Cold Rubidium Molecules Formed in a Magneto-Optical Trap

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We report the first observation of translationally cold ($\sim 90\ \mu$K) Rb_2 molecules. They are produced in a magneto-optical trap in their triplet ground state. The detection is performed by selective mass spectroscopy after two-photon ionization into Rb_2^+, resonantly enhanced through the intermediate $a\,^3\Sigma_u^+ \rightarrow 2\,^3\Pi_g$ molecular band. The two rubidium isotopes present very different types of behavior that are interpreted in terms of their respective collisional properties.

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The availability of cold molecular samples, at temperatures in the μK–mK range, and the possibility of storing them in traps on a time scale of seconds are opening up new perspectives in chemistry, metrology, and quantum physics. Nevertheless, their production is still limited. While laser cooling is routinely used to cool alkali, alkaline-earth metals, and metastable inert atomic vapors down to temperatures in the μK range or below, their application to molecules is not straightforward, due to the absence of closed optical transitions. Molecules irradiated with laser light would be optically pumped into "dark" levels well before translational cooling is completed [1].

Photoassociation (PA) of two cold colliding atoms into an excited dimer by the resonant absorption of a laser photon was proposed [2] as a possible source of translationally cold molecules. In fact, photoexcited molecules can spontaneously decay into their ground states while remaining translationally cold. Notwithstanding the rather small Franck-Condon factors expected for the spontaneous decay step, the process is quite efficient. A group in Orsay [3] reported the first observation of cold Cs_2 molecules produced in a magneto-optical trap (MOT) through photoassociation into long-range excited states [4]. Recently, PA of cold potassium atoms led to the observation of cold K_2 molecules, produced in deeply bound levels of the singlet ground state [5].

Optical trapping of cold cesium molecules has subsequently been demonstrated [6] in a dipolar trap with a CO_2 laser. In a very different approach, not based on laser cooling, CaH molecules have been cryogenically cooled to a few hundred mK through collisions with helium buffer gas, and loaded into a magnetic trap [7].

While the PA mechanism has demonstrated the production of cold molecules for both cesium and potassium, Cs cold molecules have been observed in a MOT even under the absence of a specific PA laser [3,6]. Two different mechanisms were proposed for the molecule formation: photoassociation induced by the trapping and repumping lasers [3] and three-body recombination [6]. While the former process relies on laser excitation, in the latter case no laser photons are required. However the mechanism that produces molecules directly in a Cs MOT has not yet been completely clarified.

In this Letter we report the first observation of translationally cold rubidium molecules for both ^{85}Rb and ^{87}Rb isotopes, at a temperature $T_{\mathrm{mol}} \sim 90\ \mu$K. They are produced by photoassociation in a MOT in their radiatively-stable $a\,^3\Sigma_u^+$ triplet ground state, and are observed by photoionization into molecular Rb_2^+ ions, detected after a mass selection. Furthermore, while ^{85}Rb cold molecules are also directly produced during the MOT operation without any additional PA laser, the reverse is true for ^{87}Rb trapped in similar conditions. The dependence of the molecular formation rate on the atomic density shows the important role of a three-body process. We interpret these results in terms of the different collisional properties of the two isotopes.

The present experiment is performed with rubidium, which is widely used in laser cooling and led to the first demonstration of a Bose-Einstein condensate (BEC) in a dilute medium [8]. The collisional properties of Rb at ultralow temperatures are very well studied, and accurate scattering length data for both rubidium isotopes are now available [9]. Moreover, the long-range part of the first excited molecular potential curves has been carefully investigated by photoassociation spectroscopy [10].

A magneto-optical trap [11] is produced in the high vacuum environment of a stainless steel cell. Several windows allow for optical access and for positioning of a channeltron multiplier near the MOT [12]. The background Rb pressure is kept typically in the 10^{-8} torr range. The trap is created at the center of a quadrupolar magnetic field, produced by two coils in an anti-Helmholtz configuration, where three pairs of counterpropagating, retroreflected laser beams (1 cm diam) in the standard σ^+/σ^- configuration orthogonally cross each other. The trapping laser is a 50 mW cw diode laser (SDL5401-G1), injection locked by an extended cavity diode laser. This master laser has a linewidth below 1 MHz and is frequency locked 12 MHz to the red side of the $F_g = 3 \rightarrow F_e = 4$

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hyperfine transition of the ^{85}Rb D_2 line at 780 nm (or $F_g = 2 \rightarrow F_e = 3$ transition for ^{87}Rb). Light from another diode laser (ML64114R), tuned to the D_1 line, is split into two beams and superposed onto the trapping beam in two arms. This avoids optical pumping into the lower hyperfine level of the ground state. By simply changing the master and repumping laser locking points, it is possible to switch from a ^{85}Rb trap to a ^{87}Rb one, while maintaining the same optical alignment and thus similar trapping conditions. An acousto-optic modulator allows a fast shutting of the trapping laser.

