Atomic coherence-state phase conjugation in optical coherent transients
by Zachary Cole

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in Physics
Montana State University

Abstract:
Atomic coherence-state phase conjugation is studied via optical coherent transient phenomena, in Tm+3: YAG. The theoretical framework is a semi-classical perturbative approach to time-domain wave mixing in an inhomogeneously broadened two-level atomic system. Phase conjugation occurs between coherence-state pathways associated with the stimulated photon echo and the so-called 'virtual' echo. The pathway associated with the virtual echo is experimentally shown to exist for causal time domains through its coherence decay and optical processing relations to the stimulated photon echo pathway. A conjecture is presented which links phase conjugation to a correspondence symmetry between spatial and frequency holography.

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APPROVAL

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Zachary Cole

This thesis has been read by each member of the thesis committee and has been found to be satisfactory regarding content, English usage, format, citations, bibliographic style, and consistency, and is ready for submission to the College of Graduate Studies.

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The scientist does not study nature because it is useful; he studies it because he delights in it, and he delights in it because it is beautiful.

– Henri Poincaré

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ABSTRACT

Atomic coherence-state phase conjugation is studied via optical coherent transient phenomena, in Tm^{+3}: YAG. The theoretical framework is a semi-classical perturbative approach to time-domain wave mixing in an inhomogeneously broadened two-level atomic system. Phase conjugation occurs between coherence-state pathways associated with the stimulated photon echo and the so-called 'virtual' echo. The pathway associated with the virtual echo is experimentally shown to exist for causal time domains through its coherence decay and optical processing relations to the stimulated photon echo pathway. A conjecture is presented which links phase conjugation to a correspondence symmetry between spatial and frequency holography.

CHAPTER 1

INTRODUCTION

The investigation of resonant interactions between light and matter has illuminated dynamics which make it possible to store and process the spectral (energetic) character of light in a crystalline environment[1-9]. This is achieved through a quantum mechanical analog to traditional spatial holography, know as spatial-spectral holography (S-S-H), or time and space domain holography. The spectral information of a coherent 'object' light pulse is stored in the absorption spectrum of an inhomogeneously broadened two-level atomic system (IBA) by interfering it with a brief 'reference' pulse inside the medium. A spatial-spectral population grating ensues, which may be illuminated at a later time by a brief 'recall' pulse. The recall pulse stimulates a coherence-state evolution that emits a time-domain replica of the object pulse. This spatial-spectral holographic event is known as the stimulated photon echo (SPE)

Motivation

In the mathematical formulation where the SPE is considered an optical coherent transient (OCT), the coherence-state stimulated by the recall pulse contains multiple terms that are conjugate in their phase evolution. These terms correspond to pathways of the medium's response to waveform perturbations. One set of the coherence state pathways corresponds to the SPE event, while the other set appears to promote an echo event prior to the recall pulse, known as the 'virtual' echo (VE). The VE, being a non-causal event, does not exist. This set of coherence-state paths, called here the virtual coherence (VC), was first postulated in spin echo studies of magnetic resonance phenomena[10].

Although the VE does not exist, the research presented in this thesis indicates that after pulse 3, the medium evolves in every way as if the virtual echo had occurred.

A quantified relationship between the phase conjugate sets of coherence state pathways has, we believe until now, not been demonstrated in a solid material. In an aqueous medium it is possible to heterodyne (phase) detect portions of both the SPE and VE[11]. However, these methods are fundamentally limited in their ability to detect full echo events associated with the conjugate coherence-state pathways and are not in general applicable to a solid-state medium[12].

In this study, echoes resulting from the conjugate paths are recorded and characterized in a crystal lattice environment. Unlike the measurements of reference [11], fully rephased stimulated echo signals are detected. This is achieved through a six wave-mixing technique that introduces another pulse into the medium after the SPE. This has the effect of inverting the phase evolution of the coherence-state pathways such that echo signals associated with all pathways are produced. The conjugate pathways are characterized through their six-wave mixing signals.

