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Phase-sensitive microwave optical double resonance in an N system

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Abstract – An experimental investigation of a Microwave Optical Double Resonance (MODR) phenomenon is carried out in a four level N system of 85Rb atoms, at room temperature. This N system consists of a closed three level Λ subsystem irradiated with two optical fields and one microwave field. The MODR response is investigated in a separate probe field which drives a resonant transition from one of the ground states of the Λ system to a fourth level. We find that, under two-photon resonance condition for the optical fields, the MODR becomes a function of the relative phase between the beat frequency envelop of the optical fields and the microwave field. The variation in MODR is shown to be correlated with the phase-sensitive variation of the EIT phenomenon seen in such microwave-connected closed Λ systems. We envisage that this phase-sensitive variation in the MODR, can be utilized for a phase-sensitive manipulation of non-linear optical phenomena in N systems.

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Introduction. – Creation of CPT states [1] or dark states has now become a standard starting point for studying coherence based linear and non-linear interaction between laser light and dilute atomic vapour. The atom light coherences lead to phenomena like Electromagnetically Induced Transparency (EIT) [2–5], Lasing without Inversion (LWI) [6] and resonant enhancement of non-linear optical phenomena [7–9]. The enhanced group refractive index associated with EIT has given rise to a variety of experiments investigating slow light phenomenon [10]. This in turn has led to optimal storage and retrieval of coherences between matter variables and optical fields. Optimal storage and retrieval of light pulses by tailored pulse shapes and CPT coherences, has been demonstrated using RF phase modulated light fields [11]. In an N level configuration of levels, CPT coherences are responsible for the creation of giant Kerr non-linearities with negligible single-photon absorption [9]. N level systems are central to proposals for realisation of laboratory quantum simulators [12], quantum phase gates [13] and few photon Kerr non-linearities [14]. The N system has also been studied extensively with non-degenerate ground-state configurations in the context of enhancement of four-wave mixing signals [15,16]. In all the above-mentioned effects arising in the N system, the Λ subsystem with two optical fields connecting ground states to a common excited state has been responsible for generation of coherences between the ground states.

Of recent, both experimental and theoretical studies have focused on the effect of connecting the two ground states of a regular Λ system, by a third field, whose frequency for alkali metal atoms is in the microwave regime [17–20]. The papers [18,19] were amongst the first to show the dependence of ground state coherence on the relative phase of the participating fields. These and other recent studies show that the EIT line width and peak value is dependent on the relative phase between the beat envelop of the EIT forming optical fields and the microwave field connecting the ground states. Such a phase-sensitive response is expected in closed level configurations which are addressed by coherent fields [21]. It has also been shown in the past [22,23], that a ground-state RF coupling can create enhanced non-linear responses even in a non-closed configuration of levels.

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We know that, traditionally, a Microwave Optical Double Resonance (MODR) phenomenon has been used routinely for feedback controlled atomic frequency standards [24]. In recent years, CPT coherences are used in these systems [25], for enhanced short term frequency stability [26].

In this paper, we have combined the closed nature of level configurations in a \( \Lambda \) system and a microwave optical double resonance, by choosing an \( N \) level system with a three level closed \( \Lambda \) subsystem. As seen in fig. 1, the transition denoted by \( \Omega_{DR} \), along with those of the closed \( \Lambda \) subsystem makes an \( N \) level system. We show that, in this special \( N \) system, a three-optical-electron resonance condition, is equivalent to satisfying a condition necessary for a microwave optical double resonance transition. This transition is seen in the optical field \( \Omega_{DR} \). This equivalence introduces a microwave control on the non-linear optical three-photon process of the \( N \) system. Such a phase-dependent control for a reactive non-linear process has not been demonstrated so far. This experiment thus demonstrates the possibility of phase control of giant Kerr non-linearities seen in such systems [27]. Additionally, this also simultaneously establishes the phase-sensitive control of a double resonance clock transition (MODR absorption resonance). As elucidated in [17], this phase, can be connected to the phase difference between the microwave dressed state and the dressed (dark) state created by the optical fields. Thus the MODR response can potentially become a non-destructive measurement tool for finding relative phases between dressed eigenstates. We envisage that this aspect will have far-reaching experimental consequences.

