the Bose–Einstein condensation. In agreement with Bohn,[11] the coupling is mediated by the field whose intensity, the Rabi frequency, results in a time scale much shorter, approximately 150 ps, than the time scale of the spontaneous emission and, hence, the spontaneous emission broadening of the Feshbach resonance is negligible compared to its field-induced width.

The manipulation of the scattering properties can be checked in other applications. For example, photoassociation spectroscopy[28] can be carried out using cold alkali atoms with modified scattering properties. The correspondence between the scattering length and the nodal structure of the Franck–Condon factors suggests a direct experimental test of the proposed schemes employing a CW field with intensities obtainable by a diode laser radiation source. The technique can be used to modify existing experiments done with Rb (Ref. 29) or Cs.[20,21]

The change of the scattering length from negative to positive results in decreasing the population density at short internuclear distance by several orders-of-magnitude. This will decrease the probability for cold molecule formation mediated by three-body collisions.[30] As a result, the reduction in the Franck–Condon overlap and transition dipole matrix elements will decrease the spontaneous decay loss in the cold molecule formation via photoassociation.[20,29,31,32]

Recently, Söding *et al.*[33] measured very high inelastic collision rates of the spin-flip of ground state cesium atoms. They result from the negative scattering length. The attractive interaction enhances the ground state population in the inner part of the potential and makes the Bose–Einstein condensation unstable. Our calculations show that the positive scattering length due to the proposed manipulation schemes will reduce the short range population at least by a factor of 30. This decreases the spin-flip scattering rate by almost three orders-of-magnitude, removing another obstacle to Bose–Einstein condensation in cesium.

In conclusion, the present calculation reproduces the measured oscillations in the $0_g^-(6S+6P_{3/2})$ photoassociation spectrum of ^{133}Cs by making a small adjustment to the $^3\Sigma_u^+(6S+6S)$ potential. For this potential, we calculated the scattering length of the $^3\Sigma_u^+(6S+6S)$ state to be -350 bohr (for $C_6=-6331$). Three different schemes to change the scattering length from negative to positive were studied. The scattering length can be modified by a laser field either off-resonant with the $6S\rightarrow 6P$ transition or on-resonance with one of the lowest vibrational levels of $6S+6P$ potentials. The third scheme consists in replacing the ^{133}Cs by the ^{135}Cs isotope. For this case, the scattering length is 165 bohr. Another obstacle to the Bose–Einstein condensation, the large rate of inelastic collisions, can be overcome by tuning the scattering length.

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