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Published in:
Measurement Science and Technology

DOI:
10.1088/0957-0233/2/7/007

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
1991

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Simultaneous long-time frequency stabilization of two dye lasers for a two-step atomic excitation

H A J Meijer, K Zinsmeister, T Kraft, M-W Ruf and H Hotop
Fachbereich Physik, Universität Kaiserslautern, Postfach 3049, 6750 Kaiserslautern, Federal Republic of Germany

Received 8 January 1991, accepted for publication 8 March 1991

Abstract. We describe a straightforward and accurate method for the frequency stabilization of two dye lasers on frequencies $\nu_1$ and $\nu_2$, fitting two atomic transitions from the ground state to the first electronically excited state and from there to a higher lying state. We make use of the Doppler shift by monitoring atomic fluorescence when the laser beams are crossed with an atomic beam under angles $90^\circ - \Delta \alpha$ and $90^\circ + \Delta \alpha$, respectively. The laser frequency can be stabilized to better than $\pm 1$ MHz for the first and $\pm 2$ MHz for the second transition for periods of hours. We apply this stabilization method to collision experiments between two excited Na(n) atoms.

1. Introduction

In the past few years there has been increased interest in the literature for collision processes involving electronically excited atoms, which are produced by two step (cw) laser excitation [1–8]. This development is, of course, closely connected to the availability of advanced single mode cw dye lasers. In order to get the optimum excitation ratio, i.e. the percentage of atoms actually excited to the several nl-excited states, one has to make sure that the laser frequency is tuned resonantly to the transitions continuously during the data acquisition. Only long-time exact tuning will produce the highest possible signal-to-background ratio in the experimental results, and only with exact tuning is it possible to say something quantitatively about the excitation ratios. In cases where the alignment and orientation of the excited states are also important, even continuous precise tuning of the laser becomes crucial, since both alignment and orientation are very sensitive to the laser frequencies [8]. The laser frequency tuning becomes especially critical for the second step because, generally, natural linewidths decrease for higher lying transitions.

Whereas several methods have been developed for active frequency stabilization of the first transition [9–12], up to now no work has become known to us concerning the second, except for a short remark in [6]. Actually, all experiments involving two excitation steps must rely on the intrinsic frequency stability of the laser system, with the close attention of the experimentalist, who has to interrupt the measurement for manual frequency optimization now and then (i.e. if the experiment itself offers a signal strong enough to permit laser tuning). This procedure is especially unsatisfactory in automated experimental set-ups, planned for continuous data collection for several hours. Furthermore, it causes the signal-noise ratio to be worse than necessary, it is an important source of error in the calculation of excitation ratios (and, e.g., in absolute cross sections), and can eventually wipe out polarization effects to a considerable extent. One needs, therefore, for both transitions, an active frequency stabilization system. Moreover, the system should also produce a 'laser frequency okay' signal, which inhibits the measurements in case the laser frequency is not resonant to the transition within clearly definable limits.

To achieve this goal we further developed the frequency stabilization method for one transition used by Meijer et al [10], which again was an improved version of the original system exploiting the Doppler effect used by Jitschin [9]. The most remarkable feature shared by the above mentioned systems is their 'static' stabilization character: unlike many simpler stabilization systems, which wiggle the laser frequency continuously around resonance (the 'dynamical' method), the frequency is fixed in the static systems, thus avoiding artificial broadening of the laser line width.

We used our apparatus with success for the excitation of Na atoms, in the first step from the ground state (3S_{1/2}, F=2) to the lowest excited state (3P_{3/2}, F=3, \lambda = 589 nm), and for the second step either to 3d_{5/2} (820 nm), 5s_{1/2} (616 nm), or 4d_{5/2} (569 nm). The method, however, is not at all restricted to these specific cases, nor to Na.
2. Method

The principle of the method we use is illustrated in figure 1. A well-collimated atomic beam is intersected at position (A) by the beam of the laser driving the first atomic transition. The crossing is not perpendicular, but at an angle $90^\circ - \Delta \alpha$. Then the laser is reflected by a retroreflector, so that it crosses the atomic beam again, but now at a different position and at an angle $90^\circ + \Delta \alpha$.