The organization of this thesis is as follows: in chapter 2, theoretical considerations, beginning with a general overview of spatial and spatial-spectral holography, are introduced. The remainder of chapter 2 develops time-domain four-wave mixing theory (TD4WM). This approach illuminates the multiple pathways of the medium's response to optical waveform interaction. Experimental techniques and considerations are presented in chapter 3 along with results. Chapter 4 concludes the thesis.

CHAPTER 2

THEORY

Spatial Holography

In classical (spatial) thin film holography, the interference pattern of two beams (object and reference), is recorded in the absorption spectrum of a holographic medium[13] (see Figure 2.1).

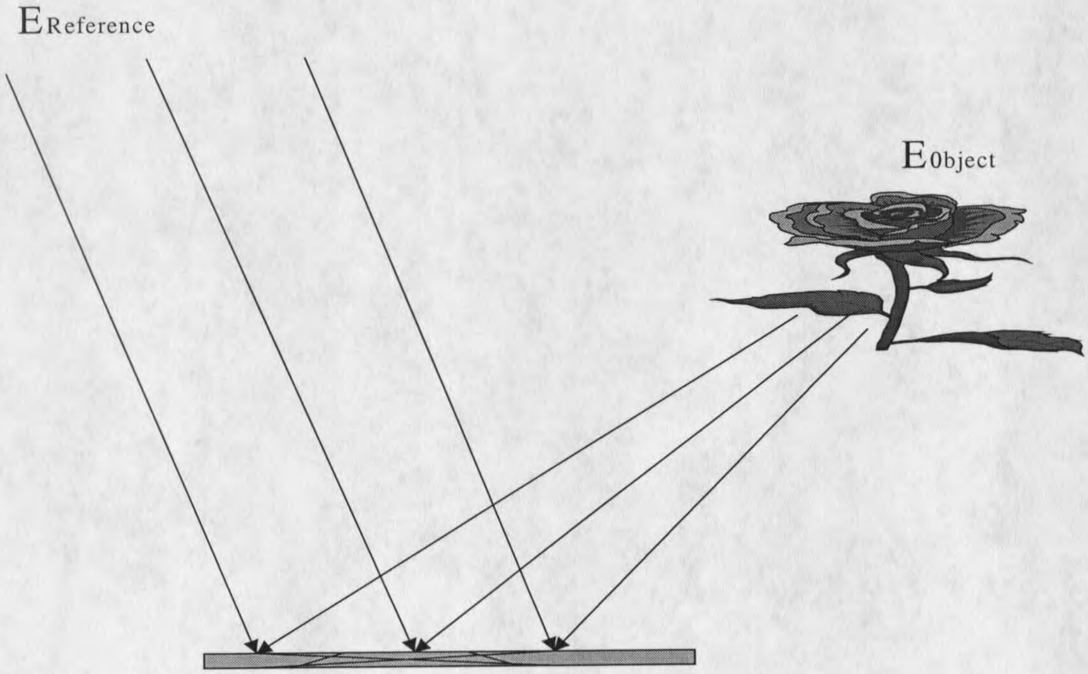

Figure 2.1: The interference between object and reference waveforms is recorded in the holographic medium.

The primary distinction between holography and photography is that the hologram is able to spatially store the phase relationship between the two beams. The interference pattern is stored in the intensity profile of the medium. In complex notation the interference terms of the intensity profile are conjugate in phase, ensuring that the intensity and recorded interference grating is real:

$$\begin{aligned} \text{Intensity} &= \left| E_{reference} + E_{object} \right|^2 \\ &= E_r^{\ 2} + E_o^{\ 2} + E_r E_o e^{i\varphi_{relative}(\mathbf{r})} + E_r E_o e^{-i\varphi_{relative}(\mathbf{r})} \end{aligned} \quad (1)$$

The holographic images are formed by scattering a third 'recall' beam (identical to the reference beam) through the medium. This beam will scatter into two first order waveforms representing a recreation of the object wave and a wavefront that is phase conjugate to the object.

