ROLE OF FIELD FLUCTUATIONS IN NONLINEAR ABSORPTION

D. S. Elliott, Rajarshi Roy* and S. J. Smith†
Joint Institute for Laboratory Astrophysics, University of Colorado and National Bureau of Standards, Boulder, Colorado 80309

The effect of finite laser bandwidth on nonlinear optical transitions has recently been the focus of theoretical interest [1-4]. No fully quantitative measurements to serve as benchmarks for any of this theoretical work have been provided, however. We have undertaken the development of a technique for superimposing statistically defined phase and frequency fluctuations onto a cw laser beam, thereby making the laser broadband in a fully controllable manner. A systematic quantitative study of interactions of atoms with intense fluctuating fields may then be carried out.

The method being developed is based on modulating the laser beam with random phase and frequency variations, as shown in the block diagram (figure 1). Low-frequency noise is used to randomly frequency modulate a voltage controlled oscillator, and these frequency fluctuations are transferred to the laser beam via an acousto-optic modulator. A traveling wave electro-optic phase modulator is used to place high frequency fluctuations onto the laser beam. Control of the laser bandwidth and bandwidth depends upon accurate shaping and amplitude control of the noise used for these modulation processes. Although fluctuations could in principle be superimposed onto the laser beam with the frequency modulation process alone, or alternatively the phase modulation process alone, major technical problems, which we discuss later, arise, making use of the hybrid system necessary.

*Present Address: School of Physics, Georgia Institute of Technology, Atlanta, GA 30332.

†Staff Member, Quantum Physics Division, National Bureau of Standards.
Fig. 1. Schematic diagram of the hybrid (acousto-optic and electro-optic) system developed to impose fully controlled lineshapes onto highly stabilized laser beams. Bandwidths range up to 35 MHz, with wings fully controlled to 1 GHz from line center.
This technique is capable of generating a laser lineshape (of up to 35 MHz FWHM) which follows a Lorentzian to frequencies greater than 1 GHz from line center. A limited form of the hybrid phase/frequency modulation scheme has been successfully demonstrated, and the complete process should be operational soon. The system will then be applied to a saturated two-level atomic sodium transition in a double optical resonance experiment.

In a double optical resonance experiment, two optical fields, at frequencies \( \omega_a \) and \( \omega_b \) are near resonance with atomic transitions \( |0> + |1> \) and \( |1> + |2> \), respectively. The transition \( |0> + |2> \) is not dipole allowed. Of interest here is the case where the field at \( \omega_a \) is an intense saturating field, and the field at \( \omega_b \) is a weak probe. The ac Stark splitting of the atomic state \( |1> \) results in a double-peaked structure (in photoionization current or in fluorescence) when the probe laser is tuned through the \( |1> + |2> \) transition. In the limit of large detuning of the saturating laser from resonance \( |0> + |1> \), one of these peaks corresponds to a two-photon excitation of state \( |2> \), \( \omega_a + \omega_b = (E_2-E_0)/\hbar \), and the other peak to a two-step excitation via an intermediate state \( |0> + |1> + |2> \). For monochromatic radiation, Whitley and Stroud [5] showed that the two-photon process should dominate and that the peaks should be equal height only when the strong laser is on resonance. The asymmetry in peak height as observed by Moody and Lambropoulos [6], however, was reversed for the range of laser detunings used. Hogan, Smith, Georges and Lambropoulos [7] extended the observations to detunings of several laser bandwidths where they found the peak height asymmetry reverted to normal.

Georges and Lambropoulos [2] were able to explain the reversed asymmetry in terms of laser bandwidth. They used a phase diffusion model with \( \delta \)-correlated frequency fluctuations, finding that when the bandwidth of the Lorentzian laser was larger than the natural atomic line width, the peak height asymmetry should be reversed for all detunings. This enhancement of the two-step process is a result of the overlap of the broadened laser power spectrum with the level \( |1> \), causing direct population of this state. Zoller and Lambropoulos [3] and Dixit, Zoller and Lambropoulos [4] revised the laser model to include exponentially correlated frequency
fluctuations. Since the wings of this power spectrum fall off faster than those of a Lorentzian, the peak height asymmetry, which is reversed for small detunings, reverts to normal for large detunings.

