

Measurement of the spin-forbidden decay rate $(3s3d)^1D_2 \rightarrow (3s3p)^3P_{2,1}$ in ^{24}Mg

K. T. Therkildsen,^{*} B. B. Jensen, C. P. Ryder, N. Malossi, and J. W. Thomsen

The Niels Bohr Institute, Universitetsparken 5, 2100 Copenhagen, Denmark

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We have measured the spin-forbidden decay rate from $(3s3d)^1D_2 \rightarrow (3s3p)^3P_{2,1}$ in ^{24}Mg atoms trapped in a magneto-optical trap. The total decay rate, summing up both exit channels $(3s3p)^3P_1$ and $(3s3p)^3P_2$, yields $196 \pm 10 \text{ s}^{-1}$ in excellent agreement with recent relativistic many-body calculations of Porsev *et al.* [Phys. Rev. A **64**, 012508 (2001)]. The characterization of this decay channel is important as it may limit the performance of quantum optics experiments carried out with this ladder system as well as two-photon cooling experiments currently explored in several groups.

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I. INTRODUCTION

The interest in alkaline-earth-metal systems has increased significantly over the past years. One of the main interests in these systems is their applications to high resolution spectroscopy and atomic frequency standards, as they offer very narrow electronic transitions [1]. Another attractive feature of the two-electron systems is their very simple energy level structure, for bosonic isotopes, free of both fine and hyperfine structures. This simple structure opens for detailed comparison between theory and experiment [2]. Recent advances in *ab initio* relativistic many-body calculations of two-electron systems have provided very accurate values for atomic structure and properties such as the electric-dipole transitions of Mg, Ca, and Sr [3]. However, in the case of Mg relatively few experiments have been reported in literature motivating the studies in this Brief Report.

In a series of papers two-photon cooling in ladder schemes has been proposed to lower the temperature below the Doppler limit of alkaline-earth-metal elements [4–6]. The magnesium and calcium systems are of particular interest since cooling is limited to a few millikelvin. Experimental evidence for two-photon cooling has been established, but the effect is rather modest contrary to theoretical predictions [7,8]. One important mechanism to take into account in these studies are leaks from the ladder system, limiting the number of atoms that eventually can be cooled.

In this Brief Report we present measurements of the spin-forbidden decay from the magnesium $(3s3d)^1D_2$ state to $(3s3p)^3P_1$ and $(3s3p)^3P_2$. Using atoms trapped in a magneto-optical trap we optically prepare the atoms in the $(3s3d)^1D_2$ state and measure the decay to $(3s3p)^3P_{2,1}$ manifold by trap loss measurements. This enables us to deduce the absolute decay rate for these transitions and compare our results to state-of-the-art theoretical calculations.

II. TRAP LOSS MEASUREMENTS

In Fig. 1, we show the relevant energy levels and transitions in our experiment. The 285 nm transition is used for cooling and trapping the atoms, while the 881 nm transition

is used to populate the $(3s3d)^1D_2$ state. From this state the atoms may decay to the $(3s3p)^3P_1$ state (407.6 nm) with rate Γ_{21} or to the $(3s3p)^3P_2$ state (408.3 nm) with rate Γ_{22} or simply back to $(3s3p)^1P_1$. By neglecting decay to the $(3s3p)^3P_0$ state, which is weaker by orders of magnitude, no other decay channels are open. When the 881 nm laser is switched off we express the steady-state number of atoms N_0 as the ratio of the load rate to linear losses L/α . Here we neglect intra-MOT (magneto-optical trap) collisions since we are working at relative low atom densities. The equation for the number of atoms trapped in the MOT with 881 nm light present can be written as

$$\dot{N} = L - \rho_{aa}(\Gamma_{22} + \Gamma_{21})N - \alpha N. \quad (1)$$

Experimentally we determine the total loss rate, $\Gamma_{22} + \Gamma_{21}$. In our case the linear loss rate α is dominated by resonant photoionization of $(3s3p)^1P_1$ at 285 nm [9]. Photoionization from the $(3s3d)^1D_2$ level is below 1 Mb and will not be considered here [10]. The steady states become

$$N^{\text{on}} = \frac{L}{\rho_{aa}(\Gamma_{22} + \Gamma_{21}) + \alpha^{\text{on}}} \quad (2)$$

