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Circular Rydberg-state spectroscopy

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We report the first observation of a resonance induced by a submillimeter wave between circular atomic Rydberg states of maximum angular momentum ($n=25, m_l=24 \rightarrow n=24, m_l=23$ transition in lithium). This experiment is the first step towards a possible measurement of the Rydberg constant directly in frequency units. The Stark shift of the resonance has also been studied and its quadratic character put in evidence.

Circular Rydberg states,[1] corresponding to a large principal quantum number n together with the maximum possible orbital angular momentum projection along the quantization axis ($|m_l|=n-1$) are quasiclassical atomic systems with very peculiar properties. The valence electron distribution in these states has the shape of a thin torus of radius n^2a_0 and width na_0 (a_0 is the Bohr radius) perpendicular to the quantization axis and centered on the atomic core. Because the Rydberg electron practically does not penetrate this core, circular states of all elements have nearly the same valence-electron wave function and their binding energy differs from the hydrogen one, $E_n=-e^2/2a_0n^2$, by an exceedingly small shift[2] $\delta E_n=-\alpha_0' e^2/2a_0^4n^8$ (α_0' ion-core polarizability). Furthermore, since the valence electron moves in a quasi-two-dimensional orbital, these atoms do not have any dipole along the quantization axis and are thus relatively insensitive to electric field components perpendicular to the orbit plane (their Stark effect is quadratic in this direction). Moreover, all circular states radiatively decay only through $\Delta n=-1$, $\Delta m_l=-1$ circularly polarized radio frequency transitions and have very long spontaneous-emission lifetimes τ_n, scaling as n^5 ($\tau_n \sim 2$ ms for $n=30$).[3]

The above characteristics—among many other interesting ones—make these systems very promising for high-resolution microwave spectroscopy in view of a determination of the Rydberg constant directly in frequency units.[1] One can indeed expect very narrow resonances between circular states, with spectral lines only quadratically sensitive to stray electric fields and frequencies depending only slightly upon the atom ionic-core properties and being easily related to the hydrogen frequencies via the determination of very small quantum-defect corrections.

In this Rapid Communication, we report the first observation of a circular to circular Rydberg-state resonance induced in the millimeter wave domain ($n=25, m_l=24 \rightarrow n=24, m_l=23$ transition at 447.749 GHz). This experiment, whose resolution is still moderate, demonstrates the feasibility of circular Rydberg-state spectroscopy and points out the difficulties to overcome in order to reach a metrological resolution in the Rydberg-constant measurement.

Our experiment is performed on an atomic beam of lithium and the circular Rydberg states are prepared by the adiabatic microwave transfer method (AMTM) first demonstrated by Hulet and Kleppner.[1]

The lithium energy levels relevant for our experiment are shown on Fig. 1(a) and the experimental setup sketched on Fig. 1(b). The atomic energy excitation is performed via a stepwise process involving 3-ns long dye laser pulses tuned in resonance with the $2s \rightarrow 2p$, $2p \rightarrow 3d$, and $3d \rightarrow n=25$ transitions, respectively, at the wavelengths indicated on Fig. 1(a) (upwards solid line arrows on the figure).

Following the laser excitation, AMTM is used to bring the Rydberg atoms into a circular orbit. This method requires a homogeneous electric field F_1 (~ 200 V/cm in our experiment) produced by the stack of equally spaced metal-