Finally, Nitz, Smith, Levenson and Smith [8] observed double optical resonance with a 17 MHz FWHM laser which had very broad weak wings. In this case, the peak asymmetry was reversed for all detunings observed, but when the wings were eliminated by means of a Fabry-Perot etalon, the asymmetry was reversed only for small detunings.

Our immediate goal is to present quantitative experimental measurements on the effect of a single-mode broadband laser on saturated transitions. In particular, we are studying the $3S_{1/2}, F = 2, m_F = 2 \rightarrow 3P_{3/2}, F = 3, m_F = 3$ transition in sodium. With the broadband saturating laser held at a fixed detuning from resonance, a weak probe laser is tuned through the $3P_{3/2} \rightarrow 4D_{5/2}$ region. The population of the $4D$ state is monitored via the $4P_{3/2} \rightarrow 3S_{1/2}$ fluorescence, and we study the characteristic two-peak structure as a function of laser bandwidth, bandshape and detuning from resonance. We have complete control over both parameters of a phase diffusion type laser field (the amplitude of the frequency fluctuations and the timescale of the frequency fluctuations) and will provide measurements for direct comparison to theoretical results. This paper describes major recent modifications made to a previously reported technique [9] which improve our ability to control the laser power spectrum.

The previous report described a method of broadening a laser power spectrum by randomly frequency modulating a voltage controlled oscillator (VCO) and transferring the fluctuations to the laser using an acousto-optic modulator. It has previously been shown [10] that the process of frequency modulating with Gaussian noise which follows a power spectrum of the form $W_N(f) = W_0[1 + (\omega RC)^2]^{-1}$ results in a broadened carrier described by

$$W_{FM}(\omega) = \frac{A_0^2}{2\pi} \int_0^\infty dt \cos(\omega - \omega) \exp\left[-\frac{(2\pi b)^2}{4} \frac{W}{\sigma} \left[\nu + \nu / \nu_c (e^{-\nu / \nu_c} - 1)\right] \right] (1)$$
where $D$ is the slope of the frequency versus voltage response of the VCO (Hz/volt), $W_0 = 4RC V_{rms}^2$ is the spectral density of the noise at low frequencies (V$^2$/Hz) and $\omega_0$ is the carrier frequency. This modulated power spectrum can be controlled, therefore, by controlling two parameters: (1) the root-mean-square deviation frequency, $DV_{rms}$, and (2) the 3 dB roll-off frequency ($1/2\pi RC$) of the noise power spectrum. The Fourier transform in equation (1) can be evaluated in the two limiting cases of either of these noise parameters dominant. When the root-mean-square deviation frequency is large compared to the 3 dB down frequency, the frequency modulated power spectrum is Gaussian with a FWHM of $\sqrt{2 \ln 2} DV_{rms}$. Note that in this case the power spectrum is independent of noise bandwidth. When $DV_{rms} << 1/2\pi RC$ the laser modulated power spectrum follows a Lorentzian to frequency $1/2\pi RC$ from the carrier, beyond which the power density decreases rapidly. The FWHM in this case is $\pi D^2 W_0$. The model described by Eq. (1) is consistent with that used in the theoretical work of Refs. [3] and [4]. A similar process has been successfully demonstrated as reported in Ref. [9], with two limiting features. These two problems are: (1) the spectrally broadened laser beam is also spatially broadened since the diffraction angle of the acousto-optic modulation depends upon the acoustic frequency, and (2) the HWHM bandwidth of the acousto-optic modulator determines the maximum modulation frequency to which this technique can be applied. These problems have now been overcome, and the solutions are reported here.

Double Pass AOM

Since the angle at which light is diffracted by the moving acoustic waves in an AOM depends upon the acoustic frequency, modulating the acoustic frequency affects the spatial quality of the output of the AOM. This unwanted characteristic can be eliminated by directing the light back into the AOM for a second pass [11]. Light which was diffracted at a large angle, for instance, on the first pass will be diffracted by a large angle on the second pass, as well, exiting anti-parallel to the incident beam (see figure 2). The useful beam can be separated from the incident beam
Fig. 2. The acousto-optic double pass technique. The angle at which the traveling acoustic wave diffracts the optical wave depends upon the acoustic frequency. On the second pass through the crystal, the light returns anti-parallel to the incident beam independent of acoustic frequency.

by introducing a displacement in the direction normal to the wave vectors of the incident light and the acoustic waves [11], or by a polarization technique [12].

