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Multiphoton intrashell resonances in Rydberg atoms: Bloch-Siegert shifts and widths

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We measure for the first time, to our knowledge, N-photon intrashell resonances, widths, and Bloch-Siegert shifts up to $N=23$. Rydberg atoms of Li ($n=25$) in a rotating electric field and a static, in-plane electric or magnetic field are used. The measurements, in conjunction with theoretical estimates, precisely determine the characteristics of the resonances (shifts and widths) and their dependence on the strength of the rotating field.

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Nonperturbative and high-order nonlinear processes constantly challenge our understanding of the fundamental photon-atom interaction [1]. Bound-bound transitions where many photons are needed to bridge the energy gap between the states is one example. In the theoretical description, the photon-atom Hamiltonian can be split into right- and left-hand circularly polarized components. One of these shows resonance behavior while the other gives rise to ac Stark shifts of upper and lower energy levels and a shift of the resonance position known as the Bloch-Siegert (BS) shift [2].

The BS shift is of essential importance in a broad variety of different branches of physics. For a long time it has been a subject of concern in nuclear magnetic resonance [3,4] which besides numerous biological and medical applications is the only realization of a (small) quantum processor [5]. Furthermore, BS shifts may play a crucial role in electron paramagnetic resonance spectroscopy [6], light emission by laser-driven atoms [7–9], including Raman lines [10], and polychromatic fields [11]. BS shifts were observed in electromagnetically induced transparency [12], included into theory of lasers [13], micromasers [14], as well as quantum optics [15–17]. More recently the importance of BS shifts was pointed out in the theory of multiphoton far-infrared interband transitions in condensed matter [18] and in theory of continuously driven dissipative solid-state qubits [19] within quantum information.

Measurements of BS shifts for the absorption of one or three photons were performed some time ago [20], but higher photon numbers N were not seen. Similar landmarks were achieved in a completely different physical realization within classical electrodynamics when interaction of two modes with orthogonal polarizations was considered in an optical ring resonator [21]. For *intershell* transitions in Rydberg atoms (gigahertz domain of driving frequency Ω) resonances with N up to 28 were observed but the shifts were not detected [22,23]. The effect is proportional to Ω^{-2}, hence it may become large for *intrashell* transitions in the megahertz domain. Multiphoton intrashell resonances were previously observed only indirectly in microwave ionization spectra which showed resonant multiphoton enhancement when the dc Stark splitting of the shell $\frac{3}{2}nF_{\mathrm{dc}}$ is close to an integer number of field quanta $N\Omega$ ($N\leqslant 5$) [24], but BS shifts were not exposed. While microwave ionization is a complicated process which is difficult to understand in full detail [25], the intrashell resonance dynamics responsible for the observed resonances may be treated analytically with very few limitations [26].

In this work, we demonstrate that intrashell transitions in carefully prepared Rydberg atoms offer a unique possibility for studying multiphoton processes of high order in a controllable way. The unambiguous identification of high-N shifted and broadened resonances poses a special challenge which is met by carefully tracing the evolution of resonances with increasing field strength and with the help of nonperturbative calculations. A special field configuration with ac components both along and perpendicular to the dc field was employed to enhance the resonance effect.

For the pure Coulomb problem, the stationary states are described by two independent pseudospins of constant magnitude $\frac{1}{2}(n-1)$ quantized in two independent directions. Each of the two pseudospins can be decomposed into $n-1$ independent pseudospin-$\frac{1}{2}$ components. The intrashell dynamics of hydrogenic Rydberg states in time-dependent external electromagnetic fields may thus be reduced to the dynamics of two of these independent spin-$\frac{1}{2}$ particles in two independent magnetic fields [27]. This description is very attractive due to its simplicity and accuracy. It was used recently in analysis of state control by external fields [28], experimental data on microwave ionization resonances [26], adiabatic or near-adiabatic state transformations [29–32], and intrashell single-photon resonances for circularly polarized fields [33]. It is used here to derive analytical expressions for positions and widths of intrashell multiphoton resonances. The general problem of laser excitation of atoms or molecules can in principle be treated by perturbation theory, but it becomes intractable even for relatively small photon orders. By contrast, our analytical results do not have this restriction, and the present comparison with experimental data provides fur-

