Quantum Electrodynamic Shifts of Rydberg Energy Levels between Parallel Metal Plates

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The energy level shifts of Rydberg atoms in confined space were investigated by passing the atoms between two parallel metal plates. The shifts were measured by Doppler-free two-photon absorption in combination with the two-field Ramsey method. Radiative shifts induced by blackbody photons could be observed. At very low temperatures, the shift was determined being on the order of 100 Hz. [S0031-9007(98)08042-9]

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Since the pioneering work of Casimir and Polder [1] many authors have tried to improve the understanding of vacuum effects (see [2] for a review), especially shifts of atomic energy levels in a parallel plate geometry [3,4]. The amount of theoretical work available is in contrast to the total absence of efforts on the experimental side even though the planar cavity is considered to be the prototype geometry. Its special feature is the extreme selectivity in respect of the wave vectors **k** and the creation of a field cutoff frequency ν_{cutoff} for a particular polarization, with the result that only a discrete spectrum of the radiation field with frequencies greater than ν_{cutoff} is present inside the cavity. The modification of the field between two parallel plates allows one to isolate the contribution to the atomic frequency shift resulting from a single **k**. This modification is resonantly enhanced when the plate distance reaches either $d = \lambda_0/2$ (cutoff) or small odd multiples of $\lambda_0/2$, λ_0 being the transition wavelength to a neighboring atomic level. The restriction of the measurement to a particular contribution presents a special challenge owing to the tiny level shift expected.

There have been experimental attempts to determine radiative shifts for other cavity geometries [5–7]. In [5], a low-Q concentric resonator was used to observe a resonant frequency shift of 1 MHz in the fluorescence spectra of an optical transition of Ba atoms inside the resonator. In [6], ground-state Na atoms were transmitted through a wedge. From the atomic opacity of the wedge the authors derived information on the potential and used that to determine the energy shift. In [7] the authors used an open resonator with curved mirrors. The shift obtained from the dispersive interaction between a superposition of circular states in Rb was measured for different cavity detunings. Although the experiment was carried out at low temperature, the outcome included a non-negligible thermal influence which had to be corrected.

In the present work the cavity geometry is made up of two plane parallel copper plates. Rydberg atoms are sensitive probes since the transitions to neighboring levels are in the microwave region, resulting in a resonant plate distance in the mm range. With analysis limited to atomic states between $20S$ and $30S$, these atomic level changes are expected to be a few hundred Hz when thermal radiation at room temperature is additionally present within the cavity and is resonantly coupled to the relevant Rydberg transition. In a cavity without thermal radiation the effect is only of the order of 100 Hz [4]. Measurement of these cavity-induced resonances represents the main aim of this work.

The spectroscopic technique involves Doppler-free two-photon absorption in combination with the method of successive oscillatory fields [8]. This high-precision spectroscopy needed to measure the expected small shifts requires a highly frequency-stabilized laser system. It is based on the phase modulation method and has a remaining frequency noise on the Hz level [9].

The experimental scheme is shown in Fig. 1 [10]. A beam of rubidium atoms is produced via an atomic oven collimated via liquid nitrogen cooled circular apertures 200 μm in diameter and then passes through a final collimation aperture of 150 μm which is approximately 2.6 cm from the first laser interaction region (beam waist approximately 500 μm in diameter) formed by a standing wave. ^{85}Rb atoms are excited into a superposition of the $5S_{1/2}$ ground state and the $nS_{1/2}$ excited state with $n \geq 22$. Afterwards the beam passes through the center of the region containing the plate structure (grey rectangles in the picture) and then enters the second interaction region. Both excitations are in the low-saturation regime. After the second standing wave laser field the atoms are detected by field ionization, with the ejected electron being multiplied by means of a channeltron. A spectrum is recorded by counting the channeltron pulses via a computer which also controls the laser frequency. The field ionization region is within a Faraday cage. This ensures that no electric fields from there can leak into the region with laser interaction and plate structure. Additionally, the whole area is shielded from external electric and magnetic fields by a copper tube surrounded by two layers of mu metal. The copper tube is in contact with a cryostat that can be cooled