In typical operating conditions, the MOT, which is continuously monitored by a photodiode and a CCD camera, is loaded with about $N \sim 10^7$ rubidium atoms in an almost Gaussian distribution of 0.8 mm diam, leading to a maximum peak density $n \sim 3 \times 10^{10}$ cm^{-3}. The atomic temperature, measured by the release and recapture method, is 120 ± 40 μK.

Two additional lasers can be applied to the MOT. A pulsed dye laser, pumped by the second or third harmonic of a Nd:YAG laser (7 ns pulse duration and 10 Hz repetition rate), is the ionizing source. We used pulse energies of about 1 mJ and focused the laser beam onto the trap to a waist of the order of 1 mm, i.e., slightly larger than the MOT itself. Its emission wavelength λ_1 is tuned using several dyes to cover most of the 460–750 nm excitation range. The PA laser is a cw free running diode laser (Sanyo DL-7140-201) of 70 mW maximum output power, with a linewidth of about 20 MHz. Its emission wavelength λ_2, centered near 781 nm, is continuously tuned by varying the diode temperature, and is monitored by a λ-meter (NIST LM-10) and a Fabry-Perot interferometer, which provide, respectively, an absolute measure of the laser wavelength and a finely calibrated relative scale. The PA laser is focused to the MOT position in a spot smaller than the atomic cloud.

The first observations were performed on ^{85}Rb without PA laser. When the ionizing pulsed laser is applied to the MOT, a signal due to atomic ions is detected by the channeltron, with a time-of-flight delay $t_{\rm at} = 1.8$ μs, and is recorded by a boxcar integrator, averaging typically over 30 laser pulses. Atomic ions are produced by three-photon ionization of ground-state atoms and by two-photon ionization of excited-state ones. When the dye wavelength λ_1 is tuned into the 600–609 nm spectral region, a second well-defined peak appears at a delay $t_{\rm mol} \simeq \sqrt{2}\, t_{\rm at}$, showing evidence for rubidium dimer detection. The molecular ion signal corresponds to 2–3 detected ions per shot and is due to Rb_2 molecules produced inside the MOT.

To test the electronic state in which molecules are produced, we scanned the ionizing pulsed laser wavelength λ_1. The spectrum of the molecular ions, reported in Fig. 1a, shows a complex band structure. The spectral region corresponds to a "diffuse" band of Rb_2 connecting the ground triplet state $a\,^3\Sigma_u^+$ to the excited $2\,^3\Pi_g$ state, studied in thermal alkali samples some years ago [13]. This

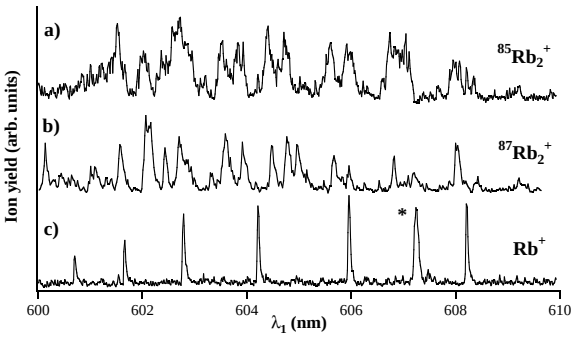

FIG. 1. Ion spectra for a Rb MOT as a function of the dye laser wavelength λ_1. The boxcar integrator is gated at the molecular (a, b) and at the atomic (c) times of flight, respectively. In case (b) a PA laser, red detuned by ~10 GHz from the $F_g = 2 \rightarrow F_e = 3$ ^{87}Rb resonance, is present. Peaks in (c) are due to atomic two-photon resonances to Rydberg states ($18D-13D$, from left to right); the peak labeled with (*) corresponds to the $5P_{3/2} \rightarrow 5F$ transition.

correspondence, and the absence of the molecular signal in other spectral regions of the covered range, allows us to identify the observed $^{85}Rb_2$ molecules as being essentially in their triplet ground state [14].

The translational molecular temperature is measured by switching off the trapping laser for a period of 20 ms and by detecting the number of molecular ions as a function of the delay in the application of the ionizing pulse (Fig. 2). From the comparison of these results with the outcome of a simple model of the production and release of molecules in the MOT, and ionization within a given volume [15], we deduce a molecular temperature of 90 ± 50 μK, consistent with the atomic one within the experimental error. This result ensures that the observed rubidium molecules are cold and long lived.