**Experimental setup.** – Three optical beams, namely, the drive with strength \( \Omega_D \), the probe with strength \( \Omega_P \) and the MODR probe with strength \( \Omega_{DR} \), are derived from a single diode laser which has a typical long time linewidth of 1 MHz. The diode laser is tunable over the entire frequency region around the D2 manifold of \( ^{85}\text{Rb} \) and its frequency is monitored using a reference saturation absorption spectroscopy arrangement. These three beams address an \( N \) level around the D2 manifold in \( ^{85}\text{Rb} \) as shown in fig. 1. The drive beam is derived from the laser directly and it connects the upper hyperfine ground state \( 5\!P_{1/2}, F = 3 \) (\( |c\rangle \)), to the excited state \( 5\!P_{3/2}, F = 3' \) (\( |a\rangle \)). The probe beam which connects the lower hyperfine ground state \( 5\!P_{1/2}, F = 2 \) to the excited state \( 5\!P_{3/2}, F = 3' \) (\( |b\rangle \) to \( |a\rangle \)), is derived from the drive, by phase modulation of the drive, through an Electro Optic Modulator (EOM). The EOM is driven at 3.035 GHz which is the ground hyperfine separation between \( 5\!S_{1/2}, F = 2 \) and \( F = 3 \) states. This creates two sidebands S1 and S2 centered about the drive frequency (see fig. 1) with the upper sideband S1, acting like the probe (P) beam. The other sideband S2 is far away from single-photon resonance and is not in two-photon resonance with any of the other participating beams. Thus in contrast to other experiments [28], this beam is assumed to have negligible effect [20] on CPT coherence. The MODR probe (DR) beam is also derived from the drive, by shifting the drive beam frequency upwards by 121 MHz using an AOM. Part of this beam is used as a local oscillator for heterodyne measurement of the EIT probe beam [29]. In fig. 1, \( \delta_P \) denotes the detuning of the EIT probe beam from the \( a \rightarrow b \) transition and \( \delta_D \) denotes detuning of the drive from the \( a \rightarrow c \) transition. The DR beam addresses the \( 5\!S_{1/2}, F = 3 \) (\( |c\rangle \)) to \( 5\!P_{3/2}, F' = 4 \) (\( |d\rangle \)) transition in the \( N \) level scheme (see fig. 1). Its detuning is denoted by \( \delta_{DR} \). In addition to these optical fields driving transitions between the ground hyperfine states and excited hyperfine states, the ground states \( 5\!S_{1/2}, F = 2 \) and \( F = 3 \) are connected by a microwave field whose detuning from the \( c \rightarrow b \) transition is denoted by \( \delta_m \). Since an electric dipole transition is forbidden between the ground states, the microwave field drives a magnetic dipole transition. Due to the inherent weakness of magnetic field coupling to atomic levels, our rubidium \( ^{85}\text{Rb} \) atoms are placed inside a cylindrical microwave cavity to enhance coupling efficiency. This is shown in the schematic of the experimental setup in fig. 2. Enriched \( ^{85}\text{Rb} \) held in a 5 cm long glass cell is placed inside the cylindrical microwave cavity. To achieve the best \( Q \) value for the cavity, we selected the \( TE_{011} \) mode, which shows a high \( Q \) at a diameter equal to the length of the cavity and gives uniform field over the desired spatial region. The length is 13.5 cm and the diameter is 6.5 cm. The measured \( Q \) value of the cavity is around 1800.
A signal generator generates microwaves at 3.035 GHz. This is coupled into the microwave cavity using a loop antenna after being amplified by a low noise microwave amplifier. Both the drive and the probe beam (P) are copropagating after the EOM, and they pass through the cavity which contains the rubidium atoms. In order to discriminate the drive and probe beam, they are mixed with a local oscillator (LO) beam which is shifted from ω_D by 121 MHz on a beam splitter. All three beams fall on a high bandwidth photodiode where they generate beat frequencies at ±121 MHz around 3.035 GHz. The lower sideband at 3.035 GHz - 121 MHz carries the EIT probe signal. The beat signal from the photodiode is frequency analyzed using a spectrum analyzer. The microwave optical double resonance probe (ω_DR) passes counter to the drive and the probe beam (P). The counter geometry was chosen for easy discrimination of the DR probe from the co-propagating drive and probe beams. The transmission of the DR probe is detected through a photodiode and recorded on an oscilloscope. The entire schematic of the experimental setup is given in fig. 2.

The three optical fields and the microwave field connect the N level system as shown in fig. 1. The microwave signal fed to the cavity and to the EOM, are derived from the same signal generator as is done in [20]. The experiment consists of scanning the microwave frequency over a range of 10 MHz around 3.035 GHz. Consequently the EIT probe frequency also scans around |b⟩ → |a⟩ transition by the same amount. At all times during the experiment, the drive beam which is σ_+ polarized addresses resonantly the |c⟩ → |a⟩ transition (δ_D = 0) and the DR probe which is σ_- polarized addresses resonantly the |c⟩ → |d⟩ transition (δ_DR = 0). During each scan of the microwave frequency, the EIT probe P which is also σ_+ polarized and the DR probe are recorded in a spectrum analyzer and an oscilloscope, respectively.