The resulting fluorescence intensities $I_1$ and $I_2$ of the atoms at the two places are detected independently. Atoms having velocity $v$ are in resonance with the laser at the first crossing point at a laser frequency $v_1 = v_0 \left[1 + (v/c)\sin(\Delta \alpha)\right]$, and at the second at a frequency $v_2 = v_0 \left[1 - (v/c)\sin(\Delta \alpha)\right]$. Therefore we obtain two laser-frequency-dependent absorption profiles $I_1(v)$ and $I_2(v)$, the shapes of which are identical and which can be mirrored into each other along the frequency $v_0$ of the first transition. Figure 2 shows absorption profiles calculated for a Maxwell-Boltzmann velocity distribution of the atomic beam (together with the measured ones), which show this behaviour. Clearly, at (or very close to, see section 4) the frequency $v_0$ the two profiles cross each other and the difference signal $\Delta I(v)$ has a zero crossing at this frequency. This $\Delta I(v)$ is well suited as a feedback signal for a frequency stabilization system.

The method described above is in fact a simplification of the more general case described in [10]. There it was possible to choose the two crossing angles independently by means of two mirrors, thereby enabling the choice of the zero crossing of $\Delta I$ at any frequency $v_0$ within $\pm 120$ MHz away from $v_0$. In the experiments for which we built our present system, however, there was no need for this freedom.

![Figure 1. Semi-schematic view of the apparatus. The Na oven produces two separate, well-collimated effusive beams, lying one closely above the other. At (A) the upper one is crossed with laser 1 which must be stabilized to the first transition. At (B) the lower one is used to stabilize laser 2 to the second transition. Laser 1 is also needed at (B), to produce atoms in the first excited state. The coordinate frame used throughout this paper is also shown. BS = beam splitter, Ch = chopper, D = diaphragm, F = optical fibre, IF = interference filter, M = mirror, PhT = phototransistors, R = retroreflector.](image)

![Figure 2. (a) Fluorescence signals (3p→3s, 589 nm) of the two phototransistors at (A), as a function of the frequency detuning of the first laser. The full lines are the experimental results, the broken lines are computed for an effusive beam, crossed at angles 89.6°/90.4°. (b) The difference signal of the two upper curves. Around 0 MHz the signal has a zero crossing with a steep slope, which makes this signal well-suited for feedback purposes. The window marks the 'laser frequency okay' settings.](image)

At position (B) the second transition is stabilized. Firstly, of course, we have to create atoms in the first excited state. This is done by intersecting the atomic beam again twice with the first laser, again using a retroreflector but now at 90°, in order to create the highest number of excited atoms. Then, the second laser crosses with angles $90^\circ - \Delta \beta$ and $90^\circ + \Delta \beta$ (using a third retroreflector), overlapping the atomic beam-laser beam crossing points. For the second transition we will now find similar absorption profiles as described above for the first transition. If we were able to detect only the fluorescence light coming from the higher excited atoms, we would again have a difference signal $\Delta I_2(v)$ with a zero crossing at the desired stabilization frequency $v_0$ as in figure 2. In order to separate this fluorescence from the (bright) fluorescence of the first transition, which has to be suppressed, one would need optical filters, e.g., an interference filter. In practice this is not a convenient solution. Firstly because a change in the second transition necessitates a change of optical filters, which are situated within the vacuum system of the stabilization apparatus, and secondly because normally there is no single wavelength at which the higher excited state emits: rather it 'branches' over many intermediate excited states, and selecting the fluorescence of only one transition causes considerable signal loss.

For these reasons we did not detect the fluorescence of the second transition, but rather the fluorescence decrease of the first. This decrease is caused by the depletion of the first excited level in favour of the second, and has a frequency dependence $\propto 1 - d I_1(v^2)$, where $d$ is the depletion ratio. In fact the $\Delta I_1(v^2)$ based on this has exactly the same shape as $\Delta I_2(v^2)$. The signals are
illustrated in figure 3 (again with the, now much broader, measured ones).