and

$$N^{\text{off}} = \frac{L}{\alpha^{\text{off}}}. \quad (3)$$

The ratio of signal with 881 nm turned on and off is proportional to ρ_{bb}^{on} and ρ_{bb}^{off} , respectively. We find finally

$$\frac{S^{\text{on}}}{S^{\text{off}}} = \frac{\alpha^{\text{off}} \rho_{bb}^{\text{on}} / \rho_{bb}^{\text{off}}}{\rho_{aa}(\Gamma_{21} + \Gamma_{22}) + \alpha^{\text{off}} \frac{\rho_{bb}^{\text{on}}}{\rho_{bb}^{\text{off}}}} \quad (4)$$

as the α coefficient is dominated by resonant photoionization and thus proportional to the fraction of atoms in the $(3s3p)^1P_1$ state. So far we assumed that all atoms decaying to the $(3s3p)^3P_J$ manifold are lost. This is indeed the case for the $(3s3p)^3P_2$ final states due to their long lifetime exceeding 2200 s [11]. On the time scale of the experiment, being only a few seconds, we consider the atoms from this state lost. However, for the $(3s3p)^3P_1$ states atoms in the $|J=1, m_J=+1\rangle$ magnetic substate will be trapped in the magnetic quadrupole field of the MOT and most likely be recaptured when decaying back to the $(3s^2)^1S_0$ ground state. Atoms in

^{*}kasptt@fys.ku.dk

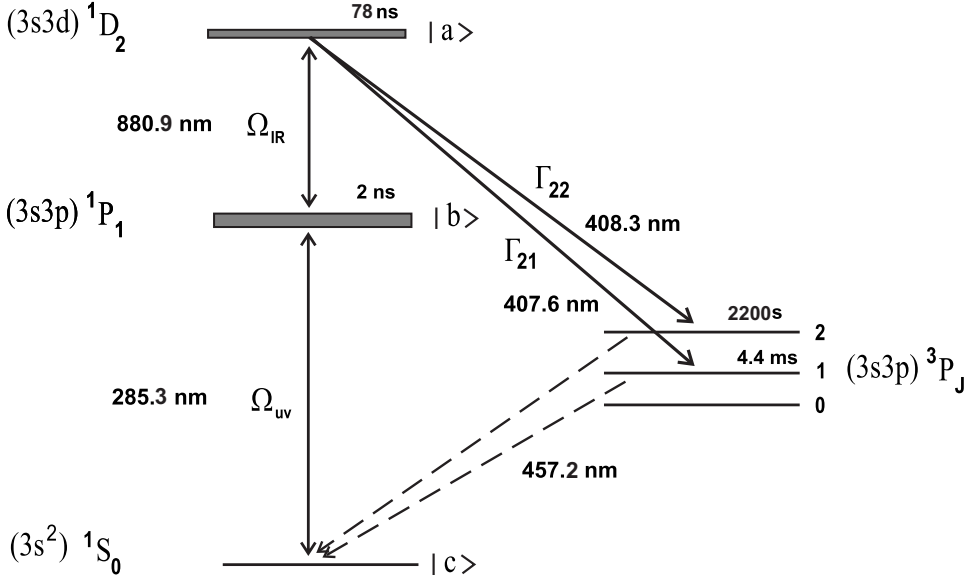


FIG. 1. Energy levels relevant for the $(3s3d)^1D_2 \rightarrow (3s3p)^3P_{2,1}$ decay measurement in ^{24}Mg .

$|J=1, m_j=0, -1\rangle$ magnetic substates are expected to be lost since the rms velocity of the trapped atoms exceeds 1 m/s leading to a rms traveled distance of more than 5 mm, which is large compared to our MOT beam diameter of only a few millimeters. The total decay rate will thus be modified to $\gamma\Gamma_{21} + \Gamma_{22}$. The actual value of γ depends strongly on the polarizations of the 285 nm beam and the 881 nm beam.

