V. QUANTUM ELECTRONICS

A. Laser Applications

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Our interest is in the general area of precision measurements related primarily to spectroscopy and the interaction of radiation with matter. Since many of the measurements that we perform require lasers with high spectral purity and excellent long-term stability, considerable effort is devoted to improving laser performance.

1. FREQUENCY STABILIZATION OF MULTIWATT CONTINUOUS-WAVE ARGON LASERS

U. S. Air Force – Office of Scientific Research (Contract F44620-71-C-0051)

Shaoul Ezekiel, Lloyd A. Hackel, Richard P. Hackel

This research is motivated by the need for long-term stabilized lasers for applications in earth strain seismometry, optical communication, precision spectroscopy and fundamental measurements in experimental relativity.

During the past few years we investigated the use of an I_2 molecular beam as a reference for the long-term stabilization of an argon ion laser. A long-term stability of one part in 10^{13} in an integration time of 200 seconds has been achieved.[1] The argon lasers used in the experiments were homemade with power output of the order of milliwatts.

Recently, we have extended our effort to the stabilization of commercially made multiwatt argon lasers such as the 15 W Spectra-Physics laser. The short-term absolute jitter of ~30 MHz that is inherent in such a laser was reduced to less than 50 kHz by locking the laser frequency to an external Fabry-Perot cavity. A wide bandwidth (1 MHz) feedback loop was necessary, and was made possible by using an intracavity electro-optic phase shifter. Long-term stabilization was accomplished by locking the reference Fabry-Perot to a hyperfine transition observed in an I_2 molecular beam.

The performance of the laser was measured by heterodyning two high-power argon lasers, each independently stabilized to adjacent I_2 lines. A stability of 1 part in 10^{13} was achieved in an integration time of 300 seconds.

The residual short-term jitter of 50 kHz is set by the jitter of the reference Fabry-Perot and not by the bandwidth of the fast feedback loop. The laser jitter relative to the reference Fabry-Perot is less than 10 kHz. At present, the long-term stability is limited by the stability of the alignment of the argon laser cavity and this is being investigated.

Such a stabilized laser was used to perform ultrahigh resolution spectroscopy in I_2 (one part in 10^{10}) and to measure spacings between hyperfine lines with a precision of several parts in 10^{12}.

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2. ULTRAHIGH-RESOLUTION SPECTROSCOPY USING MOLECULAR BEAMS

U. S. Air Force — Office of Scientific Research (Contract F44620-71-C-0051)

Shaoul Ezekiel, Lloyd A. Hackel, Richard P. Hackel

The high spectral purity of well-controlled lasers makes it possible, in principle, to observe atomic and molecular spectra with extremely high resolution. Since Doppler broadening is typically several hundred megahertz in the visible region, techniques for reducing Doppler and collisional broadening must be employed. The use of molecular beams is a convenient way of substantially reducing both Doppler and collisional broadening.

Using laser molecular-beam techniques, we have measured the complete hyperfine structure of the P(13) and R(15) 0-43 transitions in I_2^{127} with unprecedented precision. We have observed all 21 of the $\Delta F = \Delta J$ (strong) components and 16 of the 18 $\Delta F = 0$ (weak) components in the I_2 hyperfine structure. The frequency spacings of the lines were measured by heterodyning two argon lasers that are independently stabilized to different I_2 hfs components. The data were fitted with a standard deviation of 6 kHz to a theoretically calculated spectrum. We have included in the hfs Hamiltonian terms arising from nuclear electric quadrupole, magnetic spin-rotation, and tensor nuclear spin-spin, scalar nuclear spin-spin and nuclear magnetic octopole interactions.

The observation of the magnetic octopole interaction, which to our knowledge is the first such observation in excited electronic states, determines a coupling strength that is the difference between upper and lower state values. An improvement in the data of at least an order of magnitude would be necessary in order to determine the constants for each state. Such an improvement is certainly possible, since the present long-term stability of the molecular beam-stabilized argon laser is 1×10^{-13} for integration times of 200 seconds.

