12. Transport at optical frequencies

Electromagnetic waves and trains of photons

An external electromagnetic wave leads to the motion of charged particles, electrons and nuclei. Let us assume the harmonic time dependence of the electric field intensity at a given location in space,

$$ \tag{12.1} $$

E_0 is its amplitude. The usual simplification consists in a simple space-time dependence of the electric field, e.g., the plane wave of the wavector k_0:

$$ \tag{12.2} $$

The basic characteristics of the wave are:

electric intensity	E (V/m),
frequency	$f = \omega/2\pi$ (Hz),
wavelength	$\lambda = c/f$ (mm, μm, nm),
wavenumber	$W = 1/\lambda$ (cm^{-1}).

Quantum behavior of the wave requires adding the

photon energy	$\hbar\omega$ (eV),
momentum	$\hbar\omega/c$ (eVs/m).

Conversions of the wavelength – photon energy – wavenumber:

(12.3)

The interaction of charged particles with the electromagnetic wave is governed by the electric intensity E. However, the signals of the detectors of the optical wave are proportional to the intensity I, which is the time average of the power flow (Poynting vector of the classical description of electromagnetic waves),

(12.4)

An instructive numerical example:

the wave with the electric field intensity of 1 V/m has the intensity of 1.33 mW/m^2;
the wave with the intensity of 1 mW/μm^2 has the electric field amplitude of 8.68E5 V/m. This is to be compared with the electric field intensity of an elementary charge at the distance of 0.1 nm, which amounts to 1.44E11 V/m.

The classical picture of the electromagnetic waves fails rather frequently. Taking the quantum nature of the waves leads to the intensity I and power P of an optical beam given by

(12.5)

Numerical example:

the red line of HeNe laser with the wavelength λ = 632.8 nm consist of the 1.959 eV quanta;
the power of 1 mW of this laser beam is carried by 3.19×10^{18} photons per second;
state-of-the-art detectors are limited by the dark noise of about 10 elementary charges, which can be generated by a slightly larger number of photons; consequently, it is relatively easy to observe the linear detector response, proportional to the number of incoming photons.

Electric polarization of matter

The motion of electrons and nuclei is typically asynchronous with the driving force of the electric field, comprising a "phase shift" between the electric field intensity and the induced dipolar moment (and the corresponding electrical current). The polarization is typically strongly dependent on the frequency ω. Suitable complex quantities can contain information on both amplitudes and phases.

The response to weak fields is linear:

$$ \tag{12.6} $$

We use the symbol of D for electric displacement, P for polarization, χ for susceptibility, ε for permittivity ("dielectric function"), j for the induced current density, and σ for conductivity.

The basic response functions are (complex) permittivity and conductivity. Their values for a given frequencies are usually termed "optical constants". In the physical models of the response, the preferred optical constants are the real part of conductivity, or the imaginary part of permittivity, since they are proportional to the absorbed energy (resulting from the product of photon energy and the probability of it being absorbed). On occasion, a suitable quantity is also the negative inverse of the permittivity – in particular, in the description of the collective plasma excitations. The propagation of light waves is most conveniently described using the (complex) refractive index, which is the square root of the permittivity. Its imaginary part quantifies the damping of waves.

Interrelations between response functions ("optical constants")

Optical Constant (symbol)	Real part	Imaginary part
conductivity $(\sigma = \sigma_1 + i\sigma_2)$	$\sigma_1 = \omega\varepsilon_0\varepsilon_2$	$\sigma_2 = -\omega\varepsilon_0(\varepsilon_1 - 1)$
dielectric function $(\varepsilon = \varepsilon_1 + i\varepsilon_2)$	$\varepsilon_1 = 1 - \sigma_2/(\omega\varepsilon_0)$	$\varepsilon_2 = \sigma_1/(\omega\varepsilon_0)$
	$\varepsilon_1 = n^2 - k^2$	$\varepsilon_2 = 2nk$
refractive index $(N = n + ik)$	$n = \sqrt{(\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2})/2}$	$k = \sqrt{(-\varepsilon_1 + \sqrt{\varepsilon_1^2 + \varepsilon_2^2})/2}$
	$n = \varepsilon_2/(2k)$	$k = \varepsilon_2/(2n)$
negative inverse of dielectric function $(-\varepsilon^{-1})$	$-\varepsilon_1/(\varepsilon_1^2 + \varepsilon_2^2)$	$\varepsilon_2/(\varepsilon_1^2 + \varepsilon_2^2)$

physical models of optical response
propagation of optical waves
collective (plasma) phenomena

