Accurate Determination of the Scattering Length of Metastable Helium Atoms Using Dark Resonances between Atoms and Exotic Molecules

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We present a new measurement of the s-wave scattering length a of spin-polarized helium atoms in the $2\,^3S_1$ metastable state. Using two-photon photoassociation spectroscopy and dark resonances, we measure the energy $E_{v=14} = -91.35 \pm 0.06$ MHz of the least-bound state $v = 14$ in the interaction potential of the two atoms. We deduce a value of $a = 7.512 \pm 0.005$ nm, which is at least 100 times more precise than the best previous determinations and is in disagreement with some of them. This experiment also demonstrates the possibility to create exotic molecules binding two metastable atoms with a lifetime of the order of 1 μs.

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The s-wave scattering length, which characterizes the interactions between ultracold atoms, is essential to describe the structure and the dynamics of Bose-Einstein condensates (BEC). After BEC was achieved for metastable helium atoms in the $2\,^3S_1$ state [1,2], several estimates for their s-wave scattering length a were derived (see Fig. 1), which lack accuracy and are not all in good agreement with each other. We present in this Letter a new determination of a, at least 100 times more precise than the best previous ones, solving unambiguously all the discrepancies in the literature. This important constant is critical to interpret previous or future experiments dealing with the hydrodynamical regime in the ultracold gas [3], the metastable helium BEC in an optical lattice, or ^{4}He-^{3}He mixtures [4]. To measure a, we use two-photon photoassociation (PA) and dark resonances to perform a spectroscopic measurement of the binding energy E_{b_2} of the least-bound molecular state b_2 ($v = 14$) in the interaction potential $^5\Sigma_g^+$ between spin-polarized metastable helium atoms. The precise value of E_{b_2} that we obtain can also be used to check the predictions of *ab initio* recent quantum chemistry calculations of the interaction potential, which can be very accurate for an atom as simple as ^{4}He having only two electrons and no hyperfine structure. Another interesting point is that the two-photon process which we use prepares an exotic molecular state binding two atoms with a very high internal energy of about 20 eV. From the line shape analysis of our spectra, we are also able to estimate the lifetime of this exotic state to about 1 μs. This information could stimulate theoretical models trying to understand the effect of Penning processes between two metastable atoms interacting in a bound state rather than in a free collisional state.

The two-photon scheme is sketched in Fig. 2: it involves a pair of colliding atoms in a state 0 with energy E_∞ and two molecular bound states b_1 and b_2; here b_1 is the $v = 0$ level in the purely long range 0_u^+ potential previously studied by our group [5]. Such Λ-type excitation schemes were used with alkali atoms to realize the spectroscopy of molecules in the electronic ground state in ultracold gases [6–8], to find scattering lengths [9], and very recently to observe subnatural linewidth quantum interference features [10]. In some other experiments molecules in the electronic ground state have been coherently formed from a BEC [11,12]. Oscillations between atomic and molecular quantum gases have recently been observed [13]. The results obtained here with metastable ^{4}He atoms are simpler to interpret theoretically because of the absence of hyperfine structure, which reduces the number of collision channels involved in the light-assisted collisional process. Here, we do not use a BEC of metastable ^{4}He atoms in order to avoid mean-field shifts. Finally, we use ultracold temperatures such that the dispersion k_BT of scattering energies E_∞ is very small compared to the

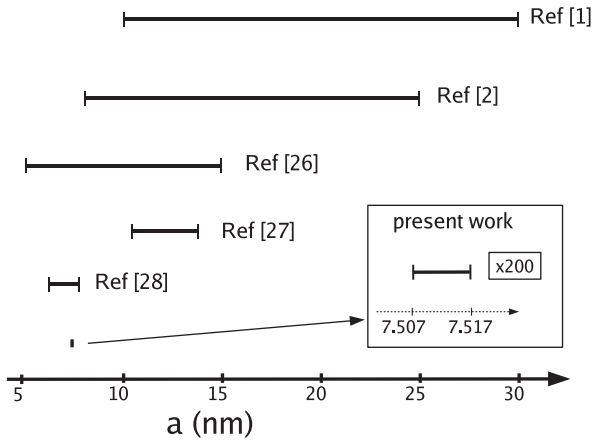

FIG. 1. Comparison of the present result with previous experimental determinations of the s-wave scattering length a of spin-polarized metastable helium: from the expansion of the condensate [1,2], from the evaporative cooling rate [26], from the observation of inelastic collisions [27], and from light-induced frequency shifts in one-photon photoassociation [28].

