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Single-shot real-time spectroscopy of permanent structural and chemical change in solids

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Time-resolved measurements of solid samples in the process of irreversible change pose special challenges since the common practices of signal averaging and measurement repetition with different temporal delays or windows are often impractical. Ideally, the entire time-dependent sample response can be recorded in real time, during a single event. Here we present two types of time-resolved measurements of solid-state structural and chemical evolution that are conducted in real time, on a single-measurement basis. The first is femtosecond time-resolved transient absorption or reflection spectroscopy, using a single excitation laser pulse and 400 distinct probe pulses with different temporal delays. Real-time measurements of ultrafast, permanent structural and chemical rearrangements in semimetals, semiconductors, and organic molecular crystals have been conducted using this method, which is based on the use of orthogonally oriented staircase “echelon” elements to split an incident probe beam into a 20 x 20 array of spatially and temporally distinct pulses that are passed through the sample and then imaged onto a CCD camera for data extraction. The second measurement, conducted on a single-shot basis using a GHz-bandwidth photodetector and oscilloscope for nanosecond time resolution, involves optically generated and monitored acoustic waves whose properties reveal the mechanical state of a sample. Both of these measurements are of particular interest in the case of shock loading. Measurements on samples under laser or gas gun driven shock loading will be discussed.