If we assume a standard one-dimensional model for the MOT we can determine the influence of the constant γ . This is a good approximation as the 881 nm beam is directed along one of the uv MOT beams but not retroreflected. Consider atoms on the $-z$ side (see Fig. 2). These atoms will mainly populate the $|J=1, m_j=0, -1\rangle$ state. Further excitation of the collinear 881 nm laser will drive the atoms to the

left [blue (dashed) line] or right [red (solid) line] depending on the helicity of the 881 nm laser. This creates an anisotropy in the decay to the $(3s3p)^3P_1$ state. Atoms driven along the blue (dashed) line, i.e., with the same polarization helicity as the MOT beams, will only populate nontrapped states. Experiments performed in these configurations are thus insensitive to the retrapping dynamics as described above. Using the Clebsch Gordan coefficients shown in Fig. 2 we find the rate $[1/2 \times 1 + 1/2 \times (1/2 + 1/2)]\Gamma_{21} = \Gamma_{21}$, as expected. For the opposite polarization we find a reduction $[1/2 \times 1/2 + 1/2 \times (2/3 + 1/6)]\Gamma_{21} = 2/3\Gamma_{21}$. We now consider a more general case where all three magnetic substates $|J=1, m_j=+1, 0, -1\rangle$ are populated. The populations are denoted n_+, n_0 and with $n_+ + n_0 + n_- = 1$. We find the effective decay constant

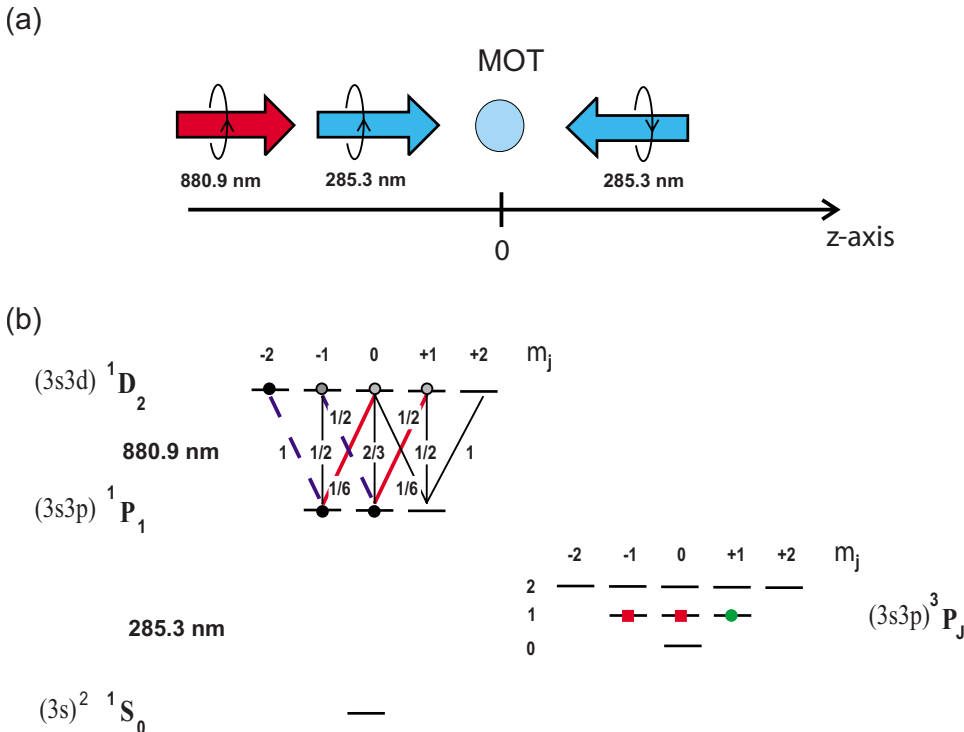


FIG. 2. (Color online) (a) Experimentally the 881 nm beam is collinear with one of the MOT beams. The polarization state can be selected similar to the MOT helicity, as shown in the figure, or opposite. (b) Polarization dependent pumping of atoms in the MOT. Assuming a one-dimensional representation of the MOT the 881 nm polarization, represented by blue (dashed) line (=MOT helicity) or red (solid) line, optical pumping will drive the atoms such that anisotropy in the decay rate will emerge. The green dots represent atoms trapped in the MOT quadrupole magnetic field, while red squares represent atoms in nontrapped states. The Clebsch Gordan coefficients shown will also apply to the $(3s3d)^1D_2 \rightarrow (3s3p)^3P_1$ decay.