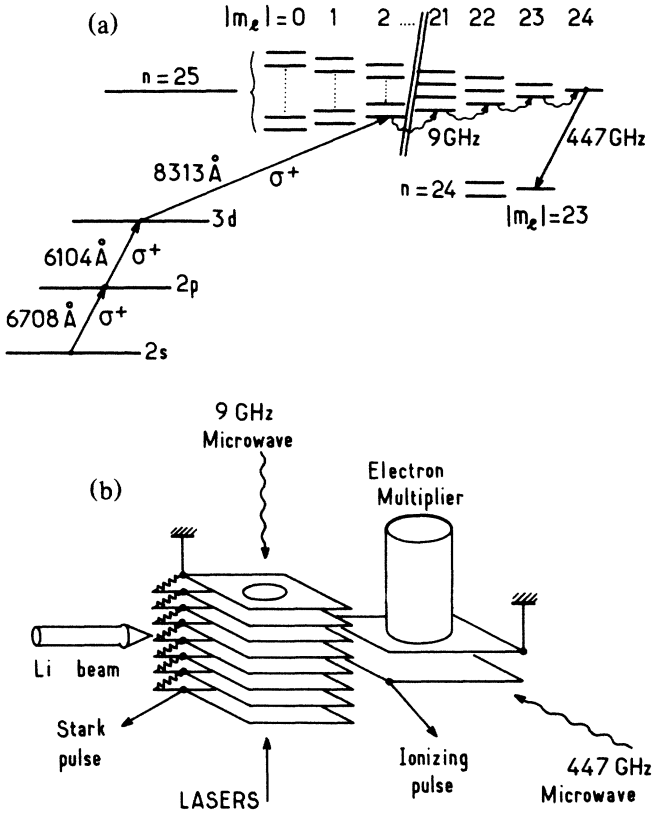

FIG. 1. (a) Energy levels of lithium relevant for the experiment. Some of the Stark states of the $n=25$ and $n=24$ levels are represented on the right, the various m_l manifolds being displayed as vertical energy scales. The route followed by an atom during an experimental sequence is marked by arrows: optical excitation (upwards arrows), adiabatic centimeter microwave transitions (wavy lines, there are 22 such transitions), and submillimeter wave-induced resonance (solid line downwards arrow). (b) Sketch of experimental setup. The laser excitation and circular state preparation take place in the multiple condenser at left. The millimeter wave is sent inside the condenser at right, where the field-ionization takes place. Atoms experience a nonvanishing vertical electric field throughout their path from one zone to the other.

lic plates shown on Fig. 1(b), and a microwave field (9.2-GHz frequency in our experiment) polarized perpendicular to F_1 and inducing transitions among the Stark sublevels produced by F_1. The direction of F_1 defines the axis along which the angular momentum of the Rydberg states is quantized. The stark sublevels in this field are represented on the right upper side of Fig. 1(a). They can be ordered according to their m_l value: For each m_l, one has $n-|m_l|$ quasiequidistant Stark substates making up a vertical scale of levels and labeled by parabolic quantum numbers n_1, n_2 with $n_1+n_2+|m|+1=n$ ($n_1=0$ is the lower level in each m_l manifold).

Fine tuning of the 8313-Å laser pulse frequency enables us to select a given n_1 state in the m_l manifolds whose excitation is permitted by the optical polarization selection rules. We have used three σ_+ polarized light beams propagating along F_1 [see Fig. 1(b)] and have prepared the $n_1=0$ level of the $m_l=2$ manifold (the absorption of three circularly polarized photons from the $m_J=-\frac{1}{2}$ ground state enables us to excite an $m_J=\frac{5}{2}$ Rydberg state which has a nonzero projection onto an $m_l=2$ orbital angular momentum state: m_J refers to the total orbital plus spin angular momentum).

Once this state is optically prepared, F_1 is adiabatically swept down (from 200 V/cm to ~ 192 V/cm in about 10 μs), while the 9.2-GHz, 50-μW power microwave field is kept constant. In this way, a fast adiabatic passage of the Rydberg atom is performed along the succession of $\Delta m_l=+1$ microwave transitions marked by the wavy arrows on Fig. 1(a). At the end of this process, nearly all the Rydberg atoms are carried into the $n=25$, $m_l=24$ level, the angular momentum being fed into the atoms at nearly constant energy by the soft microwave photons. The Rydberg states angular momentum then points along the direction of F_1. The microwave field is subsequently switched off (~ 10 μs after the laser pulses).