As the precision increases further, we could consider the hexadecapole interaction as the next significant term in the Hamiltonian. In fact, we have already calculated the matrix elements for this interaction and have estimated the coupling strengths. Preliminary attempts to fit the data by including the hexadecapole term showed that the effect of this interaction is indeed small. The shift in the line positions is expected to be of the order of a few hundred hertz so that an improvement of one to two orders of magnitude in the data would be required to observe the effect of the hexadecapole.

With the advent of dye lasers, it should now be possible to perform similar highly precise measurements over the entire optical region of the spectrum. We have already

observed the dye laser excited hyperfine structure in the molecular beam of I_2 with a resolution of one part in 10^9. The measurement of the coupling strengths that are due to higher order nuclear moments would therefore make possible a more complete study of electric and magnetic field distributions in excited electronic states of atoms and molecules.[1-3]

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3. K. H. Casleton, L. A. Hackel, and S. Ezekiel, "Magnetic Octupole Interaction in I_2," Proc. Second Laser Spectroscopy Conference, Megève, France, June 23-28, 1975.

3. SINGLE-FREQUENCY CONTINUOUS-WAVE DYE LASER

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Joint Services Electronics Program (Contract DAAB07-75-C-1346)

Shaoul Ezekiel, Robert E. Grove, Frederick Y-F. Wu

The primary objective in this program is the development of an extremely stable, low-jitter, single-frequency cw dye laser for use in a variety of applications such as optical communication and ultrahigh-resolution spectroscopy, and for studying fundamental interactions between radiation and matter.

We have been investigating the use of a free-flowing jet stream of rhodamine 6G dissolved in ethylene glycol as a gain medium. The dye laser cavity is of the astigmatically compensated variety and single-frequency operation is accomplished with the aid of one intracavity tunable etalon and one fixed etalon.

After a careful examination of the causes of laser jitter, we were successful in reducing the laser linewidth to less than 200 kHz. This narrow linewidth was demonstrated by a scanning Fabry-Perot interferometer with a 1-MHz instrument width. We used the tunable dye laser to excite extremely narrow resonances (700 kHz FWHM) in a molecular beam of I_2 which, as far as we know, is the highest resolution reported with a dye laser. Furthermore, we locked the dye laser to one of the hyperfine transitions in a molecular beam of I_2 and achieved a long-term stability of 6 parts in 10^{13} for an integration time of 25 seconds.[1, 2]

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4. HIGH-RESOLUTION MEASUREMENTS OF THE SPECTRUM OF RESONANCE FLUORESCENCE INDUCED BY A MONOCHROMATIC FIELD

Joint Services Electronics Program (Contract DAAB07-75-C-1346)

Shaoul Ezekiel, Robert E. Grove, Frederick Y-F. Wu

There is considerable interest, both theoretical[1,2] and experimental,[3,4] in the spectrum of resonance fluorescence induced by a monochromatic field. We have conducted experiments to measure the fluorescence spectra with improved resolution for both on- and off-resonance excitation. We have observed also the spectrum of elastically scattered light under weak field excitation.

The experimental arrangement is a single-frequency cw dye laser orthogonally exciting an atomic beam of sodium, which is collimated to 2.5 mrad. The laser-induced fluorescence from the $3\,^2P_{3/2}(F=3) \rightarrow 3\,^2S_{1/2}(F=2)$ transition is collimated to 4 mrad and analyzed by a Fabry-Perot interferometer with 2 MHz width along a mutually orthogonal direction. The arrangement is similar to that used by Schuda et al.[3] The dye laser used in the present experiments[5] produces 30 mW of single-frequency power at 5890 Å. The spectral width of the laser is less than 250 kHz and the amplitude stability is better than 2%.

At the interaction region, the laser beam has a Gaussian field distribution with a waist diameter of 2 mm, and is unrestricted by apertures.

The orthogonality of the atomic beam with respect to the direction of observation was carefully determined by sending a laser beam along that direction and observing the sodium absorption linewidth. The alignment was adjusted at low laser power until the linewidth was within 1 MHz of the natural width of 10 MHz.

