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Atomic spectroscopy by resonance scattering

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Resonance scattering techniques are very useful for high-resolution atomic spectroscopy. The applicability of these techniques has been much extended, particularly through the rapid development of tunable-laser technology. The use of a narrow-band tunable laser, acting on a collimated atomic beam, gives a direct method enabling, for example, hyperfine structure and isotope shift studies. The intensity of lasers allows stepwise excitations to be performed, and with the two-photon absorption technique, Doppler-free measurements on thermal gases are also possible. By using pulsed lasers, time-resolved measurements yielding radiative life-times and structural information can be performed. The basic resonance scattering methods can be combined with radiofrequency and coherence techniques to yield a resolution, limited only by the uncertainty relation. Optical double resonance and level-crossing techniques, not requiring a narrow-band light source, have been extensively used. Several examples of the application of resonance scattering methods are given.

1. Introduction

For a long time, resonance scattering techniques have been used to obtain information on the energy structure and radiative properties of free atoms. Although normal emission spectroscopy has been extremely useful in the acquisition of our basic understanding of atomic systems, and still is very important in the study of, for example, highly ionized atoms, resonance scattering spectroscopy offers advantages in many cases. The Doppler broadening of the spectral lines, emitted by normal atomic light sources, often prevents the study of features like fine and hyperfine structure splittings, Stark splittings and isotope shifts. In contrast, optical resonance methods and laser–spectroscopic methods, incorporating the concept of resonance scattering, yield a resolution, essentially limited only by the Heisenberg uncertainty relation. Optical resonance methods, like optical pumping, optical double resonance and level-crossing spectroscopy, were introduced during the 1950s and have enabled much precision work. The methods and results have been discussed in several review articles (Budick 1967; zu Putlitz 1969; Happer 1972; Arimondo *et al.* 1977). The rapid progress in tunable-laser technology has enabled an enormous development to take place in the field of laser spectroscopy. Several new Doppler-free techniques have been introduced, and older techniques have become much more powerful through the application of laser light sources. Laser spectroscopy allows not only a very high spectral resolution, which is of great importance to the spectroscopist, but also very strong signals, which previously were generally incompatible with a good resolution. Several reviews of the new field of laser spectroscopy are available (Walther 1976; Shimoda 1976; Letokhov & Chebotaev 1977).

In the following, different kinds of resonance scattering techniques for studies of atomic systems are briefly discussed. Clearly the field is very vast, and for the convenience of the author, examples are chosen from studies performed mainly at Chalmers University of Technology. For a full account of the work performed in this fascinating field of research we refer to the excellent reviews already mentioned.

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2. PRINCIPLES OF RESONANCE SCATTERING

We will discuss the basic principles of resonance scattering using the specific example of the cesium atom. In figure 1 an energy level diagram for this atom is given. With a single outer electron, sequences of S, P, D, F... doublets are obtained. In a Cs light source, the different energy levels are populated, e.g. through collisional processes, and in the decay of these levels the Cs spectral lines are emitted in electric dipole transitions, according to the quantum
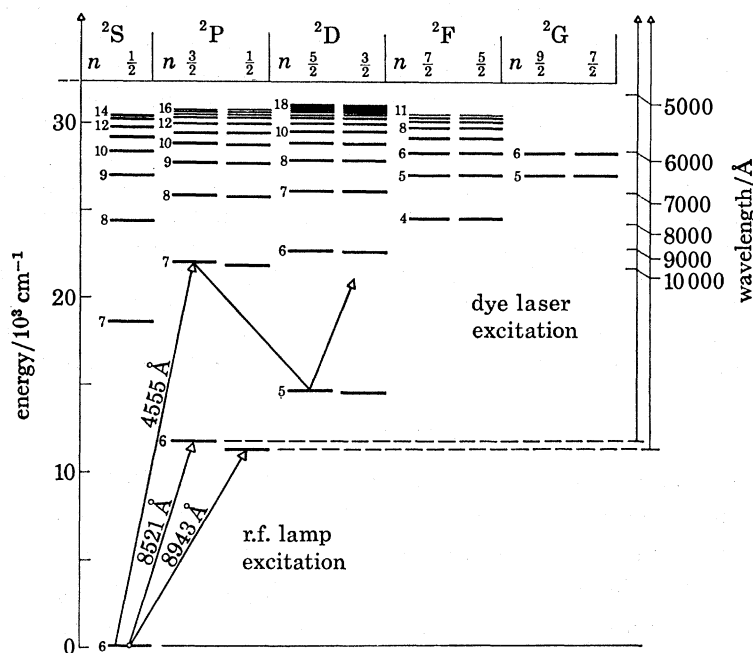

