Coherent Population Trapping Probed by Charge Exchange Reactions

M. Gieler and F. Aumayr
Institut für Allgemeine Physik, Technische Universität Wien, Wiedner Hauptstrasse 8-10, A-1040 Wien, Austria

L. Windholz
Institut für Experimentalphysik, Technische Universität Graz, Petersgasse 16, A-8010 Graz, Austria
(Received 8 September 1992)

Coherent trapping of atomic population in sodium induced by two-frequency laser light has been studied by using a charge exchange reaction for probing the upper level population. When the two-photon resonance condition occurs, the signal due to electron capture from excited $Na^*(3p)$ atoms by He^{2+} projectile ions exhibits the characteristic "black resonance" dip, indicating (by nonoptical means) that the fraction of Na atoms in the excited state actually drops to zero.

PACS numbers: 32.80.Bx, 32.80.Pj, 34.50.Rk, 34.70.+e

The phenomenon of coherent trapping of atomic population (CT) is known to occur, e.g., in a three-level Λ atomic system, where two atomic lower levels are connected to a common upper level by two resonant laser fields [1–4]. Theoretical treatments of this situation have shown [1,3] that then two different coherent superpositions of the two lower states exist, one being coupled to the excited state by the combined laser fields, and the other being entirely decoupled although still populated by spontaneous emission from the upper level. Therefore the atomic population becomes trapped in the nonabsorbing linear combination of the lower states, and the (steady state) population of the upper state vanishes. Experimentally, CT is usually observed as a sharp frequency dip in the fluorescence light, the so-called "black resonance." Early observations of the black resonance effect were made in the late seventies by Alzetta *et al.* [1] and Gray, Whitley, and Stroud [3], and have been interpreted to be a coherence effect by Arimondo and Orriols [2]. In the last few years the CT phenomenon has regained interest since it is closely related to proposed "lasing without inversion" schemes [5], electromagnetically induced transparency [6], and laser cooling of atomic beams [7].

This Letter reports the results of an experiment testing the CT effect independently from optical means by using a charge exchange reaction for probing the upper level population. For this purpose, state-selective single electron capture into different final $He^+(n)$ projectile states in collisions of He^{2+} ions with a mixed target of ground-state $Na(3s)$ and laser-excited $Na^*(3p)$ atoms has been used. This technique—although being considerably more complicated than fluorescence detection—has the advantage of measuring the population of the upper level in a direct (but nonoptical) way and therefore allows us to distinguish between the well established quantum optics interpretation (cf. above) and the following "intuitive" but (as will be shown by our measurements) wrong picture of the coherent population trapping phenomenon: For the observation of the black resonance (vanishing spontaneous emission) one could think of another (classical) explanation. If the atom is in resonance simultaneously with a two-frequency laser field of sufficient intensity, the vanishing fluorescence could be caused by excitation into the upper level by one laser frequency and by consecutive (fast) deexcitation via stimulated emission due to the second laser frequency (before spontaneous emission and therefore fluorescence occurs) and vice versa. So we were interested in probing the upper level population directly by using collision processes, since in this case the probing time is much faster than the spontaneous lifetime (of 10^{-8} s).

The measurements have been carried out by means of translational energy spectroscopy, with the setup described in detail in [8–10]. $^4He^{2+}$ ions produced in a Nier-type ion source were accelerated to about 10 keV, magnetically mass selected, and crossed under 90° with a highly collimated Na atomic beam. The charge exchanged He^+ ions have been separated from the primary ions by a deceleration lens, and the related translational energy spectra (TES) measured by means of a hemispherical energy analyzer. In this way the involved principal reaction channels could be identified via the related energy losses or gains of the projectiles during the inelastic reaction.