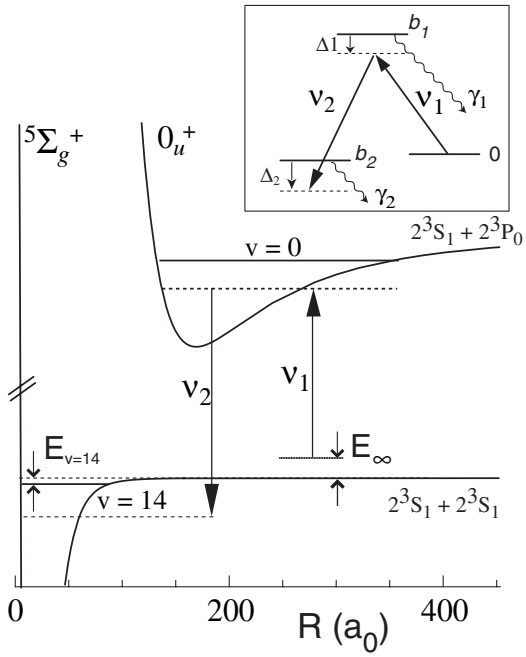

FIG. 2. Levels involved in the two-photon photoassociation experiment. ${}^5\Sigma_g^+$ is the interaction potential between two spin-polarized $2\,{}^3S_1$ helium atoms, $v = 14$ is the least-bound state in this potential. Laser L_1 at frequency ν_1 operates on the free-bound transition with a detuning Δ_1, and laser L_2 at frequency ν_2 drives the bound-bound transition with a detuning Δ_2 from the two-photon transition. The levels b_1 ($v = 0$) and b_2 ($v = 14$) are characterized by their decay rates γ_1 and γ_2.

widths $\hbar\gamma_1$ and $\hbar\gamma_2$. As a consequence, our signal is not affected by thermal averaging [14].

The metastable helium sample is confined in a magnetic trap of three-coil Ioffé-Pritchard-type with a bias field of 3 G and operated slightly above condensation at a temperature of a few μK and at a density of a few 10^{13} cm^{-3}. The PA beams of lasers L_1 and L_2 are derived from a distributed Bragg reflector diode laser operating in an external cavity (linewidth of order 600 kHz) at 1083 nm. Its frequency is tuned close to the 0-b_1 transition, 1.4 GHz red of the $2\,{}^3S_1$-$2\,{}^3P_0$ atomic transition. The frequencies ν_1 and ν_2 are generated from this laser using acousto-optical modulators, which ensures that both laser beams are coherent and eliminates the frequency jitter of the lasers. The two superimposed laser beams are focused through the same optical fiber (waist size of 300 μm) on the atomic cloud, which is much smaller (a few tens of μm). The pulse duration is adjusted to optimize the PA signals. We registered signals with a fixed value of the frequency ν_2 of laser L_2 and used laser L_1 as a probe, scanning its frequency ν_1 in the vicinity of the free-bound transition 0-b_1. We measured the temperature rise of the cloud, as well as the decrease of optical density and the loss of atoms. The three methods give the same resonance positions, but the temperature rise signal has the best quality [5].