FIGURE 1. Energy level diagram for the Cs atom. Stepwise excitation schemes using the first or second excited ^{2}P state as an intermediate level are indicated.

mechanical selection rules. Spectral lines, emitted in the visible region, have a Doppler linewidth of about 0.01 Å† or 1 GHz. In a Cs vapour cell, almost all atoms are in the ground $6\,^2S_{\frac{1}{2}}$ state. Atoms can be excited to higher P states by resonance absorption of photons of the right frequency, produced, for example, in a Cs radiofrequency discharge lamp. Only P states can be populated because of the selection rules. The excited state will decay exponentially with a typical time constant, the natural radiative lifetime τ. The $6\,^2P_{\frac{3}{2}}$ state, excited by 8521Å light, can only decay back to the $6\,^2S_{\frac{1}{2}}$ state emitting *resonance* radiation at the same wavelength. This absorption–emission process is called *resonance scattering*. The next higher $^2P_{\frac{3}{2}}$ state can be excited by absorption at 4555 Å and resonance radiation of the same wavelength is obtained in the decay. In addition, this state can decay, e.g. to the $5\,^2D_{\frac{5}{2}}$ state. The light emitted at a longer wavelength is called *fluorescence* light.

When discussing resonance scattering we will here use a generalized definition that includes all processes where atoms are transferred to higher energy states and subsequently decay to lower-lying levels, in every case through allowed electric dipole transitions. The processes can be stepwise both in absorption and emission. While the decay can naturally follow in many steps by cascading, stepwise excitation has become possible, mainly through the use of intense

† 1 Å = 10^{-10} m = 10^{-1} nm.

