Coherent two-dimensional spectroscopy of a Fano model¹ FE-LIPE POULSEN, Department of Chemistry, University of Copenhagen, DANIEL FINKELSTEIN-SHAPIRO, TNU PULLERITS, Division of Chemical Physics, Lund University, THORSTEN HANSEN, Department of Chemistry, University of Copenhagen — The Fano line shape arises from the interference of two excitation pathways to reach a continuum. Its generality has resulted in a tremendous success in explaining the line shapes of many one-dimensional spectroscopies: absorption, emission, scattering, conductance, photofragmentation applied to very varied systems: atoms, molecules, semiconductors, and metals. Unraveling a spectroscopy into a second dimension reveals the relationship between states in addition to decongesting the spectra. Femtosecond-resolved two-dimensional electronic spectroscopy (2DES) is a four-wave mixing technique that measures the time evolution of the populations and coherences of excited states. It has been applied extensively to the dynamics of photosynthetic units, and more recently to materials with extended band structures. In this paper, we solve the full time-dependent third-order response, measured in 2DES, of a Fano model and give the system parameters that become accessible.

¹Lundbeck Foundation