In a first series of experiments, we used large intensities I_2 such that laser L_2 induces a coupling (Rabi frequency Ω) between b_1 and b_2 larger than the linewidth $\gamma_1/2\pi =$ 3 MHz of the excited state b_1 ($\Omega \gtrsim \gamma_1$). In Fig. 3(a) the frequency ν_2 is set close to resonance with the b_1-b_2 transition ($\Delta_1 \approx \Delta_2$). The Rabi frequency is $\Omega/2\pi =$ 4 MHz. We clearly identify a double-peak structure corresponding to an Autler-Townes splitting [15]: the two molecular bound states b_1 and b_2 are dressed with the light field of laser L_2, forming a doublet probed by laser L_1. We checked that the separation between the two peaks increases linearly with the square root of the intensity I_2 according to the Rabi frequency Ω. We also studied the case when laser L_2 is set farther from resonance with the b_1-b_2 transition ($\Delta_1 \neq \Delta_2$). One of the two peaks gets closer to the frequency of the 0-b_1 transition while the second one, shown in Fig. 3(b), becomes asymmetric and is associated with a stimulated Raman process between 0 and b_2. When increasing $\Delta_1 - \Delta_2$, one progressively goes from the purely Autler-Townes case to the stimulated Raman effect. In the case where ν_2 is set such that the two peaks are symmetrical, the abscissa of the middle point between the two peaks in Fig. 3(a) can be used to deduce the energy of the b_1-b_2 transition, from which the energy E_{b_2} can be derived. An extrapolation to zero laser intensity is necessary to take light shifts into account.

In order to derive a very precise measurement of E_{b_2}, we performed another series of experiments at smaller intensity I_2 ($\Omega \lesssim \gamma_1$) taking advantage of the phase coherence between the two lasers in our setup. Here, the intensity I_2 is not high enough to induce an Autler-Townes splitting. We find dark resonances occurring when the Raman resonance condition $E_\infty + h\nu_1 - h\nu_2 = E_{b_2}$ is satisfied. They are interpreted as an interference feature: a linear superposition of states 0 and b_2 is created such that a destructive interference occurs between the absorption amplitude of photon 1 on the 0-b_1 transition and of photon 2 on the b_1-b_2 transition. The photoassociation process is thus blocked, and a very narrow dip appears at the center of the main PA line when laser L_1 is scanned [see Fig. 3(c)]. The dark resonance observed in our experiment does not result in a complete suppression of the PA signal, due to the finite lifetime of level b_2. This method offers several advantages. The position of the dark resonance is independent of the position of the excited state b_1 and is not affected by a Zeeman shift, as both 0 and b_2 states are expected to have nearly the same magnetic moment [16]. The three solid lines of Fig. 3 represent a fit based on the theory of [17,18], taking into account only the heating by the decay product of b_1. The detailed analysis of the asymmetric line shape of Fig. 3(b), typical of a Fano profile (see [19] and references therein), shows that the heating due to b_2, which would give rise to a symmetrical profile with a similar width, is negligible [see Fig. 4(a) of [20]]. We have also checked that a small but non-negligible

