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14. High-Resolution Spectroscopy at Short Wavelengths Using Pulsed Dye Lasers

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With 5 Figures

The invention of the dye laser [14.1, 2] 25 years ago has brought about an enormous development of optical spectroscopy (for reviews see, e.g., [14.3, 4]). The tunability and high spectral intensity of these sources in combination with the possibility to achieve extremely narrow bandwidths for cw lasers, or pulses of extremely short duration, have greatly facilitated many spectroscopic investigations and made entirely new types of experiments possible.

In the present chapter we will discuss high-resolution laser spectroscopy of free atoms and especially focus on the UV and VUV wavelength region. (For a more detailed discussion of this topic we refer to [14.5]). Dye lasers do not operate at such wavelengths, but their radiation can be frequency converted to this spectral region using nonlinear optics techniques. Such conversion can most conveniently be performed with pulsed lasers, that, however, necessarily have a much larger linewidth than the single-mode cw systems available at longer wavelengths.

The topic of this chapter is primarily to discuss how a resolution, limited only by the Heisenberg uncertainty relation, can still be obtained by combining intense, broadband excitation with "classical" high-resolution techniques such as optical double-resonance (ODR), level-crossing (LC) and quantum-beat (QB) spectroscopy. Such techniques proved very useful at an earlier stage of development of dye lasers. At the beginning of the 1970s only multi-mode cw lasers were available and in a very limited wavelength range. Such lasers, used for stepwise excitations and combined with ODR and LC spectroscopy, allowed a substantial extension of high-resolution alkali-atom spectroscopy (see e.g. [14.6, 7]). At the same time, QB spectroscopy was shown to effectively extend the wavelength range of high-resolution laser spectroscopy [14.8].

While awaiting the development of broadly tunable, narrow-band cw laser sources allowing the full utilization of Doppler-free laser spectroscopy methods using collimated atomic or ionic beams, spectral holeburning, two-photon absorption and cooled atoms or ions in traps, the ODR, LC and QB methods can provide precision spectroscopic data on atoms and ions. However, it must be remembered that these resonance and coherence methods are only useful for the measurement of level *splittings* due to, e.g., fine and hyperfine interactions, Zeeman and Stark effects. Measurements of isotopic shifts, scalar Stark interaction, and of course investigations of the absolute wavelength/frequency of the optical transition, still require the narrow-band laser sources. For selected wavelengths such radiation can be achieved, e.g., for Lamb-shift/Rydberg

constant measurements or for optical frequency standards (see [14.5] for references).

14.1 Generation of UV/VUV Radiation

A variety of methods is available for the generation of short wavelength coherent radiation. Most commonly tunable radiation in the UV or VUV spectral region is generated using nonlinear optics frequency shifting of dye laser radiation.

14.1.1 Sum-Frequency Generation in Crystals

Frequency doubling or mixing in non-linear crystals is an efficient way to generate radiation in regions where the crystals are transparent and phase-matching conditions can be obtained. A review of modern materials and techniques can be found in [14.9]. Of particular importance is β-barium borate (BBO), which provides frequency doubling down to 205 nm. Excimer-pumped blue dye lasers can very efficiently be doubled in this way. Frequency tripling of red dye-laser radiation, by first doubling in KDP and subsequent mixing with the residual fundamental in BBO, can be performed with high efficiency with a frequency cut-off at 197 nm [14.10]. We have been able to generate almost 10 mJ of tunable radiation in the region around 200 nm using the radiation from a Nd:YAG pumped dye laser.

14.1.2 Frequency Conversion in Gases

Gases are transparent at shorter wavelengths than crystals and different non-linear techniques can be used to attain VUV radiation. A simple technique not requiring phase matching is stimulated Raman scattering in high-pressure H_2 gas [14.11]. By the generation of successively higher anti-Stokes components, a photon energy increase of $4155 \, cm^{-1}$ or $\approx 0.5 \, eV$ can be gained in each step. This means that, using a primary laser wavelength of 200 nm, the first anti-Stokes Raman component will be at 185 nm and the second anti-Stokes component at 170 nm.

