The observation of dark-resonances in the two-electron atom barium and their influence on optical cooling is reported. In heavy alkali earth atoms, i.e. barium or radium, optical cooling can be achieved using $n^1S_0-n^2P_1$ transitions and optical repumping from the low lying $n^1D_2$ and $n^3D_{1,2}$ states to which the atoms decay with a high branching ratio. The cooling and repumping transition have a common upper state. This leads to dark resonances and hence make optical cooling less inefficient. The experimental observations can be accurately modelled by the optical Bloch equations. Comparison with experimental results allows us to extract relevant parameters for effective laser cooling of barium.

The pioneering work on optical cooling and trapping of neutral atoms as well as important advances made since then have mainly been carried out with atomic systems which have a strong optical transition connected to the ground or metastable state. Although a two-level system would be ideal, even optically rather simple accessible atoms, such as stable alkali isotopes, have hyperfine structure. This requires (re-)pumping of the atomic population into the state of highest angular momentum in order to realize an effective two-level system.

Some atoms with complex atomic structure have a unique potential for precision tests of fundamental symmetries [1,2]. This can be exploited best with cooled and trapped atoms. Among these systems is the radium (Ra) atom, which offers a strong enhancement of a potential permanent electron Electric Dipole Moment (EDM) due to the unique close proximity of the $7s6d^3P_1$ and $7s6d^3D_2$ levels [3]. In addition it provides an excellent opportunity to find a potential nucleon EDM in some isotopes where the nuclei are octupole deformed [4].

Levels [3]. In addition it provides an excellent opportunity to find a potential nucleon EDM in some isotopes where the nuclei are octupole deformed [4]. The radioactivity of all Ra isotopes and the availability of rather small cooling force. Unfortunately for the heavy alkali earth elements barium (Ba) and Ra the branching of the decay of the excited $^1P_1$ state to the $^3S_0$ state respectively sum of the $^1D_2$ and the $^3D$ states is 1:330(30) (Ba) and 1:50(80) (Ra). In the lighter alkali earth atoms Ca and Sr the ratio is 1:100000(1000) and 1:52000(500), and the transition to the $^1D_2$ provides the strongest of the leaks. Therefore effective repumping of the population trapped in the metastable D-states to the cooling cycle will be essential for a scheme involving the strong transition from the ground state. In addition, repumping needs to go via the same excited $^1P_1$ state, otherwise the atoms will be distributed over even more metastable states. We observe dark resonances, which connect the ground state and the metastable states through coherent interaction of the cooling laser and the repump lasers. These Raman transitions decrease the efficiency of the cooling process.

Since their first experimental observation in 1976 in sodium [6], coherent dark resonances have been observed e.g. in alkali atoms between different hyperfine states, in chromium [7] and in samarium vapor [8]. A review on the developments and status of the coherent dark state spectroscopy can be found in references [3,10]. We have studied such processes in the heavy alkali earth atom Ba which has a number of stable isotopes and is accessible with commercial lasers for all relevant transitions. We expect that the results can be transferred directly to Ra because of the similar atomic level scheme.

The relevant part of the Ba atom energy level scheme is displayed in Fig. 1. The $6s6p^1P_1$ excited state decays to the $6s^2^1S_0$ ground state and weakly to the $6s5d^3D_2$ metastable state. If the atom is exposed to a bichro-
matic laser field with copropagating green and infrared laser beams at λ₁=553.7 nm and λ₂=1500.4 nm, the scattered light contains a strong component λ₁ and a small component at λ₂.

The atomic beam is modelled as an atomic three-level Λ-system, which coherently interacts with two light fields. The formalism applied here is similar to the semi-classical scheme reported to describe the Ba atoms [12,13]. The atom is treated quantum mechanically in the Heisenberg picture and characterized by the 6 parameters |i⟩⟨j| (i,j=1,2 and 3). The coherent laser fields are treated as classical electromagnetic waves E₀ sin(ω₀t) for λ₁ and Eᵣ sin(ωᵣt) for λ₂. The two transitions have a linewidth Γ₀ and Γᵣ respectively. Spontaneous emission of photons is treated as decays from |2⟩ to |1⟩ and |3⟩ with rates Γ₂₁ and Γ₂₃.