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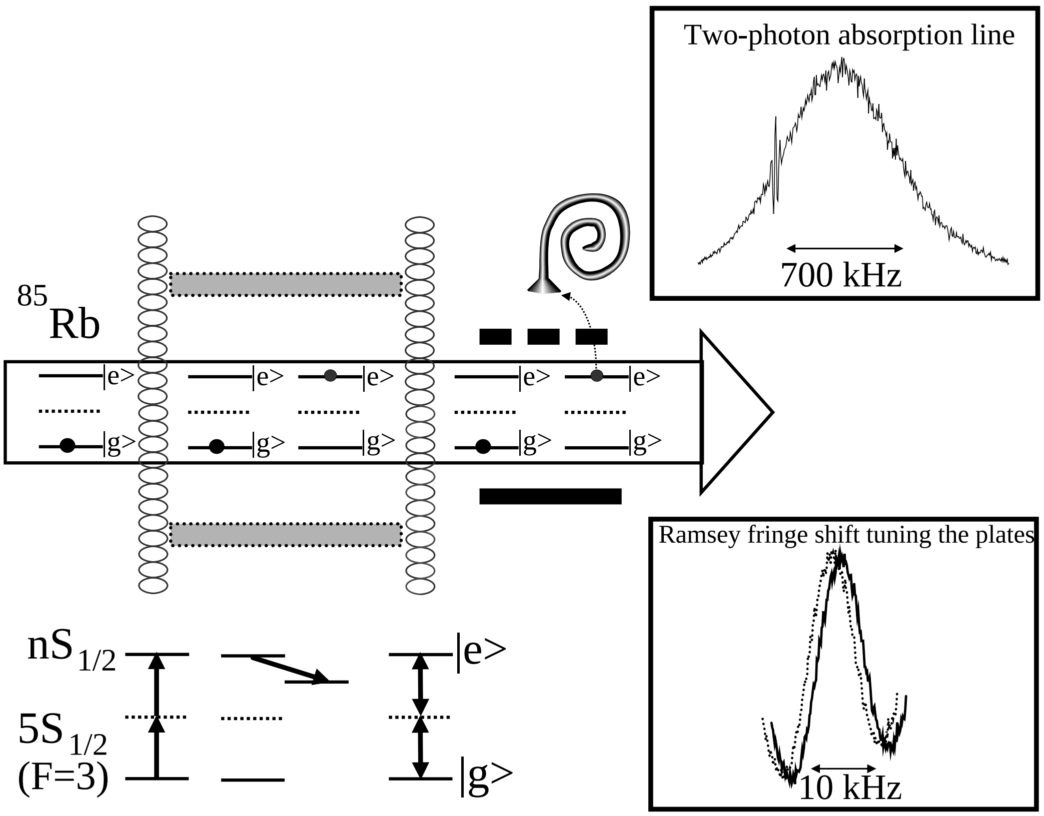

FIG. 1. Experimental setup. The insets show the Doppler-free two-photon absorption line with the Ramsey fringe (top) and the displacement of the Ramsey fringe on changing the plate distance (bottom). Note that the Ramsey fringe is not in the center of the two-photon absorption line because of its different ac Stark shift (measured to be 8 Hz for a reference intensity of 1 W/cm^2) compared with that of the Ramsey peak (220 mHz for the same reference intensity). This difference results from the fact that in the Ramsey two-field method the atom passes through the laser-free region most of the time.

to either 77 or 4 K. Inchworms (piezo-driven devices) can move the plates perpendicular to the direction of the atomic beam in a precise and reproducible manner which allows for exact symmetrical positioning of the plates relative to the beam. The motors are connected to the plates via rods made of zirconium oxide having a low thermal conductivity of 2.5 W/K m. The interplate distances, ranging from 0.5 to 3 mm, can be monitored by means of an interferometric measuring device with a maximum uncertainty of ± 1 μm. The inchworm motion is dependent on both its load and its temperature; therefore special care was taken to control the parameters of the piezo motors.

