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Terahertz Nonlinear Optics in Semiconductors

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***Abstract*—We demonstrate the nonlinear optical effects – self-phase modulation and saturable absorption of a single-cycle THz pulse in a semiconductor. Resulting from THz-induced modulation of Drude plasma, these nonlinear optical effects, in particular, lead to self-shortening and nonlinear spectral breathing of a single-cycle THz pulse in a semiconductor.**

I. INTRODUCTION AND BACKGROUND

We present the nonlinear optical effects such as saturable absorption (SA) and self-phase modulation (SPM) of a single-cycle THz pulse in a doped semiconductor. The nonlinearity arises from the electron plasma response to the ponderomotive potential of the strong-field THz pulse, which leads to electron heating and intervalley electron scattering. This produces ultrafast modification of electron plasma frequency via increase of average electron effective mass (see Fig. 1). The complex-valued dielectric function $\hat{\varepsilon}(\omega)$ of a semiconductor in the presence of free carriers is described by a well-known Drude model:

$$\hat{\varepsilon} = \left(n + \frac{i\alpha c}{2\omega}\right)^2 = \varepsilon_{dc} - \frac{\omega_p^2}{\omega^2 - i\omega/\tau} \qquad (1)$$

where n and α are frequency-dependent refractive index and power absorption coefficient, ω_p – plasma frequency, and τ is electron momentum scattering rate. Plasma frequency

$$\omega_p = (Ne^2/\varepsilon_0 m)^{1/2} \qquad (2)$$

where N is free carrier density, e is elementary charge, ϵ_0 is the vacuum permittivity, and m is the effective mass.

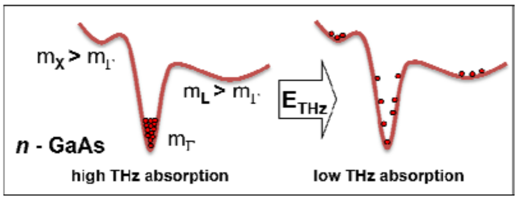

Fig. 1. Mechanism of THz nonlinearity in doped semiconductors by carrier heating in THz field.

It is apparent from eqs. (1) and (2), that at constant carrier density, the increase of the electron effective mass leads to reduction of the plasma frequency, and hence to the reduction of absorption coefficient and to the change in the refractive index. The heating of low-effective-mass Γ-valley electrons in the ponderomotive potential of a strong-field THz pulse leads to intervalley scattering in the energy and momentum space – the population of higher-energy satellite valley (L- and X-valleys) featuring larger effective masses. This naturally leads to an increase in the *average effective mass* of the electron population, and hence to a reduction of the plasma frequency of a semiconductor. The principle of such as THz-induced nonlinearity in a doped semiconductor is illustrated in Fig. 1.
As a result, the complex dielectric function of a semiconductor undergoes dramatic modification *during* the interaction with the THz pulse, leading to its nonlinear propagation. In this study the n-GaAs was chosen as a sample, however any semiconductor with complex band structure will produce the THz nonlinearity via the same mechanism.

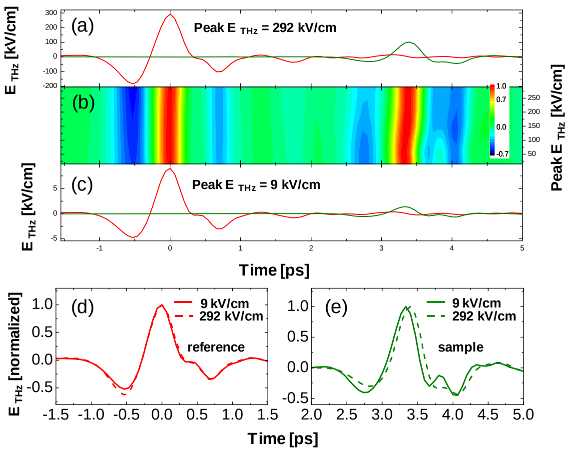

Fig. 2. THz SPM in n-GaAs in the time domain. Peak E_{THz} of reference THz pulse in (a) is 292 kV/cm, and in (c) is 9 kV/cm. (b) Reference and sample pulses normalized to their maxima, for the whole range of peak E_{THz} of 9 - 292 kV/cm. (d,e) Normalized reference (d) and sample (e) pulses for the cases of peak E_{THz} of 9 kV/cm and 292 kV/cm, demonstrating the SPM.

