Two-dimensional Spectroscopy of 'Forbidden' Raman Transitions by Laser-Induced Coherence Transfer

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We demonstrate a new spectroscopic technique that makes it possible to obtain information from 'forbidden' Raman transitions. The technique, which is based on time-resolved two-dimensional spectroscopy, uses laser-induced transfer of sublevel coherence from the Raman-inactive transition to an adjacent transition that can be observed in a Raman-process.

The interaction between atomic systems and radiation is often described in terms of a model that treats the atom as a system of two energy levels interacting with a classical electromagnetic wave [1]. In reality, however, most atomic systems have considerably more complicated level structures. The electronic ground state of atomic sodium, e.g., consists of eight energy levels grouped into two hyperfine multiplets. As is well known, this multiplicity leads to many interesting effects, like optical pumping, that cannot be explained in terms of the simple two-level model. We are interested primarily in the dynamics of such multilevel systems coupled to optical fields. Our experiments involve therefore primarily time-resolved experiments, in contrast to the more conventional steady-state experiments [2].

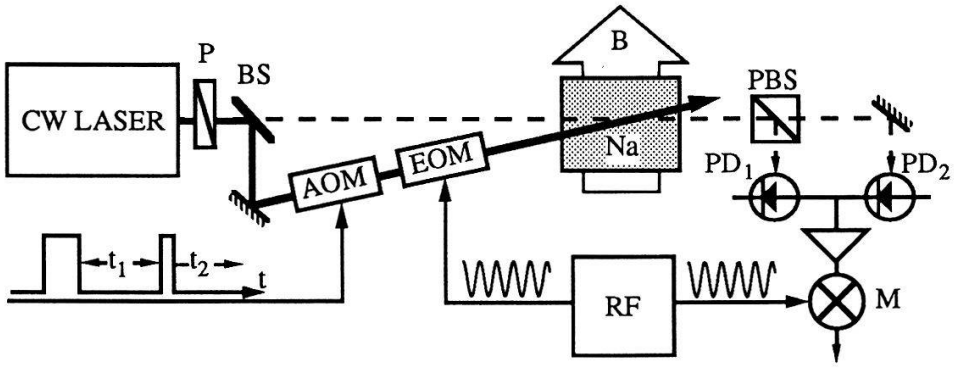

Figure 1: Experimental set-up used for time-resolved pump-probe experiments in atomic multilevel systems. P = polarizer, BS = beam splitter, AOM = acoustooptic modulator, EOM = electrooptic modulator, RF = radio-frequency synthesizer, B = magnetic field, PBS = polarizing beamsplitter, $PD_{1,2}$ = photodiodes, M = mixer.

The experimental setup used for this type of experiment is shown schematically in figure 1. The laser beam, which is derived from a single mode ring dye laser, is split into two parts, a pump beam with an intensity of the order of 100 mW/mm^2, and a probe beam with an intensity of the order of 1 mW/mm^2. The amplitude of the pump beam is controlled with an acoustooptic modulator (AOM) and its polarization can be modulated with an electrooptic modulator (EOM). For an efficient excitation of the atomic medium, which is located in a transverse magnetic field, the polarization of the pump laser beam is modulated between opposite circular polarizations at a frequency close to the Larmor frequency [3]. Pump and probe beam overlap in the sample region, where the metallic sodium is heated to a temperature of ~140°C in the presence of 200 mbar of Ar. The argon is used as a buffer gas to suppress the inhomogeneous Doppler broadening of the optical resonance line and to increase the time the atoms spend inside the laser beam. Behind the sample cell, the probe laser beam is passed into a polarization-selective detector . With the arrangement shown in figure 1 and the polarization of the incident probe beam rotated by 45° with respect to the axis of the polarizing beam splitter, the detector produces a signal proportional to the angular momentum component of the Na atoms parallel to the laser beam.

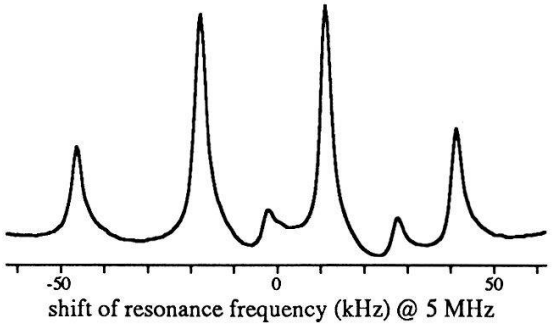

Figure 2: Raman spectrum of the Na ground state.

