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Excitation of coherent Raman beats in rare earth solids with a bichromatic laser field

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Abstract

A new method is presented for measuring coherent Raman beats in systems with a wide spectral range that works even for weakly allowed optical resonance lines. A bichromatic pump laser field is used to resonantly pump a coherent excitation in the material, and a monochromatic test laser field, whose frequency coincides with that of one component of the pump laser beam.

1. Introduction

Scattering light from coherent excitations of quantum mechanical systems [1] provides an attractive extension of Raman spectroscopy. This process, which is known as coherent Raman scattering, is closely related to stimulated Raman scattering; the main difference is that the coherent excitation in the medium enhances the coupling between light and material qualitatively: the Raman field becomes linear in the incident field. To obtain this increased coupling efficiency, it is of course necessary to create the coherent excitation in the medium. This can be done continuously, e.g. by a radio-frequency [2] or microwave [3,4] irradiation, or in a transient mode [5–8], where the coherent excitation is prepared before the actual Raman process. In the latter case, the coherent excitation in the medium decays after the end of the pulse under the influence of radiative decay and collision processes. The Raman signal, which is usually observed as a beat signal between the probe laser and the Raman field, exhibits therefore a similar decay which is known as a coherent Raman beat. This signal contains information about energy level splittings, as well as the collision processes that cause the decay of the excitation.

The method has therefore been applied to study Raman transitions in molecular [7,9] and atomic vapours [10,11]. Applications to crystals of rare earth ions have also been demonstrated [12,13]. These crystals, which have been identified as interesting candidates for optical data storage [14], have been investigated with various kinds of high-resolution coherent optical spectroscopy [15,16]. Coherent Raman beat experiments could provide an ideal way for measuring the hyperfine structure at a resolution that is independent of laser jitter. However, the early experiments showed, that the weak transition strengths that make the narrow linewidths possible, make a straightforward application of the Raman beat technique difficult: with the optical power that is available from commercial cw dye lasers, the excitation of coherences with evolution frequencies more than a few MHz is very inefficient [13].

In this communication, we present an experimental procedure that allows the application of the Raman beat technique to the investigation of much wider spectra without requiring excessive laser power.

For this purpose, we use a bichromatic laser field for the excitation process and a probe laser field whose frequency can be shifted independently of the pump laser frequency. The conditions for the applicability of the technique are the same as for the conventional method, except that the requirements on the laser intensity and the pulse duration are relaxed significantly. The signals which we obtain contain the same information, and we demonstrate the method for a purely optical determination of the nuclear quadrupole splittings in the excited and in the ground state of $Pr^{3+}:YAlO_3$.

Fig. 1 explains the principle of the technique for a three-level atom in a V-configuration with an electronic ground state $|g\rangle$ and two non-degenerate sublevels of an electronically excited state. During the excitation process of the Raman beat experiment, a laser pulse excites both optically allowed transitions simultaneously. It excites therefore a coherent superposition of the states $|1\rangle$ and $|2\rangle$, the coherent excitation from which the Raman scattering occurs. This excitation can only proceed efficiently if the excitation laser is simultaneously resonant with both optical transitions. If the laser linewidth is smaller than the splitting of the two sublevels, this is only possible if the excitation pulse is short enough that the Fourier transform of the pulse shape has frequency components at both frequencies.

The alternative, which we propose here, is to use a bichromatic laser field, whose frequency components are resonant with the two optical transition frequencies. This allows the excitation of wide spectra with low optical power, and the duration of the pulses can be adjusted for an optimal excitation of the system. In our experiments, we use different modulation schemes to create the bichromatic field with frequency components over some tens of MHz. Similar schemes should allow an extension into the GHz range.

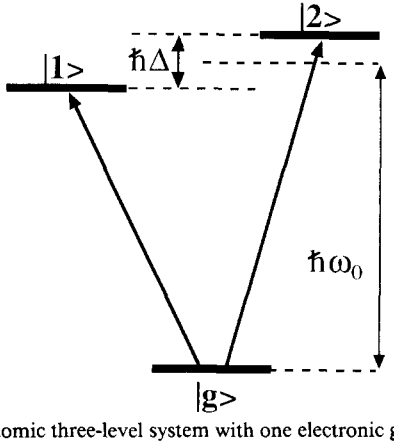

Fig. 1. Atomic three-level system with one electronic ground state $|g\rangle$ and two sublevels ($|1\rangle$, $|2\rangle$) of the electronically excited state. The resonance frequencies of the two optical transitions are $\omega_0 \pm \Delta/2$.