laser sources tuned to the correct frequencies. For the specific case of Cs, a radiofrequency lamp is adequate to excite the first P state using the very strong $6\,^2S_{\frac{1}{2}}$–$6\,^2P_{\frac{3}{2},\frac{1}{2}}$ transitions. The state has a lifetime of only 30 ns, but with a c.w. dye laser it is possible to excite a substantial number of atoms further, to a highly excited S or D state. If the laser is tuned to 5414 Å, the $12\,^2D_{\frac{5}{2}}$ state will be populated in transitions from the $6\,^2P_{\frac{3}{2}}$ state. In the radiative decay of this state, a large number of lines will be emitted in the cascade down to the ground state. In figure 2 the fluorescence spectrum following this selective excitation is shown. With the Cs atoms contained in a cell, the absorption lines, as well as the emission lines, will be Doppler-broadened. We will now discuss the high-resolution techniques briefly mentioned in the introduction, by which the Doppler broadening problems can be eliminated.
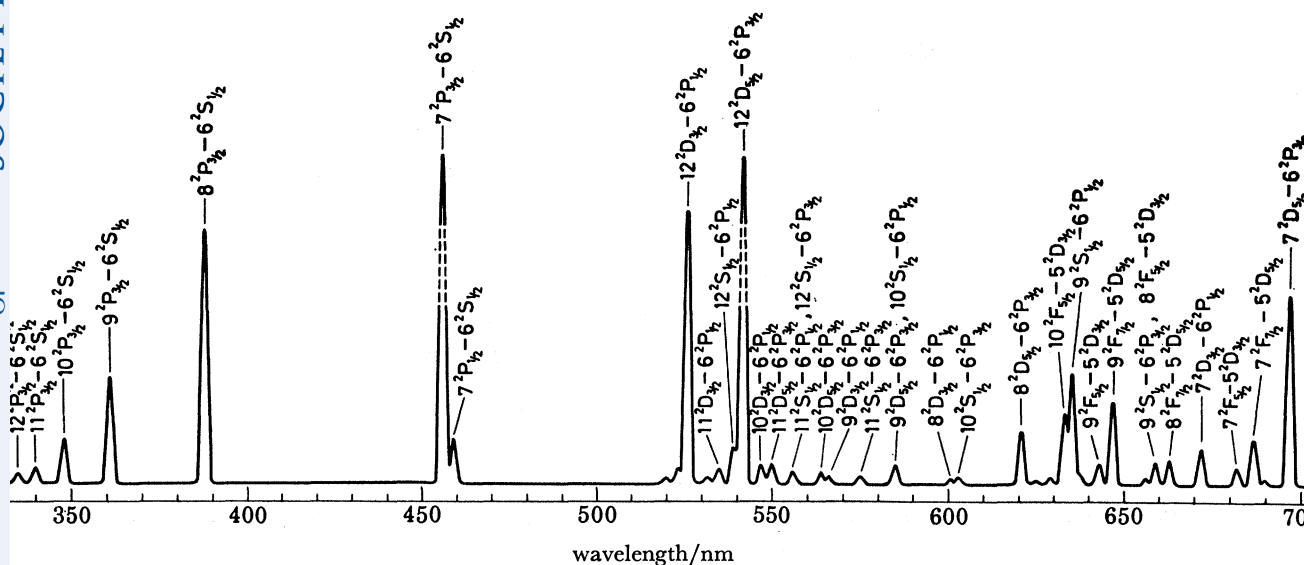

FIGURE 2. Fluorescence-light spectrum obtained in the decay of the selectively excited $12\,^2D_{\frac{5}{2}}$ state of Cs, the excitation being $6\frac{1}{2}\,^2S \to 6\,^2P_{\frac{3}{2}} \to 12\,^2D_{\frac{5}{2}}$. Some of the lines occur because of nearly resonant collision processes (after Belin *et al.* 1976).

3. COLLIMATED-ATOMIC-BEAM SPECTROSCOPY

A particularly straightforward type of laser spectroscopy is the use of a well collimated atomic beam in conjunction with a narrow-band laser beam, incident at right angles. Commercial single-mode dye lasers with a linewidth of about 1 MHz are available. By using a sufficiently small collimation ratio for the atomic beam, the linewidth will be given by the natural radiation width $\Delta\nu_N = 1/2\pi\tau$ of the excited state. The recording of hyperfine structure in the sodium D_2 line, shown in figure 3 was obtained by using this technique. We observed the resonance light released when the laser was successively tuned to the individual hyperfine transitions, shown in the inserted energy level diagram. The observed linewidth of 12 MHz is only slightly larger than the 10 MHz natural width, corresponding to the 16 ns lifetime of the $3\,^2P_{\frac{3}{2}}$ state. This Na transition corresponds to the 8521 Å line of Cs indicated in figure 1.

The technique with collimated atomic beams can also be used in conjunction with stepwise excitations. Recently, we have performed an investigation of fine-structure splittings in highly excited Cs F states (Fredriksson *et al.* 1978). In figure 1 the excitation scheme used in this case is indicated. Using a c.w. multimode dye-laser operating at 4555 Å, the $7\,^2P_{\frac{3}{2}}$ state of the atoms in

