RF-dressed Rydberg atoms in hollow-core fibres

C Veit1, G Epple1,2, H Kübler1, T G Euser2,3, P St J Russell2 and R Löw1,4

15. Physikalisches Institut and Center for Integrated Quantum Science and Technology, Universität Stuttgart, Pfaffenwaldring 57, D-70569 Stuttgart, Germany
2Max Planck Institute for the Science of Light, Günther-Scharowsky-Str. 1/Bldg. 24, D-91058 Erlangen, Germany
3Cavendish Laboratory, JJ Thompson Ave, University of Cambridge, Cambridge, CB3 0HE, UK

E-mail: r.loew@physik.uni-stuttgart.de

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Abstract
The giant electro-optical response of Rydberg atoms manifests itself in the emergence of sidebands in the Rydberg excitation spectrum if the atom is exposed to a radio-frequency (RF) electric field. Here we report on the study of RF-dressed Rydberg atoms inside hollow-core photonic crystal fibres, a system that enables the use of low modulation voltages and offers the prospect of miniaturised vapour-based electro-optical devices. Narrow spectroscopic features caused by the RF field are observed for modulation frequencies up to 500 MHz.

Keywords: Rydberg, HC-PCF, RF-dressed Rydberg atoms

(Some figures may appear in colour only in the online journal)

Introduction

The large polarizability of Rydberg states [1] enhances the electro-optical response of a thermal Rydberg gas. The combination of thermal vappors and the spectroscopically narrow features of electromagnetically induced transparency involving Rydberg states [2] allows electro-optical effects to occur at very small electric field amplitudes. The interplay between Rydberg atoms and radio-frequency (RF) or microwave electric fields is especially interesting, since its nature strongly depends on many different parameters such as the modulation frequency, the Kepler frequency, the excitation Rabi frequencies and potentially the mutual interaction strength between the Rydberg atoms themselves. All these parameters can be made comparable in their magnitude in the experiment and tuned over a wide range, permitting exploration of different manifestations of the field-atom interaction. Ac-dressed Rydberg atoms have been employed to study microwave excitation and ionization [3, 4], to demonstrate phase modulation of light [5], to measure atomic polarizabilities [6, 7] and to observe St¨uckelberg oscillations in cold atomic ensembles [8]. Regarding potential future applications, it is especially interesting that RF-dressed Rydberg atoms have been shown to exhibit enhanced dc electric field sensitivity [9] and that Rydberg atoms also allow accurate sensing of microwave electric fields [10–12], strong RF fields [13, 14] and RF noise [15, 16].

Adopting the approach presented in [9], we study RF-dressed Rydberg states and reduce the size of the initial macroscopic vapour cell experiment to the lengthscales of a hollow-core photonic crystal fibre (HC-PCF) [17, 18]. In this way we can not only minimise the modulation voltage and the power consumption but also increase the bandwidth. The performance of the system is analysed by the position and magnitude of sidebands in the Rydberg excitation spectrum that appear when the RF field is applied. Throughout this article we discuss a regime in which the applied modulation frequencies and powers justify a perturbative Floquet analysis of the spectroscopic features [6, 8, 9].

Theoretical description

The extreme sensitivity of highly excited atoms to external fields allows modification of the absorptive and dispersive behaviour of a thermal gas by weak applied electric fields. In
the following we consider the effect of a RF field (with dc offset)

\[ E(t) = E_{dc} + E_{ac} \cos \omega t, \]  

(1)
on a long-lived Rydberg state \( (r_{\text{ryd}} \ll \omega) \). We restrict the analysis to a regime where the modulation frequency \( \omega \) is larger than the Rabi frequencies of the excitation lasers but also much smaller than the transition frequencies to neighbouring Rydberg states. This implies that the atom-light interaction cannot follow the energy modulation of the Rydberg state, enabling us to treat the external field as quasi-static. It is therefore justified to apply time-independent perturbation theory at sufficiently low field amplitudes. In this regime, the Rydberg state is split into equidistant quasi-stationary Floquet states separated by multiples of the modulation frequency \([9]\). The considered \( P \)-state of caesium experiences the quadratic Stark effect causing the energy of the Rydberg state \( \varepsilon(t) \) to shift according to:

\[ \varepsilon(t) = \varepsilon_0 - \frac{1}{2} \alpha E(t)^2. \]  