2. Theory

The Raman beat experiment can be separated into two largely independent parts: the excitation of the coherent excitation in the medium and the coherent Raman scattering process. The two processes are independent of each other also in the experiment: while it is possible to use the same laser beam for excitation and detection [13], we use separate pump and test laser beams. This separation allows to independently control the frequency and intensity of the laser during preparation and detection. We start our discussion with a description of the excitation process.

The simplest energy level scheme for the description of the experiment is the V-type three-level system of Fig. 1. The ground state $|g\rangle$ can be coupled via an optical transition to any of the two sublevels $|1\rangle$ and $|2\rangle$ of the electronically excited state. The energy difference between both states is much smaller than the optical frequency. The unperturbed Hamiltonian $\mathscr{H}_0$ of this system can be written as

$$\langle g|\mathscr{H}_0|g\rangle = 0, \quad \langle 1|\mathscr{H}_0|1\rangle = \hbar(\omega_0 - \Delta/2),$$
$$\langle 2|\mathscr{H}_0|2\rangle = \hbar(\omega_0 + \Delta/2). \qquad (1)$$

To describe the excitation process, we assume first a monochromatic laser field on resonance with the atomic transition ($\omega_{\text{laser}} = \omega_0$). Within the rotating wave approximation, the evolution of the system is governed by the Hamiltonian of Eq. (1) with $\omega_0 = 0$ and an interaction Hamiltonian $\mathscr{H}_{\text{int}}$

$$\langle 1|\mathscr{H}_{\text{int}}|g\rangle = \hbar V_1, \quad \langle 2|\mathscr{H}_{\text{int}}|g\rangle = \hbar V_2, \qquad (2)$$

that describes the coupling with the laser field. In rare earth ionic crystals, where the sublevel splitting is due to nuclear quadrupole interaction, both transitions are allowed due to the different orientation of the electric field gradient tensor in the ground and electronically

excited states. In the case of $Pr^{3+}:YAlO_3$, the two transition matrix elements differ by approximately an order of magnitude.

Assuming that the system is initially in the ground state, we write its wave function as $|\Psi_0\rangle$. A short laser pulse with frequency ω_0 then drives both optical transitions. If the duration τ of the pulse is short enough ($\tau\Delta \ll 1$), we can approximate the resulting state of the system in lowest order perturbation theory as

$$|\Psi\rangle = |\Psi_0\rangle + iV_1\tau \exp(-i\Delta\tau/2)|\Psi_1\rangle + iV_2\tau \exp(i\Delta\tau/2)|\Psi_2\rangle . \quad (3)$$

The excited state coherence $|\rho_{12}|$ is then proportional to $V_1\tau V_2\tau$. For short optical pulses, the coherence should thus increase with the square of the pulse duration. However, if the duration of the pulse exceeds the inverse level splitting, $\tau\Delta > 1$, coherence packets that have been created during different parts of the pulse start to accumulate different phase factors. Destructive interference between them results in a decrease of the observed signal [13,17]. A large excitation of the system under these conditions requires therefore short pulses with high intensity. On the other hand, the pulses should selectively excite only a narrow portion of the inhomogeneously broadened optical transition, a condition which can be met only with cw lasers. With the intensities that are available from commercial cw ring lasers, these conditions can be satisfied only over a relatively narrow frequency range comparable to the optical Rabi frequency (of the order of some tens of kHz).

Our approach to circumvent this problem uses a bichromatic laser field with frequency components near $\omega_0 \pm \Delta/2$. With this procedure coherence packets that are created at different times during the pulse add coherently instead of interfering destructively, as in the case of monochromatic excitation. The restriction $\tau\Delta < 1$ on the pulse duration is thus removed and it becomes possible to extend the pulse duration until a significant population of the excited state is achieved.

To realise the bichromatic laser field in the excitation period we use the modulation sidebands, which result from an intensity modulation of the pump beam at a frequency ω_{mod}. For a fully modulated beam, the frequency spectrum of the laser amplitude contains two sidebands with equal amplitudes at the frequencies $\omega_{las} \pm \omega_{mod}/2$. In the experiment a coherent superposition of sublevels $|1\rangle$ and $|2\rangle$ (compare Fig. 1) is created, if this modulation frequency matches the energy splitting between the two sublevels: $\omega_{mod} = \Delta$.