To understand the double pass process, it is useful to analyze the interaction between a (single) frequency modulated acoustic wave and the light. The notation used here is the same as that used by Yariv [13] in his analysis of acousto-optic modulation for a fixed frequency process.

The change in refractive index of the crystal due to the acoustic wave is described by

\[ \Delta n(x,t) = \Delta n \cos(\omega_s t - \hat{k}_s \cdot \hat{r} + \delta \sin(\omega_m t - \hat{k}_m \cdot \hat{r})) \]

\[ = \Delta n \sum_{j=-\infty}^{\infty} J_j(\delta) \cos[(\omega_s + j\omega_m)t - (\hat{k}_s + j\hat{k}_m) \cdot \hat{r}] \]

where \( \omega_s, \hat{k}_s, \omega_m, \) and \( \hat{k}_m \) are the carrier frequency, carrier wave vector, modulation frequency and modulation wave vector, respectively. \( \delta \) (=deviation frequency/modulation frequency) is the modulation parameter, \( J_j \) is the Bessel function of order \( j \), and \( \Delta n = (1/2) (\Delta \varepsilon /\varepsilon_0) \) is the amplitude of the refractive index modulation. A macroscopic polarization of the form
\[ \Delta p(r,t) = \frac{2\sqrt{\varepsilon\varepsilon_0}}{\Delta n(r,t)} e(\hat{r},t) \]

does not serve as a source term for the diffracted wave in the wave equation for a
sourceless nonconducting medium:

\[ \nabla^2 e(\hat{r},t) = \mu \frac{\partial e}{\partial t} + \frac{\partial}{\partial t} \left( \frac{\partial}{\partial \hat{r}} \Delta p(\hat{r},t) \right) . \]

For a plane wave

\[ e(\hat{r},t) = \frac{1}{2} E_i(\hat{r}) e^{i(\omega t - \hat{k} \cdot \hat{r})} + c.c. \]

we obtain, describing the gain in the diffracted beam Fourier amplitude, \( E_d \),

\[ -i k \frac{\partial}{\partial r} E_d(r) e^{i(\omega t - \hat{k} \cdot \hat{r})} = \frac{1}{2} \sqrt{\varepsilon\varepsilon_0} \Delta n \sum_{j=-\infty}^{\infty} (\omega_i + \omega_j + j \omega_m)^2 J_j(\delta) \]

\[ \exp\left\{ i[(\omega_i + \omega_j + j \omega_m) t - (\hat{k}_i \cdot \hat{k}_j + j \hat{k}_m) \cdot \hat{r}] \right\} \]

\[ = \frac{1}{2} \sqrt{\varepsilon\varepsilon_0} \Delta n E_i \omega_i^2 \sum_{j=-\infty}^{\infty} J_j(\delta) \]

\[ \exp\left\{ i[(\omega_i + \omega_j + j \omega_m) t - (\hat{k}_i \cdot \hat{k}_j + j \hat{k}_m) \cdot \hat{r}] \right\} \]

\[ = \frac{1}{2} \sqrt{\varepsilon\varepsilon_0} \Delta n E_i \omega_i^2 \exp\left\{ i[(\omega_i + \omega_j ) t - (\hat{k}_i \cdot \hat{k}_j + j \hat{k}_m) \cdot \hat{r}] + \delta \sin(\omega_m t - \hat{k}_m \cdot \hat{r}) \right\} \]

or

\[ k \frac{\partial}{\partial r} E_d(r) = \frac{1}{2} \sqrt{\varepsilon\varepsilon_0} \Delta n \omega_m^2 E_i(r) \]

with a similar expression describing the loss in the incident wave \( E_i \).