Besides the careful shielding of external fields, it is also necessary to avoid any potential differences between the plates. To eliminate any shift from contact potentials, the two plates and their connection via thin and deformable lamellas were machined from a single piece of copper. To avoid adsorbates, the plates could be baked during the preparation stage of the experiment.

A folded optical resonator with active stabilization provided a stable phase between the two Ramsey zones. This was achieved by a piezo-mounted mirror and the phase modulation method (similar to the method used to stabilize the dye laser). Additionally the resonator enhanced the laser power in the two interaction zones.

Scanning the laser frequency and detecting the excited-state atoms reveals a two-photon peak with Ramsey fringes, as displayed in the upper inset of Fig. 1. The width of the central peak of the Ramsey structure is limited by the time of flight between the two zones. The mean time of flight in our case was 28 μs, which is slightly greater than the lifetime of the excited Rydberg states. Only the zero-order fringe is seen because a thermal atomic beam source was used, and higher-order fringes were therefore washed out owing to the longitudinal velocity distribution. The position of this Ramsey fringe was monitored as a function of the relative distance between the plates.

The experimental protocol was as follows. The plates were initially set at a position which served as a reference distance d_{ref} and the laser frequency was scanned across the central Ramsey fringe. Then the plates were moved to a new distance d and the laser was scanned again. This process was reiterated for other distances d relative to the same reference distance d_{ref}. The central fringe position was determined by a least-squares fitting routine which detected the peak center with an accuracy between approximately 20 and 100 Hz, depending on the Ramsey fringe contrast, which was limited by the background pressure at the different temperatures.

Careful control of the drift of the reference frequency (which is provided by the supercavity in the locking scheme of the dye laser) in the course of the measurement had to be exercised. Because the measurement of the level shift is given relatively to a reference frequency, its drift has to be considered. Although there were two temperature isolation stages the residual thermal length change of the supercavity resulted in a drift ranging between zero and 100 Hz/s. A measure of this drift as a function of time could be derived from the data obtained from the Ramsey spectra at plate distance d_{ref}.

Before proceeding, it is useful to point out that retardation effects in the interaction of the atomic dipole with the cavity walls are dominant only when the distance between the plates is larger than the wavelength of the relevant Rydberg transition, i.e., $d > n^3 a_0/\alpha$, where n is the principal quantum number, a_0 the Bohr radius, and α the fine-structure constant. In this regime the round-trip time for a photon leaving the atom, reaching the cavity walls, and returning to the atom is larger than the oscillation period of the relevant transition, and the shift is caused by the interaction of the atom with the cavity field. In the opposite case, $d < n^3 a_0/\alpha$, the atom would interact with its mirror image in the cavity walls, giving rise to a van der Waals interaction. Therefore the condition $d > n^3 a_0/\alpha$ has to be fulfilled.

