Destruction of Darkness: Optical Coherence Effects and Multi-Wave Mixing in Rubidium Vapor

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Motivated by the desire to understand atomic coherence effects in multilevel atoms, we have been studying characteristic lineshapes in Lambda and cascade 2-photon transitions with variations in system parameters. We continue to be surprised by the richness of phenomenon that occur when even low-power laser beams interact with reasonably dense atomic vapors. Better knowledge and control of these systems should allow us to take advantages of the novel coherence effects (CPT, EIT, gain without inversion etc.) to develop advanced sensors and sources of radiation, such as: microwave frequency references, short wavelength lasers, and media with high index of refraction or tailored group velocities. Significant research efforts have been directed recently toward the studies of these effects in similar systems, with very interesting results from groups of Arimondo, Hemmer, Ducloy, Haensch, Wynands, Wellegehausen and others. One aspect that distinguishes this work is that we typically use low power cw sources, very high spectral resolution and atomic vapors that are relatively dense. Optically-thick atomic vapors offer potential advantages of long effective interaction lengths, bigger signals and sometimes narrower linewidths. If we can achieve good transparency at high densities then more atoms can contribute to the signal, and the spectral features could be narrowed due to exponential absorption through the cell. In addition, with long interactions dispersive and other nonlinear processes become important and can significantly affect the signal, sometimes interfering with, sometimes enhancing and often creating new structure within the lineshape. This paper focuses on how these nonlinear interactions affect three different experimental configurations.

The first case we consider is the usual lambda-CPT system with an optically thick cell and co-linear propagation. As the optical thickness increases the lineshape of the "dark-line" resonance shows the clear signature of the interfering effects resulting from additional fields generated in the cell by a double-lambda type interaction. The effects of optical pumping and coherences around closed loop double-lambda paths are very important. Theoretical models provide excellent agreement with the experimental results.

More dramatic effects become apparent in this lambda-CPT system if even a small amount of counter-propagating beam (for example, reflection from the cell window) travels back through the vapor cell. With a single laser field incident on the cell, the transmitted beam develops

strong AM modulation at the frequency corresponding to the ground state hyperfine structure. Thus, a 10 mW laser beam tuned near resonance shows large AM modulation sidebands that appear on the transmitted laser beam at the 3.0 GHz ⁸⁵Rb hyperfine splitting. The AM sidebands have narrow linewidths relative to the incident laser carrier (300 Hz FWHM at 3.0 GHz). We have seen as 4% of the transmitted power contained in the frequency shifted sidebands. Higher-order sideband(s) are also observed under some conditions. In contrast to our usual experience with a single laser bean and Rb vapor, the atoms do not get pumped into a "dark state". In fact the system becomes bright. The net effect is to scatter photons from the input laser frequency to upper and lower sidebands.

The second experimental configuration consists of two co-propagating beams near resonance with a cascade 2-photon transition. The goal of this work is to experimentally study the feasibility of short-wavelength sources of coherent radiation. We find that with two input laser fields near 2-photon resonance (780nm + 776nm) the Rb atoms generate a coherent, unidirectional, blue beam out of the vapor cell. This blue beam at 420 nm originates from the second resonance line. This is not the configuration in which we would expect LWI, but this type of signal has been previously observed and studied in some detail using high-power short-pulse lasers sources. We detect as much as 12 microwatts of narrow-band coherent blue output power for 2 x 10mW input power. Detuning the frequencies of the input lasers produces additional coherent output beams at wavelengths corresponding to other Rb transitions, these including: 1.3 microns and 5 microns. We have identified these signals as resulting primarily, from forward 4-wave mixing around closed loop paths of the Rb energy levels.

The third case that we have studied consists of counter-propagating beams near cascade 2-photon resonance. Here we use higher cell temperatures, a strong field on the upper part of the two photon transition propagating in the forward direction, and a weak probe-field on the resonance line propagating in the backward direction. The probe only penetrates a short distance in to cell, but the coherent effects are detected on this beam in reflection from the cell window. This configuration might be attractive for observing large index of refraction or low group-velocity effects. Near resonance, with high atom density near the cell window surface we see the well know "selective reflection" signals, which are again affected by additional multi-wave mixing terms. These two types of interactions can be separated experimentally and we find regimes where the multi-wave mixing terms can be much larger and much narrower than the selective-reflection effects.

In all three of the cases above, we observe very significant variations of the usual 2-photon coherence lineshapes when the cells are optically thick. These effects result mainly from multilevel, multiphoton coherent interactions that cycle around closed loops in the atomic energy levels. In atoms such as Rb, the additional nonlinear terms can dominate the usual 2-photon coherences even at relatively low input power levels. Though different in outcome and manifestation, these effects can, in some sense, be viewed as multi-wave mixing (predominantly multiple 4-wave mixings) enhanced by the underlying 2-photon coherence.