3. Experiment

We performed an electron spectrometric study of Na(nl)—Na(n'f) collisions in a single effusive beam (to be described elsewhere [13]). For this experiment, we applied the described stabilization method and used a separate small vacuum chamber for this goal (see figure 1). An Na-oven (T = 250 °C), having two slits of 0.5 x 3 mm (X x Y) one close above the other, produced two effusive beams (in the Z-direction). A collimating diaphragm at 90 mm distance from the oven orifice formed these beams into two well-collimated beams, one close above the other, but still separated. Doing this, we could be sure that the atoms interacting with the laser beams at position (B) were not 'reused' at position (A). In this way we avoided an interplay between the two regions. Because of the relatively low operating temperature of the oven the Na beam intensity remained constant over long periods, and could be reproduced accurately, even after several months and at two atmosphere-vacuum cycles. Furthermore, the charge of the oven, 6 g, lasts for over 2000 hours of operation.

The interaction zone (B) (see figure 1), where the lasers interacted with one of the atomic beams, was at a distance of about 145 mm from the oven orifice and interaction zone (A), where interaction took place with the other Na beam, was at about 235 mm. We estimate Na densities of $2 \times 10^5$ mm$^{-3}$ at (B) and $8 \times 10^4$ mm$^{-3}$ at (A). The mean divergence of both beams was $\varepsilon \approx 1.8$ mrad in the X-direction. Both Na beams were chopped by a home-built tuning-fork-type chopper, with a frequency of $\approx 38$ Hz. This enabled us to use lock-in techniques for the amplification and further processing of the phototransistor signals. The typical response time was 0.5 s.

The first laser was a frequency stabilized single mode linear cw dye laser (Coherent CR 599-21). A fraction of its output power was transported to the stabilization apparatus via a single mode fibre. At position (A) the light of this laser, tuned to the $F=2\rightarrow F'=3$ component of the Na($3^2S_{1/2}\rightarrow 3^2P_{3/2}$) transition (wavelength 589 nm, natural linewidth 9.7 MHz, and the $F'=2$ hyperfine component separated by 59 MHz [14-17]), was intersected at angles of $\alpha_1=89.6^\circ$ and $\alpha_2=90.4^\circ$. The laser spots had sizes of $\approx 73\mu$m, and the average irradiance was 0.4 mW/mm². Both laser beams interacted with different (central) parts of the Na beam, again to minimize mutual influence. The two crossing points were about 7 mm apart (in the Z-direction), and therefore the fluorescence light of both points could easily be detected separately by means of two BPY62 III phototransistors with a built-in lens. Typical currents were 5 nA, on a 250 nA background.

The two signals were amplified to the same peak voltage and the difference ('error') signal was then built which is well-suited for the stabilization of the laser frequency (see figure 2). The theoretical curves have been calculated according to [10, equation A8], and they are in good agreement with the measured curves. The frequency scale of the measured curves here has been calibrated to the well known hyperfine splitting frequency of the Na ground state [18]. The error signal $\Delta f$ is also shown, as well as the electronically installed voltage window of $\pm 150$ mV, which corresponds to a frequency width of $\pm 0.8$ MHz. While the error signal lies within these limits the laser is regarded to be in stabilization, and a 'frequency okay' signal is generated to be used by the data acquisition system. Of course, the error signal would be zero if there were no fluorescence at all. Therefore we also generated the sum $I_1^I$ of both signals, which must be above a certain preselected value at the same time in order to generate the 'frequency okay' signal. The frequency window could not be narrowed much further due to the intrinsic limitations of the used laser.

At position (B) the situation was somewhat more complicated because here we had to use both lasers: the second one to be stabilized on the chosen transition $3p\rightarrow nl$, and the first one to produce Na(5p). For this purpose, laser 1 was crossed with the Na beam twice under $90^\circ$. This angle could be found very accurately by comparing the frequencies at which the signal was maximal for the direct and for the reflected beam (see [10]). For practical reasons (lack of space) these beams were off the $X$–$Z$ plane by $45^\circ$.

The second laser to be stabilized on the chosen $3p\rightarrow nl$ transition was a frequency-stabilized single mode
cw ring dye laser (Coherent CR699-21). Also in this case light was split off the main beam and was directed to the stabilization apparatus by means of an optical fibre. This light crossed the Na beam at the same positions as that of laser 1, but at angles $90^\circ - \Delta \beta$ and $90^\circ + \Delta \beta$, and lying in the X-Z plane. The sizes of the laser beams were comparable to those at (A). The irradiance of the second laser was typically 0.6 mW/mm², depending somewhat on the actual transition. The value of $\Delta \beta$ depended on the frequency and the effective linewidth of the specific $3p \rightarrow n\ell$ transition. It was always chosen such that the crossing of the signals was at $\approx 2/3$ of the full modulation depth (see figure 3).

