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Experimental study of Stokes fields linewidth in resonant four-wave mixing in Rb vapour

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Abstract. We report transmission linewidths for a weak anti-Stokes component and generated Stokes component during resonant four-wave mixing in atomic Rb vapour. We consistently observe larger linewidth of the generated field relative to the input field.

1. Introduction

Interest in narrow resonances in electromagnetically induced transparency (EIT) is connected with applications to high-resolution spectroscopy. Recently, the absorptive width of EIT resonances has been studied both experimentally and theoretically [1–8]. Furthermore, the steep dispersion associated with EIT plays a key role for experiments that have produced ultra-slow light propagation [9–11] and have enhanced nonlinear optical processes by orders of magnitude [12–15].

However, in media with high optical density, the generated Stokes fields introduce significant changes and must be taken in account. We have found that as a function of probe laser (anti-Stokes) power the linewidth of the generated Stokes field measured at the output of the cell is significantly broader than that of the probing anti-Stokes field. This result may be surprising in light of earlier work [16]. The study of Stokes harmonics properties, such as resonance width and dependence on drive laser power, is the subject of this paper.

2. Experiment

2.1. Setup

A schematic of the experiment is shown in figure 1. Two external cavity diode lasers (ECDL) were used, one of them as a strong drive laser and the second as a relatively weak probe laser. The drive laser was tuned to the $S_{1/2}(F = 2) \rightarrow$ $P_{1/2}(F = 2)$ transition of the ⁸⁷Rb D₁ resonance line ($\lambda = 794.7$ nm). The probe laser was phase-locked to the drive laser with a frequency offset approximately equal to the ground-state hyperfine splitting (6.835 GHz), so its frequency was close to the $S_{1/2}(F = 1) \rightarrow P_{1/2}(F = 2)$ transition. This is shown in figure 2.

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Figure 1. Experimental setup.

The co-propagating beams of both lasers were combined and passed through a polarization-preserving single-mode optical fibre in order to obtain good spatial overlap and spatial modes. The beam width was adjusted with a telescope and diaphragm and the light was circularly polarized with a quarter-wave plate just before it entered the ⁸⁷Rb cell.



Figure 2. Level scheme in Rb.

The cell contained a mix of isotopically enhanced ⁸⁷Rb and N_2 buffer gas at a pressure of 3 torr. Different concentrations of Rb vapour were achieved by adjusting the temperature of the cell. The cell was placed inside a three-layer magnetic shield to screen the earth's magnetic field.

2.2. Width measurement procedure

The nonlinear production of a Stokes (or 'new') field in this system has previously been reported [10, 16]. As described there, the 'new' field is generated by nonlinear four-wave mixing of the drive and probe fields. The difference between the probe (anti-Stokes) and drive field frequencies is equal to the difference between the drive and new field frequencies (see figures 2 and 3).

Rather than measure the DC component of the transmitted light, which contains drive, probe, and new fields, we detect the AC component near the beat frequency of the probe and drive lasers. This allows clean measurement of both probe and generated (Stokes) field amplitudes as reported previously [10]. As described there, the amplitude of the beat signal at the frequency difference of the probe and drive fields (which is near the hyperfine frequency of 6.835 GHz) contains a contribution from the beat between the probe and drive fields, and a contribution from the beat between the drive and new fields (see figure 4).



Figure 3. Diagram showing relative frequency of the laser fields present in the experiment. The drive field is tuned to the $S_{1/2}(F = 2) \rightarrow P_{1/2}(F = 2)$ transition of the ⁸⁷Rb D₁ resonance line ($\lambda = 794.7$ nm). The probe laser is tuned near the $S_{1/2}(F = 1) \rightarrow P_{1/2}(F = 2)$ transition.



Figure 4. Diagram showing the beat frequencies present in the experiment.

To separate the individual frequency components, another field is introduced on the detector which is derived from the drive field but shifted up slightly (50 MHz) in frequency by an acousto-optical modulator (figure 1). This introduces sidebands in the beat spectrum. Because the shifted field does not pass through the cell these sidebands are just proportional to the transmitted field amplitude of the probe and new fields. The beat spectrum is shown in figure 4. Measurement of the transmitted probe field (anti-Stokes component) gives information about the EIT resonance. Similarly, measurement of the transmitted new field (Stokes component) provides information about the resonance of the new field.

2.3. Experimental results

We have measured the difference in the resonance width of the probe and new field. Representative data is shown in figure 5. The new field transmission resonance width was noticeably greater then the EIT resonance width of the probe field. Also we report linear growth of the resonance width with increasing drive field power for constant probe field. This data is shown in figure 6. Similarly, we observe linear increase of the resonance width with increasing probe field power when the drive laser power is held constant, as seen in figure 7.

3. Theory

Since the driving field has circular polarization, it transfers all populations of the magnetic sublevels to the one with maximal value of m_F optical pumping and to the ground state. Thus, effectively, the population ends up in the one state. A simple model to describe this physical situation is a three-level Λ -system obtained from the real one (figure 2) by neglecting the magnetic sublevels emptied by optical pumping.

3.1. Simple model

The Hamiltonian of the simplified system (depicted in figure 2) is



Figure 5. Representative data showing the shape of the EIT resonance for probe and new fields. Temperature is 89.6° C. Probe power is $3 \mu W$.



Figure 6. Data showing dependence of EIT width on drive power for probe and new fields. Temperature is 89.6° C. Beam diameter is 5 mm. Probe power is $3 \mu W$.