The atoms leave the multiplate condenser in which they have been prepared and enter into a second condenser in which they undergo the millimeter wave transition at 447 GHz [solid line downwards arrow on the right of Fig. 1(a)] and are detected by the selective-field-ionization method[4] (SFIM). The submillimeter wave source is a frequency-stabilized backwave oscillator[5] (Thomson-C.S.F. carcinotron) which emits a microwave power of the order of 10 mW in the frequency interval from 415 to 485 GHz. The SFIM consists of applying to the atoms an ionizing field $F_2(t)$ parallel to the quantization axis and raising linearly from 0 to 2600 V/cm in about 8 μs. This field starts ~ 30 μs after the laser excitation. The threshold for ionization of the various Rydberg states relevant to our experiment are thus reached at different times in the field $F_2(t)$ ramp and a time resolution of these levels is achieved.

It is very important for the stability of the circular states between the excitation and the detection region that the atoms experience along their trajectory a nonvanishing electric field. If the field vanishes or abruptly changes direction along the atom path, it can be shown theoretically and verified experimentally[6] that the circular atoms rapidly loose their angular momentum and are transferred back towards small $|m_l|$ states. We have thus taken care to maintain a residual field of a few V/cm in the intermediate region between the two condensers.

Figure 2 shows the time resolution ionization signals obtained under various conditions. Figure 2(a) corresponds to

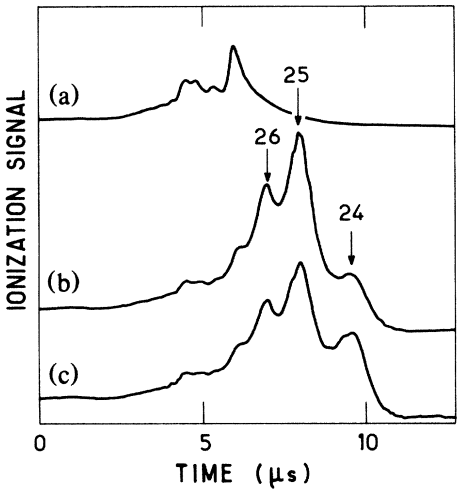

FIG. 2. Time-resolved ionization signals: (a) signature of the $n=25$, $m_l=2$ low angular momentum state (9- and 447-GHz microwaves off); (b) signature of $n=25$, $m_l=24$ circular Rydberg state together with $n=24$, $m_l=23$, and $n=26$ states populated by blackbody radiation transfer (9-GHz centimeter microwave on); (c) signal showing the resonant transfer towards the $n=24$, $m_l=23$ level induced from the prepared $n=25$, $m_l=24$ level, by the 447-GHz submillimeter wave.

the case when the 9- and 447-GHz microwave fields are switched off. The main peak in this figure corresponds to the $n=25$, $m_l=2$, $n_1=0$ Stark state ionizing at the time when F_2 reaches 1430 V/cm, in good agreement with the theoretically computed ionizing field for this level (the features appearing in weaker fields correspond to $m_l=0$, 1, and $m_l=3$ states also excited by the laser beams).

When the 9-GHz microwave field is switched on, three new features appear in the ionizing signal [Fig. 2(b)]. The largest peak, occurring in a 2000-V/cm field corresponds to the $n=25$, $m_l=24$ circular state, whereas the two smaller peaks at 2400 and 1730 V/cm can be assigned to the $n=24$, $m_l=23$, and $n=26$ ($m_l=25$, 24, and 23) adjacent states populated by blackbody radiation transfer from the $n=25$, $m_l=24$ level. The calculated blackbody transfer time between nearby circular states at room temperature (~ 62.5 μs) is of the order of the atom flight time in our experiment, which explains the large population of the $n=26$ and $n=24$ circular states. Note also the strong increase in the signal from Fig. 2(a) to Fig. 2(b), which comes from the increase of the Rydberg atom lifetime when they are transferred from a small to a large angular momentum state.

Figure 2(c), finally, shows the ionization signal obtained when the 447-GHz submillimeter wave is tuned to the $n=25\rightarrow n=24$ circular-circular Rydberg transition. We notice a resonant enhancement of the $n=24$ feature.