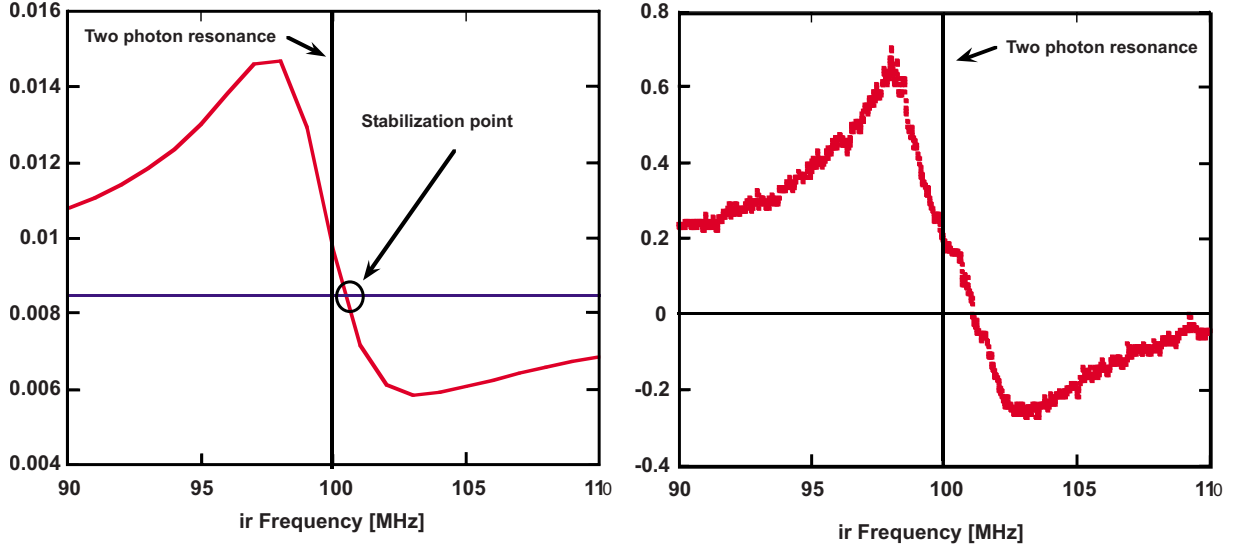


FIG. 3. (Color online) The 881 nm laser is frequency locked to the two-photon resonance of the ladder system $\nu_{881} = -\nu_{285} = 100$ MHz. This ensures a maximal transfer of population to the $(3s3d)^1D_2$. Left part of the figure shows the $(3s3p)^1P_1$ excited state fraction obtained from the optical Bloch equations. Right part shows the experimental lock signal.

as $(1 - n_+/6)\Gamma_{21}$ for MOT polarization. The n_+ coefficient is expected to be small compared to the sum of the two other terms. However, even in an extreme case where all substates are populated equally Γ_{21} is affected only by about 5%.

With an 881 nm helicity opposite to that of the MOT beam, marked in red color on Fig. 2, will ultimately result in a destruction of the trapping force as the 881 nm laser intensity increases. This effect is expected as a result of destructive reshuffling of the atoms into “wrong” magnetic substates.

III. EXPERIMENTAL SETUP AND RESULTS

The main part of the experimental setup has been described in [12]. In our experiment the 285 nm MOT is probed by 881 nm light produced by a Ti:sapphire laser. The 881 nm light has power and frequency controlled by an acousto-optic modulator (AOM). A $\lambda/4$ plate controls the helicity of the light. Presence of the 881 nm light will change the 285 nm light level recorded by a photo-multiplier (not sensitive to 881 nm light). We use this to lock the 881 nm laser frequency close to the two-photon resonance $\nu_{881} = -\nu_{285} = 100$ MHz during the experiments (see Fig. 3). Increasing the Rabi frequency of the 881 nm light causes a minor change in the locking point. In our power range this corresponds to about 1 MHz and not considered important at this level of accuracy.

Typically 10^7 atoms are captured in the MOT, with a rms diameter of 1 mm and temperatures from 3 to 5 mK. The Doppler cooling limit for the MOT is 2 mK as sub-Doppler cooling is not supported in this system.

The number of emitted 285 nm photons when the IR laser was on and off was measured for each of the two circular polarization states. The 881 nm light was sent through the AOM, which was modulated with a square wave at 1 kHz, with a duty cycle of 20%. The photomultiplier signal was

measured and averaged over 32 periods with the light on and off, thus providing values for S^{on} and S^{off} , respectively. At the same time these signals provided a lock of the Ti:S laser to the two-photon resonance (see Fig. 3). This measurement was repeated for different IR intensities (Rabi frequencies), while the MOT was running in steady-state mode.