II. RESULTS

In our studies we have used the nonlinear THz time-domain spectroscopy (NL THz-TDS) based on the strong-field THz emitter - the tilted pulse front pumped lithium niobate [1]. Our experiments were performed in a traditional transmission configuration, and for each measurement two THz waveforms – reference (THz propagation through vacuum) and sample (THz propagation through vacuum and sample) were measured. The THz peak field strength was controllably attenuated in the range 9 – 292 kV/cm using crossed wire-grid polarizers.

The optical nonlinearities are observed directly in the time domain (see Fig. 2), and characterized in the frequency domain (Fig. 3).

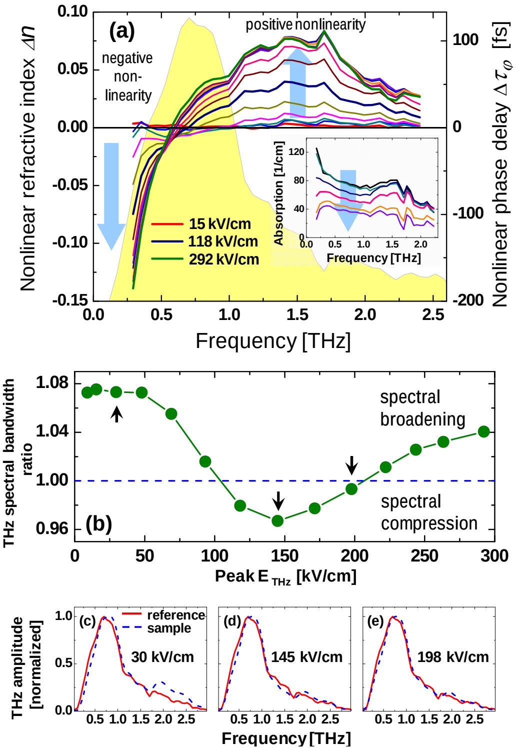

Fig. 3. (a) THz SPM and saturable absorption in n-GaAs in spectral domain. Nonlinear contribution to refractive index Δn, and corresponding nonlinear phase delay as a function of frequency and peak E_{THz}. Inset: spectrally resolved saturable absorption. The amplitude spectrum of the reference THz pulse is shown as a background. (b-e) Nonlinear spectral broadening and compression of a THz pulse in *n*-GaAs. (b)The ratio of effective bandwidths of the sample and reference pulses, as a function of peak E_{THz}. (c-e) Examples of spectral dynamics at selected peak THz field values.

In particular, we present the effects of *self-shortening* of a single-cycle THz pulse [2], and *THz self-phase modulation* (SPM) observed on a single-cycle waveform, with sub-cycle time resolution [3], as shown in Fig. 2. The THz SPM, similar to the nonlinear optics demonstrated in the infrared and visible spectral ranges, results in *nonlinear spectral breathing* of the THz waveform. Further, we have found that the sign of the THz-range refractive index nonlinearity in doped semiconductor can be both positive and negative. In fact, we have discovered the co-existence of positive and negative *refractive index nonlinearity* within the spectral bandwidth of a single-cycle THz waveform [3]. This is quite a unique situation in nonlinear optics, though hardly unexpected given an inherently ultrabroadband nature of any single-cycle waveform.

All our findings, including the co-existence of index nonlinearity of different signs within the spectrum of the same THz waveform, can be well described within the Drude plasma model. For example, we have found that the point of zero THz refractive index nonlinearity is defined by (but is not equal to) the electron momentum relaxation rate τ in the semiconductor, as a result of the transition between low-frequency Hagen-Rubens regime featuring a larger refractive index, and higher frequency conductivity featuring a reduced values of *n* in the presence of free carriers.

In summary, the effects of saturable absorption, self-phase modulation, pulse self-shortening, and nonlinear spectral breathing were demonstrated in nonlinear propagation of a single-cycle THz waveform through a doped semiconductor. The THz nonlinearity is caused by the nonlinear response of the free carriers to the strong THz fields, leading in particular to inter-valley scattering.

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