The equations of motion for the density matrix ρ of the atom (Bloch equations) contain terms that model the coherent coupling with the external light fields by effective Rabi frequencies Ω₁₂ and Ω₁₃, which denote the strength of the coupling between the atom and the electric field of the lasers.

\[
\Omega_{ij} = \frac{-eE_0}{\hbar} <j|\hat{r}|i> \quad (1)
\]

where e is the charge of the electron, E₀ is the electric field of the electromagnetic wave, h is Planck's constant and the last term is the transition dipole matrix element between states j and i. The rotating wave approximation, which neglects the fast oscillating terms, is applied [14]. The optical Bloch equations where we use the abbreviation \( \hat{\rho}_{ij} = \rho_{ij} e^{i\Delta_{ij}t} \) is

\[
\frac{d\rho_{22}}{dt} = -(\Gamma_{21} + \Gamma_{23}) \rho_{22} + \frac{i}{2} \Omega_{12}^{*} \hat{\rho}_{21} - \frac{i}{2} \Omega_{32} \hat{\rho}_{32} + c.c.
\]

\[
\frac{d\rho_{33}}{dt} = \Gamma_{23} \rho_{22} + \frac{i}{2} \Omega_{32}^{*} \hat{\rho}_{23} + c.c.
\]

\[
\frac{d\rho_{12}}{dt} = -[\Gamma_{12} + \Gamma_{23}] \rho_{12} - i \Delta_{12} \rho_{12} + \frac{i}{2} \Omega_{12}^{*} \hat{\rho}_{32} + i \hat{\rho}_{13} - \frac{i}{2} \Omega_{13}^{*} \hat{\rho}_{13} + \frac{i}{2} \Omega_{32} \rho_{22} + \frac{i}{2} \Omega_{12} \rho_{32} + c.c.
\]

\[
\frac{d\rho_{13}}{dt} = -i(\Delta_{12} - \Delta_{32}) \rho_{13} - \frac{i}{2} \Omega_{12} \rho_{33} + \frac{i}{2} \Omega_{32} \rho_{22} + \frac{i}{2} \Omega_{32} \rho_{33} + \frac{i}{2} \Omega_{13} \rho_{22} + c.c.
\]

\[
\frac{d\rho_{21}}{dt} = -i(\Delta_{12} - \Delta_{32}) \rho_{21} - \frac{i}{2} \Omega_{12} \rho_{33} + \frac{i}{2} \Omega_{32} \rho_{22} + \frac{i}{2} \Omega_{13} \rho_{33} + \frac{i}{2} \Omega_{32} \rho_{33} + c.c.
\]

\[
\frac{d\rho_{31}}{dt} = -i(\Delta_{12} - \Delta_{32}) \rho_{31} - \frac{i}{2} \Omega_{13} \rho_{33} + \frac{i}{2} \Omega_{32} \rho_{22} + \frac{i}{2} \Omega_{13} \rho_{33} + \frac{i}{2} \Omega_{32} \rho_{33} + c.c.
\]

The equations are solved numerically for our experimental conditions. In additions we take the moving of the atoms through the laser fields into account by considering the finite time the atoms spent in the respective laser beams. A Maxwell-Boltzmann velocity distribution at temperature T_{Ba} for the atoms in an atomic beam is assumed.

Depending on the detuning Δ₂ of the laser at λ₂ from the 6s²¹S₀ - 6s6p¹P₁ transition and the detuning Δ₁ of the laser at λ₁ from the 6s²¹S₀ - 6s6p¹P₁ transition the atoms are driven coherently between the ¹S₀ and the ¹D₂-states without passing through the ¹P₁ state. For the resonance condition Δ = Δ₁ = Δ₂ and the detuning Δ much larger than the Rabi frequencies Ω₁₂ and Ω₁₃ the effective Rabi frequency for the Raman transition can be written as

\[
\Omega_{13} = \frac{\Omega_{12} \Omega_{32}}{2\Delta} \quad . (3)
\]

For an intensity of I = 1 mW/cm² of both lasers the estimated Rabi frequencies (Eq.1) are given in Table I together with the spontaneous transition probabilities. However, for effective cooling Δ₁₂,3₂ are both on the order of the linewidth of the transitions and Eq. (3) is not applicable. Thus the full optical Bloch equations need to be evaluated.

The fluorescence spectrum observed shown in Fig. 3 is obtained by scanning the frequency of the laser at λ₁ in the absence of the laser at λ₂. Contributions from different isotopes of Ba are visible. The solid line in the figure

| Transition | λ_{ik} [nm] | Ω_{ik} [rad/s] | Γ_{ik} [10⁸ s⁻¹] | Ω_{ik} [10⁸ s⁻¹] |
|------------|-------------|----------------|-----------------|----------------|
| 6s²¹S₀ - 6s6p¹P₁ | 553.7       | 1.19(1)        | 22 · 10⁶        |
| 6s5d¹D₂ - 6s6p¹P₁ | 1500.4     | 0.0025(2)      | 4.7 · 10⁶        |
| 6s5d³D₁ - 6s6p¹P₁ | 1130.6     | 0.0011(2)      | 2.0 · 10⁶        |
| 6s5d³D₁ - 6s6p¹P₁ | 1107.8     | 0.000031(5)    | 0.32 · 10⁶    |
FIG. 2: A beam of atomic Ba with natural isotope composition emerges from an oven heated to ≈ 800 K. The two counter-propagating laser beams cross the atomic beam at right angle 0.6 m downstream, limiting the divergence of the atomic beam to 30 mrad. Fluorescence from the 6s6p 1P1 state is detected with a photomultiplier tube (PMT).