After the preparation pulse, the coherence decays with the lifetime of the excited state T_1 and evolves as

$$\rho_{12}(t) = \rho_{12}(0) \exp(i\Delta t) \exp(-t/T_1) . \quad (4)$$

If a probe laser field at frequency ω_{probe} interacts with the atom, it excites a coherent Raman field

$$E_R = c_R \rho_{12}(t) E_t , \quad (5)$$

where E_t is the amplitude of the test laser field and the coefficient c_R describes the efficiency of the Raman process, which depends on the coupling strength as well as on the optical path length [18]. The Raman field has a frequency of $\omega_t + \Delta$ and propagates collinear with the probe laser field. As the two fields with different frequencies interfere, they give rise to a heterodyne beat signal at the difference frequency Δ on a quadratic photodetector.

This signal can of course be obtained only if the test laser interacts with the same atoms within the inhomogeneously broadened optical resonance line as the pump laser field; it must therefore match one of the two sidebands of the pump laser field and must be shifted with respect to the unmodulated pump laser frequency. If the sublevel splitting exceeds the homogeneous optical linewidth and the laser linewidth, this requires an independent control of the test laser frequency.

3. Experiment

Fig. 2 illustrates the experimental setup. The experiments were performed in a $5 \times 5 \times 1$ mm $YAlO_3$ (YAP) crystal, doped with 0.1% Pr^{3+}-ions. The crystal was mounted on the cold finger of a He flow cryostat and the two linearly polarized laser beams propagated along the crystallographic c axis at an angle of intersection of 5 mrad. The laser frequency was resonant with the transition between the $^3H_4$ ground state and the $^1D_2$ excited state ($\lambda = 610.6$ nm). The frequency of the actively stabilised ring dye-laser was

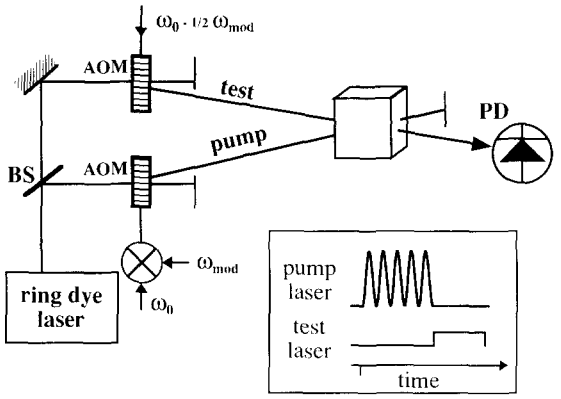

Fig. 2. Experimental setup. BS = beam splitter, AOM = acousto-optic modulator, ω_0, ω_{mod} = carrier and modulation frequency of the AOM driving signal, PD = photodiode. The inset shows the time-dependence for the intensities of pump and test laser beam.

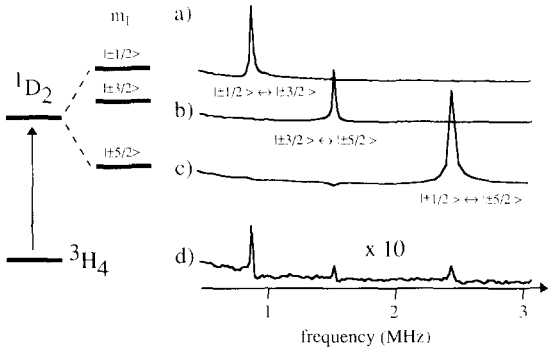

Fig. 3. The left side shows the relevant level scheme for Pr^{3+}:YAP with three doubly degenerate nuclear spin sublevels ($m_I = \pm 1/2$, $\pm 3/2$ and $\pm 5/2$). The sublevel spectra of the excited state shown on the right side were obtained using modulated excitation at (a) 0.914 MHz, (b) 1.6 MHz and (c) 2.5 MHz. Trace (d) shows the reference spectrum obtained with single-frequency excitation. Note the expanded scale.

close to the line centre of the inhomogeneously broadened line with a width of 5 GHz (fwhm).