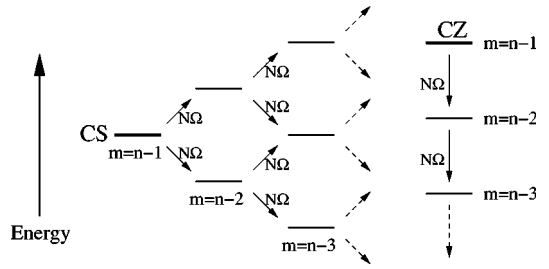

FIG. 1. Hydrogenic energy levels in electric (left) and magnetic (right) fields, respectively, where m is the magnetic-field quantum number. Spectral positions of initial circular Stark (CS) and Zeeman (CZ) states are marked. Multiphoton quanta $N\Omega$ connect neighboring states.

ther strong evidence for the validity of the pseudospin formalism.

The experimental arrangement was described in Ref. [29]. In short, a Stark state of maximum polarization [a parabolic state with quantum numbers $|n_1,n_2,m\}=|24,0,0\}$ or a coherent elliptic state (CES) of eccentricity $e=1$ [32]] was formed by laser excitation in a strong electric field (145 V/cm), then transformed by the adiabatic crossed-field method (ACFM) into a nearly circular CES ($e\simeq 0$) stabilized by a static field [32], and finally exposed to a homogeneous, rotating electric field of frequency Ω and amplitude F_Ω for a time T. The rotation vector of the field, $\vec{\Omega}$, was perpendicular to the static field which in turn was perpendicular to the orbital plane of the circular state. The static electric and magnetic fields ($\vec{F}$ and $\vec{B}$, respectively) produced a constant Stark-Zeeman splitting of the shell described by frequencies $\omega_\pm=|\vec{\omega}_L\pm\vec{\omega}_S|$, with $\vec{\omega}_S=\frac{3}{2}n\vec{F}$ and $\vec{\omega}_L=\frac{1}{2}\vec{B}$ [27]. The Stark frequency of the rotating field is $\omega_\Omega=\frac{3}{2}nF_\Omega$. The parameters $\Omega/2\pi$, $\omega_\Omega/2\pi$, and $\omega_\pm/2\pi$ were varied within the intervals 12–30 MHz, 0–200 MHz, and 30–370 MHz, respectively. These frequencies are much smaller than the orbital frequency of the $n=25$ shell (421 GHz). The transformation was completed in <10 μs and the exposure to the rotating field lasted for $T=2$ μs with switching times of a few field cycles [29]. A ramped electric field of slew rate 420 (V/cm)/μs triggered about 50 μs after laser excitation was used to analyze the final distribution of Rydberg states by selective field ionization (SFI) [22], and to determine the probability that the Rydberg atom remained in the original state, i.e., responded adiabatically to the rotating field.

In one version of the ACFM, $F_i=145$ V/cm was switched off in the presence of a weak magnetic field to produce a CES of $e=0$ oriented relative to $\vec{B}$ with spherical quantum numbers $|n,\ell,m\rangle_B=|25,24,24\rangle_B$. We call this a circular Zeeman (CZ) state. It is nondegenerate and at the top of the n manifold (Fig. 1). In another version [29,30] the electric field rotated while it approached a constant final value $\vec{F}_f$ perpendicular to $\vec{B}$. This was arranged such that $3n\vec{F}(t)\simeq\vec{B}$ at an intermediate time $t=t_c$ when sharp avoided crossings of quasienergy curves were encountered and purely diabatic transitions brought the CES into a new type of CES. As $\vec{F}$ subsequently rotated and increased towards $\vec{F}_f$ this new CES