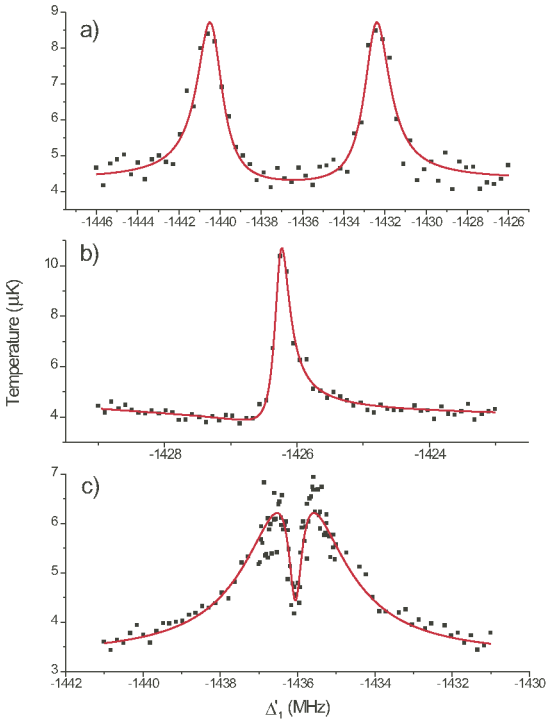

FIG. 3 (color online). Two-photon PA experiments with metastable helium. Temperature of the cloud as a function of frequency detuning Δ_1' of laser L_1 from the atomic resonance for a fixed frequency ν_2 of laser L_2 [detuning from the atomic resonance $\Delta_2' = -1345.0, -1335.0, -1344.6$ MHz in (a), (b), (c), respectively]. Laser intensities: $I_1 = 7\ \mathrm{mW \cdot cm^{-2}}$, $I_2 = 330\ \mathrm{mW \cdot cm^{-2}}$ in (a), $I_1 = 140\ \mathrm{mW \cdot cm^{-2}}$, $I_2 = 33\ \mathrm{mW \cdot cm^{-2}}$ in (b), and $I_1 = 7\ \mathrm{mW \cdot cm^{-2}}$, $I_2 = 7\ \mathrm{mW \cdot cm^{-2}}$ in (c). Data are remarkably well fitted using the theory of Refs. [17,18] (see text). (a) The Autler-Townes doublet, (b) the Fano profile of the Raman signal for $\Delta_1 - \Delta_2 \approx 10$ MHz, and (c) a narrow dip attributed to a dark resonance signal. The Zeeman shift of the one-photon transition and the temperature shift have to be added to the value of Δ_1' to compare it with the binding energy $E_{v=0} = -1418.1$ MHz quoted in [29]. Note the differences between the horizontal scales in the three figures.

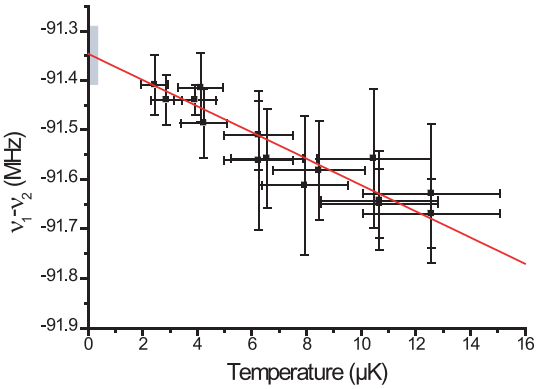

FIG. 4 (color online). Position $\nu_1 - \nu_2$ of the dark resonance signal [see Fig. 3(c)], as a function of temperature T. The slope of the straight line is compatible with a dependence of $3k_BT/2$ in unit of h (see text). On the vertical axis we indicate the final error bar, which we attribute to the binding energy of the molecule in the $v = 14$ state.

contribution of b_2 to the heating would not change the center of the symmetrical dark resonance of Fig. 3(c) used to measure $E_{v=14}$.

Several dark resonance spectra were recorded for atomic clouds prepared with a temperature between 2 and 10 μK while keeping the atomic density in the range of a few 10^{13} cm^{-3}. The temperature T is evaluated with an estimated precision of 20% by fitting the cloud expansion in free flight. Since k_BT is small enough ($k_BT \lesssim \hbar\gamma_2$), the calculation of the thermal averaging of the line shape shows that the width and shape of the resonance do not depend on T, but its position is shifted by an amount equal to $3k_BT/2$. This prediction has been checked experimentally: Fig. 4 shows a linear decrease of the resonance parameter ν_1-ν_2 as a function of T, with a slope that matches $3k_B/2h$ within 15%. For the very low intensities used here, we have checked that various corrections associated with light shifts, mean-field interaction, and laser frequency measurement are smaller that the error bar given for our result. We found no Zeeman shift of the two-photon transition when varying the magnetic field between 0.5 and 6 G. For the extrapolated position at zero temperature we thus adopt the value and the conservative error bar indicated on the vertical axis in Fig. 4. Finally, we find the value $E_{v=14} = -91.35 \pm 0.06$ MHz for the binding energy of level b_2 ($v = 14$).