Experimental values for the ratio $\frac{S^{\text{on}}}{S^{\text{off}}}$ as a function of the 881 nm Rabi frequency are shown in Fig. 4. As expected the fraction starts out at 1 and gradually decreases as the Rabi frequency is increased. In Fig. 4 the 881 nm polarization corresponds to the MOT helicity [blue (dashed) line in Fig. 2] and polarization opposite to the MOT helicity. The solid curves are results from a simulation based on the optical Bloch equations including the MOT magnetic field. Experiments with this helicity clearly support a value of $\Gamma = 196 \pm 10 \text{ s}^{-1}$ in good agreement with calculations performed in [3], where $\Gamma = \Gamma_{22} + \Gamma_{21} = 57.3 + 144 \text{ s}^{-1}$. For the opposite 881 nm polarization [red (solid) line in Fig. 2] we observe a reduced decay constant of $\Gamma = 150 \pm 15 \text{ s}^{-1}$. Using values in [3] and the coefficient deduced above we obtain $\Gamma = \Gamma_{22} + 2/3\Gamma_{21} = 153 \text{ s}^{-1}$ in favor of the one-dimensional model described above. From this we find the decay constants $3s3d^1D_2 \rightarrow 3s3p^3P_1$ and $3s3d^1D_2 \rightarrow 3s3p^3P_2$ to be $\Gamma_{21} = 138 \pm 10 \text{ s}^{-1}$ and $\Gamma_{22} = 58 \pm 4 \text{ s}^{-1}$.

IV. CONCLUSION

In conclusion we have measured the total spin-forbidden decay rate $(3s3d)^1D_2 \rightarrow (3s3p)^3P_{2,1}$ in ^{24}Mg . We find a rate of $\Gamma = 196 \pm 10 \text{ s}^{-1}$ supporting state-of-the-art relativistic many-body calculations. Assuming a simple one-dimensional model for the MOT we find the differential decay constants $(3s3d)^1D_2 \rightarrow (3s3p)^3P_1$ and $(3s3d)^1D_2 \rightarrow (3s3p)^3P_2$ as $\Gamma_{21} = 138 \pm 10 \text{ s}^{-1}$ and $\Gamma_{22} = 58 \pm 4 \text{ s}^{-1}$ supporting the above mentioned calculations.

With a total loss rate of 196 s^{-1} the magnesium MOT lifetime can be limited to millisecond time scale or less near