The response to optical waves is causal (the induced polarization cannot occur before the arrival of the wave. This leads to the validity of the following integral relations between the real and imaginary parts of the response functions:

$$\varepsilon_1(\omega) - 1 = \frac{2}{\pi} \int_0^\infty \frac{\Omega \varepsilon_2(\Omega)}{\Omega^2 - \omega^2} d\Omega \ , \quad \varepsilon_2(\omega) = \frac{\sigma_0}{\omega \epsilon_0} - \frac{2\omega}{\pi} \int_0^\infty \frac{\varepsilon_1(\Omega) - 1}{\Omega^2 - \omega^2} d\Omega$$

<div align="right">(12.7a)</div>

$$\sigma_1(\omega) = \frac{1}{\pi} \int_{-\infty}^\infty \frac{\sigma_2(\Omega)}{\Omega - \omega} d\Omega = \frac{2}{\pi} \int_0^\infty \frac{\Omega \sigma_2(\Omega)}{\Omega^2 - \omega^2} d\Omega$$

$$\sigma_2(\omega) = \frac{-1}{\pi} \int_{-\infty}^\infty \frac{\sigma_1(\Omega)}{\Omega - \omega} d\Omega = \frac{-2\omega}{\pi} \int_0^\infty \frac{\sigma_1(\Omega)}{\Omega^2 - \omega^2} d\Omega$$

<div align="right">(12.7b)</div>

$$n(\omega) - 1 = \frac{2}{\pi} \int_0^\infty \frac{\Omega k(\Omega)}{\Omega^2 - \omega^2} d\Omega \ , \quad k(\omega) = \frac{-2\omega}{\pi} \int_0^\infty \frac{n(\Omega) - 1}{\Omega^2 - \omega^2} d\Omega$$

<div align="right">(12.7c)</div>

Several models of the optical response

Classical damped harmonic oscillator (bound charges)

We describe the polarization via the displacement of charges u, using the damping force proportional to the velocity. The resulting spectral profile is Lorentzian.

The equation of motion reads

$$(12.8)$$

It is solved by the displacement

$$(12.9)$$

having the amplitude

$$(12.10)$$

which leads to the polarizability

$$(12.11)$$

Assuming the volume density of the oscillators of N , the susceptibility and permittivity of their ensemble are

$$(12.12)$$

$$(12.13)$$

Both functions resonate around the eigenfrequency of the oscillator in the case of weak damping ($1/\tau << \omega_0$).

Classical Drude model (free carriers)

The absence of the restoring force in Eq. (12.8) leads to the response of free carriers; this is achieved by setting the resonance frequency ω_0 to zero. The „resonance" of the free carrier gas occurs at zero frequency, following the dielectric function

$$(12.14)$$

whose imaginary part diverges for zero frequency. Note that the real part remains finite,

$$(12.15)$$

and assumes negative values for large N and/or τ. The low-frequency behavior is described more conveniently using the complex conductivity. Its imaginary part vanishes at zero frequency, and the real part is finite (see 10.10, be careful when using different unit systems):

$$\sigma_1(0) = \frac{e^2\tau}{m} \ . \tag{12.16}$$

Quantum description – transitions due to harmonic perturbation

The energy taken from a harmonic electromagnetic wave in a unit volume per unit of time is linked to the imaginary part of the dielectric function,

$$\text{(12.17)}$$

proportional to the time average of the power of the wave. Its macroscopic form is usually the (Joule) heat.

We will use small areas (of the volume *V*, with their dimensions much smaller than the wavelength of the optical field). In these areas, the electric field intensity of the wave is independent of the position; we retain solely the harmonic time dependence

$$\text{(12.18)}$$

starting from the time t_o=0 of switching the perturbation on. We neglect the magnetic component of the light wave.

The perturbative part of hamiltonian can be expressed via the operator of dipole moment (charge times its displacement) as

$$\text{(12.19)}$$

is a unit vector in the direction of force. The force performs work due to the displacement of the charge, equal to the scalar product of the vectors of force and displacement.

