



FRONTIERS IN OPTICAL COHERENT AND ULTRAFAST SCIENCE

A NATIONAL SCIENCE FOUNDATION PHYSICS FRONTIER CENTER AT
THE UNIVERSITY OF MICHIGAN AND THE UNIVERSITY OF TEXAS AT AUSTIN

RANDALL LABORATORY, 500 EAST UNIVERSITY, ANN ARBOR, MI 48109-1120
PHONE (734)763-4932 FAX (734)764-5153

<http://www.umich.edu/~focuspc>

DIRECTOR
PHILIP H.
BUCKSBAUM
phb@umich.edu
ASSOC. DIRECTOR
PAUL BERMAN
pberman@umich.edu

ADMINISTRATOR
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mamurn@umich.edu

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A nugget from FOCUS:

Calculation of Nonlinear optical signals in liquid solutions via the generalized quantum master equation.

Being J. Ka, Ming-Liang Zhang and Eitan Geva

The goal of this ongoing project is the development of a general methodology for simulating the quantum dynamics in liquid solutions and its ultrafast spectroscopic signature. To this end, we have introduced a new and general theoretical framework for simulating the dynamics of the electronic state in processes that involve dynamics on multiple electronic surfaces [J. Chem. Phys. **119**, 12063 (2003)]. The central quantity in this new approach is the memory kernel, which accounts for the effect of the nuclear degrees of freedom (“the bath”) on the electronic degrees of freedom (“the system”). Once the memory kernel is known, it can be substituted into the generalized quantum master equation (GQME), which is subsequently solved to obtain the dynamics of the electronic state. The new approach represents an important breakthrough because of its ability to deal with arbitrary system-bath coupling strengths, while still restricting the input required for calculating the memory kernel to two-time correlation functions.

We have made several important advances in this research project over the last year:

1. We have been able to generalize and simplify the theoretical framework, so as to make it applicable for arbitrary choices of initial state and projection operator. This development makes significantly extends the range of applicability of the methodology.
2. We have developed a new GQME-based framework for calculating nonlinear spectroscopic signals, and used it for simulating different four-wave mixing experiments, such as two and three pulse photon echo experiments, in liquid solutions.

Ongoing and future work will focus on applications to coherent control experiments in liquid solutions.