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Ultrafast Dynamics in Vanadium Dioxide: Separating Spatially Segregated Mixed Phase Dynamics in the Time-domain[1]

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In correlated electronic systems, observed electronic and structural behavior results from the complex interplay between multiple, sometimes competing degrees-of- freedom. One such material used to study insulator-to-metal transitions is vanadium dioxide, which undergoes a phase transition from a monoclinic-insulating phase to a rutile-metallic phase when the sample is heated to 340 K. The major open question with this material is the relative influence of this structural phase transition (Peirels transition) and the effects of electronic correlations (Mott transition) on the observed insulator-to-metal transition. Answers to these major questions are complicated by vanadium dioxide's sensitivity to perturbations in the chemical structure in VO_2. For example, related V_xO_y oxides with nearly a 2:1 ratio do not demonstrate the insulator-to- metal transition, while recent work has demonstrated that W:VO_2 has demonstrated a tunable transition temperature controllable with tungsten doping. All of these preexisting results suggest that the observed electronic properties are exquisitely sensitive to the sample disorder. Using ultrafast spectroscopic techniques, it is now possible to impulsively excite this transition and investigate the photoinduced counterpart to this thermal phase transition in a strongly nonequilibrium regime. I will discuss our recent results studying the terahertz-frequency conductivity dynamics of this photoinduced phase transition in the poorly understood near threshold temperature range. We find a dramatic softening of the transition near the critical temperature, which results primarily from the mixed phase coexistence near the transition temperature. To directly study this mixed phase behavior, we directly study the nucleation and growth rates of the metallic phase in the parent insulator using non-degenerate optical pump-probe spectroscopy. These experiments measure, in the time- domain, the coexistent phase separation in VO_2 (spatially separated insulator and metal islands) and, more importantly, their dynamic evolution in response to optical excitation.

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