Assume the system in a stationary state i (with the energy E_i) at the initial time t_o. The probability of a transition to a stationary final state f (with the energy E_f) at the time T (which is the squared modulus of the probability amplitude) is

$$(12.20)$$

For $T \to \infty$ the function F can be replaced by the Dirac δ-function:

$$(12.21)$$

The transition $i \rightarrow f$ is enabled by the work of the light field, which (per unit volume and time) reads

$$ \tag{12.22} $$

The work vanishes whenever the resonance condition is not fulfilled, and diverges otherwise. This is a consequence of the stationary initial and final states. Quasistationary states have finite lifetimes, leading to the uncertainty of their energies and relaxation of the resonance condition of the above Fermi golden rule.

Introducing the lifetime τ of a quasistationary state, inversely proportional to the energy parameter Γ,

$$ \tag{12.23} $$

we arrive at the probability density of finding the energy of the Lorentzian (Breit-Wigner, Cauchy) form

$$ \tag{12.24} $$

After convolving the spectral dependence of Eq. (12.22) with the above probability density, the contribution of the i-f transition to the imaginary part of the dielectric function assumes the following form,

This can be rewritten as

actually avoiding the necessity to calculate the real part of permittivity from its imaginary part using the Kramers-Kronig relations.

In weak optical fields, the probabilities of different transitions are not mutually influenced, and the resulting dielectric function is just a sum over all possible transitions,

(12.27)

where

Note the Lorentzian form of the individual contributions in the sum of Eq. (12.27). This is the consequence of our choice in the description of the involved quasistationary states.

In particular, Eq. (12.27) is useful for the transitions within the one-electron picture of crystalline matter, that do not involve other excitations (such as phonons). As the momentum is proportional to the k–vector of the Bloch states, the matrix element is zero for states of different k (the state vectors are orthogonal). The allowed transitions are called "direct" (in k-space). This selection rule can also be interpreted as the requirement of momentum conservation, since the momentum of the involved photon is negligible.

Adding independent contributions from all available pairs of initial and final states provides the total response. In the picture of transitions between different electronic bands, occupied l and unoccupied l', the result based on the Fermi golden rule and conservation of momentum implies the dielectric function (12.27) in the form

$$(12.28)$$

Evidently, the energy differences between different bands ("interband energies") resonate with the energy of incoming photons.

Joint density of states (JDOS) is usually very instrumental in dealing with most of the crystalline matter. In fact, neglecting the k-dependence of the oscillator strength in (12.28), the frequency dependence of the probability of a photon being absorbed is proportional to the number of available energy differences between the occupied initial (band l) and free final (band l') electron states.

JDOS changes strongly with frequency in the neighborhood of the critical points (minima, saddle points, maxima) of the interband energy.

Discussion of the response functions of crystals

The translational symmetry leads to the use of the wave vector k to index the energy bands, restricted typically to the first BZ; combined with the point symmetry, we use the group symbols for high symmetry points and directions in the reciprocal space.

In the phonon band structure of GaAs, we distinguish acoustic and optic branches, and identify the degeneracy forced by the crystal symmetry.
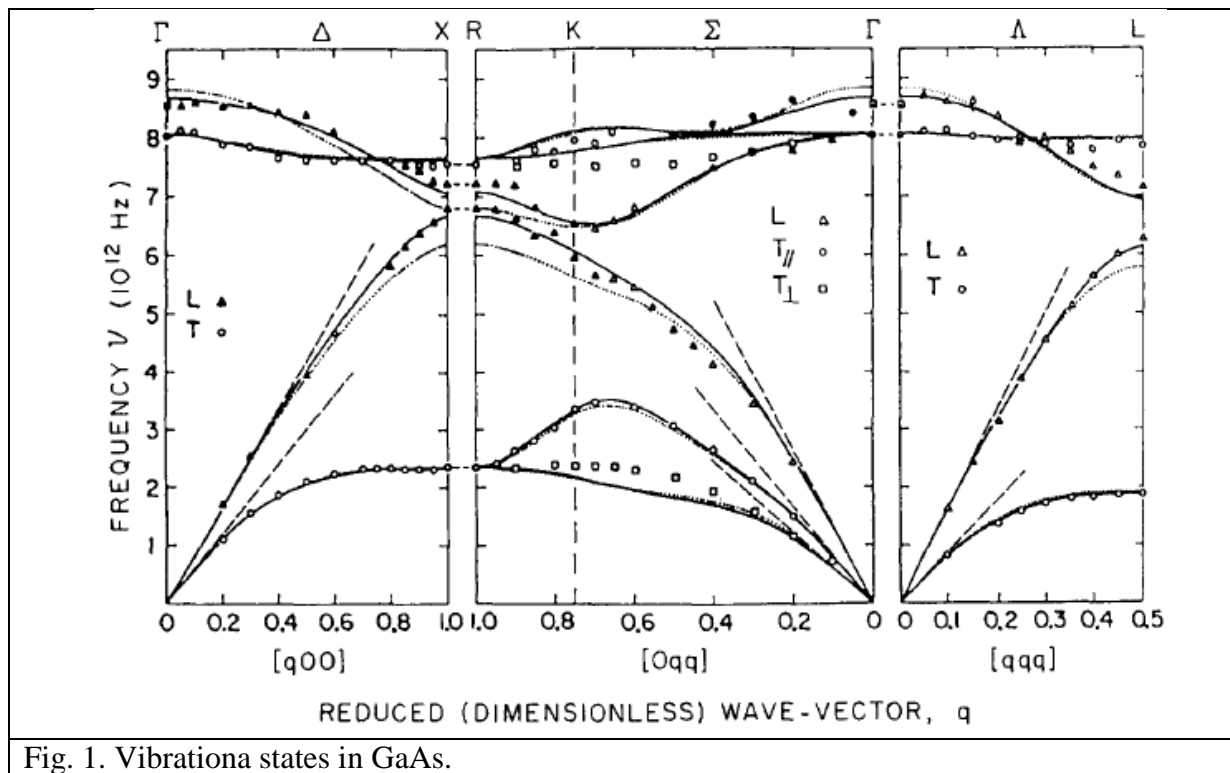