It is of great interest to investigate the temperature influence on the energy shift in order to discriminate both the thermal and vacuum contributions to the shift itself. This aim was pursued by scanning the plates at three different temperatures $T = 292$, 77, and 4 K over interplate distances close to $d_{\text{res}} = 3\lambda_0/2 = 1.173$ mm, where λ_0 is the wavelength associated with the transition $24S_{1/2} \rightarrow 23P_{3/2}$. The average number of thermal photons nth per mode is slightly above 15 at room temperature, and thus a large thermal content is expected, whereas at $T = 4$ K, nth is only 0.01. Figure 2 shows the experimental result. It is evident that a resonant effect decreasing with temperature is present and is in agreement with the expected on-resonance interplate distance. Some small mismatch for d_{res} is mainly due to the uncertainty in the reference distance between the plates. It is noteworthy that the scatter of the off-resonance points fits quite well with the general uncertainty, calculated to be around 150 Hz at room temperature and around 40 Hz at low temperature. The main contributions stem from the uncertainty in evaluating the drift motion of the supercavity and in determining the central frequency of the Ramsey fringe. By comparing Figs. 2a and 2c it can be deduced that the thermal contribution to the shift amounts to about 150 Hz. This value is a factor of almost 4 smaller than the expected value calculated from [11]. The discrepancy can be explained by the fact that the plates are not a perfect black-body radiator. In addition, their conductivity and their geometrical extension is finite. Figure 2c shows a shift of about 110 Hz. Note that the measured value $\Delta\nu$ corresponds to a relative accuracy of $\Delta\nu/\nu \approx 2 \times 10^{-13}$.

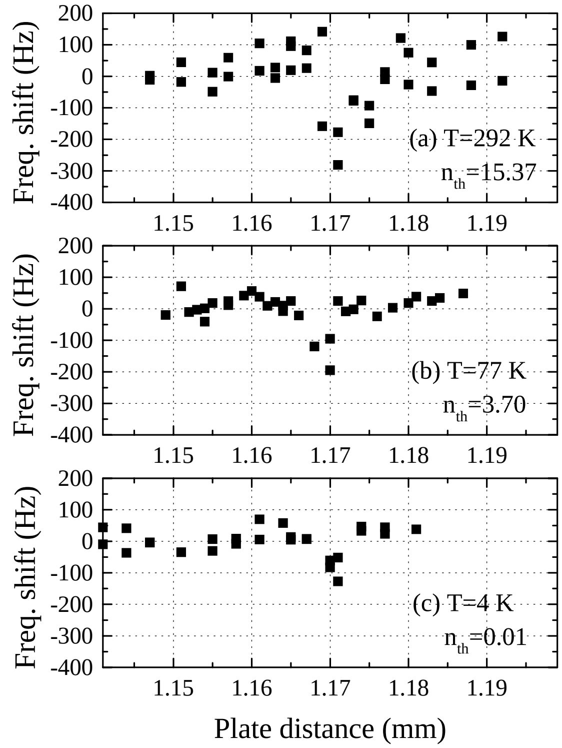

FIG. 2. Experimental results on the level shift for three different temperatures. The transition chosen was $24S_{1/2} \rightarrow 23P_{3/2}$. The number of thermal photons per mode is denoted by nth.

A closer investigation of the level shift at low temperature has been additionally carried out. Two different experiments using the transitions $22S_{1/2} \rightarrow 21P_{3/2}$ and $26S_{1/2} \rightarrow 25P_{3/2}$ were prepared at 4 K. The frequency shift as a function of the plate distance for both cases is shown in Fig. 3. The resonance corresponding to 3/2 of the wavelength of the transition $26S_{1/2} \rightarrow 25P_{3/2}$ has a peak value of about 100 Hz. In Fig. 3b the measured resonance for the level $22S_{1/2}$ shows a shift of 120 Hz.

The theoretical treatment of quantum electrodynamic level shifts between parallel conducting plates in resonance with atomic transitions is very involved. According to [4,12] the relevant dependences of the shift Δ on the distance and principal quantum number n are given (in atomic units) by the approximate formula

$$\Delta \approx \frac{4\alpha^2}{3d} \sum_{n'<n} E_{n'n}^2 R_{n'n}^2 \ln\left\{2\left[\cos^2\left(\frac{E_{n'n}d}{2\hbar c}\right) + \Gamma^2\right]^{1/2}\right\}, \tag{1}$$

where $E_{n'n}$ is the energy difference between the states n and n', $R_{n'n}$ is the radial matrix element between the same states. $\Gamma = \sinh(\beta/2)$ expresses the damping due to the attenuation of the field upon one reflection so