Sum and difference frequency mixing can be performed in noble gases such as Kr and Xe, and also in metal vapours such as Hg and Mg. The efficiency is greatly improved if resonance enhancement through two-photon resonances in the gas can be obtained. For sum frequency mixing, phase matching must be achieved. The index of refraction of the gas can be manipulated by adding noble gas to the converting metal vapour. Most of the range 100–200 nm can be covered in this way. Even shorter wavelengths can be achieved by high (odd) harmonic generation in the noble gases. The field of non-linear frequency conversion in gases is covered in [14.12–14].

Recently, very high odd overtones have been generated in the interaction of intense laser radiation with noble gases [14.15, 16]. The highest overtone generated so far is the 97th (8.3 nm) of a titanium–sapphire solid-state laser operating at about 807 nm [14.17]. In Lund, a tunable titanium–sapphire terawatt laser system suitable for high overtone generation is now being assembled.

14.2 High-Resolution Laser Spectroscopy Using Pulsed Dye Lasers

We will now illustrate how pulsed dye laser radiation at short wavelengths can be combined with ODR, LC and QB spectroscopy to produce precision data on radiative lifetimes, Landé g_J factors and hyperfine interaction constants. The three methods will be illustrated by recent work at the Lund Institute of Technology, with examples from silver and ytterbium. Finally, some hybrid experiments on nitrogen atoms that are produced by photodissociation of parent molecules using the resonant spectroscopic laser pulse are described.

14.2.1 Optical Double Resonance Experiments

In the ODR method [14.18–20] polarised light is used to excite the different substates of the excited atomic level, and the decay exhibits a spatial anisotropy and certain polarisation properties. By inducing radio-frequency transitions between the sublevels the population differences are leveled out and the angular and polarisation pattern changes. A sufficiently high rf amplitude is needed to bring about the transfer during the limited time available in the short-lived excited state. The signal linewidth directly reflects the level broadening due to the uncertainty relation. A useful experimental set up for pulsed ODR experiments is described in [14.21], discussing measurements on the 6p ^{2}P states of Ag reachable with 206 nm radiation. A diagram of the magnetic sublevels of the 6p ^{2}P$_{3/2}$ state is shown in the upper part of Fig. 14.1. Silver has two stable isotopes, ^{107}Ag and ^{109}Ag, both with nuclear spin $I = 1/2$. In the Paschen–Back region $2I + 1$ rf signals are expected, symmetrically around the position for the spin-zero case. Such signals are shown in the lower left part of Fig. 14.1, also featuring the narrowing down of the signals when detection is restricted to "old" atoms, i.e. when the detection system is switched on after a certain delay. Recordings of this kind yielded $a = -9.05(25)$ MHz for the magnetic dipole interaction constant and $g_J = 1.336(2)$ for the Landé factor [14.21].