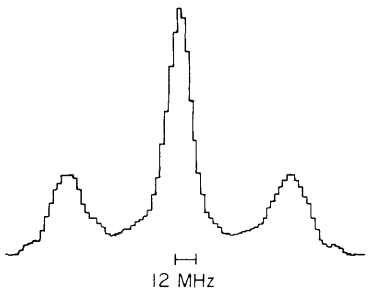

Fig. V-1.

Spectrum of fluorescence in sodium atomic beam induced by circularly polarized light. Frequency scale: 12.5 MHz/box.

Fluorescence spectra with linearly polarized excitation have been included elsewhere.[6] Figure V-1 shows the on-resonance fluorescence spectrum at high field intensities for circularly polarized light. The predicted three-peak spectrum is clearly observed. Our data will be compared with the results of other groups,[3,4] in particular with regard to any asymmetry in the observed spectra. The high resolution of the present data permits a more precise measurement of the widths of the sidebands and the ratio of elastic-to-inelastic scattering as a function of laser intensity.

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5. NEW OPTICAL RATE SENSOR

Joint Services Electronics Program (Contract DAAB07-75-C-1346)

Shaoul Ezekiel, Salvatore R. Balsamo

We propose to develop a new optical rate gyroscope using a passive ring Fabry-Perot interferometer as the rotation sensing element, based on the Sagnac effect. The clockwise and counterclockwise lengths of the cavity, which depend on inertial rotation, will be measured by means of two independently controlled laser frequencies. One laser is locked to the center of the cw resonance and the other to the ccw resonance of the cavity.

To eliminate the effect of laser-frequency jitter, we propose to use one laser whose output is shifted by two independently controlled acousto-optic frequency shifters.

For a square ring of 10 cm on a side, a cavity resonance width of 1 MHz and laser power of 1 mW, it should be possible to detect earth rate in an integration time of 0.5 ms and milliearth rate in several hundred seconds.

Aside from applications in navigation, we propose to examine the possibility of measuring earth rotation to better than one part in 10^8, using a cavity 10 m on a side and a 10-W argon laser. Such measurements would give information on polar wobble, continental drift, and changes in the length of the day. The connection between earthquakes and earth wobble may also be examined. Application of such a device in experiments related to general relativity will also be considered.

V. QUANTUM ELECTRONICS

B. Gaseous Lasers

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1. ULTRAVIOLET LASERS

Joint Services Electronics Program (Contract DAAB07-75-C-1346)

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Ultraviolet lasers utilizing Raman anti-Stokes emission from rare gas metastable states offer high energy storage capacity by eliminating the radiative decay channel. We are investigating the possibility of obtaining stimulated vacuum ultraviolet radiation in atomic xenon. Our recent results[1] on the interaction of a CO_2 laser with a helium plasma indicate that our theory of laser-induced optical satellites correctly predicts the experimentally observed spectra. Using these results, an experiment is now in progress in which a CO_2 laser interacts with an argon-xenon plasma formed in a narrow capillary discharge tube. Energy is collisionally transferred from Ar_2^* molecules to the Xe $^1P_1$ level which decays to the metastable Xe $^3P_2$ level. This process, along with other energy transfer mechanisms in rare gas systems, has been studied previously.[2]

A CO_2 laser operating on the R(22) transition of the 10.6 μm branch induces transitions between the Xe $^3P_2$ and $^3P_1$ levels accompanied by the anti-Stokes emission of an ultraviolet photon at 1470 Å with the xenon atom returning to the $^1S_0$ ground state. Calculations show that a CO_2 laser power of 1 MW cm^{-2} and a population inversion of 10^{15} cm^{-3} should result in a gain of 1% cm^{-1} at 1470 Å.