(2)
Here \( \varepsilon_0 \) is the energy and \( \alpha \) the polarizability of the unperturbed state. Inserting the electric field from equation (1) we obtain the expression

\[ \varepsilon(t) = \varepsilon_s - \alpha E_{dc}E_{ac} \cos \omega t + \frac{E_{ac}^2}{4} \cos 2\omega t, \]  

(3)
consisting of a static contribution

\[ \varepsilon_s = \varepsilon_0 - \alpha (2E_{dc}^2 + E_{ac}^2)/4, \]  

(4)
and terms oscillating at \( \omega \) and \( 2\omega \). If the admixture of neighbouring Rydberg states is small, the electron wave function \( \Psi(r, t) \) can be separated into a time-independent part \( \Psi(r) \) and a temporally varying part determined by equation (3). The time-dependent Schrödinger equation can then be integrated to obtain

\[ \Psi(r, t) = \sum_{n=-\infty}^{\infty} A_n(x, y) \exp\left(\frac{i}{\hbar} \varepsilon_n t + in\omega t\right)\Psi(r), \]  

(5)
where the coefficients

\[ A_n(x, y) = \sum_{m=-\infty}^{\infty} J_{n-2m}(x)J_m(y) \]  

(6)
are determined by Bessel functions of the first kind and

\[ x = \frac{\alpha E_{dc}E_{ac}}{\hbar \omega} \quad \text{and} \quad y = \frac{\alpha E_{ac}^2}{8 \hbar \omega}. \]  

(7)
Equation (5) predicts not only a constant shift of the originally unperturbed Rydberg state but also the formation of sidebands with a fixed phase relationship and energies

\[ \varepsilon_n = \varepsilon_0 - n\hbar \omega = \varepsilon_0 - \alpha (2E_{dc}^2 + E_{ac}^2)/4 - n\hbar \omega. \]  

(8)
If optically probed, these sidebands, illustrated in figure 1(a), appear in the absorption spectrum and can be interpreted as being caused by multi-photon excitation involving optical excitation photons and \( n \) RF photons. Correspondingly, stimulated emission involving various numbers of RF photons can lead to modulation of the excitation light, which as a result acquires sidebands. It is worth noting that even for a pure ac field \( (E_{dc} = 0) \), the root-mean-square amplitude of the \( n \) sidebands, for example, the relative sideband amplitude is sideband at a given driving frequency. For the second-order sidebands, the sum in equation (6) reduces to a single term, since \( E_{dc} = 0 \). This is consistent with equation (3), which predicts modulation of the energy at \( \omega \) only in the presence of a dc field.

For \( E_{dc} = 0 \), it is particularly easy to determine the ac field amplitude that maximises the amplitude of a specific sideband at a given driving frequency. For the second-order sidebands, for example, the relative sideband amplitude is given by \[ |A_2(0, y)|^2 = \frac{1}{2} |J_2(y)|^2 \] and reaches a maximum of \(~33.9\%\) at \( y \approx 1.84 \). For the \( 3P_{3/2} \) \( (m_j = 1/2 \) state (considered later) and a modulation frequency of \( 500 \) MHz, this corresponds to an ac field amplitude \( E_{ac} \approx 16.5 \) V cm\(^{-1}\). Since \( y \) is at the same time proportional to the inverse of the modulation frequency \( \omega \) and \( E_{ac}^2 \) (equation (7)), the RF voltage required to achieve efficient modulation scales with the square root of the modulation frequency.

**Experimental setup and excitation scheme**

We study RF-dressed Rydberg atoms inside a vapour-filled kagomé-type HC-PCF (core diameter 60 \( \mu \)m see figure 2(c)). By confining both excitation light and atoms, these band-guiding fibres offer an elegant way to interface atomic vapours with light. The fibre is mounted inside a CF63 vacuum cube connected to a flexible metal bellow acting as a caesium reservoir. After pumping and baking out the vacuum system, the valve connecting the chamber to the turbo pump.
was closed and a caesium ampoule inside the reservoir bellow was broken. The caesium then diffused into the main chamber and on to the open ends of the fibre. By changing the reservoir temperature, we are able to control the vapour density inside the main chamber. As explained later in more detail, due to slow diffusion of the atoms the vapour densities inside and outside the fibre are not at equilibrium.