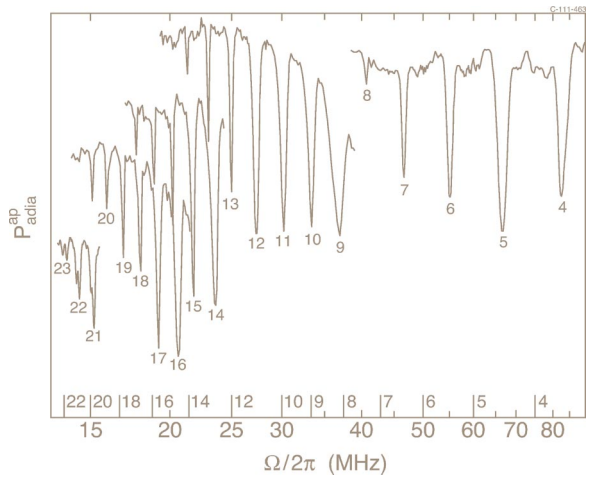

FIG. 2. Multiphoton resonances vs frequency $\Omega/2\pi$ for (almost) circular Stark states. $F=6$ V/cm (300 MHz), $B=15$ G (21.4 MHz), $F\perp B$, $\omega_+=\omega_-\simeq 300$ MHz, and $e=0.07$. Curves for different ω_Ω are displaced vertically for clarity. Photon numbers N are given next to each curve and unshifted resonance positions are indicated. The resonances $N=21-23$ are split (see text).

developed into a circular Stark (CS) state quantized along the direction of $\vec{F}_f$, i.e., $|n,\ell,m\rangle_F=|25,24,24\rangle_F$. The eigenenergy of the CS state is highly degenerate and located in the middle of the n manifold (Fig. 1).

The nearest-neighboring states of the initial CZ or CS state could not be distinguished by the SFI so only apparent adiabatic probabilities P^{ap}_{adia} were obtained. In the pseudospin description, transition to an adjacent state corresponds to a flip of one of the constituent spins. Therefore, not only one but at least two spin flips were required for having a detectable change of the SFI spectra.

We first measured the dependence of P^{ap}_{adia} on Ω for various splittings $\omega(\omega_+=\omega_-)$ and strengths ω_Ω to have an overview of the resonances, and then in detail to determine BS shifts and resonance widths of selected resonances. The overview is shown in Fig. 2 for initial CS states. Similar results were seen for initial CZ states, so the resonance structures do not depend on location within the manifold or degree of degeneracy. Some resonances were split ($N=21,22$, and 23). This happened when $\vec{B}$ and $\vec{F}$ were not exactly perpendicular. One component corresponds to ω_+ and the other to ω_-. The splitting could always be eliminated by adjusting the direction of $\vec{F}$ or $\vec{B}$. It was left in one case to expose the phenomenon. The resonance frequency Ω_{res} is clearly shifted relative to $\Omega=\omega/N$ and it depends on ω_Ω. The correct assignment of the high photon numbers was made difficult by this shift and required careful measurements of $\Omega_{res}(\omega_\Omega)$ for many N.

For the theoretical description, an explicitly time-dependent multicycle Landau-Zener model (MLZ) [34] as well as an adiabatic time-independent Floquet theory (AD) [35,36] have been developed. The spin-flip probability $p(\Omega)$ is generally oscillatory with a Lorentzian envelope function approaching one at resonance unless $q=\omega_\Omega/\omega$ is very small. For the BS shift ($N>1$) of the resonances both theories give

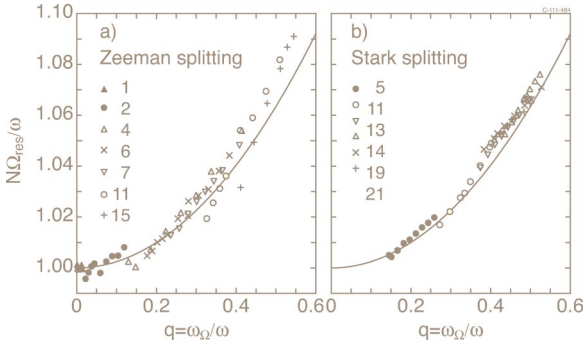

FIG. 3. Scaled resonance frequency $N\Omega_{\rm res}/\omega$ vs scaled harmonic field strength $q=\omega_\Omega/\omega$ for different photon numbers N. Full curve, Bloch-Siegert expression (1). (a) Static Zeeman splitting. (b) Static Stark splitting.

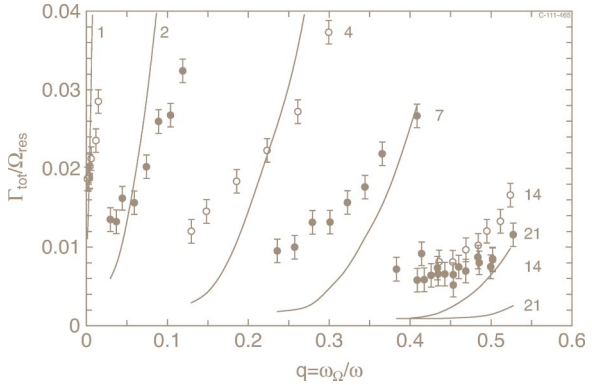

FIG. 4. Widths $\Gamma_{\rm tot}/\Omega_{\rm res}$ (FWHM) vs scaled harmonic field strength $q=\omega_\Omega/\omega$ for different photon numbers N ($N=21$ covers $N=19$ and 21). Bars show statistical errors; full curves, theory.