Figure 7. Data showing dependence of EIT width on probe power for probe and new fields. Temperature is 77.7° C. Beam diameter is 5 mm. Drive power is $300 \,\mu W$.

$$\hat{H} = \hbar \Omega(|a\rangle \langle c| + |a\rangle \langle b|) + \hbar \alpha |a\rangle \langle b| + \hbar \beta |a\rangle \langle c| + adj$$
(1)

where Ω , α and β are the driving, probe and new fields, levels $|a\rangle$ and $|b\rangle$ correspond to ground state levels F = 1 and F = 2 respectively, and $|c\rangle$ corresponds to the excited level F = 2 (figure 2). The corresponding density matrix equations for coherences in the steady-state approximation are

$$\Gamma_{ab}\sigma_{ab} = in_{ab}\alpha - i\Omega\sigma_{cb} \tag{2}$$

$$\Gamma_{ca}\sigma_{ca} = \mathrm{i}n_{ca}\Omega + i\alpha\sigma_{cb} \tag{3}$$

$$\Gamma_{cb}\sigma_{cb} = \mathbf{i}(\sigma_{ca}\alpha - \Omega\sigma_{ab} + \sigma_{ca1}\Omega - \beta\sigma_{ab1}) \tag{4}$$

$$\Gamma_{ab1}\sigma_{ab1} = in_{ab}\Omega - i\beta\sigma_{cb} \tag{5}$$

$$\Gamma_{ca1}\sigma_{ca1} = \mathrm{i}n_{ca}\beta + i\Omega\sigma_{cb} \tag{6}$$

where $\Gamma_{ab} = \gamma + i\omega$, $\Gamma_{ca} = \gamma + i\omega_d$, $\Gamma_{cb} = \gamma_{cb} + i(\omega - \omega_d)$, $\Gamma_{ab1} = \gamma + i(\Delta - \omega_d)$, $\Gamma_{ca1} = \gamma + i(\Delta - \omega)$, $n_{ab} = \sigma_{aa} - \sigma_{bb}$, $n_{ca} = \sigma_{cc} - \sigma_{aa}$. The equations for the populations are

$$r_a - \gamma_{cb}n_a + \gamma n_a - 2\operatorname{Im}\left(\sigma_{ab1}\Omega + \sigma_{ab}\alpha\right) = 0 \tag{7}$$

$$r_c - \gamma_{cb} n_a + \gamma n_a - 2 \operatorname{Im} \left(\sigma_{ca1} \beta + \sigma_{ca} \Omega \right) = 0$$
(8)

$$n_a + n_b + n_c = \frac{r_a + r_c}{\gamma_{cb}} \tag{9}$$

Let us consider the case of a strong driving field, $|\Omega|^2 \gg \gamma_{bc} W_D$, and weak probe and new fields, $|\Omega| \gg \alpha, \beta$. Under such conditions, all atomic populations should be in state $|b\rangle$, and we then find the solution to the above equations is

$$\tilde{\Gamma}_{cb}\sigma_{cb} = -\frac{\alpha\Omega}{\Gamma_{ab}} - \frac{\beta\Omega^*}{\Gamma_{ca1}}$$
(10)

$$\sigma_{ab} = -i\frac{\alpha}{\Gamma_{ab}} + i\frac{\alpha|\Omega|^2}{\tilde{\Gamma}_{cb}\Gamma_{ab}^2} + i\frac{\beta\Omega^{*2}}{\tilde{\Gamma}_{cb}\Gamma_{ca1}\Gamma_{ab}}$$
(11)

$$\sigma_{ca1} = -i \frac{\alpha \Omega^2}{\tilde{\Gamma}_{cb} \Gamma_{ab} \Gamma_{ca1}} + i \frac{\beta |\Omega|^2}{\tilde{\Gamma}_{cb} \Gamma_{ca1}^2}$$
(12)

where

$$\tilde{\Gamma}_{cb} = \gamma_{cb} + \frac{|\Omega|^2}{\Gamma_{ab}} + \frac{|\Omega|^2}{\Gamma_{ca1}} + \frac{|\alpha|^2}{\Gamma_{ca}}$$

To compare with the observed results we need to take into consideration the propagation effects

$$\frac{\partial \alpha}{\partial z} = i\eta \sigma_{ab} \qquad \frac{\partial \beta}{\partial z} = i\eta \sigma_{ca1}$$
 (13)

where $\eta = 3\lambda^2 N/8\pi\gamma$.

3.2. Theoretical results

We calculate the fields propagated through a dense medium. At the frequency of two-photon resonance, the probe and drive fields acting together induce the low-frequency atomic coherence σ_{cb} that plays a crucial role in establishing EIT. Increasing the two-photon detuning decreases the coherence σ_{cb} and increases absorption of the probe field. Thus, the probe field has a narrow window at the vicinity of two-photon resonance, with a width that depends on the rate of coherence relaxation and power broadening. On the other hand, the low-frequency coherence σ_{cb} is also responsible for a new field generation, and the intensity of the new field has a similar resonance behaviour to the transparency of the probe field. The important difference is that the new field has a large detuning from onephoton resonance and, consequently, has practically no absorption.

Our numerical calculation showed similar dependence for the width of the resonance versus drive laser power (see figure 6). The simulation are in good agreement with the experimentally observed data, because the experimental range of driving field intensities are in the range for which the simplest theory developed above is valid. From the simple theory we can see that the difference in the widths of the EIT window and the new field generated in the cell appears from the propagation. Increasing the probe field leads to redistribution of the population between levels b and c, which changes the effective power broadening of the EIT window width while the change of the width of the generated new field is not that strong (figure 7).

4. Summary

We have shown that the transmission resonance for the Stokes component is wider than the resonance for the low power anti-Stokes (probe) field. Also we report that both resonance widths of the Stokes component resonance and the probe field have linear dependences on the drive and probe laser power when the power of the second laser (probe and drive correspondingly) is kept constant.

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