非線形時間領域2次元テラヘルツ分光：実験の提案と理論解析モデル

Nonlinear Time-Domain Two-Dimensional Terahertz Spectroscopy: Proposal of Experiments and a Theoretical Model

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Recent progresses in generation of intense terahertz (THz) pulses have enabled us to observe nonlinear THz response of various materials. We will soon see experiments of various types of time-resolved nonlinear THz spectroscopy. In this presentation, I summarize expected features of signals of nonlinear two-dimensional time-domain THz spectroscopy (2D THz-TDS) using a simple phenomenological and classical model [1].

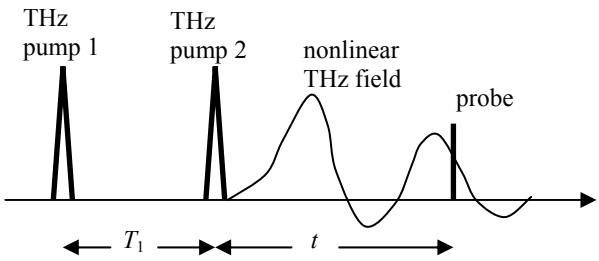
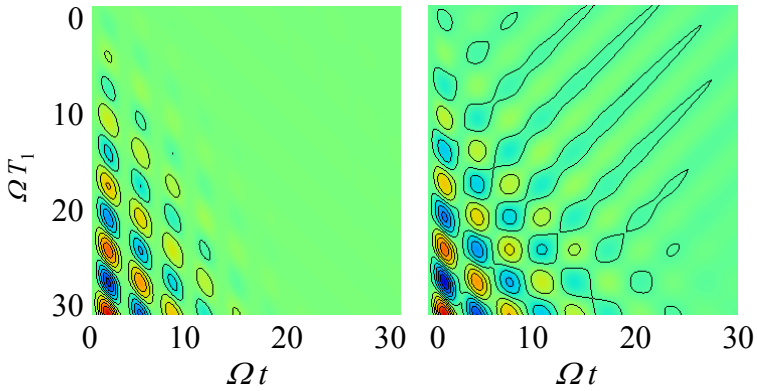


Fig. 1 Time sequence of two-dimensional time-domain THz spectroscopy measurements

Time sequence of experiments assumed is shown in Fig. 1, where short THz pulses excite the sample collinearly at times $-T_1$ and 0, and 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. Second-order nonlinear processes in noncentrosymmetric media and third-order one in centrosymmetric media were considered. The theoretical treatment is closely related to that of fifth-order Raman scattering [2], and a brief description of 2-D THz spectroscopy was already given [3]. Examples of the results for a homogenous and inhomogeneous system with the same linear spectrum are depicted in Fig. 2. The figure clearly shows that the 2D THz-TDS can clarify the inhomogeneity, which is not accessed by a linear spectroscopy. The present theoretical treatment can easily be extended to include effects of multiple modes and mode coupling.



(a) Homogeneous system (b) Inhomogeneous system

Fig. 2 Example of two-dimensional signals of a homogenous and inhomogeneous system with the same linear spectrum with a resonance angular frequency Ω excited by two delta-function THz pulses.

[1] T. Hattori, J. Chem. Phys. **133**, 204513 (2010).
[2] Y. Tanimura and S. Mukamel, J. Chem. Phys. **99**, 9495 (1993).
[3] K. Okumura and Y. Tanimura, Chem. Phys. Lett. **295**, 298 (1998).