In order to record a spectrum, we have measured the variations of the $n=24$ peak as a function of the millimeter wave frequency, the result being shown on Fig. 3(a). The resonance is found to be strongly asymmetrical, with a sharp edge on the high-frequency side and a 50-MHz wide tail on the low-frequency one. This shape is characteristic of a quadratic Stark inhomogeneous broadening. In this preliminary experiment, the submillimeter wave field is interacting with the atoms everywhere along the beam and excites some atoms still perturbed by the relatively large field leak-

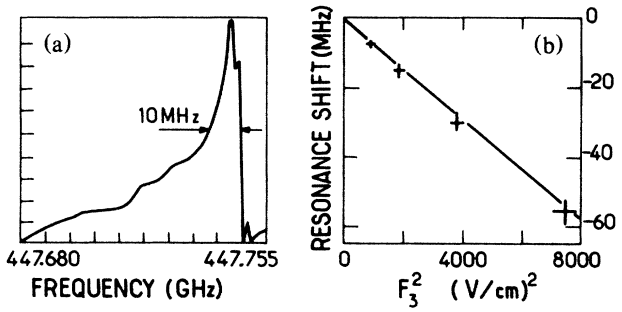

FIG. 3. (a) Spectrum of the $n=25$, $m_l=24 \rightarrow n=24$, $m_l=23$ resonance obtained by recording the $n=24$, $m_l=23$ level ionization-peak amplitude vs the submillimeter frequency: The inhomogeneous quadratic Stark broadening is clearly visible. (b) Resonance frequency shift as a function of the electric field squared. Solid line is theoretical.

ing from the condenser plates. Residual inhomogeneous fields of the order of 50 V/cm account for the observed line shape. The fact that the broadening occurs only on the low-frequency side of the line is a clear indication of a purely quadratic Stark shift, as expected from circular Rydberg states in a field parallel to the quantization axis.

The position of the sharp high-frequency edge of the line provides a measure of the unperturbed Rydberg frequency. We find $\nu_{25-24}=447\,749.5(5)$ MHz, in good agreement with the formula

$$\nu_{25-24}=R\,(1/24^2-1/25^2)=447\,749.028 \text{ MHz} ,$$

the Rydberg constant in ^{7}Li being

$$R=3.289\,584\,70(1)\times 10^9 \text{ MHz} .$$

The present resolution does not, of course, enable us to observe the small $\delta E_n-\delta E_{n+1}$ shift due to the Li^+ core polarization ($\delta E_{24}-\delta E_{25}\simeq 1$ kHz).

In order to study the circular static Stark shift, we have applied a third electric field F_3 across the detection condenser plates, kept constant during each experimental sequence, and plotted the shift of the resonance line sharp edge as a function of F_3^2. Figure 3(b) shows the measured frequency shifts, which are in agreement with the calculated quadratic Stark shift (solid line), $\Delta\nu=-7.11\ F_3^2$ [kHz/(V/cm)^2].

In order to improve on these very preliminary results, it is essential to confine the millimeter wave field in a region where the electric field amplitude is well controlled. A shielded cm-size microwave cavity tuned at the Rydberg-Rydberg frequency should be used for that purpose. The time of flight across the cavity should limit the linewidth to about 100 kHz. Cooling to liquid-helium temperature will suppress the blackbody radiation perturbations. In order to avoid inhomogeneous Stark broadenings at the linewidth level, the electric field in the cavity should be set to a known value in the 100-mV/cm range, which is not too difficult to achieve. (A well defined residual field, parallel to the direction of F_1 is indeed required as explained above to preserve the circular character of the Rydberg atoms in the cavity).

In a later stage of the experiment, the use of the Ramsey method of separated oscillatory fields[7] with two cavities separated by ~ 1 m (the average length of flight of the excited atoms which have a 1500-m/s thermal velocity) together with a control of the electric field down to the 10-mV/cm domain should permit reduction of the linewidth to the natural limit (240 Hz for $n=25 \rightarrow n=24$ transition) and hopefully to measure the Rydberg in a part in 10^{11}. At this precision level, the ion-core perturbation becomes significant and the experiment will eventually have to be carried out in hydrogen, and a systematic comparison between hydrogen and alkali Rydberg frequency will have to be performed.

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