Also at point (B) we detected the fluorescence from the first excited state: $3p \rightarrow 3s$. In order to prevent the phototransistors from observing light from any other transition, a 589 nm interference filter had been installed in front of them. The phototransistor currents were similar in value to those at (A). (The transmission losses of the interference filter were compensated by the higher Na density at (B).)

The signal handling at these points was similar to that described at (A) (cf figures 2 and 3), except for three differences. The most important difference is that in this case the electronic amplification of both signals could only be used to make the maximum signal voltage equal, i.e. the signal without depletion of the 3p level. However, we also needed the modulation of both signals to be equal. This was done by deliberately misaligning the second laser for that crossing where the highest modulation depth was achieved (usually the crossing with the direct laser beam), thereby decreasing the modulation until it fitted to the other one.

The other two differences were that the logic of the sum signal had to be the other way around: a ‘frequency okay’ signal was produced only when also the sum signal $I_2$ was lower than a certain preselected value. Furthermore, the second stabilization unit was only allowed to send its error signal to the second laser if the first laser had ‘frequency okay’ (so-called ‘master–slave’ arrangement).

We stabilized the second laser to three different transitions. The modulation of the signal, i.e. the percentage with which the $3p \rightarrow 3s$ signal could be reduced obviously depended very much on which transition was chosen. In our case of Na the modulation is not only caused by part of the 3p population being transferred to the higher excited level but also (and in some cases mainly) by the higher excited state also being able to decay to the $F = 1$ hyperfine ground state of Na, from where the atoms can no longer be excited (the so-called trapping effect, see e.g. [19–21]). In the first case we used the transition $3^2P_{3/2}, F = 3 \rightarrow 3^2D_{3/2}, F = 4(3,2)$ (wavelength 820 nm, natural linewidth 8.1 MHz, the hyperfine splitting much smaller than the natural linewidth [22]). The maximum obtainable modulation was well over 50%, which clearly shows the influence of trapping. However, this was not a preferable setting, since the trapping effect causes a considerable broadening of the depletion profile, which makes the $\Delta I$ feedback signal less pronounced and thus the method less accurate. In figure 3 one can see that with smaller modulations the experimental curves are also broader than the theoretical ones. The frequency scale here has been calibrated to the fine structure splitting $3^2D_{5/2} - 3^2D_{3/2}$ [22]. The crossing angles were 86.6° and 91.4°. The voltage window of ±35 mV corresponds to ±14 MHz. Here, as in all cases for the second-step stabilization, signal jitter rather than intrinsic laser stability was the limiting factor.

We also stabilized on the transition $3^2P_{3/2}, F = 3 \rightarrow 4^2D_{3/2}, F = 4(3,2)$ (wavelength 569 nm, natural linewidth 3.0 MHz, again with the hyperfine splitting smaller than the natural linewidth [24,25]). Here we reached a modulation depth of $\approx 45\%$, and a frequency window of ±16 MHz, for crossing angles 91.1° and 88.9°.

Finally, figure 4 shows the measured and computed results for the $3^2P_{3/2}, F = 3 \rightarrow 5^2S_{1/2}, F = 2$ transition (616 nm, natural linewidth 2.0 MHz, the transition to the $F = 1$ component ($\approx 150$ MHz away [26]) is forbidden). One again sees the broadening due to the trapping effect in the experimental signals. The experimental frequency scale was assumed to be the same as in the nearby 4D case (which was calibrated to the fine splitting $4^2D_{5/2} - 4^2D_{3/2}$ [24]). The crossing angles were 91.0° and 89.0°. The stabilization was somewhat less accurate, because of the low modulation of only 20%. The window corresponds to ±2.2 MHz. The low modulation is caused by the less effective pumping scheme and the longer lifetime of the 5s state.

4. Evaluation

We have used the apparatus with great success during two months of experiments on Na(nl)–Na(n'l') collisions. The first laser was stable within the required + Note that in our theoretical description we include Doppler and saturation broadening effects, but we assume the two transitions to be independent; i.e. we neglect the Autler–Townes effect (see e.g. [23]). This is still a good assumption at the laser irradiances we have used.
frequency window almost 100% of the time. By means of the error signal we could even trace the main stability-disturbing instrument to be a roughing pump of an adjacent apparatus. The stability of the second laser was also very satisfactory, with 'laser frequency okay' times from >90% for the 'easy' 3p→3d transition to ≥70% for the 'difficult' 3p→5s transition.