**Dressed state analysis.** – The closed Λ subsystem in fig. 1, with two optical fields and one microwave field, has been both experimentally and theoretically studied before [17,20]. In such closed systems, the dynamics becomes dependent on the phases of the laser fields and that of induced atomic dipoles. Let Ω_P, Ω_D and Ω_μ represent the Rabi frequencies of the EIT probe field, the drive field and the microwave field, respectively. When Ω_μ = 0, at two-photon resonance with δ_D = δ_P, the drive and probe field drive the system to the well-known population trapped dark state given by

\[
\Omega_D \sqrt{\Omega_D^2 + \Omega_P^2} e^{ik_D z} - \frac{\Omega_P}{\sqrt{\Omega_D^2 + \Omega_P^2}} e^{ik_P z} |c⟩. \tag{1}
\]

Here k_P and k_D represent the wave numbers of the drive and probe fields, respectively.

Conversely, with only the microwave field, the microwave field pumps the atoms into one of the dressed states spin states |S_±⟩ given by

\[
|S_±⟩ = |b⟩ \pm e^{iφ_μ} |c⟩. \tag{2}
\]

There is no microwave propagation phase because of the standing wave nature of the microwave field inside the cavity. Here φ_μ denotes the phase of the microwave field.

When all the three fields are present, the dynamics becomes sensitive to the relative phase φ between the microwave and optical fields. This phase is given by [21]

\[
φ = z(k_D - k_P) - φ_μ. \tag{3}
\]

We note here that the difference between λ_P and λ_D is exactly equal to the ground hyperfine separation at two-photon resonance. As shown in [21], only for special values of φ, (φ = 0, π), there exists dressed states which are decoupled from the excited state. In addition, in a closed Λ system, a time-independent steady-state dynamics is achieved only when [20]

\[
δ_P - δ_D - δ_μ = 0. \tag{4}
\]

At φ = 0, the decoupled state is the dark state of equation (1). This is verified in the experiment reported in [17]. When φ = π, the decoupled state can be either of the spin dressed states |S_±⟩. This results in the absorption of the EIT probe field.

We describe below the consequences of incorporating such a closed Λ system in our N system. In a traditional N system [30] where the microwave is absent, a three-photon resonance condition given by

\[
δ_P - δ_D + δ_DR = 0 \tag{5}
\]
needs to be satisfied for a simultaneous absorption of a probe (P) photon and a DR photon (also known as the signal photon in conventional N systems) through a $\chi^{(3)}$ process. In the case of our N system with a closed $\Lambda$ subsystem, this results in the condition

$$\delta_\mu + \delta_{DR} = 0,$$

which is the microwave optical double resonance condition for the absorption of a microwave photon and an optical photon from the $\Omega_{DR}$ laser. Thus we see that, in an N system with a closed $\Lambda$ subsystem, an MODR absorption resonance captures the essential non-linearity of a three-optical-photon process and renders it phase sensitive. This equivalence is one of the main results of this paper.

**Experimental results.** – A typical experimental run consists of scanning the microwave field over a width of 10 MHz around $c \rightarrow b$ resonance. Since the same microwave source drives the EOM, the EIT probe beam also scans around its resonance $a \rightarrow b$ by the same amount. Thus the detuning $\delta_P$ of the probe field $P$ and that of the microwave field $\delta_\mu$ are equal in magnitude throughout the experiment. During this scan, the transmission of the EIT probe is recorded in a spectrum analyzer. Simultaneously the transmission of the counter DR probe is recorded in an oscilloscope. The typical values for Rabi frequencies for various beams during such an experimental run are, $\Omega_\mu = 0.01, \Omega_P = 0.1, \Omega_D = 1$ and $\Omega_{DR} = 0.05$ with detunings $\delta_\mu = 0$ and $\delta_{DR} = 0$. The microwave Rabi frequency is calculated from the formula [31]

