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Spectroscopy of atoms confined to micrometric pores in glass

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Abstract: We present two-photon spectroscopy of rubidium vapor contained in the interstitial cavities of porous glass. We report on the first observation of Rydberg atoms under 3D spatial confinement in these micrometric size dielectric cavities.

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1. Introduction

With the initial motivation of studying atomic vapors under a 3D spatial confinement, during the last few years we have been studying atomic spectroscopy of alkali atoms in the micrometric interstices of porous glass. Porous glass is a highly scattering medium for light which can appear as detrimental for many applications. However, randomness and disorder could be sometimes a welcome property for some applications as random lasers or alignment insensitive spectroscopical references [2].

The light field interacting with atoms contained in pores deep inside the sample has a speckle structure in which the polarization, amplitude and phase have rapid fluctuations over distances of the order of one wavelength. Due to the atomic thermal velocity, an atom explores large variations of the light field during its interaction time with the light. As a consequence of this motion induced field averaging, coherence effects are difficult to observe in the porous samples while unusual spectral lineshapes are observed for incoherent processes.

The transmission spectrum of Rb contained in the porous glass is unusual since for low enough atomic density and low confinement, the expected Rb absorption lines are highly suppressed. This is a consequence of the diffusive propagation of light that makes indistinguishable the detection of photons directly coming from the laser from those emitted as fluorescence. However, when the atomic vapor is spatially confined, the probability for an excited atom to collide with the dielectric surface and decay non-radiatively is not negligible. Thus confinement favors a non-radiative de-excitation channel that restores the absorption lines in the transmission spectrum [1]. Moreover, at higher atomic densities, radiation trapping increases the probability to excite an atom close to a surface and consequently the probability for non-radiative de-excitation.

Different spectral structures are observed in the light back-scattered by the porous sample in the direction of the incident laser beam. Sub-Doppler saturated absorption resonances are observed. In this configuration the incoming beam saturates the atomic medium and the back-scattered field acts as a probe field. The sub-Doppler resonances width are essentially determined by the probe wave-vector dispersion which is mainly determined by detection solid angle [2]. This experiment shows that, as would be expected, for the pores close to the incidence surface the light field has not a diffuse nature but partially preserves the direction of propagation as well as its polarization and phase [4].

Unusual spectral shapes were also observed in pump-probe spectroscopy performed with two different lasers (tuned around the D1 transitions of Rb) that are essentially due to the diffusive nature of light fields. A simple model neglecting coherent effects and considering completely randomized wave-vectors satisfactorily reproduce the full variety of lineshapes observed [3].

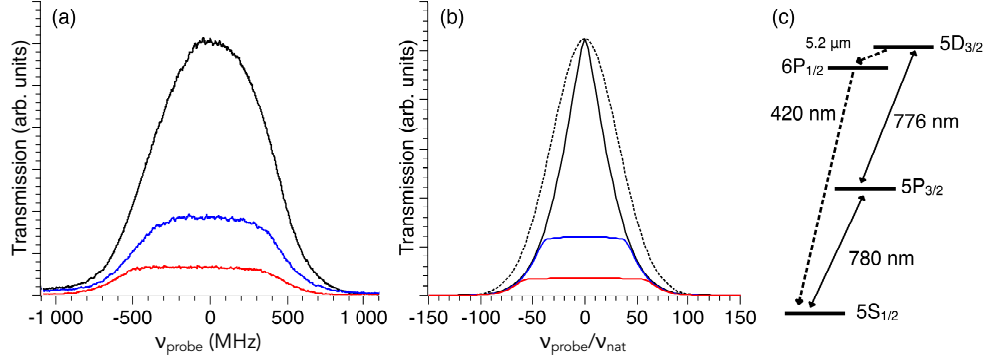


Fig. 1. (Color online) a) Measured spectra scanning the probe laser (D2) around the transition $^{85}\text{Rb}(F=2 \rightarrow F')$ at different frequencies for the coupling laser in 100 μm pores. b) Simplified model for the same conditions as a), (not considering the full hyperfine structure of level $5P_{3/2}$). The dashed curve is a Gaussian with Doppler width for comparison.

2. Highly excited energy levels

Using two-photon excitation, one can reach atomic states whose lifetime exceeds the mean time of flight of atoms in the pores. The level $5D_{3/2}$ of Rb (mean lifetime of around 240 ns) was excited using a probe laser resonant with the D2 line (780 nm) and a second coupling laser at 776 nm resonant with the transition $5P_{3/2} \rightarrow 5D_{3/2}$. The 5D level can decay through the $6P_{1/2}$ level resulting in blue fluorescence at 420 nm (Fig.1.c). We have detected the blue fluorescence through the porous medium as a measurement of the population of the 5D level. Typical spectra are shown in Fig.1.a. obtained by scanning the frequency of the probe laser (D2), for different frequencies of the coupling laser. Notice that when the coupling laser is resonant, the predicted spectra is narrower than the Doppler width. However, for a detuned coupling laser, the shape of these spectra has two remarkable properties, a flat top and symmetry around resonance.