As illustrated in figure 2(a), the fibre is equipped with five field plates, allowing individual sections along the fibre to be addressed with electric fields. The electrodes were evaporated on to 200 μm thick glass substrates as 2 mm × 22 mm silver stripes with a thickness of ∼1 μm (see figure 2(b)). A 5 nm thick layer of chromium, deposited between the glass and the silver layer, ensured a strong bond between the glass and the electrodes. For reasons of compatibility with the reactive alkali gas, the field plates were electrically connected using in-house-fabricated coaxial transmission lines consisting of a central conductor surrounded by alumina beads and a nickel-plated copper braiding. Despite the impedance mismatch between the conductor surrounded by alumina beads and a nickel-plated electrodes, for reasons of compatibility with the reactive alkali silver layer, ensured a strong bond between the glass and the thick layer of chromium, deposited between the glass and the electrodes.

The optical density of the surrounding chamber was kept below the chamber temperature. The reservoir temperature was always kept below the chamber temperature. The optical density (of the resonant probe transition) measured inside the fibre was OD₁ = 4.4 and smaller than the optical density of the surrounding chamber OD₉ = 19. Due to the mismatch in optical density, which as mentioned originates from the slow diffusion of the atoms into the fibre, we do not expect the atomic number density to be uniform along the fibre at the time of the measurements. In fact, a Stark-shifted signal was only observed when either one of the two

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**Figure 2.** (a) A hollow-core PCF with a core diameter of 60 μm (not to scale) is mounted inside a vacuum chamber exposed to caesium vapour. Five field plates allow individual sections along the fibre to be addressed with ac electric fields. In the experiments, only one field plate is modulated at a time. (b) The field plates consist of 2 mm wide silver stripes evaporated on to thin glass substrates and then positioned above the fibre. (c) Microstructure of the kagomé-PCF with a core diameter of 60 μm.

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**Results**

In the following we present spectroscopic results obtained in the hollow-core fibre while a single field plate (marked in figure 3(c)) was modulated at frequencies up to 500 MHz. The fibre had been exposed for 5 weeks to a Cs atmosphere at different chamber temperatures of up to 90 °C. The reservoir temperature was always kept below the chamber temperature. The optical density (of the resonant probe transition) measured inside the fibre was OD₁ = 4.4 and smaller than the optical density of the surrounding chamber OD₉ = 19. Due to the mismatch in optical density, which as mentioned originates from the slow diffusion of the atoms into the fibre, we do not expect the atomic number density to be uniform along the fibre at the time of the measurements. In fact, a Stark-shifted signal was only observed when either one of the two
outermost plates was modulated. Future studies may reveal whether the lack of signal from the inner sections of the fibre is due to the absence of Cs atoms or to high electric fields prohibiting Rydberg excitation. In all the measurements, the chamber and reservoir temperatures were \( T_{ch} \approx 84^\circ C \) and \( T_{res} \approx 47^\circ C \) respectively. The modulation potential was purely sinusoidal (i.e. no dc offset was applied) and the externally supplied RF voltage was \( U_{RF} = 250 \text{ mV} \) (\( \sim 1 \text{ dBm} \)). At the input face of the fibre, the beam powers were \( p_{895} = 1 \mu W \), \( p_{1359} = 8.2 \mu W \) and \( p_{780} = 25.5 \text{ mW} \). The excited Rydberg state \( 30P_{3/2} \) \( (m = \pm 1/2) \) possesses a polarizability \( \alpha_{30P_{3/2}} = 26.93 \text{ MHz/(V cm)}^{-2} \) [22].

Figure 4 shows the probe transmission (averaged over 500 traces) for four different modulation frequencies between 250 and 500 MHz as a function of \( \Delta_c \). In all the traces, the largest signal (marked by shaded area) originates from the unmodulated part of the fibre, through which the probe beam passed before it encountered the modulated section at the far end of the fibre (see figure 3(c)). Due to the high probe power employed, the signal is broadened and the depth of the central absorption dip is smaller than in the weak-probe limit. In the modulated section of the fibre, the atomic resonance (corresponding to \( n = 0 \) in equation (8)) is red-shifted. The shifted Rydberg state, which we refer to as the carrier, gives rise to a distinct and clear signal in the spectrum at negative detunings \( \Delta_c \). Setting \( E_{dc} = 0 \) we used equation (4) to estimate the field amplitude \( E_a \) from the red-shift of the carrier state. Due to the frequency response of the coaxial transmission line we obtained slightly different field amplitudes for the different modulation frequencies lying between \(~8.6 \text{ V cm}^{-1} \) at 500 MHz and \(~10.1 \text{ V cm}^{-1} \) at 250 MHz. From the geometry of the setup we conclude from these results that, at a modulation frequency of 500 MHz, approximately 70% of the input RF voltage is transferred to the field plates. This is consistent with the impedance mismatch between the air-side coaxial cable and the self-built in-vacuum transmission line.

