Abstract

In 1991 we introduced the LHD as a new method in time-resolved frequency domain spectroscopy. This method is the frequency domain equivalent of the pump/probe technique used in picosecond spectroscopy. With this method, available measurement frequencies are no longer limited by the optical detection devices (phototubes, etc.) as in the conventional frequency domain techniques used in fluorescence detection. With higher frequencies, we can study faster dynamic processes. Ultimately, we hope to achieve subpicosecond resolution. In our previous presentation, we demonstrated the possibility of heterodyning in the sample. The major strengths of the method were the high signal-to-noise ratio, and the ability to work with relatively low laser powers. Here, we present the continued development of the LHD technique. There are three main areas of development. First, we include the effect of molecular rotations on the measurement. Second, we are now using two synchronously pumped dye lasers (pulse width of 10 ps) as our light sources. These shorter pulses (previous pulse widths were 100 ps) have allowed us to extend our measured frequency range. Finally, we are now using a new sample flow system. The dye jet we used previously was ineffective for non-viscous solvents. Our new cell is usable with virtually any solvent. We show the results of our dynamical studies for rose bengal, and other samples. This work is supported by NIH grant PHS-P41-RR03155.