14.2.2 Level-Crossing Measurements

The LC method [14.20] originates in *Hanle*'s early work on magnetic depolaris-ation [14.22] which was extended to the case of high-field crossings by *Colgrove* et al. [14.23]. Using σ-light, pairs of crossing magnetic sublevels can be
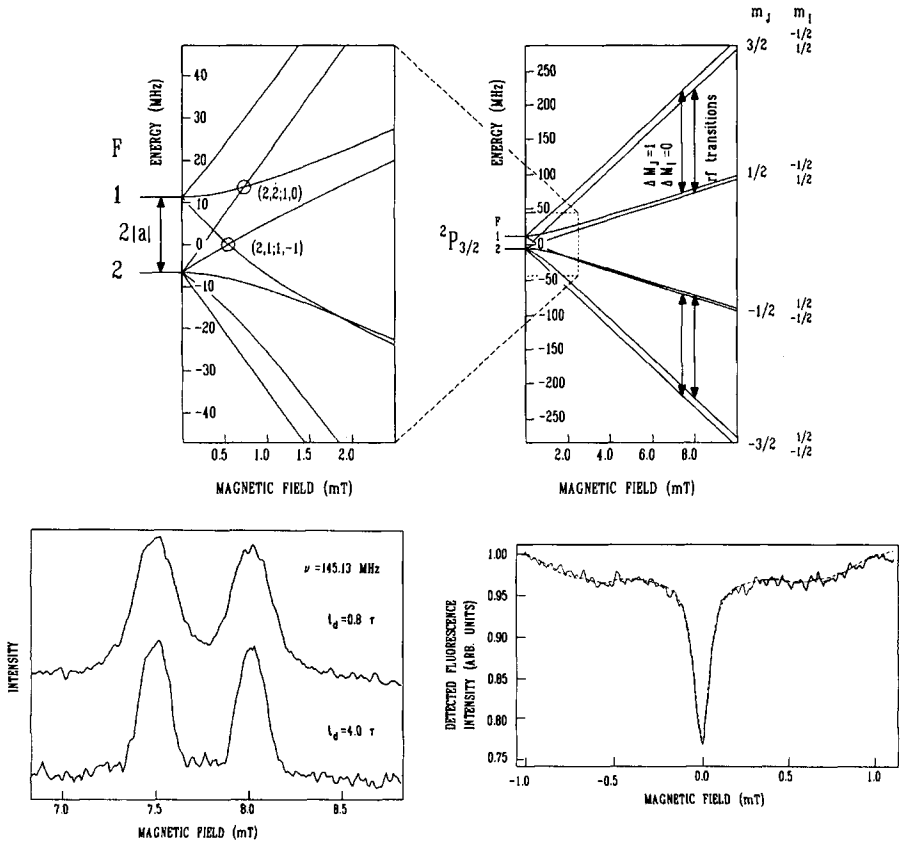
Fig. 14.1. Magnetic sublevel diagram for the 6p $^2P_{3/2}$ state of Ag (top); ODR signals (lower left); LC signal (lower right). From [14.21]

coherently excited and a redistribution in the fluorescence light can be detected that is related to the magnetic fields of level crossing. In Fig. 14.1 (top), detectable level crossings for the silver 6p $^2P_{3/2}$ state are indicated. A recording of the fluorescence light intensity as a function of an external magnetic field is included in this figure. Apart from the zero-field level crossing (the Hanle effect) the unresolved structure due to two level crossings for each of the two silver isotopes is shown. This recording was again taken for 206 nm excitation and basically yields similar information on hyperfine structure as the ODR recording in the same figure. In a previous investigation, resolved level-crossing signals were recorded for the 5p $^2P_{3/2}$ state of copper [14.24] that was excited at 202 nm.

