# Analytical Solutions to the Bloch Model for Multidimensional Coherent Spectroscopy with Gaussian Pulse Envelopes


Christopher L. Smallwood[1], Travis M. Autry[1,2], and Steven T. Cundiff[1,2,3,*]

[1]JILA, University of Colorado and National Institute of Standards and Technology, Boulder, CO 80309, USA
[2]Department of Physics, University of Colorado, Boulder, CO 80309, USA
[3]Department of Physics, University of Michigan, Ann Arbor, MI 48109, USA
* cundiff@umich.edu



We present third-order analytical solutions to the optical Bloch equations under the assumption of perturbative laser pulses with Gaussian envelopes. The treatment shows that off-resonant interactions can affect both signal amplitude and phase.


A number of theoretical models have been developed to facilitate the analysis of multidimensional coherent spectroscopy (MDCS) [1-5]. Most commonly, these are based on approximating the exciting laser pulses as delta functions to facilitate analytical manipulation, or by maintaining finite pulse envelopes and calculating response function integrals numerically. The former approach benefits from simplicity, but suffers from the fact that delta-function pulses have infinitely large bandwidth, and are consequently incapable of capturing the physics of off-resonant laser excitation. The latter approach benefits from accuracy, but its numerical aspect may inhibit the practitioner's ability to develop physical intuition.

Here we generalize the delta-function pulse approach, obtaining analytical frequency-domain solutions to the optical Bloch equations for perturbative optical fields with finite-duration, Gaussian pulse envelopes. For off-resonant excitations, phase and amplitude information can both be modified relative to the delta-function pulse solution (Fig. 1), illustrating the potentially nontrivial character of driven oscillator modes in MDCS.

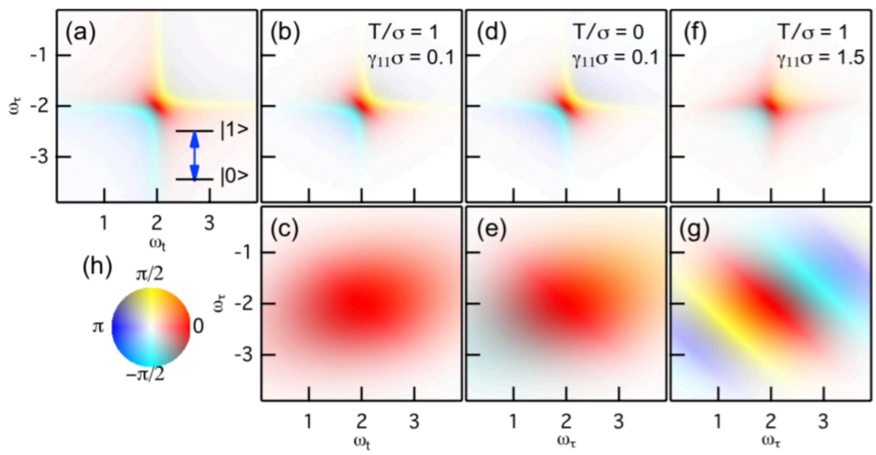


Fig. 1. Comparison between a delta-function one-quantum MDCS spectrum and a transform-limited pulse one-quantum MDCS spectrum for the interaction of light with a two-level system. **(a)** Delta-function pulses. **(b)** Transform-limited Gaussian pulses. The laser center frequency $\omega_0$ equals the atomic resonance frequency $\omega_{10}$, and has a duration $\sigma = 1$. The dephasing rate is $\gamma_{10} = \gamma_{01} = 0.2$. The inverse lifetime is $\gamma_{11} = 0.1$. The spectrum corresponds to population delay $T = 1$. **(c)** Finite-pulse effect, defined as the quotient of (b) and (a). **(d)-(e)** Same as (b) and (c), with $T = 0$. **(f)-(g)** Same as (b) and (c), with $\gamma_{11} = 1.5$. **(h)** Color scales are derived from a two-dimensional hue/lightness legend, where lightness corresponds to signal amplitude and hue corresponds to complex phase.


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