The achieved actual accuracy of the stabilized laser frequency was limited by several effects: (i) the limited accuracy of the used retroreflectors, (ii) the differences in irradiance between the direct and the reflected laser beam, causing the absorption profile at the reflected laser intersection to be less saturation broadened, (iii) the limited accuracy in the experimental normalization of the phototransistor signals, (iv) possible long-time drifts in the electronics (offset and gain), (v) the depletion of the Na(3p) level caused by the mentioned trapping effect, and, finally, (vi) the non-linearity of the phototransistor current as a function of the detected light irradiance. All possible errors and drifts in the laser frequency must be compared with the saturation-broadened natural linewidths for the transitions as they occurred in the main experiment, to the width of the 'laser frequency okay' window and to the intrinsic, short-time frequency width of the dye lasers (0.5−1 MHz FWHM).

(i) The maximum inaccuracy of the retroreflectors, as stated by the manufacturer, is 30", leading to a maximum frequency asymmetry of less than 0.2 MHz.

(ii) The difference in irradiance between direct and reflected beam was 30−40%, caused by losses in passing through the vacuum window twice and by three reflections within the retroreflector (we used a type consisting of three mirrors). Furthermore, the irradiance in the reflected spot was lower by another 25% because of the laser beam divergence (a change from ∅23 mm to ∅3.5 mm causes this 25% loss). For the theoretical curves in figure 2 the irradiance of the reflected beam was 40% of that of the direct one, leading to widths of 23.1 and 19.2 MHz (FWHM), agreeing well with the measurements. This difference caused a systematic error in the stabilization frequency of 0.6 MHz. For the stabilization frequency of laser 2 this value varied between 0.5 and 0.7 MHz for the measured and calculated profiles.

(iii) If we assume a realistic value for the normalization error of both signals of ±2%, we find a typical error in the first transition frequency of ±0.15 MHz, and for the second transition about ±0.5 MHz.

(iv) The stability in offset and gain required is typically <5 mV/hour and <0.3%/hour, respectively. Both numbers are easy to achieve with standard electronics.

(v) For the second transition we stabilized to the maximum depletion of the 3p level, which for the specific case of Na, due to the hyperfine splitting of the 3d and 4d levels, does not necessarily correspond exactly to the maximum population of the higher excited level. However, to compute possible deviations quantitatively goes beyond the scope of this paper, and the effect is most probably very small (the hyperfine splittings are about 0.5 and 0.15 MHz for 3d and 4d, respectively [22,24]).

(vi) Phototransistors generally are non-linear devices. However, in our experiment abundant background light was present from all the laser light reflections, and from fluorescence light caused by the relatively high Na background vapour pressure. This caused a 250 nA background current compared with the 5 nA signal. Correspondingly a signal on−off change only amounts to a 2% signal change of the phototransistor. Within this small range of sensitivity we expect the non-linearity effect to be negligible for the phototransistor type in use.

Table 1 gives an overview of the possible error sources, together with the estimated maximum deviation they can cause, as well as with the 'frequency okay' window and the saturation broadened linewidths of the main experiment with which they have to be compared. The results are very satisfactory.

Even better, we had a very simple external frequency checking criterion: the signals (fluorescence and ionization) in our main experiment must be maximum. In the main experiment, the two laser beams overlapped very precisely, and the crossing angle with the Na beam was (90.00±0.03)°.

For the check and optimization of the first laser frequency we used the fluorescence signal from a photodiode in the main experiment. We 'scanned' the stabilization frequency by gradually moving the zero-crossing of the ΔI signal from ≈90% of its left peak value to ≈90% of its right, by means of the offset voltages of the lock-ins. This gave a scan range of ±6 MHz around