The electric field amplitude of the output may be expressed as:

$$\begin{aligned} \text{Transmision} \propto &\ [\text{non-diffracted terms}] \\ &+ E_r^{\ 2} E_o e^{i\varphi_o(\mathbf{r}_o)} + E_r^{\ 2} E_o e^{-i\varphi_o(\mathbf{r}_o)} e^{i2\varphi_r(\mathbf{r}_r)} \end{aligned} \quad (2)$$

Equation (2) assumes that $E_{recall} = E_{reference} = E_r e^{i\varphi_r(\mathbf{r}_r)}$. The two diffracted terms of Equation (2) are phase conjugate with respect to the object wavefront. The scattered waveforms are known as the real and virtual image. The virtual image spatially diverges away fróm the hologram, appearing to converge behind the scattering center. The real image converges to produce an inverted (pseudoscopic) recreation of the object wavefront, as depicted in Figure 2.2.

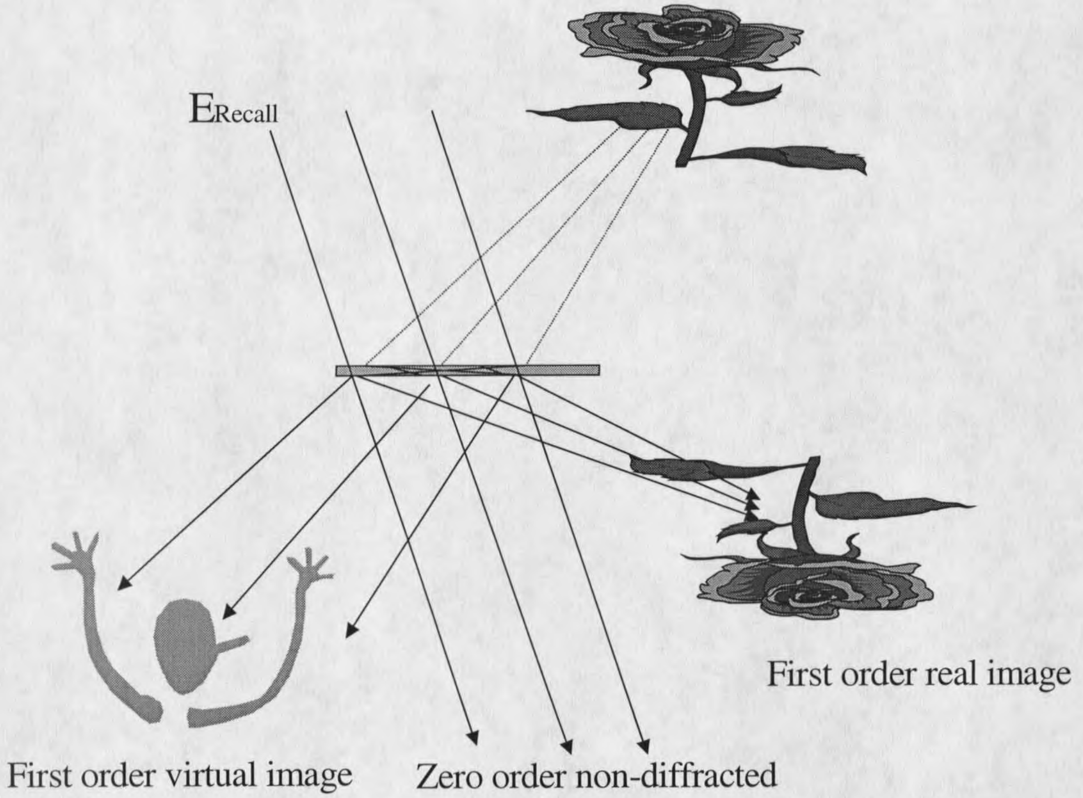

Figure 2.2: Illumination of the hologram at a later time by a 'recall' beam (identical to the reference beam) results in two first-order diffracted waveforms.

Spatial-Spectral Holography

In spatial-spectral holography (SSH), also known as time and space domain holography, the interference of object and reference pulsed waveforms is recorded as a frequency dependent absorption spectrum of the spectral holographic medium. This is achieved by separating the object and reference pulses in time by a delay, τ_{21}. At a later time, a 'recall' pulse interacts with the spatial-spectral holographic grating and a reproduction of the object pulse is emitted, τ_{21} later. The delayed waveform is known as the stimulated photon echo (SPE). The time delay behavior of the echo event is related to the spatial-spectral holographic medium being inhomogeneously broadened in its electronic absorption spectrum.