$$\Omega_\mu = \frac{\mu BB_z}{c},$$

$$B_z = \sqrt{\frac{2\mu_0 Q P}{\omega_\mu V}}.$$  

Here $\mu_B$ is the Bohr magneton, $B_z$ is the on axis magnetic field, $Q$ is the quality factor of the loaded cavity, $V$ is the volume of the cavity, $P$ is the power dissipated, $\mu_0$ is the permeability of free space and $\omega_\mu$ is the microwave frequency. Figure 3 shows the EIT probe transmission with and without the microwave field connecting the ground hyperfine states. Figure 3(b) shows the transmission of the EIT probe of our N system under two-photon resonance without the microwave field connecting the ground states. The width of the EIT transmission seen in probe is sub-natural at 2 MHz. The major contribution to this width comes from transit time broadening effect arising from the small 1 mm beam size used in our experiment. In addition, the incoherent ground-state spin-flips happening due to atomic collision with the walls of our Rb cell also contribute to the width. In [20], these spin-flips are reduced due to the presence of buffer gas in their cell volume. Figure 3(a) shows the transmission of EIT probe with the microwave field connecting the ground hyperfine states at a $z$ position where the relative phase $\phi = 0$. We see an enhancement in the peak value of EIT probe transmission as compared with fig. 3(b). We also see a reduction in the width of EIT from 2 MHz to about 1 MHz. We attribute this reduction to originate from the high $Q$ of the microwave cavity. In fig. 3(c) we present the EIT lineshape for a $z$ position where the phase difference from that in fig. 3(a) is $\pi$. We note that there is a narrow central absorption inside the EIT transmission. The narrow spectral nature of this absorption testifies the coherent nature of this feature, as is also noted in [20]. The enhancement and reduction in EIT resonance varies with a periodicity of about 10 cm which is the wavelength of the microwave when at resonance with the $c \rightarrow b$ transition. This is also equal to the frequency of the beat envelop of the optical fields at two-photon resonance.

In fig. 4, we give transmission of the DR probe of our N system for the same experimental parameters as is given in fig. 3. Figure 4(a) gives the DR absorption when both the drive and probe beams are absent. We see a small absorption whenever the microwave field becomes resonant to the ground hyperfine states. This is the familiar microwave optical double resonance involving a ladder configuration of the levels $|b\rangle$, $|c\rangle$ and $|d\rangle$. Figure 4(b) gives the DR absorption when both the drive and EIT probe connecting all the transitions of the N system shown in fig. 1. The DR probe absorption is enhanced over that of fig. 4(a). The inset in the same figure gives the corresponding transmission profile seen in the EIT probe beam. We see a central absorption dip (as in fig. 3(c)) in EIT probe transmission for this $z$ value. Figure 4(c) gives the DR probe transmission at a $z$ value which is about 5 cm away from the corresponding curve.

![Fig. 3: (Colour on-line) Transmission of EIT probe beam (P), with and without the microwave field. Here (b) gives the EIT transmission of the probe (P) in the absence of the microwave field with $\Omega_\mu = 0, \Omega_D = 1, \Omega_{DR} = 0.05$ and $\Omega_P = 0.1$. (a) and (c) give the EIT transmission of probe (P) with the microwave connecting the ground states with strength $\Omega_\mu = 0.01$. The relative position of the cavity and cell between (a) and (c) differ by 5 cm.](image-url)
with the constraint
\[ \sum_i \rho_{ii} = 1. \]  

Using the steady-state density matrix values, we then proceed to numerically integrate Maxwell equations for the slowly varying envelopes of the propagating optical fields. Thus we can find values of all the optical fields at any position \( z \). We have given in fig. 5, the numerical result of this analysis, for the parameters of fig. 4(d). We find good qualitative matching of these two results.

**Discussion.** – In closed \( \Lambda \) systems, there are two processes which form ground-state coherences. One is the microwave field and the other is the difference frequency of the optical fields. Because of the coherent nature of
these fields, the induced dipoles and the phases of the fields are in phase relationship with each other. Therefore it is possible to manipulate the steady-state populations and coherences through the relative phase of the participating fields. Exploiting this feature, the experimental work in [20] showed how the phenomenon of EIT is rendered phase sensitive. We have shown in this paper that a non-EIT probe such as our DR probe can also be rendered phase sensitive if the level configuration encloses a closed Λ subsystem.

Many experimental and theoretical studies of N systems, in the context of coherent non-linear optics at low light levels, have been carried out [7–9]. CPT coherence of the ground states have been fully exploited in such schemes. The DR probe beam in our N system is the traditional signal beam in such coherent non-linear optical schemes. The most important result of this manuscript is the elucidation of the non-linear and phase-sensitive nature of the DR probe response. Such phase-sensitive Kerr non-linearity has wide ranging applications. For example using microwaves, the quantum switching property of an N system [32], can be made phase sensitive. Also, in quantum information transfer which utilises ground-state coherences, one can detect phase changes by looking at the phase-sensitive response in a beam which is not directly taking part in the EIT effect (like our DR probe). Thus potentially, this can be used for quantum error corrections in EIT based quantum information protocols.

Conclusions. – We have experimentally shown in this paper that the presence of a closed Λ subsystem in an N system can induce phase-sensitive non-linear response in a microwave optical double resonance phenomenon. We see that the double resonance probe absorption is enhanced whenever the EIT in the closed Λ system is enhanced and it is suppressed whenever EIT is suppressed. We have thus established for the first time a phase-sensitive change in the reactive non-linear response of an optical probe. More importantly, this optical phase control is effected from an entirely different frequency domain, namely from the microwave domain. We envisage that such a phase-sensitive MODR response in an N system can be utilized for phase control of giant Kerr non-linearities arising in these systems.

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