represents the numerical solution of Eqns. (2). The spectra are normalized to the peak amplitude of the 138Ba resonance. The other isotope contributions are according to their natural abundance and their known isotope shift [15].

Fig. 4 gives the fluorescence spectrum obtained with the laser at λ2 (red) detuned by Δ2 = -13.5(5) MHz with respect to the center of the 6s5d 1D2 - 6s6p 1P1 transition in the 138Ba isotope. The frequency of the laser at λ1 is scanned across the resonance. The laser power at λ2 is 10(1) mW yielding an intensity of 320(120) mW/cm². This corresponds to a Rabi frequency Ω22 = 80(30) ·10⁶ rad/s. The laser at λ1 at a power of 2.2(1) mW, or an intensity of 24(1) mW/cm², we yield a Rabi frequency of Ω12 = 120(3) ·10⁶ rad/s. The laser linewidth for the two lasers is much less than the Rabi frequencies and the decay rate of the 6s6p 1P1 state. In the calculations a laser linewidth of Γg = 1 MHz at λ1 and Γr = 50 kHz at λ2 is assumed. The spectra are normalized to the peak amplitude. The only adjustable parameter is the ratio of the coherent Raman transition fraction to the regular fluorescence spectrum due to atoms not passing through both of the laser beams. The latter contributes 45(15)% to the 138Ba fluorescence.

The dip on the left side of the resonance in 138Ba is the two-photon Raman transition. The fluorescence is reduced because of the direct coupling of the 6s² 1S0 ground state and the 6s5d 1D2 state. The solid lie is the numerical solution with the parameter for detuning and the Rabi frequency given above. This demonstrates that the numerical approach fully describes the atomic response, and such a measurement can be used to extract the Rabi frequencies Ω12 and Ω32.

We have observed the dark resonance in a second measurement. Here, the green laser at λ1 was frequency...
FIG. 5: The measured dark resonance spectrum for the \( ^1\text{P}_1 \) state. Here the laser at \( \lambda_1 = 553.7 \text{ nm} \) is frequency locked to the transition in \( ^3\text{S}_0 \) which is much larger than the linewidth we observe unperturbed scattering from the \( ^3\text{S}_0 - ^3\text{S}_1 \) transition. When the detuning \( \Delta_{32} \) approaches the resonance the scattering rate decreases, because of coherent population transfer to the metastable \( ^6\text{S}_0 \) state. The width of the observed dip is 20(1) MHz, in good agreement with the numerical solution of the optical Bloch equations with a maximum frequency of 50 MHz for the laser to achieve the same Rabi frequency for the \( ^6\text{S}_0 - ^6\text{P}_1 \) transition the laser intensity has to be about 5 times larger and for \( ^3\text{D}_1 - ^6\text{P}_1 \) transition more than 100 times larger.

Thus we can expect that Rabi frequencies of several \( 10^7 \text{ rad/s} \) can be achieved with a laser beam diameter of 5 mm and the available laser power of 10 mW from the fiber laser. To achieve the same Rabi frequency for the \( ^6\text{S}_0 - ^6\text{P}_1 \) transition the laser intensity has to be about 5 times larger and for \( ^3\text{D}_1 - ^6\text{P}_1 \) transition more than 100 times larger.

We have reported first measurements on dark resonances in atomic Ba. The observations in a three level subsystem can be well described with a model using numerical solutions of the optical Bloch equations. The accuracy of the determination is sufficient for the evaluation of the feasibility of laser cooling of barium. For a laser cooling scheme on Ba the repumping from \(^1\text{D}_2 \) states have to be included. They will also exhibit coherent Raman transitions. The full 5-level system can be modelled with the same approach and solved numerically. In conclusion we expect that using the strong transition at \( \lambda_1 \) and the repumping through the same excited state can lead to a more effective cooling of heavy alkali earth atoms like Ba and Ra than the demonstrated laser cooling on the \( ^7\text{S}_0 - ^7\text{P}_1 \) intercombination line in Ra. This will allow for more stringent limits on permanent electric dipole moments in atomic systems.

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