14.2.3 Quantum-Beat Measurements

Quantum-beat experiments [14.25, 26] can be considered as time-resolved LC investigations. Following short-pulse coherent excitation the quantum-mechanical interference between pairs of sublevels is manifested as oscillations superimposed on the temporal decay of the fluorescence light. An experimental set-up for QB experiments with VUV laser light excitation is shown in Fig. 14.2 [14.27]. A hyperfine structure QB recording for the 7p $^2P_{3/2}$ state of silver, excited at 185 nm is shown in Fig. 14.3. By frequency tripling of the output from
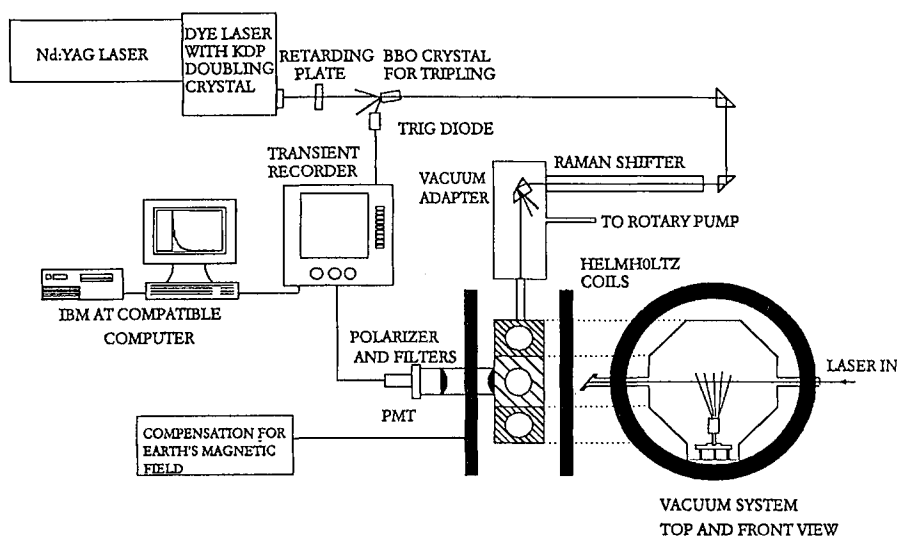
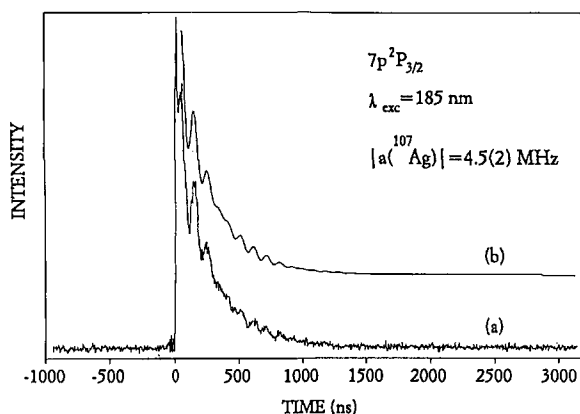**Fig. 14.2.** Experimental set-up for QB spectroscopy in the YUV region. From [14.27]

Fig. 14.3. Experimental QB recording of the structure of the 7p $^2P_{3/2}$ state of Ag. A theoretical fit to the data is also included. From [14.27]
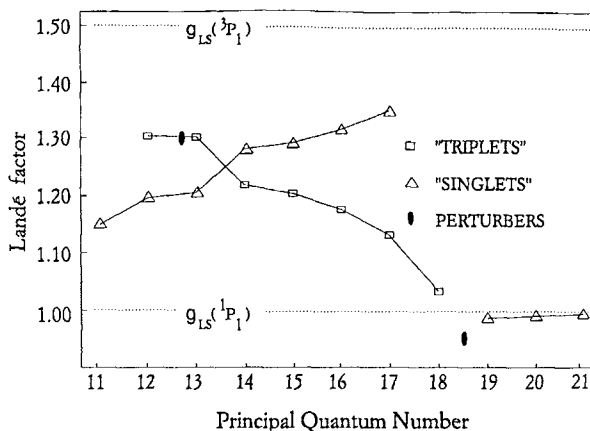
Fig. 14.4 Values of g_J for np $^{1,3}P_1$ sequences in Yb, obtained using Zeeman quantum-beat experiments. From [14.28]

a red dye laser, 200 nm was first achieved. By anti-Stokes Raman shifting the required 185 nm radiation was then generated. Because of the oxygen Schumann–Runge band absorption the Raman shifter assembly is evacuated and is directly attached to the atomic-beam vacuum chamber. In the figure a theoretically generated QB curve using a magnetic dipole interaction constant a (^{107}Ag) $= -4.5$ MHz is included. This recording provides a good example on how a very high resolution can be achieved in the VUV region using broadband pulsed lasers in combination with resonance/coherence techniques.