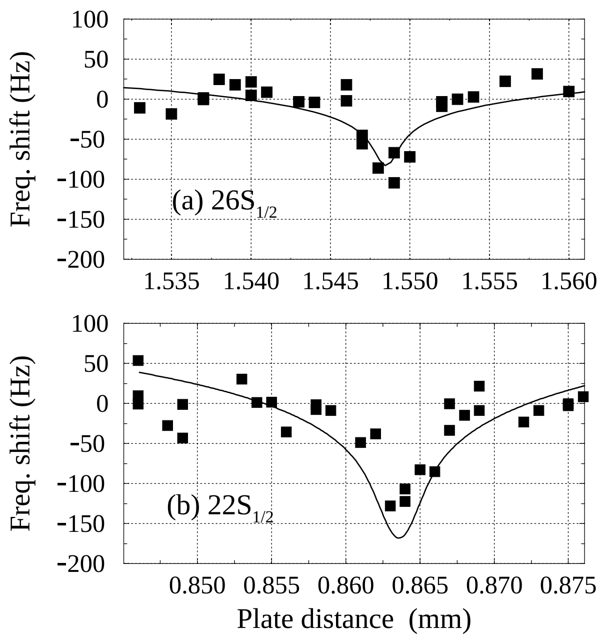

FIG. 3. Comparison of the level shift with theory for two different transitions resonantly coupled into the cavity at $T = 4$ K: (a) $26S_{1/2} \rightarrow 25P_{3/2}$ and (b) $22S_{1/2} \rightarrow 21P_{3/2}$.

that $\beta = -\ln \rho/2$ with ρ being the reflectivity of the mirrors. In Eq. (1) the main contribution to the shift comes from the transitions to the first neighboring levels and, because $E^2_{n'n} \approx 1/n^6$ and $R^2_{n'n} \approx n^4$, the changes of Δ with n and d are then given (in Hz) approximately by

$$\Delta \approx \frac{2.47 \times 10^4}{n^2 d} \ln\left\{2\left[\cos^2\left(\frac{E_{n'n} d}{2\hbar c}\right) + \Gamma^2\right]^{1/2}\right\}, \tag{2}$$

where d is measured in mm. On resonance the argument of the cosine function approaches $\pi/2$ and Eq. (2) contains a logarithmic divergence, which is smoothed to a finite value by the imperfect reflectivity of the copper mirrors [12]. The curves in Fig. 3 are drawn by averaging Eq. (2) over a Gaussian distribution of the interplate distances. The standard deviations have been determined to be roughly 0.5 and 1 μm for the $26S$ and the $22S$ states, respectively, thereby reflecting the different values of the uncertainty in the distance determination owing to the different driving distances of the piezo motors in the two measurements. The frequency-dependent reflectivities were calculated from the copper conductivity at 4 K to be better than $R = 0.9999$. With these parameters the agreement between theory and experiment is satisfactory. The widths of the experimental resonances are also well reproduced. There is a further correction due to the finite size of the atomic beam diameter Φ relative to the transition wavelength λ_0 ($\Phi/\lambda_0 = 0.26$ for the $22S$ state and 0.15 for the $26S$ state). This correction should affect more the calculation for the $22S$ state as a result of a worse pointlike beam approximation assumed in Eq. (1). Although the described model is based on a QED approach, part of the effect might also be understood following classical arguments [12].

In conclusion, we observed the change of the radiative shift by tuning the plate distance over values corresponding to three-halves of the wavelength for Rydberg transitions of the type $nS_{1/2} \rightarrow (n-1)P_{3/2}$. A level shift of a few hundred Hz was measured at room temperature for the $24S_{1/2}$. Under the same conditions at low temperature the shift reduces to 110 Hz. The shift at vanishing thermal photon number was also measured for the states $22S_{1/2}$ and $26S_{1/2}$. It shows good agreement with the presented theory.

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