<table>
<thead>
<tr>
<th>Transition</th>
<th>Systematic Error</th>
<th>Random Error</th>
<th>Hyperfine Interaction</th>
</tr>
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<tbody>
<tr>
<td>3p</td>
<td>&lt;0.2</td>
<td>±0.5</td>
<td>&lt;0.1</td>
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<tr>
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<td>&lt;0.1</td>
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<tr>
<td>4d</td>
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<td>±0.6</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>5s</td>
<td>&lt;0.2</td>
<td>±0.6</td>
<td>&lt;0.1</td>
</tr>
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Table 1. Possible error sources (systematic and random) for the stabilization frequency, compared with the 'laser frequency okay' window, and the saturation broadened width of the absorption profile in the main experiment. The last column shows the accuracy, with which the stabilization frequency could be checked in the main experiment (all values in MHz). (i) = inaccuracy of the retroreflectors, (ii) = difference in irradiance between direct and reflected laser beam, (iii) = signal normalization error, (v) = hyperfine interaction (cf section 4).
the actual stabilization frequency. We found every time that the correct setting of the stabilization apparatus (both offset voltages zero) gave the maximum fluorescence signal in the main experiment within its error bars. This check had an estimated accuracy of ±1.1 MHz, limited by the accuracy of the fluorescence signal from the main experiment (0.7%). The optimization of the second transition frequency has been done in the same way, except that in this case we had to maximize the electron spectrometer signal of an ionization reaction between Na(nl) atoms. This check on the signal had a similar accuracy to the check on the fluorescence, and here the correct setting of the stabilization apparatus also produced the maximum count rates in all cases within the frequency error bars of ±1.4–±2.1 MHz (see table 1). For the 4d and the 5s case we should in principle have been able to see fluctuations in the signal due to the fact that the frequency okay windows were larger than the frequency error bars in the main experiment. However, these differences were only marginal and such small fluctuations in the signal could just as well have been caused, for example, by Na beam density fluctuations. Furthermore, the window frequency size is not the long-time laser frequency bandwidth, but just an electronic switching point. The actual bandwidth of the laser during ‘laser okay’ was at least a factor of two smaller.

5. Conclusions

We succeeded in stabilizing two dye lasers for a two-step transition accurately and for periods of hours. The accuracy of the frequency for the first step was limited by intrinsic laser properties. The frequency accuracy for the second step was limited by the error signal quality. This quality can be improved by increasing the Na density and by reducing the laser irradiance, thereby reducing the saturation and ‘depletion’ broadening and producing a Δf signal with a steeper slope. The stabilization system is not at all restricted to Na, nor to dye lasers. The demands put upon the (cw) laser are a satisfactory intrinsic short time (<1 s) frequency stability and an external frequency scan input. Instead of Na, the system can be used for all species of which a beam can be made with densities similar to those we have been using here, and of which the fluorescence of the first transition is observable (preferably with small detectors like the phototransistors used here). Apart from the alkali and several alkaline earth atoms the system will probably also work for the excitation of metastable noble gas atoms (e.g. Ne*, Ar*, K* (3P_4, 3P_3)), for which, densities a factor of 4–10 smaller than we used with Na can be obtained [27,28], but which, on the other hand, are saturated already at low irradiances and show no trapping effects caused by hyperfine splitting. With some care the system will most probably also work in cases where the highly excited level only contains a few percent of all atoms, as is the case for excitation of Rydberg levels. Then, one could think of pumping the first excitation only weakly, such that the modulation depth is still substantial. Furthermore, the development of a similar system for use with state-selective, vibrational laser excitation in molecules [e.g. 29] is also feasible.

The apparatus is constructed such that instead of using it for stabilizing two transitions of one atomic species, two different atom sources can be installed, for both of which the first transition is stabilized. This option will be very useful in planned collision experiments between two different excited alkalis, in which we plan to measure the electron spectra in coincidence with a mass-spectrum.

Finally, the apparatus as such has also shown its use for performing pure fluorescence measurements. We have measured the effectiveness of the use of an electro-optical modulator, which turned our first dye laser into a two-mode laser, eliminating the trap in Na caused by the hyperfine splitting [30]. We found an increase of over a factor of two in the population of the 3p level (although the transition was still far from saturation), and even a factor of over 5 for the 3d_{5/2} level. By means of so-called saturation curve measurements we could determine the absolute excitation ratios rather accurately. Results will be published elsewhere [31].

Acknowledgments

We would like to thank F Ditewig and C J Wisman of the University of Utrecht for their kind help in supplying the electronics for the laser stabilization and the chopper system.

Financial support by the Deutsche Forschungsgemeinschaft through Sonderforschungsbereich 91 is gratefully acknowledged.

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