Multi-photon transitions in the ground state of cesium: applications for magnetometry and edm experiments

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Transitions involving the simultaneous absorption of several quanta of electromagnetic radiation are an important tool in atomic spectroscopy. In the optical domain two-photon absorption has become a standard powerful method for ultra-high resolution spectroscopy. Multi-photon transitions in the radio-frequency domain were intensively studied in the early days of double resonance spectroscopy on optically pumped atomic vapours and became an important corner stone for the development of the dressed atom model. The main interest of those studies was the investigation of n-quantum transitions between Zeeman sublevels with magnetic quantum numbers differing by $\Delta m = \pm 1$. The resonance frequencies of such transitions are subharmonics of the basic Larmor frequency of the system.

In this paper we report on a different type of multi-photon transitions, viz. those, where n radio-frequency photons induce a transition between states with magnetic quantum numbers differing by $\Delta m = \pm n$. In low magnetic fields (linear Zeeman effect) the resonance frequencies of these ($\Delta m = n$) n-photon transitions are all degenerate and equal to the Larmor frequency of the system. For a hyperfine level F of an alkali ground state the individual multi-photon components can be spectrally resolved when the magnetic field B is chosen large enough, so that non-linear terms in the Breit-Rabi expression for the level energies lift the degeneracy beyond the intrinsic line width. When terms of order B^3 can be neglected in the expansion of the Breit-Rabi formula lines involving an even number of photons will be centered between adjacent lines involving an odd number of photons. The highest observable order is the $2F$-photon line corresponding to a complete reversal ($m = F \rightarrow m = -F$) of the spin $\vec{F}$.

The different multi-photon orders appear as the amplitude of the driving rf-field is increased. In our experiments, performed on the magnetic sublevels of the F=4 hyperfine level of the Cs $6S_{1/2}$ ground state, one thus observes 8 one-photon lines at very low rf intensity. When the intensity is increased these lines saturate and show power broadening; at the same time 7 narrow two-photon lines appear between the broader one-photon lines. When the rf power is further increased this scenario (broadening, saturation, appearance of the next order) is repeatedly observed, until finally a single 8-photon line, superposed on a broad background, appears.

These multi-photon lines have several remarkable features with interesting applications. First of all the higher order lines have a much higher magnetometric sensitivity than the standard one-photon line, i.e. in a shot-noise limited experiment their line center can be determined with much higher accuracy. Numerical calculations show that the 8-photon line in the F=4 multiplet of cesium is more than 100 times more sensitive to magnetic field variations than the one-photon line. This property has direct applications in magnetometry and in experiments searching for permanent electric dipole moments (edm) of atoms. Moreover, some even-number multi-photon transitions, when used in edm experiments allow to suppress an important systematic effect connected with quadratic Stark shifts and imperfect electric field reversals. As these Stark shifts are proportional to m^2, the resonance lines corresponding to transitions between states m and $-m$ will not be affected by electric fields.

We have investigated multi-photon transitions in the F=4 ground state of the Cs atom with a standard optical-rf double resonance set-up using a circularly polarized single-mode diode laser locked to the optical pumping transition. Both room temperature Cs vapor contained in a paraffin-coated glass cell (D_1-line excitation and absorption detection) and Cs atoms implanted in a ^{4}He crystal (D_1-line excitation and fluorescence detection)were investigated. The measurements were performed in a magnetically unshielded environment which limited the magnetic resonance linewidths to several 10 Hz in the vapor cell and to 300 Hz in the Cs/He sample because of laboratory magnetic fields oscillating at 50 Hz. In both samples we observed all allowed multi-photon transitions up to the 8-photon transition. Despite the relatively large linewidths, an improvement of the magnetometric sensitivity by more than one order of magnitude (as compared to the one-photon transition with optimal parameter settings) could be demonstrated.