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the beam was populated. About 10 % of the P-state atoms decay to the lowest $^2D_{\frac{5}{2}}$ state, which has a conveniently long lifetime, *ca.* 1000 ns. A c.w. single-mode laser is used for a further excitation to the F state to be investigated, and fluorescent light back to the D state was observed. The overlapping laser beams crossed the atomic beam at right angles. In figure 4 the components of the $5\ ^2D_{\frac{5}{2}}$–$12\ ^2F_{\frac{7}{2},\frac{5}{2}}$ transition are shown. While the $^2D_{\frac{5}{2}}$ state is split up in six hyperfine components, the F doublet has no resolved hyperfine structure. Fringes from a 50 MHz interferometer used for monitoring the laser sweep, are also shown. The fine structure of the F state as well as the hyperfine structure of the D state was found to be inverted.
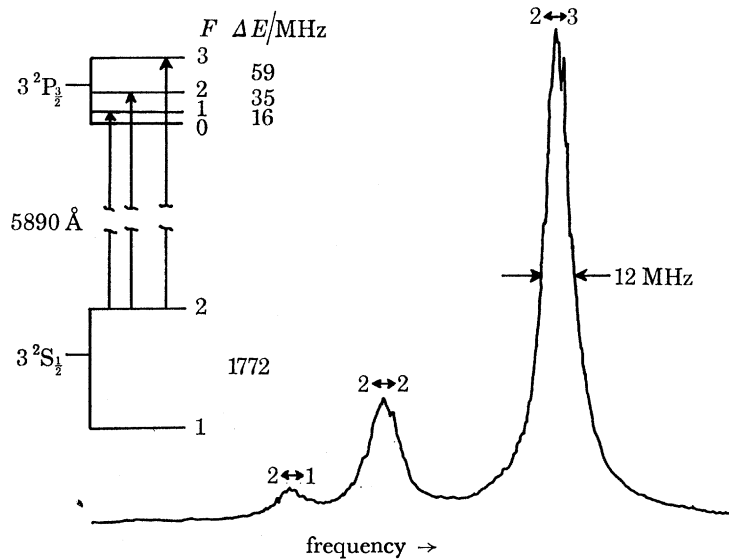

FIGURE 3. Hyperfine structure in the sodium D_2 line obtained by using a single-mode dye laser acting on a collimated atomic beam.
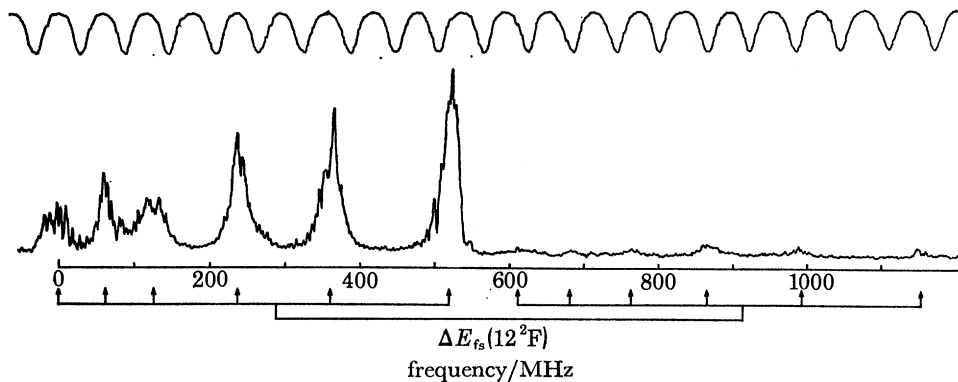

FIGURE 4. High resolution spectrum of the $5\ ^2D_{\frac{5}{2}}$–$12\ ^2F_{\frac{7}{2},\frac{5}{2}}$ transition in Cs. Calibration fringes from a 50 MHz interferometer are also included.