The amplitude and time dependence of the diffracted wave is then given by

\[ e_d = \frac{1}{2} E_i(0) \exp(i\omega_i t) (\eta) \]

\[ \exp\left\{ i(\omega_s t + \delta \sin \omega_m t) \right\} + c.c. \]

where \( \eta \) is the net efficiency of the process on passing through the crystal.
When the optical field returns to the AOM for a second pass, the diffracted field is given by

\[ e_{2d} = \frac{1}{2} E_1(0) \exp(i\omega_1 t) \left[ \eta \exp\{i(\omega_s t + \delta \sin \omega_m t)\} \right] \]

\[ \left[ \eta \exp\{i[\omega_s (t+T) + \delta \sin \omega_m (t+T)]\} \right] + c.c. \]

\[ = \frac{1}{2} E_1(0) \exp(i\omega_1 t) \eta^2 \]

\[ \exp\{i[2\omega_s t + \delta \sin \omega_m t (1 + \cos \omega_m T + \delta \cos \omega_m \sin \omega_m T)] + c.c. \]}

where T is the time required for the light to propagate from the AOM to the reflector and back to the AOM. The phase factor \( \exp(i\omega_s T) \) is unimportant and is not retained. When the light returns much faster than the acoustic frequency is able to change, \( T \ll 1/\omega_m \), we see that

\[ e_{2d} = \frac{1}{2} E_1(0) \exp[i((\omega_1 + 2\omega_s) t + 2\delta \sin \omega_m t)] + c.c. \] \quad (2)

In addition to eliminating the spatial spread of the light, the double-pass technique doubles the carrier frequency offset of the output, and doubles the effective modulation parameter of the frequency modulation process. This latter characteristic is beneficial since nonlinearities in the frequency versus voltage response of the voltage control oscillator become less critical. We have verified equation (2) experimentally.

Hybrid Frequency/Phase Modulation Technique

In order to extend the random modulation technique to frequencies greater than the HWHM response of the AOM, a scheme involving a lithium tantalate traveling wave electro-optic phase modulator is used. The ratio of thickness to width of the phase modulators is chosen such that the modulators form 50 \( \omega \) parallel plate transmission lines. Careful mounting of the crystals allows efficient modulation to frequencies greater than 1000 MHz.

Phase modulation using a power spectrum \( p(\omega)/\omega^2 \) is equivalent to frequency modulation using a power spectrum \( p(\omega) \) [14]. Due to this \( 1/\omega^2 \) term in the noise power spectrum for phase modulation, power requirements become
prohibitively high for low frequencies. We therefore employ phase modulation at frequencies greater than 20 MHz, and random frequency modulation using the AOM at frequencies less than 20 MHz. The statistical properties of the fluctuations are unaffected by the use of two noise generators shown in figure 1 since the noise is Gaussian.

Using this hybrid scheme, a Lorentzian 35 MHz FWHM laser line width can be attained using 1 W of noise power for the phase modulation process. For minimal distortion of the noise, the amplifier should have an output capability of greater than 20 W. Since a high power amplifier with a frequency range of greater than 1 GHz would be very expensive, we divide the phase modulation process into two frequency ranges. A low-frequency (20-200 MHz) high power amplifier drives one modulator, while the high-frequency region (200-1000 MHz) is covered by an amplifier of only 4 W output power capability. (Again, the statistical properties of the fluctuation are unaltered by this separation since the noise is Gaussian.) This hybrid system using the AOM and the phase modulator is capable of randomly modulating the laser over frequencies from less than 10 KHz to greater than 1 GHz. Another hybrid system for phase and frequency control has been used by Camy and Hall for just the opposite purpose — the removal of intrinsic FM noise of the laser. These workers narrowed an argon laser from 200 kHz to 6 kHz with the AOM frequency control loop alone and obtained a 2 kHz line width when the EOM was activated [15].

State of the Experiment

We have currently successfully demonstrated the hybrid technique using the AOM and the intermediate frequency phase modulator for producing a laser lineshape which follows a Lorentzian to frequencies greater than 200 MHz. The extension to 1 GHz will be implemented within a few months. Application of the controlled single-mode broadband optical field will then be applied to the double optical resonance experiment described above.
Acknowledgments

We gratefully acknowledge useful suggestions from J. Hall and L. Hollberg and technical assistance from K. Arnett. This research was supported by the Department of Energy, Office of Basic Energy Sciences.

References