From this measured value, we derive the s-wave scattering length of spin-polarized metastable helium. We use the most precise available interaction potentials derived by Przybytek and Jeziorski from *ab initio* calculations. In reference [21], a Born-Oppenheimer potential is provided, as well as its upper and lower bounds estimated from the extrapolated calculation to a complete molecular basis set. We use several potentials within these bounds by scaling the inner part of the original Born-Oppenheimer potential. For each potential the energy of the least-bound state $E_{v=14}$ and the scattering length a can be calculated, which yields a relation between $E_{v=14}$ and a. We check that the adiabatic, relativistic, and quantum electrodynamic corrections and their uncertainties as calculated in [21] introduce errors on the determination of a smaller than those due to the experimental uncertainty on $E_{v=14}$. To be conservative, the error bar on a obtained from the experimental measurement is therefore just doubled in our final result: $a = 7.512 \pm 0.005$ nm.

We also derive an estimate of the lifetime of the $v = 14$ molecular state by studying the width of the Raman peak [see Fig. 3(b)], which is directly related to γ_2 for large detunings $\Delta_2 - \Delta_1$. We additionally confirm this result using the fit of the dark resonance line shape of Fig. 3(c). The temperature and laser intensity that we use are checked to have no influence on the width. We thus find

$0.05 < \gamma_2/2\pi < 0.3$ MHz, corresponding to an intrinsic lifetime between 0.5 and 3 μs for the molecule. The main causes of inelastic decay for this molecule are likely to be spin relaxation and relaxation induced ionization [22,23], giving a roughly estimated lifetime of 4 μs quoted in [20], which is in good agreement with the presently measured linewidths. However, other decay mechanisms like atom-molecule collisions could occur. We plan to add ion detection to our setup to get more insight on the decay mechanism of b_2 since it has been shown that no ions are produced from the decay of b_1.

Our results can be compared to some very recent experiments on atom-molecule dark states. In a Bose-Einstein condensate of Rb [12], the width of the dark resonance was due to atom-molecule collisions in the limit of vanishing laser intensity. In an ultracold sample of Na atoms the linewidth was limited by the temperature [10]. In our specific case, the linewidth is not temperature limited although we are not in the condensate regime, because the ground molecular state b_2 is very short-lived compared to the alkali case.

In conclusion, we prepared exotic, doubly excited molecules in the very weakly bound state $v = 14$ in the interaction potential between two spin-polarized metastable helium atoms. We measured its binding energy $E_{v=14} = -91.35 \pm 0.06$ MHz, providing a very stringent experimental test of *ab initio* calculations [21,24,25]. The most precise theoretical value $E_{v=14} = -87.4 \pm 6.7$ MHz, which has been recently reported [21] is in very good agreement with our measurement. We deduced the value of the s-wave scattering length between two metastable atoms $a = 7.512 \pm 0.005$ nm, which is by far the most accurate experimental determination yet reported. We find very good agreement with our previous determination ($a = 7.2 \pm 0.6$ nm) based on light-induced frequency shifts in PA experiments. The discrepancy with the value found in [27] ($a = 11.3^{+2.5}_{-1.0}$ nm) remains to be explained. Finally, we gave an estimation of the lifetime of the exotic molecular state of the order of 1 μs, which could be useful for the theory of Penning ionization.

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[1] A. Robert, O. Sirjean, A. Browaeys, J. Poupard, S. Nowak, D. Boiron, C. I. Westbrook, and A. Aspect, Science **292**, 461 (2001).

[2] F. Pereira Dos Santos, J. Léonard, J. Wang, C. J. Barrelet, F. Perales, E. Rasel, C. S. Unnikrishnan, M. Leduc, and C. Cohen-Tannoudji, Phys. Rev. Lett. **86**, 3459 (2001).