The symmetry of the spectra can be explained if the contribution of coherent effects are neglected in the two-photon absorption since this results in spectral profiles centred on the atomic transition. Neglecting the coherence contribution is consistent with the phase randomization of the two fields. The flat top of the spectra is also a consequence of the diffuse nature of the optical fields. Satisfactory predictions of the measured spectra shapes (Fig.1.b) is obtained with a simplified model based on the assumptions of two-step (incoherent) two-photon excitation and isotropic distribution of the wave-vectors of the two laser fields.

It is worth mentioning that when the pore dimensions are reduced to about 10 μm , introducing then a spatial confinement that limits the mean time of flight of atoms to around 50 ns, much shorter than 5D level lifetime, no significant modifications of the spectral shapes were observed in comparison with samples with larger pores ($\sim 100\mu\text{m}$). This is in contrast with the observation in thin cells with 1D confinement and light beams propagating perpendicular to the cell windows where a narrowing of the fluorescence spectrum was observed as a consequence of the spatial confinement. For 1D confinement, the contribution of resonant atoms flying nearly parallel to the windows is favored over those that have large velocities in the direction perpendicular to the windows and consequently a short time of flight [5]. We interpret the non observation of such narrowing in the porous material as resulting from the 3D confinement in which the time of flight is mainly determined by the mean velocity modulus which is only loosely selected by the laser frequency (except for very large detunings).

2.1. Rydberg atoms under 3D confinement

We are currently exploring the excitation of Rb Rydberg atoms under a 3D confinement in micrometric pores in glass. 1D and 2D confined Rydberg atoms were previously studied in thin cells [6] and in hollow fibers [7]. In our experiment we excite Rydberg atoms using a probe laser resonant with the D1 transition of Rb, and a second coupling

laser at around 476 nm that can excite nS and $nD_{3/2}$ levels with principal quantum numbers n ranging from 30 to 40. Both lasers are superposed and focalized to a spot of $50 \mu\text{m}$ in diameter at the porous medium surface, and the backscattered light of the probe field is detected with a photodiode. The probe backscattered light was recorded scanning the coupling laser while keeping the probe laser locked to the D1 transition. Two different porous media were used having $100 \mu\text{m}$ and $10 \mu\text{m}$ pore sizes approximately. The atomic density was approximately 10^{14} atoms/cm³.

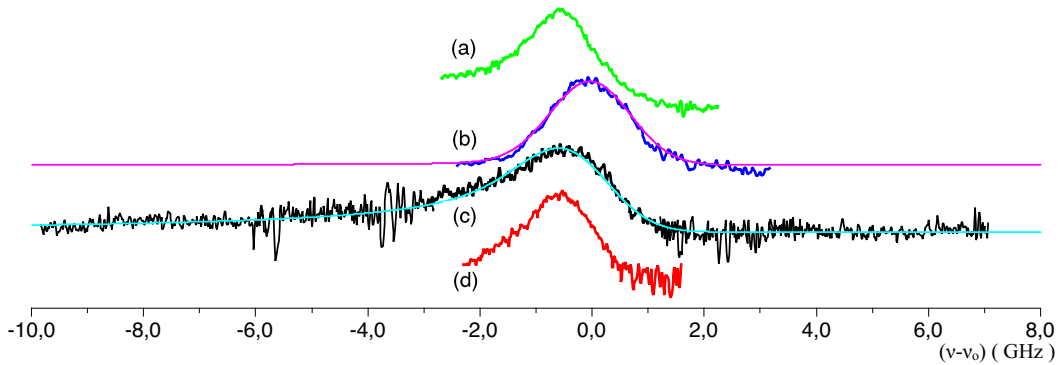


Fig. 2. (Color online) Registered spectra for $100\mu\text{m}$ pores (a,b), for $10\mu\text{m}$ pores (c,d) and for different Rydberg levels $33 D_{3/2}$ (a), $32 D_{3/2}$ (b),(c) and $31 D_{3/2}$ (d). The frequency axis is centered at the resonance frequency of the corresponding level (provided by an external reference). Continuous line: theoretical model.

Some spectra are shown at Fig.2. The spectra recorded for $100\mu\text{m}$ pores (Fig.2.a,b) have a Doppler width most probably due to the diffuse nature of light. However they have a clear asymmetry that suggest that they are also broadened by the Casimir-Polder interaction of the atoms with the dielectric walls of the pores [6]. This interaction is usually attractive (when it is not a resonant interaction [8]), introducing then a shift in the atomic level to lower frequencies. This suggestion is consistent with the measurement performed for the Rydberg atoms confined to $10\mu\text{m}$ cavities. The spectrum shown in Fig.2.c for the same Rydberg level as in Fig.2.b has an important increase in the red wing of the spectra. Also shown in Fig-2.b,c is a fit with a theoretical model using the coefficient of a Casimir-Polder atom-surface interaction as the only free parameter. The model predicts a potential corresponding to a $32 D_{3/2}$ level shift of 4 MHz at $10 \mu\text{m}$ distance, a value consistent with a non-resonant interaction. The level dependent interaction is evidenced by comparing spectra Fig.2.c-d. Further work is underway.

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