The inset in Fig. 2 shows the timing of the experiment. The intensity of the pump laser beam (typically 200–300 mW/mm^2) is modulated sinusoidally with an acoustooptic or electrooptic modulator (not shown in the figure) and gated with an acoustooptic modulator that produced pulses with a duration between 0.1 and 100 μs. The weaker probe beam (< 1 mW/mm^2) is switched on immediately after the pump pulse by a second acousto-optic modulator. The rf carrier frequency of this modulator is adjusted to shift the test laser frequency to one of the two modulation sidebands of the pump laser field. The observed beat signal is detected with a fast photodiode and averaged 500 times. The time-domain signal is then Fourier-transformed to obtain the sublevel spectrum. To avoid hole burning from the repetitive excitation pulses, we used a delay between pulses that was longer than the relaxation time of the ground state sublevels [19]. At a temperature of 8 K, we could use a repetition rate of 50 Hz.

The left-hand part of Fig. 3 shows the relevant part of the energy level structure of Pr^{3+}:YAP with a nuclear spin $I = 5/2$ in zero magnetic field. The sublevel structure with three doubly degenerate pairs with $m_I = \pm 1/2$, $\pm 3/2$ and $\pm 5/2$ is shown only for the excited state. The splitting is due to nuclear quadrupole interaction and the transition frequencies are 0.92 MHz, 1.56 MHz, and 2.48 MHz. Between these levels, it is possible to excite sublevel coherences with short laser pulses ($\tau < 1$ μs). Trace (d) at the bottom of Fig. 3 shows an example of such a spectrum. The decrease of signal amplitude at the higher frequencies illustrates the limitations of the conventional method: the excitation efficiency, as well as the detection efficiency decrease at higher frequency. Traces (a)–(c) demonstrate, how bichromatic excitation, combined with frequency-shifted detection can overcome this problem and provide significantly better signal to noise ratio. The difference is particularly striking for the line near 2.5 MHz. All four traces were measured with the same laser intensity. The pulse duration was optimised independently for all four experiments. In the case of single-frequency excitation, we found an optimal pulse-length of 0.9 μs, while it was between 20 and 28 μs for the three experiments with bichromatic excitation. The difference in the observed signal intensity is thus roughly proportional to the pulse length.

In the electronic ground state, the quadrupole splittings are significantly larger: 7.1 MHz and 14.1 MHz and 21.2 MHz. With the available laser intensity we could no longer excite these transitions with single-frequency excitation; similar problems were reported by Shelby et al. [13]. With bichromatic excitation, however, these transitions could equally be excited, as shown in Fig. 4. For these experiments, we used intensity modulation with an electrooptic modulator.

To bring the test laser frequency in resonance with the same atoms that were excited by the pump beam, we had to shift the test laser frequency to one of the

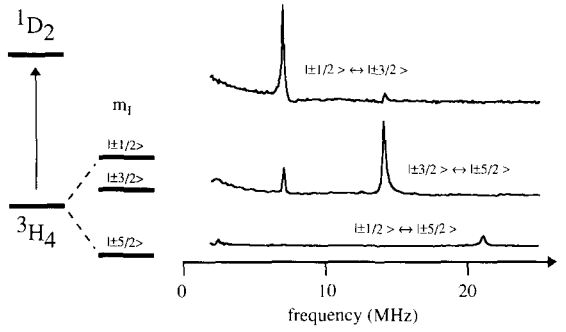

Fig. 4. Hyperfine structure of the ground state of Pr^{3+}:YAP together with three sublevel spectra obtained with bichromatic excitation. From top to bottom the modulation frequency was set to 7.06 MHz, 14.1 MHz and 21.1 MHz.

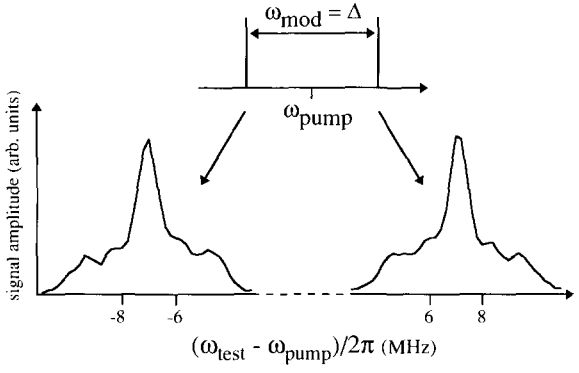

Fig. 5. Shown on the top is the frequency spectrum of the pump laser beam if the intensity is fully modulated at frequency $\omega_{mod}=\Delta$. It contains two sidebands at $\omega_{pump}\pm\Delta/2$. The lower part shows the amplitude of the Raman signal as a function of the frequency difference $(\omega_{test}-\omega_{pump})/2\pi$ between test and pump laser beam for an intensity modulation with 14.1 MHz. Due to the small homogeneous linewidth the Raman signal is only observable, if the test laser beam has the same frequency as one sideband of the pump beam.