As a further example of QB spectroscopy at short wavelengths, we choose Zeeman quantum beat measurements in highly excited ytterbium Rydberg state. From the beat frequency and the applied magnetic field value, the Landé g_J factor of the state investigated can be evaluated. A systematic study of the Zeeman effect in the np $^{1,3}P_1$ Rydberg sequences of ytterbium has been performed ($\lambda_{exc} = 207-200$ nm) and results are given in Fig. 14.4 [14.28]. In this way it is possible to study the influence of perturbing states in a similar way as previously done for the barium atom [14.29].

14.2.4 Time-Resolved Studies on Atoms Formed by Short-Wavelength Dissociation

Many of the light non-metallic elements occur naturally only as molecules. Furthermore, the resonance lines frequently fall in the deep VUV region. Both these problems can be overcome by photodissociation of molecules by short-wavelength laser radiation that is two-photon resonant with transitions in the produced free atoms. Following a study of excited-state lifetimes in oxygen atoms produced from NO_2 molecules [14.30], we have just finished corresponding studies for nitrogen atoms formed from N_2O [14.31, 32]. In Fig. 14.5 a
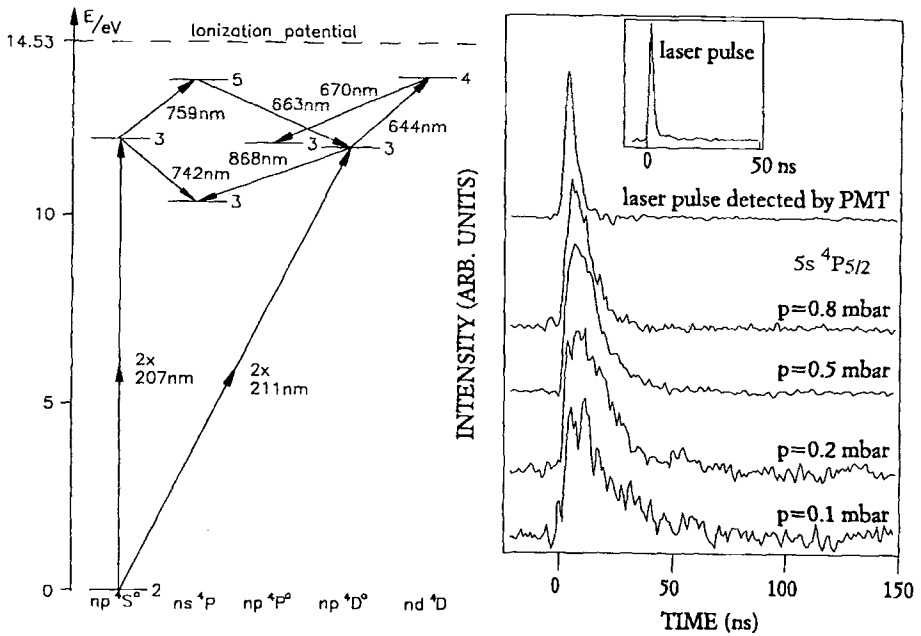
Fig. 14.5. Partial energy-level diagram for nitrogen and fluorescence decay of nitrogen atoms following step-wise excitations. From 14.32]

partial energy-level diagram for nitrogen and recordings of fluorescence light at different gas pressures following stepwise laser excitations are shown. A similar scheme can be used for many other light atoms such as C, F, Cl, P and S. Using resonance cells with molecular species enriched in odd isotopes high-resolution data on hyperfine structure can also be obtained.

14.3 Conclusion

As illustrated in the present chapter, high-resolution laser spectroscopy can readily be performed at "difficult" wavelengths combining pulsed laser radiation with resonance/coherence techniques. Basically, the ODR, LC and QB techniques provide the same type of information and the choice of method will be determined by practical aspects. QB spectroscopy requires a high time resolution on both the laser and the detection electronics. In the ODR technique, rf fields of suitable frequency must be available and at sufficient strength, which can be a limitation for short-lived states. In many respects the level-crossing method is the least demanding technique.

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