Fig. 1. Vibrationa states in GaAs.

Note the frequency splitting of the LO and TO modes at Γ, which is the consequence of the (partly) ionic bonding.
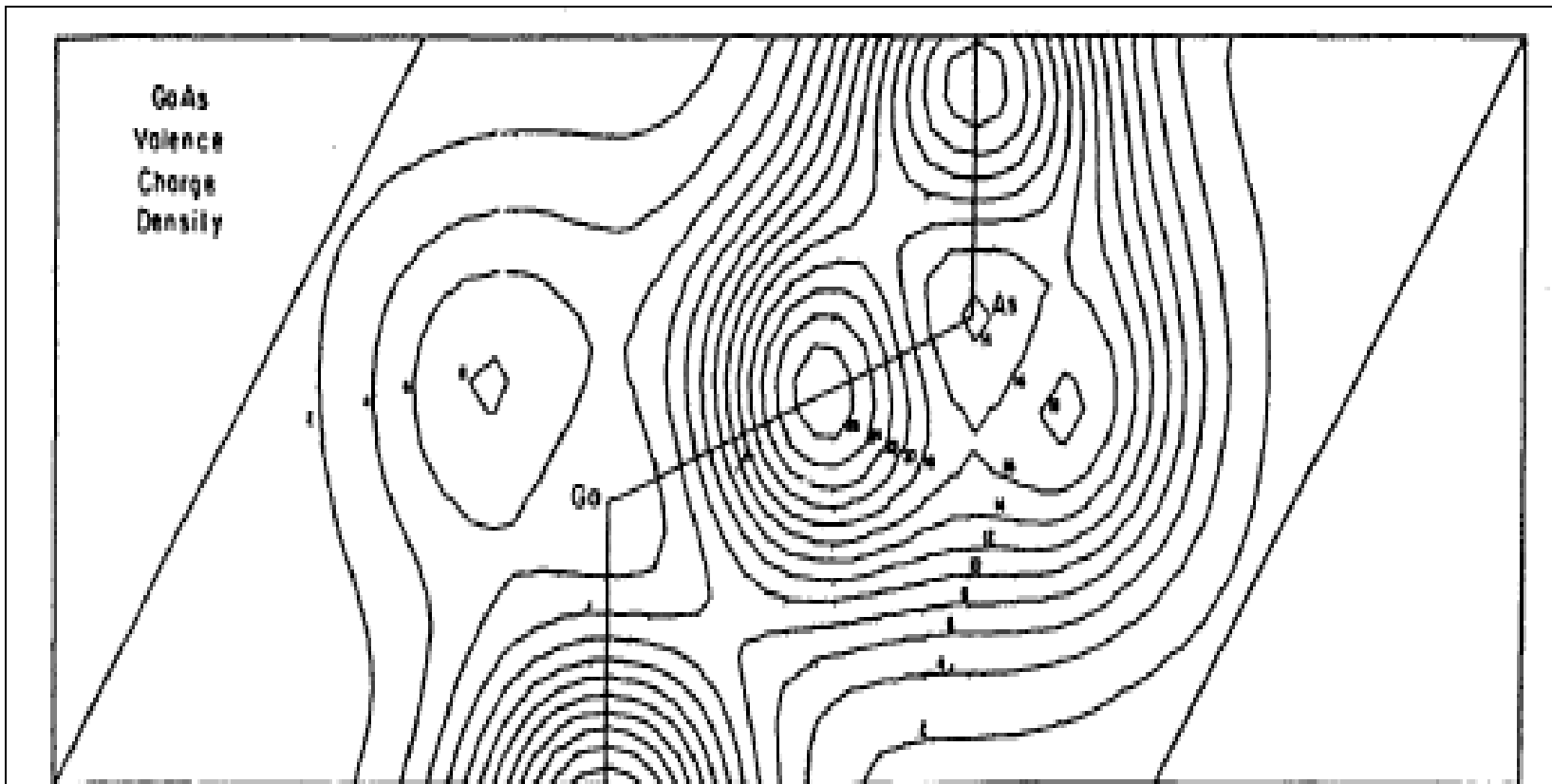
Fig. 8.29. Valence band charge density for gallium arsenide. The charge density contours are in units of electrons per unit cell volume