In our experiment the Na atomic beam (for a detailed description see [11]) was crossed under right angle with a cw dye laser beam (typically 300 mW) single-mode output power) tuned near the Na D lines, the fluorescence signal being monitored by two photodiodes. When pumping the Na D_2 $3s\,^2S_{1/2}F_l=2 \rightarrow 3p\,^2P_{3/2}F_u=3$ transition with single-mode resonant dye laser light at around 589 nm, typically only a fraction $f^* \approx 10\%$ of the Na atoms in the target beam could be prepared in the excited Na $3p\,^2P_{3/2}F=3$ state [9–11]. In a further step an electro-optical modulator (EOM) as described by Kelly and Gallagher [12] has been used to split the single-mode laser light into a number of sidebands. The microwave frequency applied to the $LiTaO_3$ crystal in the EOM and the laser frequency have then been tuned to match one of the resulting two first-order sidebands of the laser beam

© 1992 The American Physical Society

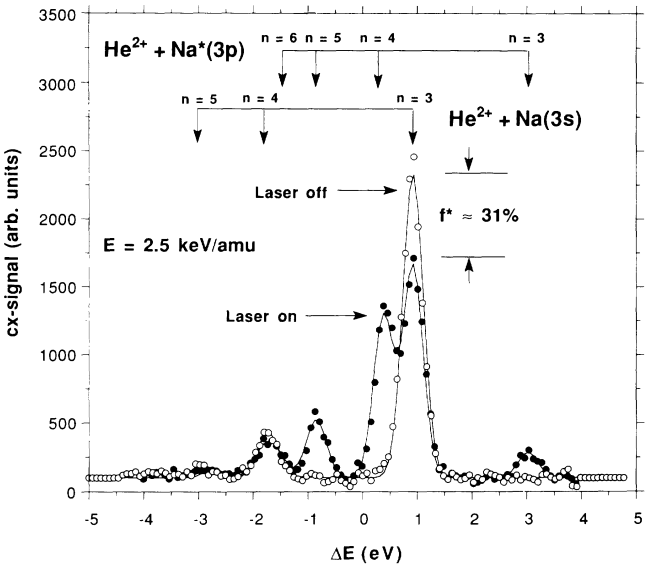

FIG. 1. Translational energy spectra (TES) for impact of 2.5 keV/amu $^4He^{2+}$ on Na [charge exchange (cx) signal] vs reaction energy defect ΔE. Comparison is made between spectra measured with a pure ground-state Na($3s$) target ("Laser off," open symbols) and a mixed target consisting of $1-f^*$ ground-state Na($3s$) and f^* excited Na*($3p$) atoms prepared with two-mode resonant laser light as described in the text ("Laser on," solid symbols). The different final $He^+(n)$ states for capture from Na($3s$) and Na*($3p$), respectively, have been indicated by vertical broken lines.

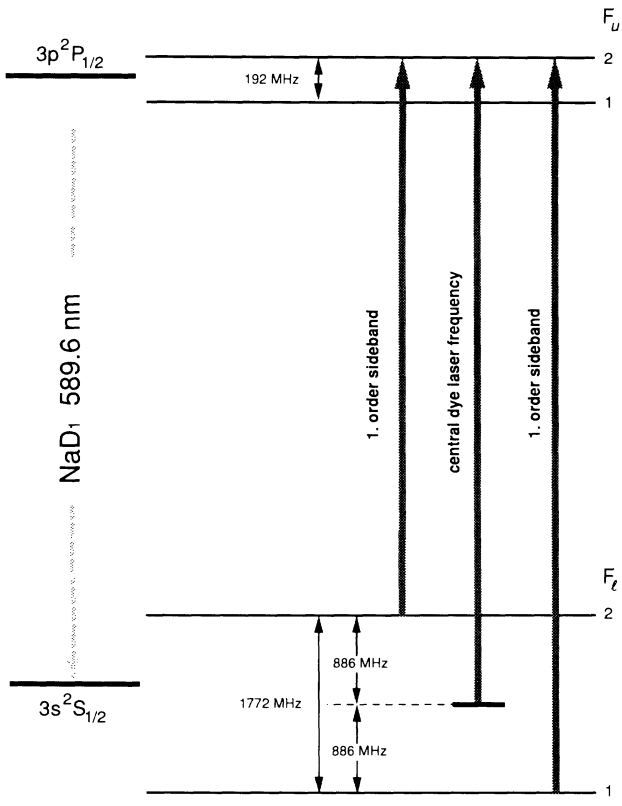

FIG. 2. Energy levels corresponding to the D_1 line of Na. Thick grey arrows indicate pump laser frequency and the two first-order sidebands produced by the electro-optical modulator (cf. text).

with the Na $3s\,^2S_{1/2}F_l=2\rightarrow 3p\,^2P_{3/2}F_u=3$ and the other one with the Na $3s\,^2S_{1/2}F_l=1\rightarrow 3p\,^2P_{3/2}F_u=2$ transition (four-level system).

