



Review

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Nonlinear optical physics at terahertz frequency

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Abstract: Terahertz (THz) waves have exhibited promising prospects in 6G/7G communications, sensing, nondestructive detection, material modulation, and biomedical applications. With the development of high-power THz sources, more and more nonlinear optical effects at THz frequency and THz-induced nonlinear optical phenomena are investigated. These studies not only show a clear physics picture of electrons, ions, and molecules but also provide many novel applications in sensing, imaging, communications, and aerospace. Here, we review recent developments in THz nonlinear physics and THz-induced nonlinear optical phenomena. This review provides an overview and illustrates examples of how to achieve strong THz nonlinear phenomena and how to use THz waves to achieve nonlinear material modulation.

Keywords: terahertz; nonlinear optics; light-matter interaction

1 Introduction

Light-matter interaction (LMI) has been a long-discussed topic in optics and condensed-matter physics [1]. In the early studies, LMI at visible and infrared frequencies were mainly discussed since lasers at such frequencies are highly developed. LMI indicates how light can polarize, heat, melt, and ablate the electrons, ions, or molecules within most materials. However, the significant difference in mass of

these microparticles causes an individual energy scale. By considering the resonant frequencies of these different particles, we find that each particle can easily interact with light at a similar frequency but is minimally affected by light far away from the resonant frequency. The Lorentz oscillator equation and energy band theory provide the fundamental basis for investigating traditional LMI that involves high frequency light.

For LMIs between high frequency light and crystal solids, the heavy ions are less possibly to contribute to such fast oscillations. Therefore, the traditional approaches assume that the ions are fixed in the crystal, only electrons can respond to such fast electromagnetic oscillations. The motion of the ions is usually taken as a perturbation to the electronic polarizations, which is known as Born-Oppenheimer approximation [2], [3]. Nevertheless, when the pump beam becomes low-frequency light such as terahertz (THz) or microwave, the ionic polarization would dominate the whole LMI process. In this case, Born-Oppenheimer approximation is not valid anymore. Especially, for the THz pump light (about 0.1 ~ 10 THz), at which the optical phonon frequency for majority crystal materials is located, strong coupling between the input THz waves and the optical phonons forms a new quasiparticle, stimulated phonon polaritons, which plays unneglected role in LMI at the THz frequency and also causes large THz nonlinearity in polar crystal solids [4]. In Figure 1, we show the dominated LMI mechanisms through various wavelengths. Notably, some physical processes, including Raman/Brillouin scattering, although the pump light could be at visible or infrared, are beyond Born-Oppenheimer approximations.

Besides, many low-dimensional materials, such as graphene, are also found to behave very large THz nonlinearity due to its Dirac electrons at the special band structures. Metamaterials/metasurfaces, plasmons, and superconductors have also been options to achieve THz nonlinearity.

In this review, we report some states of the art of the nonlinear LMI process at THz frequency. A series of nonlinear phenomena at THz frequency or induced by THz waves in recent years are displayed, including frequency conversion, Kerr effect, nonlinear absorptions, as well as THz modulating ions, molecules, and electrons in materials to realize

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