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2. LASER FREQUENCY LOCKING

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Joint Services Electronics Program (Contract DAAB07-75-C-1346)

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A TEA CO_2 laser exhibits in general a frequency chirp[1] within each pulse. When a pulsed laser oscillator is required with a spectrum that is Fourier-limited (i. e., its width conforms with that dictated by the width of the pulse) the chirp must be suppressed. At least two approaches can be pursued:

(a) The chirp has been found to be minimized when the axial TEA laser oscillates in a single axial mode and the mode is near the center of the gain line.[1,2] Through external feedback sensing of the Fabry Perot resonances of the TEA cavity by a stable cw laser, the length of the TEA cavity can be controlled so as to position the axial mode at the center of the gain line of the laser medium. A system of this kind is now being assembled.

(b) The TEA laser can be used as an amplifier in a master oscillator-power amplifier (MOPA) arrangement. This second scheme has the advantage that the frequency can be chosen through choice of the frequency of the master oscillator, and can be made phase coherent from pulse to pulse. The range of frequency control or sweep is maximized by maximizing the pressure in the cw laser. For this reason, we are investigating a capillary CO_2 laser as part of a continuing program on this laser.[3]

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V. QUANTUM ELECTRONICS

C. Nonlinear Phenomena

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1. SHORT LASER PULSES

Joint Services Electronics Program (Contract DAAB07-75-C-1346)

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Work in the past has led to a compact analytic theory of saturable absorber (passive) mode locking of lasers.[1-4] There has also been progress toward a transient theory of the buildup of mode-locked pulses. In future work we shall exploit the theory by applying it to more complicated systems of interest, and will seek experimental confirmation along the following lines.

a. Combined passive and active mode locking can produce pulses with prescribed timing. It also offers the advantage of shortness of pulses that has only been achievable with passive mode locking (see Part II, Sec. V-C.1).

b. Passive mode locking of a semiconductor laser in an external cavity will be attempted.[4]

c. We shall seek improvement of transient passive mode locking by prebleaching the saturable absorber through injection of another mode-locked laser.

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Part II. Detailed Progress Reports

GENERAL PHYSICS

V. QUANTUM ELECTRONICS

C. Nonlinear Phenomena

1. COMBINED PASSIVE AND ACTIVE MODE LOCKING

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Joint Services Electronics Program (Contract DAAB07-75-C-1346)

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Active mode locking of lasers produces optical pulses with a periodicity prescribed by the mode-locking modulation. Passive mode locking can produce much shorter mode-locked pulses than active mode locking, but has the disadvantage that the timing of the pulses is not controlled. Hence it is of interest to study the potential of combined passive and active mode locking for achieving the advantages of each. The new theories of mode locking developed by the author are suited to an analytic treatment of a laser system incorporating both kinds of mode locking.[1-3]

As expected, we find that the active mode locking can impose the timing of the pulses as long as the "natural" period of the passive mode locking is within certain limits. These limits will be established.

We start with the equations describing the modification of a pulse after one passage through the cavity[1,2]

$$v_{n+1}(t) = v_n(t-T_R) - \frac{\omega_o T_R}{2Q}\left[1 + \frac{Q}{Q_A(t)} + m(t) - g\left(1 + \frac{1}{\omega_L^2}\frac{d^2}{dt^2}\right) + \frac{g}{\omega_L}\frac{d}{dt}\right] v_n(t-T_R). \tag{1}$$

Here $v_n(t)$ is the pulse envelope at one of the end mirrors after the n^{th} passage, T_R is the cavity round trip time, ω_o is the optical carrier frequency, Q is the empty cavity Q, $1/Q_A$ is the time-dependent contribution to the inverse Q of the saturable absorber, m(t) is the modulation of the inverse cavity Q by the active mode-locking modulation, g is the normalized gain of the medium, and ω_L is the laser linewidth.

Equation 1 states that the envelope after the $(n+1)^{st}$ passage is a changed and delayed version of the envelope after the n^{th} passage. The cavity loss changes the amplitude by $-[\omega_o/2Q]\,T_R v_n$, the absorber by $-[\omega_o/2Q_A(t)]\,T_R v_n$, and the laser medium by

$$\frac{\omega_o}{2Q} T_R \left\{ g\left(1 + \frac{1}{\omega_L^2}\frac{d^2}{dt^2}\right) - \frac{g}{\omega_L}\frac{d}{dt}\right\} v_n.$$

The laser medium has dispersion causing a "diffusion" in time expressed by the second-order operator, and a (small) change of the round trip time expressed by the first-order operator.