$$\frac{N\Omega_{\rm res}}{\omega}=1+\frac{1}{4}q^2+\frac{1}{64}q^4+O(q^6), \tag{1}$$

and the two theoretical results for the widths [full width at half maximum (FWHM)] of the Lorentzian at many field cycles can be expressed as

$$\frac{\Gamma_{\rm MLZ}}{\Omega_{\rm res}}=\begin{cases}\dfrac{2}{N}\left(\dfrac{Nq}{2}\right)^N, & N\leqslant 2\\ \dfrac{2}{\pi N}\exp\left[-\dfrac{\pi}{4}\dfrac{\omega}{\Omega_{\rm res}}\dfrac{(1-q)^2}{\sqrt{q}}\right], & N>2,\end{cases} \tag{2}$$

and

$$\frac{\Gamma_{\rm AD}}{\Omega_{\rm res}}=\frac{2}{\pi N}\exp\left[-2\frac{\omega}{\Omega_{\rm res}}\frac{(1-q)^2}{1+q}\mathbf{D}\left(\frac{1-q}{1+q}\right)\right], \tag{3}$$

where **D** is the complete elliptic integral of the third kind [37]. For $q\ll 1$ Eq. (3) reduces to the anticipated power law $\Gamma_{\rm AD}\propto q^N$ [36], but does not capture the spectral width associated with the finite duration of the pulse, a contribution which correctly dominates the MLZ theory as $q\to 0$ [34], where $\Gamma_{\rm MLZ}\to 4\sqrt{2}/(NT)$.

When it is assumed that no or one spin flip leaves the SFI spectra unchanged, the apparent probability for adiabatic response to the harmonic pulse is given by $P^{\rm ap}_{\rm adia}=(1-p)^{2n-2}+(2n-2)p(1-p)^{2n-3}$ [see Eq. (14) of Ref. [27]]. $P^{\rm ap}_{\rm adia}$ saturates at practically zero value near resonance even for small p and oscillates away from resonance, but smooth resonances were observed due to smearing by inhomogeneities of ω.

Figure 3 shows measured and calculated BS shifts (1) for selected values of N. While Ω is known with high precision, the values assigned to ω and ω_Ω are sensitive to distances between electrodes, calibration factors, and unavoidable stray electric fields. The ordinate of Fig. 3 covers a total variation of only 10 % to bring out the BS shifts. This exposes some dispersion in the data due to experimental errors. With these reservations we find a very good agreement with the predictions.

Figure 4 shows measured and calculated total resonance widths $\Gamma_{\rm tot}$ for selected values of N. All experimental widths were derived from clearly split resonances to avoid apparent broadening from unresolved splitting. Widths obtained from accurate numerical calculations are shown as full curves. These are somewhat larger than values obtained from Eqs. (2) and (3) due to saturation of $P^{\rm ap}_{\rm adia}$. For each N, the data points tend to lie above theory at small q and below at large q. This is most likely due to field inhomogeneities. At small q, where $P^{\rm ap}_{\rm adia}$ is not oscillatory, inhomogeneities lead to broadening, but at large q the oscillations in $P^{\rm ap}_{\rm adia}$ are averaged and this results in a narrower profile than the envelope of the resolved oscillations. Such systematic errors make it difficult to measure the widths accurately. In spite of this, clear trends were seen and the theory reproduces these fairly well.

In conclusion, experimental positions and widths of multiphoton intrashell resonances up to $N=21$ for hydrogenic Stark-Zeeman levels of a Rydberg shell have been measured. The experimental data show multiphoton Bloch-Siegert shifts and widths in good agreement with analytical predictions based on the reduction of the intrashell dynamics to that of two pseudospin-$\frac{1}{2}$ particles in two "magnetic fields." The precise and unambiguous determination demonstrated here critically gauge effects of Bloch-Siegert shifts of importance also in related areas of fundamental physics and in high-precision technology where direct measurements cannot be performed.

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