The cold molecule production was then investigated as a function of the wavelength λ_2 of a cw PA laser. A sample of the results, which are currently limited by the power and the tunability of the available PA laser, is shown in Fig. 3,

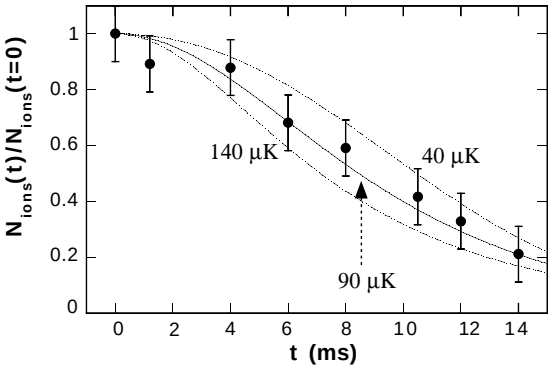

FIG. 2. Molecular ion yield as a function of the delay between the dye laser pulse and the switch-off of the trapping laser. The continuous lines are the result of a model calculation for our geometry (see text).

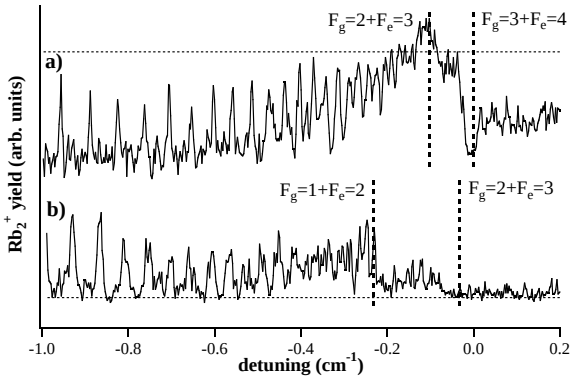

FIG. 3. Molecular ion spectra as a function of the PA laser photon energy for ^{85}Rb (a) and ^{87}Rb (b), respectively, for $\lambda_1 = 602.7$ nm. The horizontal dashed lines represent the mean number of molecular ions detected under absence of the PA laser. The vertical dashed lines indicate the positions of the excited molecular asymptotes for the two isotopes.

curve a. The application of the PA laser generally produces a clear quenching of the molecular signal [16]. Peaks in the molecular signal are, however, observed for a series of laser frequencies when molecular states are resonantly excited and partially decay into ground-state molecules. By comparing the observed peaks to published rubidium PA spectroscopic data [10,17], we can identify them as corresponding to transitions to vibrational levels of the 0_g^- long-range electronic state.

When we load ^{87}Rb into the MOT in similar trapping conditions, an important difference is observed: cold molecules are also produced through photoassociation, but no molecular signal is detected directly from the MOT alone. This difference cannot be ascribed either to a difference in the number of trapped atoms (the density being equal within 50%) or to a possible different ionizing efficiency at the chosen wavelength $\lambda_1 = 602.7$ nm (see Fig. 1). Under the action of the PA laser, instead, cold molecules are observed, and a single vibrational series is recorded in the spectrum, with a level spacing consistent with that of the 0_g^- state (Fig. 3, curve b).

The PA production mechanism in rubidium is very similar to that observed in cesium, where the $0_g{}^-$ and 1_u long-range states [4] are photoexcited and decay into ground-state molecules [3,18]. In that case, a large molecular formation rate is allowed by the peculiar double well shape of the potentials. Moreover, tunneling through the barrier enhances the molecular formation for specific vibrational levels. In rubidium, the $0_g{}^-$ state has a double well shape, with an external well-depth of ~ 28 cm^{-1}, and a minimum at $R_e \simeq 17$ Å [17] internuclear distance. In this case, tunneling effects should not be present as the internal barrier is too high to be overcome [19]. Nevertheless, the shape of its potential curve allows significant Franck-Condon factors for decay into ground-state triplet molecules, as observed.

In the absence of a PA laser, two mechanisms can contribute to the molecular formation: three-body recombination and PA by the trapping and repumping lasers. Excitation by these lasers in a binary collision takes place at very large internuclear distances ($R \sim 500$ Å), and spontaneous decay back into two free atoms is very likely to occur [20]. In order to produce ground-state dimers, either multiple excitation–spontaneous emission processes or nonadiabatic transitions in the excited state are required. Realistic modeling of these mechanisms is a very complex task, because it involves the molecular hyperfine structure. The different hyperfine structure of the two Rb isotopes could cause a difference in the molecular formation rates, as already observed for the trap loss rates [21].

