Simple Analysis of Two-Dimensional Terahertz Spectroscopy

Toshiaki Hattori
Institute of Applied Physics, University of Tsukuba
hattori@bk.tsukuba.ac.jp

Recent progresses in generation of intense terahertz (THz) pulses have enabled us to observe nonlinear THz response of various materials. We expect soon we will see experiments of various types of time-resolved nonlinear THz spectroscopy. In this presentation, I summarize expected features of signals of two-dimensional (2D) time-domain THz spectroscopy using a simple phenomenological classical model.

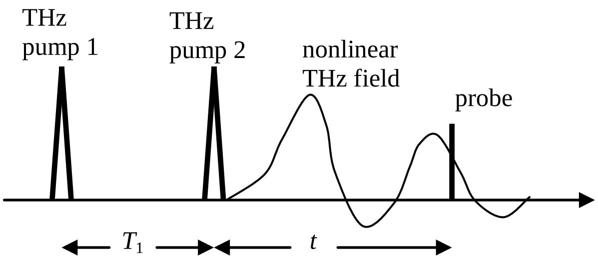

Fig. 1 Time sequence of two-dimensional THz spectroscopy measurements

Time sequence of experiments assumed are shown in Fig. 1, where the short THz pulses excite the sample at time $-T_1$ and 0, and the nonlinear THz field is observed at time t as a function of $-T_1$ and t. The resonant mode is treated classically, and three origins of nonlinearity, anharmonicity, nonlinear coupling, and nonlinear damping, are considered. In noncentrosymmetric systems, second-order processes are the lowest-order, whereas third order is the lowest in centrosymmetric systems. The theoretical treatment is closely related to that of the fifth-order Raman scattering [1], and a brief description of 2D THz spectroscopy was first given in [2]. Examples of the results for a single mode system are depicted in Fig. 2. The theoretical treatment can be easily extended to include effects of inhomogeneity, multiple modes, and mode coupling, and information that is not accessed by linear spectroscopy is expected to be observed by nonlinear measurements.

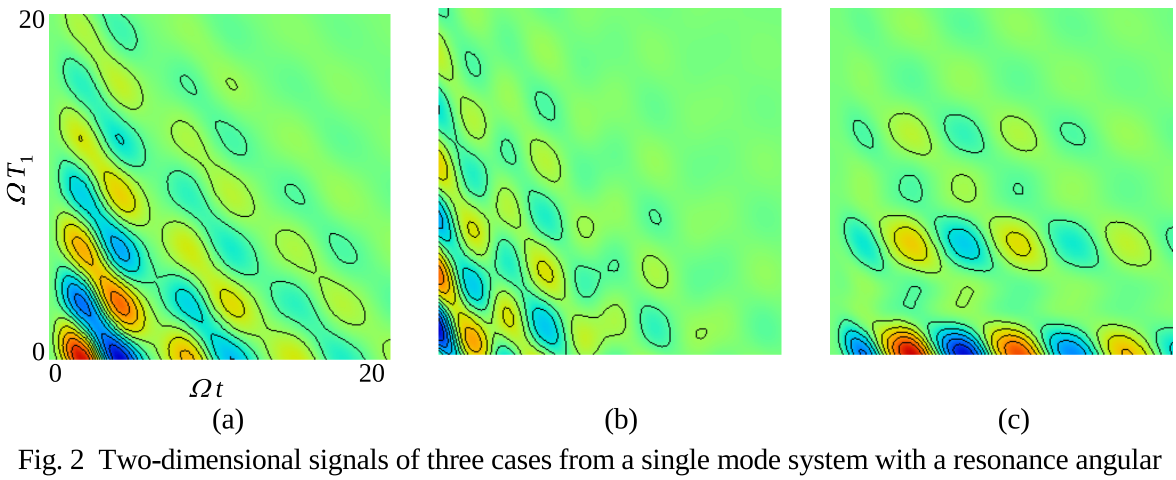

Fig. 2 Two-dimensional signals of three cases from a single mode system with a resonance angular frequency Ω excited by two delta-function THz pulses. (a) Second-order signal from a system with anharmonicity. (b) Second-order signal from a system with nonlinear coupling. (c) Third-order signal from a system with anharmonicity.

[1] Y. Tanimura and S. Mukamel, J. Chem. Phys. **99**, 9495 (1993); and succeeding papers by Tanimura's group.
[2] K. Okumura and Y. Tanimura, Chem. Phys. Lett. **295**, 298 (1998).