When a pump laser pulse is applied to the system, it initially induces transient oscillations. After a time of the order of 10 μsec, it settles into a stationary state and after the end of the pulse, the magnetization again starts to precess around the magnetic field; this precession is observed as a free induction decay (FID) signal. The interpretation of the signal can be simplified considerably if it is Fourier transformed.

The resulting spectrum, which is shown in figure 2, contains in general 6 distinct frequency components which can be assigned to the six possible Raman transitions between adjacent sublevels of the Na ground state, i.e. between sublevels whose magnetic quantum numbers m_F differ by $|\Delta m_F| = 1$.

The absence of transitions between sublevels whose magnetic quantum numbers differ by $|\Delta m_F|>1$ can be understood by considering a hypothetical Na atom with vanishing nuclear spin. The electronic ground state consists then of only two sublevels and therefore only a single transition with $|\Delta m_J| = 1$. In the limit of a weak hyperfine interaction, we expect a similar behaviour, i.e. to find only transitions with $|\Delta m_F| = 1$. This and other selection rules can be circumvented by two-dimensional time-resolved spectroscopy. This technique is used extensively in the field of nuclear magnetic resonance (NMR) [4] while applications to optical spectroscopy have not been demonstrated so far. In such an experiment, an initial laser pulse creates order in the form of population differences and coherences within the sublevel multiplets [5]. Since this laser pulse may be arbitrarily strong, multiple Raman transitions can excite coherences also in 'forbidden' transitions, e.g. in transitions with $|\Delta m_F|=2$ [6]. During the subsequent evolution period, these coherences are allowed to precess freely for a time t_1. This free precession period 'labels' the coherences with a phasefactor $\exp(i\,\omega_{ik}\,t_1)$. A second laser pulse causes an exchange of sublevel coherences between the various transitions. As a result, part of the coherence that evolved in transition ik may be transferred, together with the accumulated phase information, into a different transition. If this transition is Raman-active, it is then possible to observe the coherence and thereby measure the phase information associated with the forbidden Raman transition. The resulting signal depends then on the two independent time variables t_1 and t_2, where t_2 represents the time after the second laser pulse. Fourier transformation in both dimensions leads then to a two dimensional spectrum.

An example of a resulting spectrum is shown in figure 3 in contour plot representation. In this case, we suppressed all transitions except the $|\Delta m_F|=2$ transitions after the first pulse by using the different effect that phase shifts of the modulation frequency have on the different types of coherences [6] in order to simplify the spectrum. In the spectrum, we expect therefore along the vertical axis only frequencies corresponding to transitions between next-nearest neighbours. The corresponding frequencies are indicated in the form of a one-dimensional spectrum to the right of the two-dimensional spectrum. After the second pulse, only the 'natural' selection rule $|\Delta m_F|=1$ is active; as a result, the same frequencies as in the one-dimensional spectrum appear along the (horizontal) ω_2-axis. This experimental procedure allows therefore for the first time an observation of all possible transitions between Zeeman substates of the Na ground state.

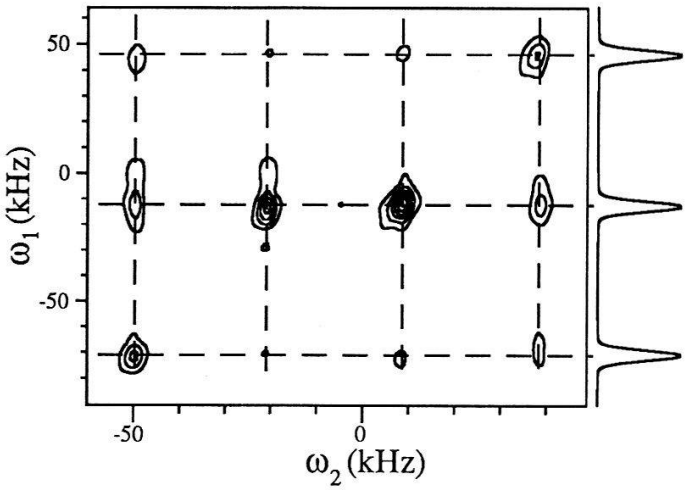

Figure 3: Example of a two dimensional spectrum demonstrating the possibility to use this method for observing 'forbidden' Raman transitions. The vertical axis represents the double quantum ($|\Delta m_F| = 2$) spectrum, while the horizontal axis corresponds to the single quantum spectrum.

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References

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