4. DOPPLER-FREE TWO-PHOTON ABSORPTION

In § 2 it was mentioned that, for example, a highly excited D state of an alkali atom can be populated by stepwise resonance transitions with the first P state as an intermediate level. For sufficiently intense light beams it is possible to populate such a state by using non-resonant two-photon absorption, where no real intermediate level is excited. The energies of the two

photons must together be equal to the excitation energy. Usually the two photons are of equal energy and obtained from the same laser. If one photon is absorbed from each of two counter-propagating beams, the first order Doppler shifts of the moving atoms will cancel, and a Doppler-free linewidth is obtained (Vasilenko *et al.* 1970; Cagnac 1977). It is possible to enhance the two-photon absorption cross-section by using two lasers of different frequencies, so that the intermediate virtual state is close to a real state (Bjorkholm & Liao 1974). In this case a total cancellation of the Doppler shifts is not obtained. The two-photon absorption process is monitored by observing fluorescence light in the decay of the excited state. Because of this we include two-photon absorption spectroscopy in this discussion. As a matter of fact, a partial cancellation of the Doppler shifts also occurs if counter-propagating beams are used in a resonant two-step excitation process (Duong *et al.* 1974). The two-step excitation process can be seen as a special case of the more general two-photon absorption process.

5. LIFETIME- AND QUANTUM-BEAT MEASUREMENTS

By using a pulsed laser tuned to a resonance transition, a large fraction of ground-state atoms can be transferred to the excited state. By monitoring the decay of the state in the resonance or fluorescence light with fast time-resolving electronics, the natural lifetime of the state can be
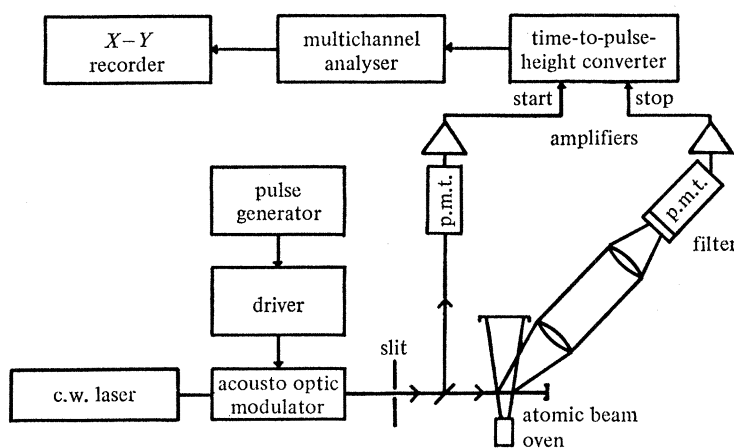

FIGURE 5. Experimental set-up for lifetime measurements with the delayed-coincidence technique, and a pulse-modulated c.w. dye laser beam.

determined. The delayed-coincidence method can be advantageous for precision work as this technique is not limited by possible nonlinearities in the detector. In this method the time lapse from the laser pulse to the arrival of the first fluorescence photon at the detector is measured. A time–pulse-height converter is used together with a multi-channel analyser to record the decay curve. As the probability of recording a photon following a laser pulse must be kept low for statistical reasons, it is highly desirable to use a high-repetition rate for the pulsed laser. We have used the arrangement shown in figure 5 to achieve short measuring times (Gustavsson *et al.* 1977). An acousto-optic modulator is used to produce a train of short pulses out of a c.w. dye laser beam. A large number of states in, for example, rare earth atoms have been studied with this efficient technique (Gustavsson *et al.* 1979).

If the excited state has an energy level substructure and the substates are coherently excited using a short laser pulse, oscillations of frequencies corresponding to certain energy splittings