If the combined active and passive mode locking is successful, the pulse after the

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$(n+1)^{st}$ passage will be a replica of the n^{th} pulse, advanced with respect to the cavity round trip delay T_R by δT:

$$v_{n+1}(t) = v_n(t-T_R+\delta T). \tag{2}$$

Note that δT will be negative if there is a delay rather than an advance.

Expanding $v_n(t-T_R+\delta T)$ to first order in δT and setting

$$\delta = \frac{2Q\omega_L \delta T}{\omega_o T_R} + g, \tag{3}$$

we have the equation for the steady-state pulse shape v(t):

$$\left[1 + \frac{Q}{Q_A} + m - g\left(1 + \frac{1}{\omega_L^2}\frac{d^2}{dt^2}\right) + \frac{\delta}{\omega_L}\frac{d}{dt}\right] v(t) = 0. \tag{4}$$

If the saturable absorber reacts instantaneously to the pulse power passing through it, we may set approximately[2]

$$\frac{Q}{Q_A} \cong \frac{Q}{Q_A^o}\left(1 - \frac{v^2}{P_A}\right) \equiv q\left(1 - \frac{v^2}{P_A}\right), \tag{5}$$

where the pulse envelope amplitude v is so normalized that v^2 is the instantaneous optical power, P_A is the saturation power of the absorber, and Q_A^o is the small-signal value of Q_A.

The active mode-locking modulation is[1]

$$m = 2M(1 - \cos \omega_M t), \tag{6}$$

where 4M/Q is the peak-to-peak modulation of the inverse Q. Equation 4, with the introduction of (5) and (6) is a nonlinear differential equation with time-variant coefficients. This equation is difficult to solve in general. We may obtain physically meaningful approximate solutions rather easily by a perturbation approach. We note that the temporal modulation of the pulse by the saturable absorber is much stronger than that by the active modulation if the pulsewidth τ_p is much less than T_R. The shape of the pulse will only be affected weakly by the active mode locking, whereas its timing will be strongly affected, provided that the active mode locking is successful in controlling the timing. We may treat (4) in two successive steps. We solve (4) in the absence of modulation, m = 0, and obtain the solution[2] of passive mode locking with a fast saturable absorber:

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$$v(t) = \frac{v_o}{\cosh \frac{(t-t_o)}{\tau_p}}, \tag{7}$$

where t_o is the time of occurrence of the peak of the pulse and

$$1 + q - g = \frac{g}{\omega_L^2 \tau_p^2} \tag{8}$$

$$q \frac{v_o^2}{P_A} = \frac{2g}{\omega_L^2 \tau_p^2} \tag{9}$$

$$\delta = \delta_o = 0 \tag{10}$$

or $\delta T_o = - \frac{g}{\omega_L} \frac{\omega_o}{2Q} T_R$. $|\delta T_o|$ is the delay of the pulse with respect to the round trip time T_R because of the laser dielectric loading. The repetition period of the laser pulse train is equal to the round trip time as modified by the laser medium. Thus δ is a measure of the deviation of the round trip time from that determined by cavity and laser medium.

Next, we introduce m as a perturbation. As a consequence, the solution v(t) will change to $v(t) + \delta v(t)$, the gain will change to $g+\delta g$ and the time advance parameter δ will cease to be zero; δ is of the order of M. This second step may be treated as the first-order term of a Taylor's series expansion in M in which the terms δv, δg, and δ are all of first order in M. Retaining only first-order terms in (4), we obtain a differential equation for δv:

$$- (1+q-g) \left[1 - \frac{6}{\cosh^2 \left(\frac{t - t_o}{\tau_p} \right)} - \tau_p^2 \frac{d^2}{dt^2} \right] \delta v - \frac{\delta}{\omega_L} \frac{d}{dt} v$$

$$+ \delta g \left(1 + \frac{1}{\omega_L^2} \frac{d^2}{dt^2} \right) v - 2M(1 - \cos \omega_M t)\, v = 0 \tag{11}$$

where we have used (8) and (9) and [1,2]

$$\delta g = - \frac{g}{1 + \frac{P}{P_L}} \frac{2 \int v(t)\, \delta v\, dt}{T_R P_L}, \tag{12}$$