[3] M. Leduc, J. Léonard, F. P. D. Santos, E. Jahier, S. Schwarz, and C. Cohen-Tannoudji, Acta Phys. Pol. B **33**, 2213 (2002).

[4] R. J. W. Stas, J. M. McNamara, W. Hogervorst, and W. Vassen, Phys. Rev. Lett. **93**, 053001 (2004).

[5] J. Léonard, M. Walhout, A. P. Mosk, T. Muller, M. Leduc, and C. Cohen-Tannoudji, Phys. Rev. Lett. **91**, 073203 (2003).

[6] B. Laburthe Tolra, C. Drag, and P. Pillet, Phys. Rev. A **64**, 061401(R) (2001).

[7] C. Lisdat, N. Vanhaecke, D. Comparat, and P. Pillet, Eur. Phys. J. D **21**, 299 (2002).

[8] U. Schlöder, T. Deuschle, C. Silber, and C. Zimmermann, Phys. Rev. A **68**, 051403(R) (2003).

[9] J. Weiner, V. S. Bagnato, S. Zilio, and P. S. Julienne, Rev. Mod. Phys. **71**, 1 (1999), and references therein.

[10] R. Dumke, J. D. Weinstein, M. Johanning, K. M. Jones, and P. D. Lett., Phys. Rev. A **72**, 041801(R) (2005).

[11] R. Wynar, R. S. Freeland, D. J. Han, C. Ryu, and J. Heinzen, Science **287**, 1016 (2000).

[12] K. Winkler, G. Thalhammer, M. Theis, H. Ritsch, R. Grimm, and J. H. Denschlag, Phys. Rev. Lett. **95**, 063202 (2005).

[13] C. Ryu, X. Du, E. Yesilada, A. M. Dudarev, S. Wan, Q. Niu, and D. J. Heinzen, cond-mat/0508201.

[14] R. Napolitano, J. Weiner, C. J. Williams, and P. S. Julienne, Phys. Rev. Lett. **73**, 1352 (1994).

[15] S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).

[16] In the absence of hyperfine coupling in ^{4}He, the only interaction that mixes different spin states and could induce a different magnetic moment for the states 0 and b_2 is the very weak spin-dipole interaction.

[17] J. L. Bohn and P. S. Julienne, Phys. Rev. A **54**, R4637 (1996).

[18] J. L. Bohn and P. S. Julienne, Phys. Rev. A **60**, 414 (1999).

[19] C. Cohen-Tannoudji and B. Lounis, J. Phys. II (France) **2**, 579 (1992).

[20] J. C. J. Koelemeij and M. Leduc, Eur. Phys. J. D **31**, 263 (2004).

[21] M. Przybytek and B. Jeziorski, J. Chem. Phys. **123**, 134315 (2005).

[22] P. O. Fedichev, M. W. Reynolds, U. M. Rahmanov, and G. V. Shlyapnikov, Phys. Rev. A **53**, 1447 (1996).

[23] V. Venturi, I. B. Whittingham, P. J. Leo, and G. Peach, Phys. Rev. A **60**, 4635 (1999).

[24] J. Stärck and W. Meyer, Chem. Phys. Lett. **225**, 229 (1994).

[25] A. S. Dickinson, F.-X. Gadéa, and T. Leininger, J. Phys. B **37**, 587 (2004).

[26] P. J. J. Tol, W. Hogervorst, and W. Vassen, Phys. Rev. A **70**, 013404 (2004).

[27] S. Seidelin, J. V. Gomes, R. Hoppeler, O. Sirjean, D. Boiron, A. Aspect, and C. I. Westbrook, Phys. Rev. Lett. **93**, 090409 (2004).

[28] J. Kim, S. Moal, M. Portier, J. Dugué, M. Leduc, and C. Cohen-Tannoudji, Eur. Phys. Lett. **72**, 548 (2005).

[29] J. Kim, U. D. Rapol, S. Moal, J. Léonard, M. Walhout, and M. Leduc, Eur. Phys. J. D **31**, 227 (2004).