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Time-Resolved Vibrational Spectroscopy

The contributions of vibrational spectroscopy, especially to the study of molecular structure, have been extremely well established throughout the twentieth century. The advent of time-resolved vibrational spectroscopy (TRVS) in the early 1970s provided the experimental basis for extending the measurement of vibrational spectra beyond stable molecules to encompass reactive species (molecules, radicals, ions, excited electronic states) having lifetimes as short as a few picoseconds. As TRVS methodologies have evolved in sophistication, the complexity of the molecular systems (e.g., proteins) and the physical processes (e.g., nonlinear phenomena) examined also has increased significantly.

The articles appearing in this special issue of the *Journal of Physical Chemistry A* provide an opportunity to view the enormous progress made in recent years in both developing TRVS methodologies and their applications in a wide variety of research areas. Much of the progress can be traced to the rapidly increasing experimental capabilities derived from new and more versatile laser systems, the quantitative measurement and interpretation of nonlinear optical phenomena, and advances in detector technology.

The breadth of applications for TRVS methodologies is no less impressive. The papers presented in this issue encompass studies of (i) vibrational dynamics derived from photophysical phenomena, (ii) photochemical reaction mechanisms in organic and inorganic molecules, (iii) structural changes in the intermediates appearing during in vitro biophysical reactions, (iv) coherent phenomena associated with nonlinear optical and molecular properties, and (v) two-dimensional imaging of reactive systems along vibrational coordinates. The diversity and sophistication of these papers not only highlight the advances made in recent years by researchers using TRVS techniques but also provide a perspective on the scientific horizon accessible to TRVS techniques in the future.

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