Fig. 2. Valence charges in GaAs.

One-electron picture of GaAs, showing possible direct transitions between occupied valence and empty conduction states.

Fig. 3. Electronic bandstructure of GaAs.

Fingerprints in the optical spectra of doped GaAs
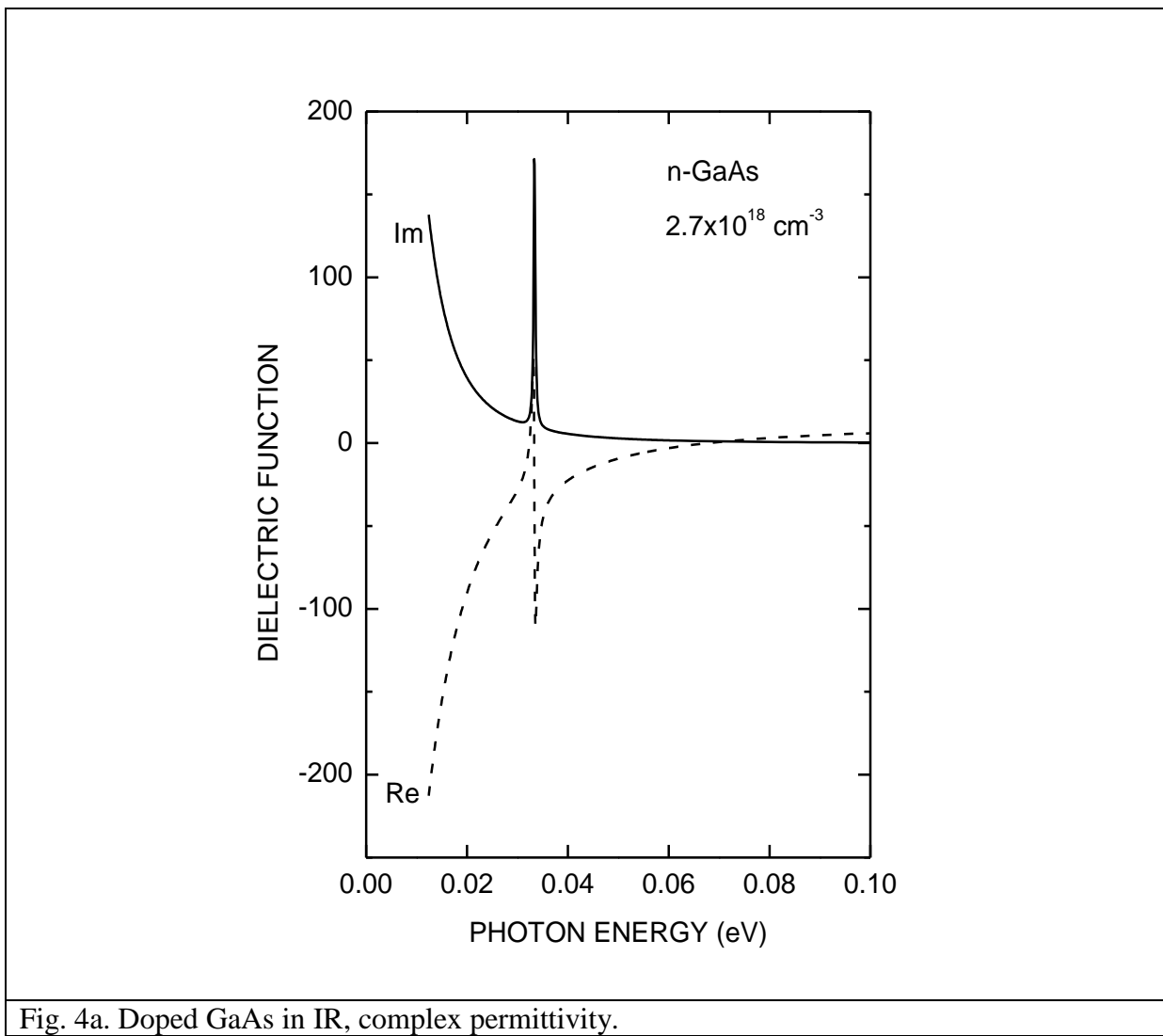
Fig. 4a. Doped GaAs in IR, complex permittivity.
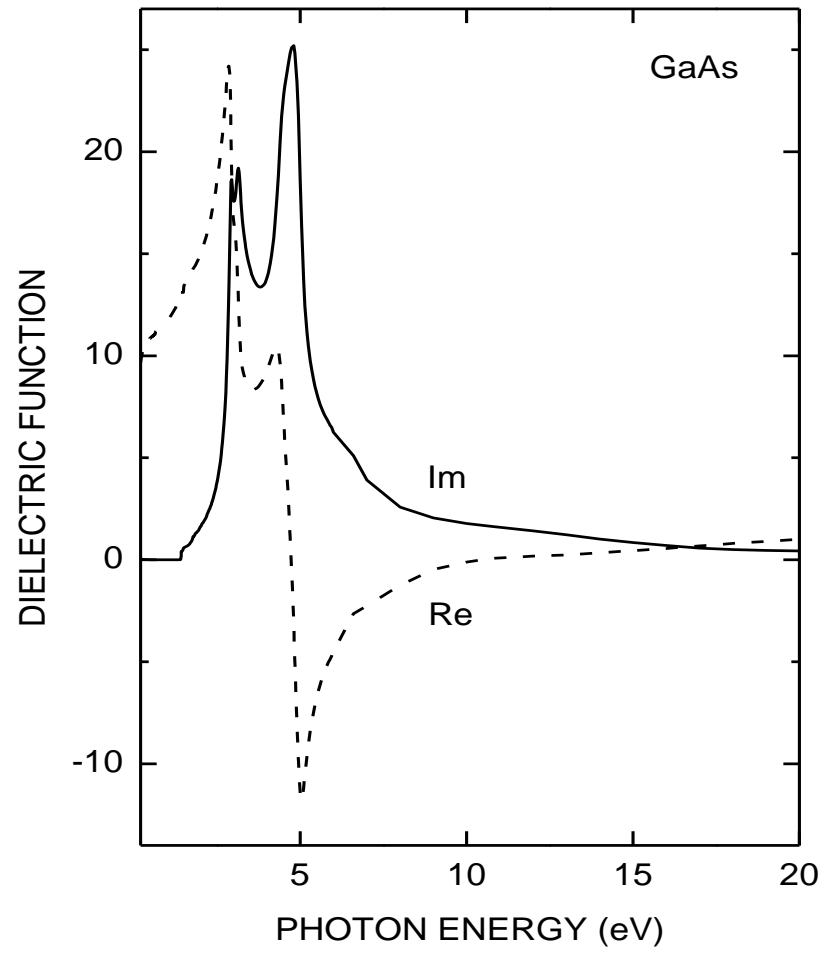
Fig. 4b. Complex permittivity in NIR-VIS-UV.
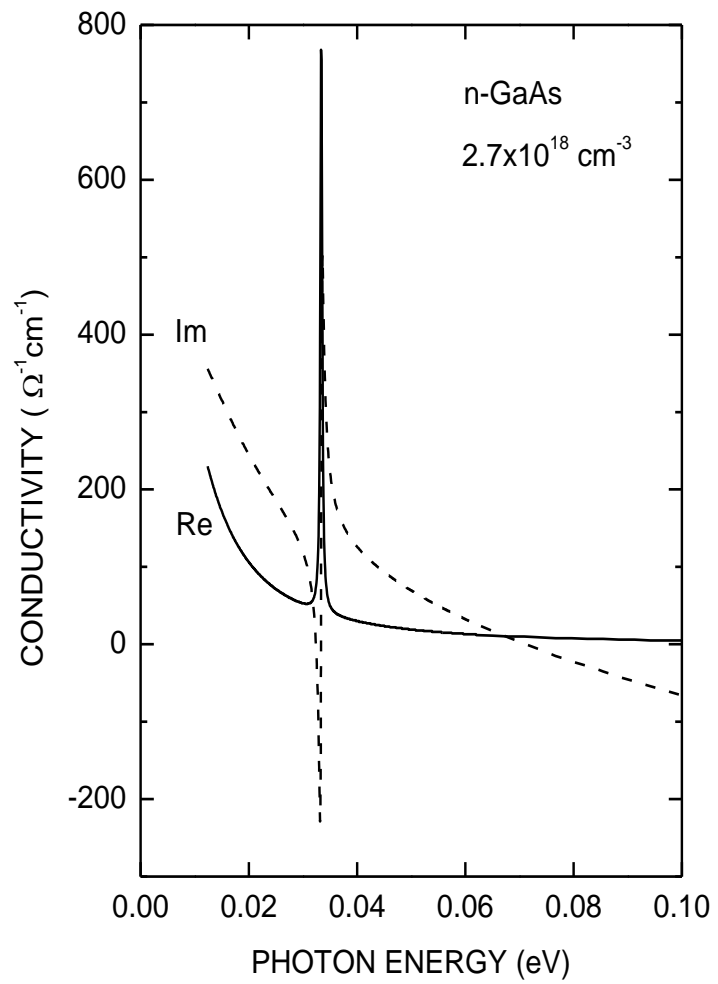