Figure 1 shows a typical TES (energy resolution ≈ 0.4 eV FWHM) for 2.5 keV/amu He^{2+} impact on Na atoms which have been pumped in this four-level configuration ("laser-on" spectrum), as compared to a TES with laser off. As known from [8–10], electron capture from ground-state Na($3s$) primarily populates $He^+(n=3)$ states. Besides this main contribution, capture into $n=4$ and $n=5$ can also be identified. In the laser-on spectrum, on the one hand, additional peaks in the TE spectrum arise which can be attributed to capture from Na*($3p$) because of their exothermic energy shift of 2.1 eV with respect to the peak positions in the laser-off spectrum [9]. On the other hand, transfer of a certain fraction of Na target atoms to the excited state manifests itself in an intensity decrease of the peak at $\Delta E=+0.91$ eV, corresponding to capture from ground-state Na($3s$) into $He^+(n=3)$ only. In using the EOM to pump both hyperfine ground states of Na($3s$), the fraction f^* of excited Na*($3p$) states could thus be increased to 30% (cf. Fig. 1).

For the CT experiment the TES peak corresponding to electron capture from Na*($3p$) into $He^+(n=5)$ (reaction energy defect about -0.86 eV) was chosen to monitor the Na*($3p$) excited-state population, because in the TES this line is well separated from reaction channels involving ground-state Na($3s$) atoms (cf. Fig. 1). To obtain a three-level Λ configuration, in this experiment the Na D_1 $3s\,^2S_{1/2}F_l=1\rightarrow 3p\,^2P_{1/2}F_u=2$ and the Na $3s\,^2S_{1/2}F_l=2\rightarrow 3p\,^2P_{1/2}F_u=2$ transitions were pumped (cf. Fig. 2) by our dye-laser–EOM system (as compared to the Na $3p\,^2P_{3/2}$ states, the Na $3p\,^2P_{1/2}$ states exhibit a hyperfine splitting which exceeds both power and Doppler broadening in our experiment). The linear laser polarization could in principle be varied in the ion-beam–Na-atom-beam plane but for these experiments was kept fixed normal to the incident He^{2+} beam direction. Care was taken to compensate residual magnetic fields in the interaction region by a set of Helmholtz coils. While keeping the dye laser frequency fixed, the microwave frequency was slowly scanned around half the value of the Na($3s$) ground-state hyperfine splitting of 1771.6 MHz. At an EOM frequency of 885.8 MHz (i.e., the three-level resonance condition) our fluorescence detector exhibited the characteristic "black resonance" dip (cf. open symbols in Fig. 3). Independent evidence for the fact that the excited-state population drops to zero (while all Na atoms are trapped in the ground state) is obtained from

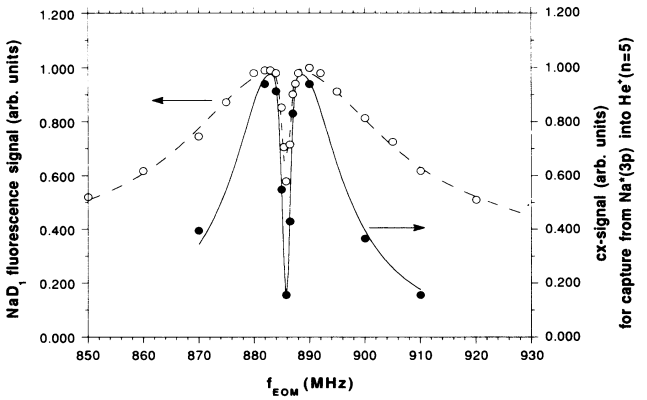

FIG. 3. Intensity of Na D_1 fluorescence and cx signals for capture into $He^+(n=5)$ from excited $Na^*(3p)$ as a function of the microwave frequency applied to the electro-optical modulator. The cx signal is proportional to the number of excited $Na^*(3p)$ atoms as seen by the probing ion beam.

the simultaneously measured charge exchanged (cx) signal (cf. solid symbols in Fig. 3). Here the black resonance effect is even more pronounced than in the fluorescence signal, which is due to the fact that the ion beam is probing just the center part of the divergent Na atomic beam while the fluorescence detector also detects light from the Doppler-shifted outer Na beam regions (the CT effect therefore being partially washed out). Other factors limiting the resolution of the black resonance are the linewidth of the involved levels and the frequency stability of the microwave generator which drives the EOM.