Three-body recombination for cold atoms depends essentially on the scattering length a of the diatomic species, which completely determines the collisional properties at ultralow temperatures [22,23]. Recent calculations [23] show that three-body recombination rates strongly depend on the sign and absolute magnitude of a. In particular, much larger rates result for atoms with negative scattering lengths. For ^{87}Rb in the spin-polarized $5S_{1/2}(F_g = 2, m_F = 2)$ state ($a \simeq +56$ Å [9]), both an experimental measurement performed in a Bose-Einstein condensate [24] and the latest theoretical calculations [23] give a recombination event rate constant $K_3 \simeq 2.2 \times 10^{-28}$ cm^6/s. Both the BEC experiment and the calculations are done in the pure s-wave limit that corresponds to an atomic temperature much lower than that of our experiment. Therefore the use of this rate constant value is not completely justified in our case. However, using this value, the density and the total number of atoms of our trap lead to a $^{87}Rb_2$ production rate lower than 0.1 mol/s. For ^{85}Rb (triplet scattering length $a \simeq -162$ Å [9]) no experimental measurements exist, but the same theoretical calculations [23] give a rate constant $K_3 \simeq 6.7 \times 10^{-25}$ cm^6/s, i.e., 3000 times larger. This rate constant would lead, in our condition, to a $^{85}Rb_2$ production rate of 200 mol/s.

The observed molecular ion signal in the absence of the PA laser is typically 2–3 ions/pulse with ^{85}Rb, and less than 0.1 ions/pulse with ^{87}Rb. Considering that the molecules remain in the ionization region for nearly 10 ms, that the repetition frequency of the pulsed laser is 10 Hz, and that the combined efficiency of the ionization and the detection processes is of the order of 0.5, the molecular production rate in the ^{85}Rb MOT is about 400 s^{-1}. This is consistent with the aforementioned calculations within the experimental uncertainty in the atomic density n and ionization probability. To verify the three-body hypothesis, we measured for ^{85}Rb the dependence of the molecular ion rate as a function of n, changed by varying the MOT magnetic field gradient. A fit of the results, shown in Fig. 4, clearly indicates a quadratic dependence on the density of the rate per atom, which is expected for a three-body recombination process. A possible effect of the trapping and repumping lasers in enhancing the flux of colliding atoms

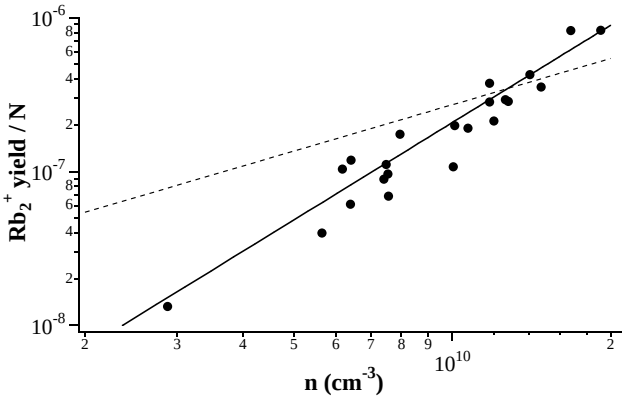

FIG. 4. Number of molecular ions per trapped atom and per pulse as a function of the peak atomic density. The solid curve is the best fitting power function to the data, having an exponent 2.1 ± 0.2. The dashed line is the best fit for a linear function. The absolute error in the density determination is about 50%, while the relative error is a factor of 2 lower.

at short internuclear distances [20] could also be present, and requires further investigation.

In summary, we have presented the first observation of translationally cold rubidium molecules at a temperature of about 90 μK. They are formed in their lowest triplet state, and are detected by resonance enhanced photoionization into Rb_2^+ ions through the intermediate $2\,^3\Pi_g$ molecular state. Cold molecules are produced in a ^{85}Rb MOT directly during the trap operation, mainly formed by three-body recombination. Molecules in the $X\,^1\Sigma_g^+$ ground state could also be produced in this way, but an appropriate ionization scheme should be used to detect them. Cold molecules resulting from photoassociation are also observed for both of the rubidium isotopes, through excitation to the 0_g^- long-range state, and PA spectra for both isotopes are reported. The use of a stronger PA laser and a denser cold atomic sample should considerably increase the molecular production rate. By trapping in either a magnetic or a dipolar trap a large number of cold triplet molecules, a wide range of experiments will become feasible.

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