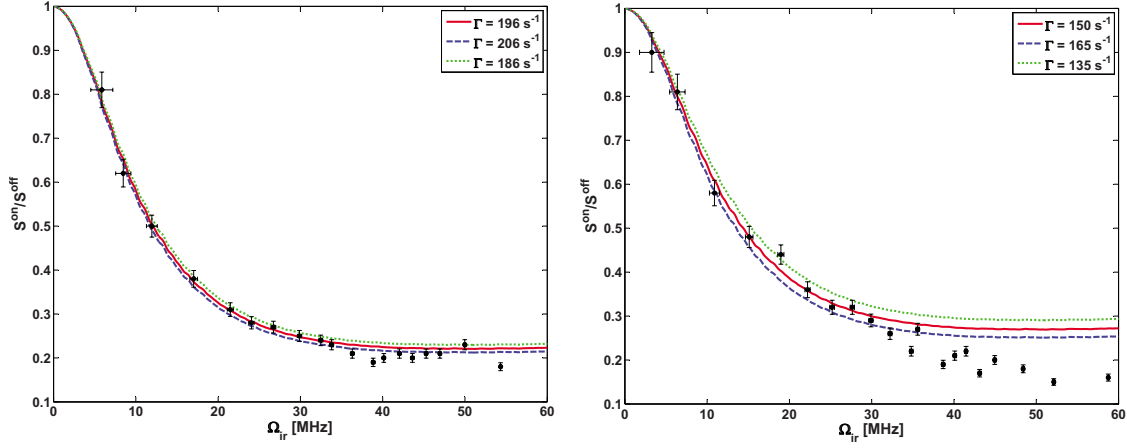


FIG. 4. (Color online) Experimental values for the ratio $\frac{S^{\text{on}}}{S^{\text{off}}}$ as a function of the 881 nm Rabi frequency. (Top) Here the circular polarization corresponds to the MOT helicity [blue (dashed) lines in Fig. 2]. The curves are results from a simulations based on the optical Bloch equations, including the magnetic field of the MOT, at different decay constants $\Gamma = \Gamma_{22} + \Gamma_{21} = 186, 196$, and 206 s^{-1} . Experiments with this circular helicity support a value of $\Gamma = 196 \pm 10 \text{ s}^{-1}$. (Bottom) Experiment with 881 nm polarization opposite to the MOT helicity. At a Rabi frequency of about 30 MHz the MOT is being destroyed by the destructive optical pumping effects of the 881 nm laser. Only data points below 30 MHz should be considered.

the two-photon resonance. To avoid significant atom losses the cooling time of a two-photon cooling scheme must be kept below 1 ms, as demonstrated in [8]. The losses from the 1D_2 state are according to [5,6] limiting the minimum temperature reachable by two-photon cooling to about 50 μK . In quantum optics experiments employing this ladder system decoherence will likewise play a role on relative short time scales.

However the decay rate Γ_{22} opens a way of populating the 3P_2 state. For our current MOT characteristics a load rate in the order of 10^5 – 10^6 atoms/s is expected. Assuming a mag-

netic trap lifetime in the order of 20 s, this will result in a 3P_2 population of 10^6 – 10^7 atoms. However, the loading rate into the 3P_2 can be increased by increasing the MOT loading rate using a Zeeman slower. The 3P_2 state is a fine starting point for further laser cooling on the 3P_2 – 3D_3 transition as done for Ca [13].

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- [1] T. P. Heavner, S. R. Jefferts, E. A. Donley, J. H. Shirley, and T. E. Parker, *Metrologia* **42**, 411 (2005); S. Bize *et al.*, *J. Phys. B* **38**, S449 (2005); L. Hollberg *et al.*, *ibid.* **38**, S469 (2005); M. M. Boyd *et al.*, *Science* **314**, 1430 (2006); W. H. Oskay *et al.*, *Phys. Rev. Lett.* **97**, 020801 (2006).
 - [2] M. Machholm, P. S. Julienne, and K.-A. Suominen, *Phys. Rev. A* **59**, R4113 (1999).
 - [3] S. G. Porsev, M. G. Kozlov, Y. G. Rakhlin, and A. Derevianko, *Phys. Rev. A* **64**, 012508 (2001).
 - [4] Victor C. Magno, Reinaldo L. Cavasso Filho, and Flavio C. Cruz, *Phys. Rev. A* **67**, 043407 (2003).
 - [5] Josh W. Dunn, J. W. Thomsen, Chris H. Greene, and Flavio C. Cruz, *Phys. Rev. A* **76**, 011401(R) (2007).
 - [6] Giovanna Morigi and Ennio Arimondo, *Phys. Rev. A* **75**, 051404(R) (2007).
 - [7] N. Malossi, S. Damkjaer, P. L. Hansen, L. B. Jacobsen, L. Kindt, S. Sauge, J. W. Thomsen, F. C. Cruz, M. Allegrini, and E. Arimondo, *Phys. Rev. A* **72**, 051403(R) (2005).
 - [8] T. E. Mehlstäubler, K. Moldenhauer, M. Riedmann, N. Rehbein, J. Friebe, E. M. Rasel, and W. Ertmer, *Phys. Rev. A* **77**, 021402(R) (2008).
 - [9] D. N. Madsen and J. W. Thomsen, *J. Phys. B* **33**, 4981 (2000).
 - [10] T. K. Fang and T. N. Chang, *Phys. Rev. A* **61**, 052716 (2000).
 - [11] S. G. Porsev and A. Derevianko, *Phys. Rev. A* **69**, 042506 (2004).
 - [12] F. Y. Loo, A. Brusch, S. Sauge, M. Allegrini, E. Arimondo, N. Andersen, and J. W. Thomsen, *J. Opt. B: Quantum Semiclassical Opt.* **6**, 81 (2004).
 - [13] C. Y. Yang, P. Halder, O. Appel, D. Hansen, and A. Hemmerich, *Phys. Rev. A* **76**, 033418 (2007).