As expected, the carrier state is accompanied by RF-induced sidebands separated by multiples of the RF frequency. At high modulation frequencies, only second order sidebands can be identified and the carrier state possesses the largest amplitude. At lower modulation frequencies, higher-order sidebands also appear and the carrier amplitude falls. For the second order sidebands, relative amplitudes of \(~5\%\) and \(~29\%\) are reached at modulation frequencies of 500 MHz and 250 MHz respectively. Strikingly, the sideband spectrum becomes asymmetric in the case of low modulation frequencies and also odd-order sidebands appear. The existence of odd-order sidebands is attributed to local electric fields inside the fibre, which are estimated to be one order of magnitude smaller than the applied ac field.
presence of local electric fields inside a kagomé-PCF with a core diameter of 60 µm. These shifts seemed to vanish after long-term exposure of the fibre to a Cs atmosphere [18].

To compare the experimental results with theoretical predictions and to estimate the local dc field inside the fibre, we fitted $|A_{ac}(x, y)|^2$ (see equation (6)) to the experimentally obtained sideband amplitudes of the data-set shown in figure 4. We then extracted the ac and dc field from the fit parameters $x$ and $y$ (see equation (7)). This procedure assumes that the ac and dc fields are parallel, while the dc field inside the fibre is likely to be randomly oriented. As a consequence, the dc field component parallel to the ac field varies over the length of the fibre and a dc field component perpendicular to the ac field exists. The latter has no influence on the amplitude distribution of the sidebands, but causes a slight broadening of the lines due to the non-vanishing tensor polarization of the $30P_{3/2}$ state [22]. For the parameter range considered here, the dc field is much smaller than the ac field. A numerical analysis taking into account a random distribution of the dc field reveals that in this regime the random nature of the dc field has negligible influence on the fitted field amplitude $E_{dc}$, and the fitted amplitude $E_{dc}$ can be interpreted as the mean or effective component of the dc field parallel to $E_{dc}$. For this reason, $E_{dc}$ only determines the order of magnitude of the dc field. Figure 5 shows both the experimental data and the fits, which are in excellent agreement with each other. Also shown are the derived field amplitudes $E_{ac}$ and $E_{dc}$. The fitted amplitudes $E_{ac}$ correspond within 4% to the values estimated from the red-shift of the carrier state. For $E_{dc}$, we obtained field strengths smaller than 0.5 V cm$^{-1}$, which is again consistent with previous results [18]. In the parameter regime investigated, the sideband spectrum becomes relatively insensitive to small dc fields at large modulation frequencies. As a result, the estimated field amplitudes $E_{dc}$ have relatively large uncertainties in the high frequency case (see figure 5). Nevertheless, it is striking that $E_{dc}$ seems to increase with decreasing modulation frequency. While this behaviour is not yet understood, further investigations are expected to yield valuable insight into the charge dynamics inside the fibre.

Outlook

In future experiments we plan to exploit the spatial resolution of patterned electrodes to obtain a more detailed understanding of diffusion, adsorption and desorption effects inside the fibre core. Also promising is the integration of the modulation electrodes in the form of conductive microwires inside the fibre [23]. This will allow even smaller modulation voltages and will lead to larger bandwidths. A more detailed insight into the direction of the electric fields and the distribution of ions potentially sitting on the walls of the hollow fibre core can be obtained by exploiting the tensor polarizability of $P_{3/2}$ states.

So far we have observed and analysed the applied radiofrequency fields only in terms of additional field-dependent spectroscopic features. If an efficient, miniaturised electro-optical device is to be realised, the back-action of the atomic system on the excitation light must be investigated. This can be carried out by measuring the amplitude of the optical sidebands (e.g., using a heterodyne setup). The kagomé-PCF system also allows the study of the atomic response in a regime where a perturbative approach is insufficient. This could be achieved either by applying microwaves resonant to Rydberg–Rydberg transitions or by increasing the power to a level where the Rabi frequencies become comparable to the transition frequencies between Rydberg states. In both cases the optical response of the system could then be studied in situations where the Rydberg electron cannot adiabatically follow the applied fields.

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