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where the $(\delta g/\omega_L)\ dv/dt$ has been incorporated in $(\delta/\omega_L)\ dv/dt$. This means that δ measures the time advance of the pulse with respect to the cavity round trip time as determined in the presence of the actual saturation of the laser medium.

If $\tau_p \ll T_R$, then the cosine function may be approximated in the time interval occupied by the pulse by $\cos \omega_M t_o - \omega_M (t-t_o) \sin \omega_M t_o$. Equation 11 is a linear differential equation with time-dependent coefficients and a "driving" term. It can be solved by expanding δv in terms of the complete set of eigenmodes[4] of the operator in the brackets, which are well known:

$$\left[1 - \frac{6}{\cosh^2\left(\frac{t-t_o}{\tau_p}\right)} - \tau_p^2 \frac{d^2}{dt^2}\right] u_k = E_k u_k. \tag{13}$$

The lowest eigenvalue is $E_o = -3$, the next is zero. The following eigenvalue is +1 and forms the start of a continuum of eigenfunctions. The eigenfunction u_o is symmetric and u_1 is antisymmetric with respect to t_o. They are

$$u_o = \sqrt{3/4}\ \frac{1}{\cosh^2\left(\frac{t-t_o}{\tau_p}\right)} \tag{14}$$

$$u_1 = \sqrt{3/2}\ \frac{\sinh\left(\frac{t-t_o}{\tau_p}\right)}{\cosh^2\left(\frac{t-t_o}{\tau_p}\right)}. \tag{15}$$

Note that u_1 is proportional to dv/dt, the time derivative of the steady-state passively mode-locked pulse. The symmetric eigenfunctions are coupled via the δg term, the antisymmetric eigenfunctions are mutually uncoupled, and a simple equation is obtained for the amplitude of each. The eigenfunction u_1, which corresponds to a time shift in the steady-state solution, has zero eigenvalue. The equation for the amplitude of this eigenfunction can only be balanced when the term

$$\frac{\delta}{\omega_L} \int_{-\infty}^{\infty} \frac{dv}{dt}\, u_1\, dt$$

cancels the term

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$$-2M\omega_m \sin \omega_M t_o \int_{-\infty}^{\infty} (t-t_o)\, u_1 v\, dt,$$

or when

$$\delta = -3\,\omega_L \omega_M \tau_p^2 M \sin \omega_M t_o. \tag{16}$$

This means physically that the antisymmetric portion of m(t), $2M\omega_M(t-t_o) \sin \omega_M t_o$, produces an antisymmetric change of the pulse. Part of this antisymmetric change is a simple time delay of the pulse. Timing control is established when the time delay of the pulse with respect to its natural repetition period is equal to the time delay required to make the pulse repetition frequency equal to the frequency of the forced modulation. If we denote by δT_M the active modulation-produced advance of the pulse as compared with the round trip time of the cavity modified by the laser medium, then from (16)

$$\delta T_M = 3\,\frac{\omega_o T_R M}{2Q}\,\omega_M \tau_p^2 \sin \omega_M t_o. \tag{17}$$

The maximum allowed advance or delay $|\delta T_{max}|$ is

$$|\delta T_{max}| = 3\,\frac{\omega_o T_R M}{2Q}\,\omega_M \tau_p^2.$$

If the detuning $\delta\omega_M (=-[\delta T_M/T_R]\omega_M)$ of the active modulation[4] is greater than $3\,\omega_M^2 \tau_p^2 \frac{\omega_o}{2Q} M$, then the period of the pulses will not be "locked." When locked, pulses occur at phase $\omega_M t_o$ of (17), deviating the more from zero the greater the "detuning" of the active modulation.

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