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will be observed in the exponential decay. The oscillations are called quantum beats. In figure 6, quantum beats due to Zeeman splittings are shown in resonance scattering (Gustavsson *et al.* 1978). Such beats have previously been observed in Hanover (Gornik *et al.* 1972). The Zeeman beats can classically be explained as due to a damped radiating dipole, Larmor-precessing in the external magnetic field. In the same way, hyperfine-structure quantum beats can be obtained (Haroche *et al.* 1973). These beats can be seen as due to an electronic dipole, precessing in the magnetic field produced by the nucleus. An example of hyperfine beats is given in figure 7, where a signal due to the $5\,^2P_{\frac{3}{2}}$ state of ^{23}Na is shown (Grundevik *et al.* 1979). The simple exponential decay has been subtracted from the experimental curve and only the beats are shown. In the Fourier-transform curve, also included in the figure, the $\Delta F = 1$ and 2 beat frequencies can be seen.
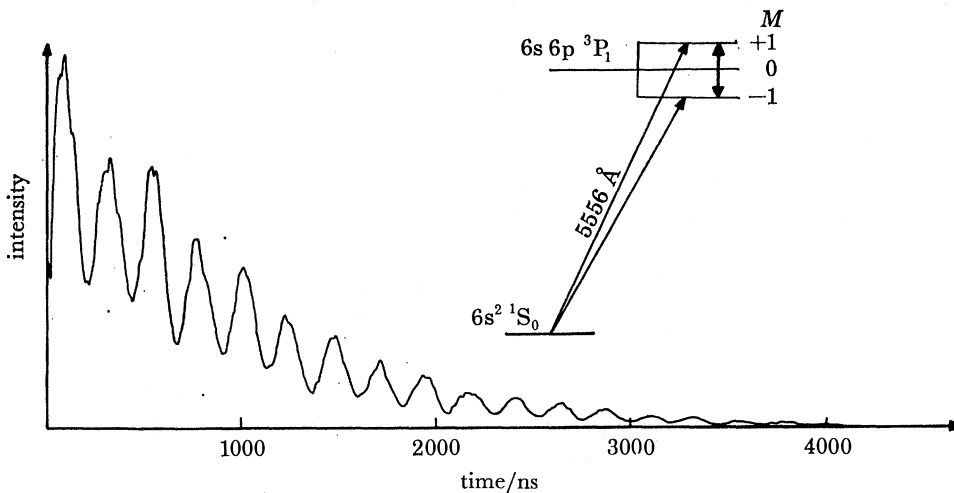

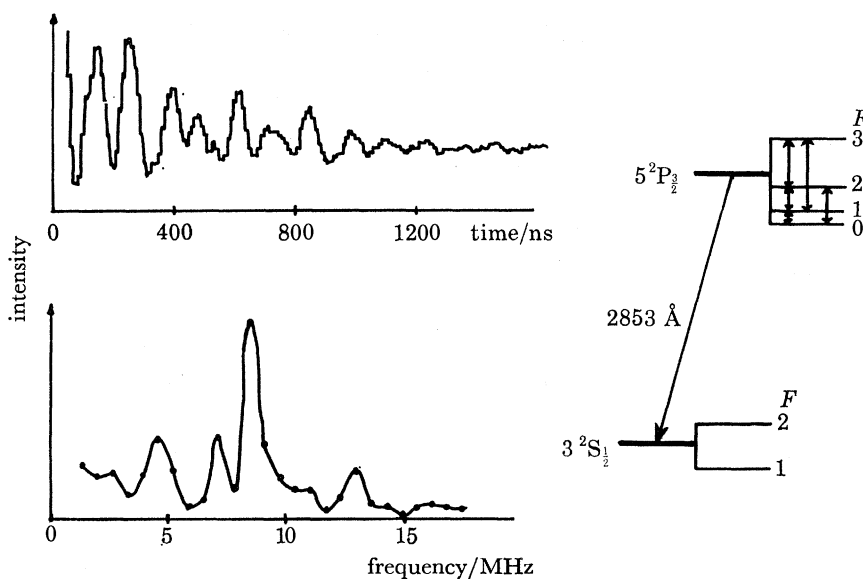FIGURE 6. Zeeman quantum beats in the decay of the laser-excited 6s6p $^3P_1$ stage of Yb. $B = 10^{-4}\,T$.

FIGURE 7. Hyperfine structure quantum beats in the decay of the $5\,^2P_{\frac{3}{2}}$ stage of ^{23}Na, excited with a frequency-doubled dye laser.