Spectral Broadening

A crystal lattice, having defects and irregularities, provides slightly different electronic potential environments to the active guest ions, serving to statically perturb their electronic energy states. The ensemble of dopant ions then have a spectral absorption profile broader than the 'natural' homogenous profile of a single ion embedded in an ideal matrix. The system is then inhomogeneously broadened as depicted in Figure 2.3.

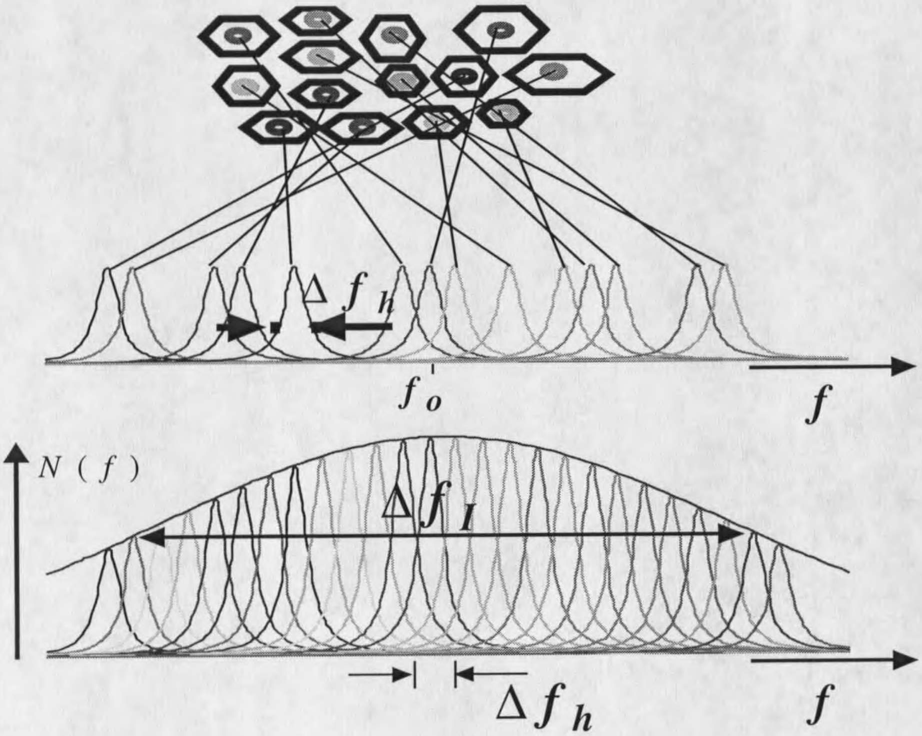

Figure 2.3: Individual absorbers experience slightly different lattice environments resulting in a shift of their resonant energy (above). The overall effect is an inhomogeneous broadening (below).

Due to the random nature of the inhomogeneous broadening, the absorption profile is modeled as a Gaussian distribution with linewidth Δf_I, whereas the homogenous profile, due to dynamic perturbations and uncertainty, is modeled as a Lorentzian distribution with linewidth Δf_H.

Both the static inhomogeneous broadening and the dynamic homogenous broadening contribute to the SPE event, but in very different ways. The homogenous contribution promotes irreversible dynamic relaxation processes and will be considered in Chapter 3. The inhomogeneous broadening promotes coherence-state dephasing (inhomogeneous dephasing) between the various frequency components of the object pulse. This inhomogeneous dephasing is crucial to the time-delayed behavior of the spatial-spectral holography. In order to understand how inhomogeneous broadening is responsible for the time delay of the echo, we view the entire process as time-domain four-wave mixing.

Time-Domain Four Wave-Mixing

The time-domain four wave-mixing process of this work introduces 3 input waveforms to interact with a nonlinear medium, which then emits a new field, the response of the system. The SPE is one such time-domain four-wave mixing (4WM) signal. A typical input scheme in a SPE experiment consists of three temporally unique, angled pulses spatially overlapping in an interaction volume of the IBA. (see Figure 2.4).