sidebands of the pump field, using an acoustooptic modulator whose carrier frequency was controlled by an rf synthesizer. The importance of this frequency control is demonstrated in Fig. 5, where we show the dependence of the signal amplitude on the test laser frequency. For this experiment, we used intensity modulation at $\omega_{mod}/2\pi=\Delta/2\pi=14.1$ MHz. The two curves show the signal close to the two sidebands at $\omega_{pump}\pm 7$ MHz. The range, over which the Raman signal could be observed, corresponds to the laser frequency jitter, which is of the order of 1 MHz for our laser (Spectra Physics model 380D). Therefore the test laser frequency must be controlled to within this range.

It is interesting to note that the spectra presented here could be obtained in the absence of a magnetic field, in contrast to Raman-heterodyne detection of NMR experiments. In these experiments two signal contributions of two non-equivalent sites in the crystal lead to a destructive interference and vanishing signal amplitude in zero magnetic field [20].

4. Discussion

Coherent Raman beat experiments are a useful tool for sublevel spectroscopy of both atomic and molecular gaseous systems. In these experiments, a laser beam is scattered off a coherent excitation of the system, thereby providing a Raman spectrum with high sensitivity and high frequency resolution, independent of laser frequency jitter. Laser pulses can be used not only for the detection process, but also to efficiently prepare the required coherent excitation of the material, provided they are simultaneously resonant with both optical transitions. In atomic vapours, this condition can usually be met easily, as the homogeneous width of the optical transition is often much larger than the sublevel splitting. In solids, however, the homogeneous linewidth can be as small as a few kHz [15] – considerably less than the sublevel splittings which are in the MHz range. Furthermore, the optical transition strengths can be many orders of magnitude smaller than in vapours. Both problems complicate the efficient excitation of sublevel coherences.

In this communication, we have demonstrated an experimental procedure that circumvents these problems: a bichromatic laser field, whose frequency components are resonant with both optical transitions, allows an efficient excitation of the sublevel coherences, for arbitrarily narrow optical resonance lines and very large sublevel splittings. In our experimental setup, we have used intensity modulation techniques to generate the sidebands with splittings from 0.9 to 21 MHz. Systems with even wider sub-

level splittings could be excited e.g. by superimposing laser beams that have been frequency-shifted: with acoustooptic or electrooptic modulators, frequency shifts up to several GHz can be generated. In principle, it should even be possible to use laser beams derived from independent sources, provided the frequency-difference can be kept stable enough. While our simple theoretical description used a V-type level system, we have demonstrated experimentally, that it is equally applicable to Λ-type systems. The theoretical model that we presented here was kept as simple as possible to show the relevant physical process. The actual experimental system has of course more than three energy levels and calculations are in progress that should allow a quantitative description of these systems. Furthermore, we are investigating excitation with more than two frequency components to allow simultaneous excitation of more than one sublevel coherence.

Modulation of the laser intensity has also been used successfully for sublevel spectroscopy of atomic vapours, both in the frequency domain [11,21] and in the time domain [22–24]. The two main differences between the vapour phase experiments and the experiments on rare earth solids presented here, are the transition strengths, which are several orders of magnitude lower in the solid, and the width of the homogeneous optical line: in the solid, the linewidth is much narrower than the sublevel splitting. In the example discussed here, the sublevel splitting is also much wider than the laser linewidth; under these conditions, care must be taken that the test laser frequency is in resonance with the same atoms as the pump laser within the inhomogeneously broadened optical transition. In our experiment, we achieved this by shifting the test laser frequency to one of the two frequency components of the pump laser beam.

Apart from the increased bandwidth of our method, the conditions for its applicability should be the same as for excitation with a monochromatic field: both excited state sublevels (for V-type systems) must be connected to the ground state by an optical transition. In the example that we used, $Pr^{3+}:YAlO_3$, the transition strengths of the two optical transitions differ by about one order of magnitude. This shows that this requirement does not significantly limit the applicability of coherent Raman beats, provided the excitation fields are close enough to resonance to allow a long enough excitation of the system. The obvious limit on the duration of the excitation process in V-type systems is the excited state lifetime.

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