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6. OPTICAL RESONANCE SPECTROSCOPY

By combining laser excitation with optical resonance methods such as radiofrequency resonance and level-crossing spectroscopy, the demands on laser linewidth and stability can be greatly reduced although a maximal resolution is retained. By using stepwise excitations, as illustrated in figure 1, studies of hyperfine and fine structures, as well as Stark effect studies for a large number of normally optically inaccessible alkali states, could be performed (Svanberg *et al.* 1973; Fredriksson & Svanberg 1976; Svanberg 1977). In optical double resonance
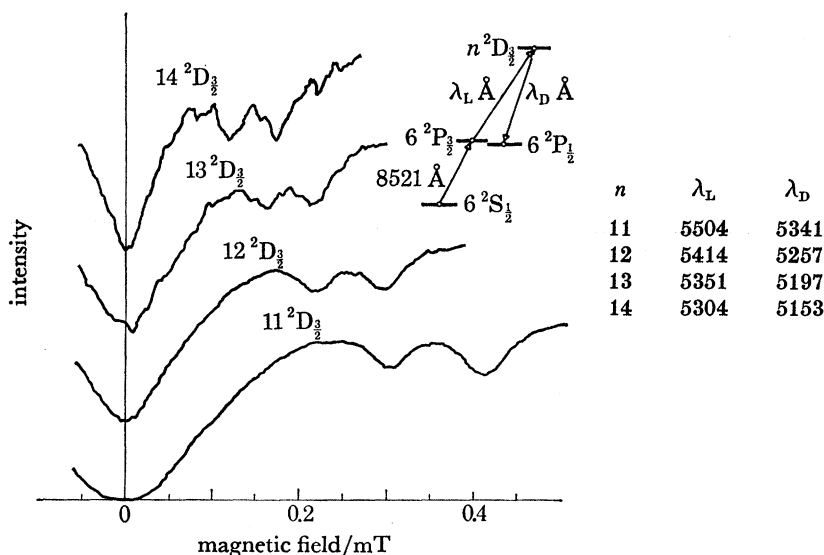

FIGURE 8. Level-crossing signals in highly excited $^2D_{\frac{3}{2}}$ states of ^{133}Cs, populated by using stepwise transitions.

experiments, polarized light is used to create unequal populations on the different substates of the excited atom. As a consequence of this the fluorescence light will be polarized and the polarization is partly retained in the cascade decay. By inducing r.f. transitions between the substates of the primarily excited state or between the substates of a lower state in the cascade, the polarization is changed (generally reduced) in the subsequent light emission. Thus an r.f. resonance experiment can be performed by polarized detection of almost any of the fluorescence lines in figure 2. When r.f. transitions are induced higher up in the decay chain, the total intensity in the Doppler broadened fluorescence line will be changed.

In level-crossing experiments, interference effects in the fluorescent light from excited states are observed when pairs of sublevels in magnetic field regions of level crossover can be coherently excited. Examples of level-crossing spectroscopy curves are given in figure 8 (Svanberg & Belin 1974). Highly excited $^2D_{\frac{3}{2}}$ states of ^{133}Cs were populated in a two-step process. At magnetic fields where certain pairs of magnetic sublevels cross, dips in the detected fluorescence light intensity are observed. From the level-crossing positions, the magnetic dipole and the electric quadrupole interaction constants can be calculated.

7. CONCLUSIONS

Resonance scattering methods that employ tunable lasers are very powerful for obtaining spectroscopic information of high precision. The alkali atoms, particulariy, have been extensively studied. These atoms are particularly suitable for comparisons with theory since they have a rather simple electronic structure, thus allowing detailed calculations. Large perturbations observed in the fine and hyperfine structure splittings can be adequately described (see, for example, Lindgren *et al.* 1976; Holmgren *et al.* 1976). The resonance scattering techniques are now being used to study a wide variety of atoms and molecules, and very interesting new information is being collected, greatly extending our basic knowledge of atomic and molecular systems.

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