In conclusion, we have applied a new nonoptical method to study coherent trapping of atomic population in sodium. This method involves probing of the upper level population by means of a charge exchange reaction. Our observation, that the "black line" shape also occurs in the charge exchange signal, establishes once more the usual quantum optics model of nonabsorbing states of the sodium atom, and rules out the above-mentioned intuitive explanation for zero fluorescence intensity.

This work has been supported by Fonds zur Förderung der wissenschaftlichen Forschung (projects No. 7006 and No. 8710) and by Kommission zur Koordination der Kernfusionsforschung at the Austrian Academy of Sciences. The authors would like to thank Professor H. O. Lutz and co-workers (Universität Bielefeld) for their help in constructing the EOM and Professor H. P. Winter (TU Wien) for his valuable comments and continuous support.

[1] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento B **36**, 5 (1976).
[2] E. Arimondo and G. Orriols, Lett. Nuovo Cimento **17**, 333 (1976).
[3] H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1976).
[4] C. Mavroyannis, Quantum Opt. **4**, 109 (1992).
[5] O. A. Kocharovskaya, F. Mauri, and E. Arimondo, Opt. Commun. **84**, 393 (1991).
[6] K. J. Boller, A. Imamoglu, and S. E. Harris, Phys. Rev. Lett. **66**, 2593 (1991).
[7] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, Phys. Rev. Lett. **61**, 826 (1988).
[8] F. Aumayr, J. Schweinzer, and H. Winter, J. Phys. B **22**, 1027 (1989); J. Schweinzer and H. Winter, J. Phys. B **23**, 3881 (1990).
[9] F. Aumayr, M. Gieler, E. Unterreiter, and H. Winter, Europhys. Lett. **16**, 557 (1991).
[10] F. Aumayr, M. Gieler, J. Schweinzer, H. Winter, and J. P. Hansen, Phys. Rev. Lett. **68**, 3277 (1992).
[11] M. Gieler, F. Aumayr, M. Hütteneder, and H. Winter, J. Phys. B **24**, 4419 (1991).
[12] J. F. Kelly and A. Gallagher, Rev. Sci. Instrum. **58**, 563 (1987).

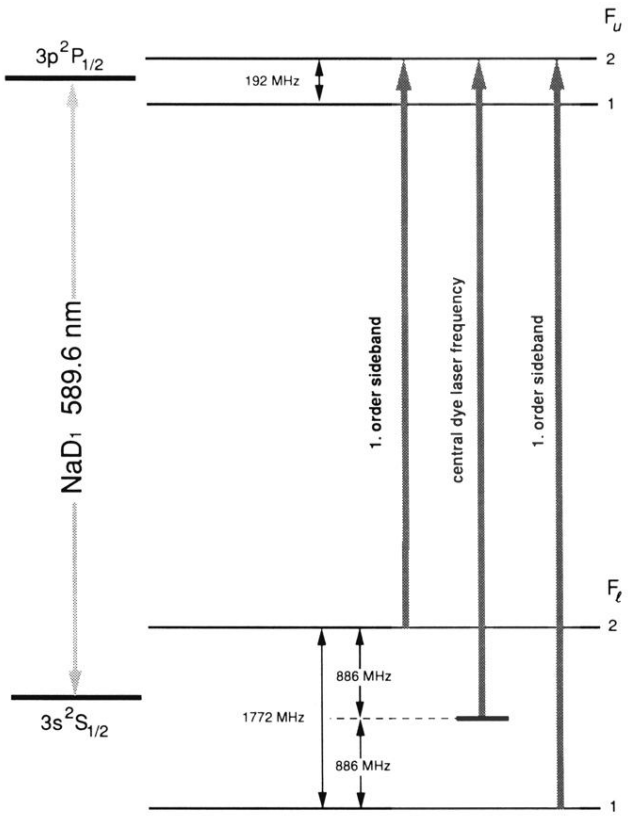

FIG. 2. Energy levels corresponding to the D_1 line of Na. Thick grey arrows indicate pump laser frequency and the two first-order sidebands produced by the electro-optical modulator (cf. text).