Fig. 5a. Complex conductivity in IR.
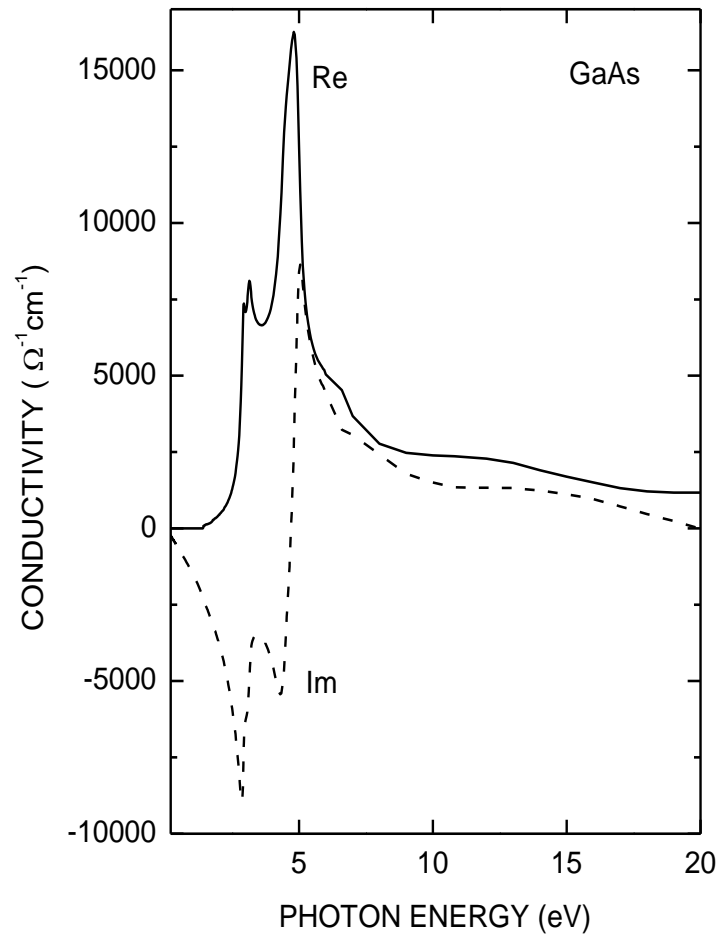
Fig. 5b. Complex conductivity in NIR-VIS-UV.
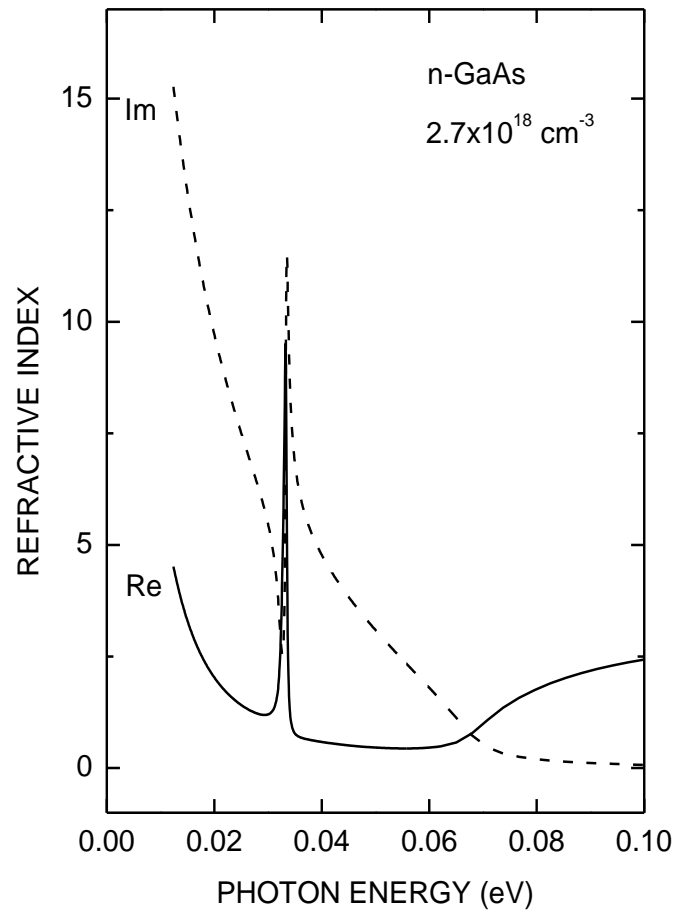
Fig. 6a. Complex refractive index in IR.
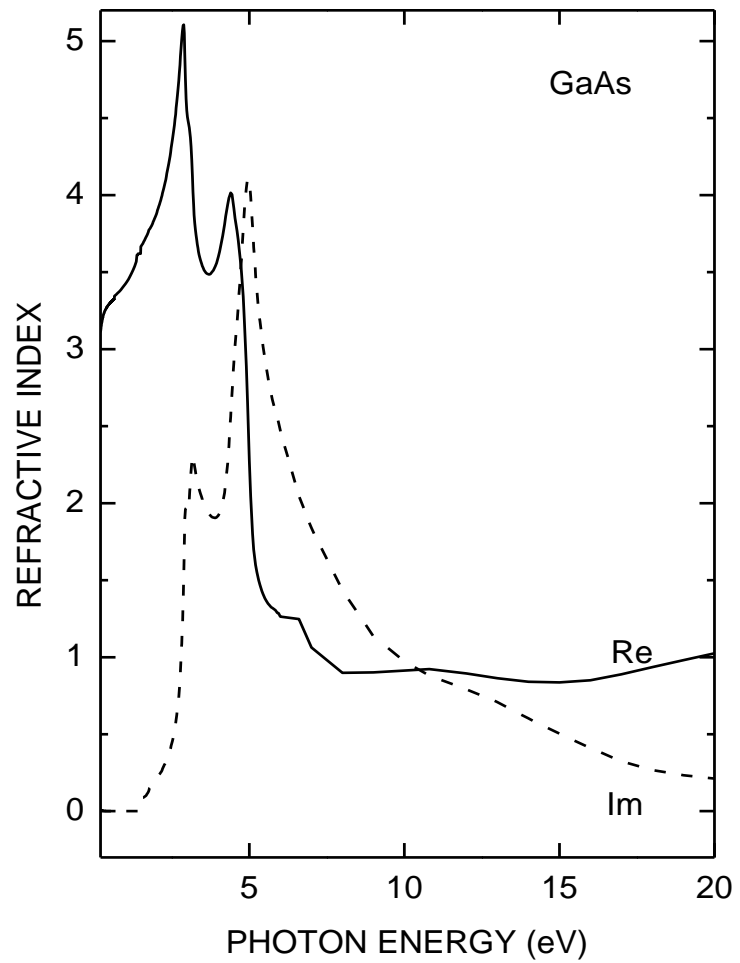
Fig. 6b. Complex refractive index in NIR-VIS-UV.

n-GaAs

2.7×10^{18} cm^{-3}

Im

Re

-1 / ε

PHOTON ENERGY (eV)

Fig. 7a. Complex -1/ε in IR.

Fig. 7b. Complex -1/ε in NIR-VIS-UV.

n-GaAs
2.7×10^{18} cm^{-3}
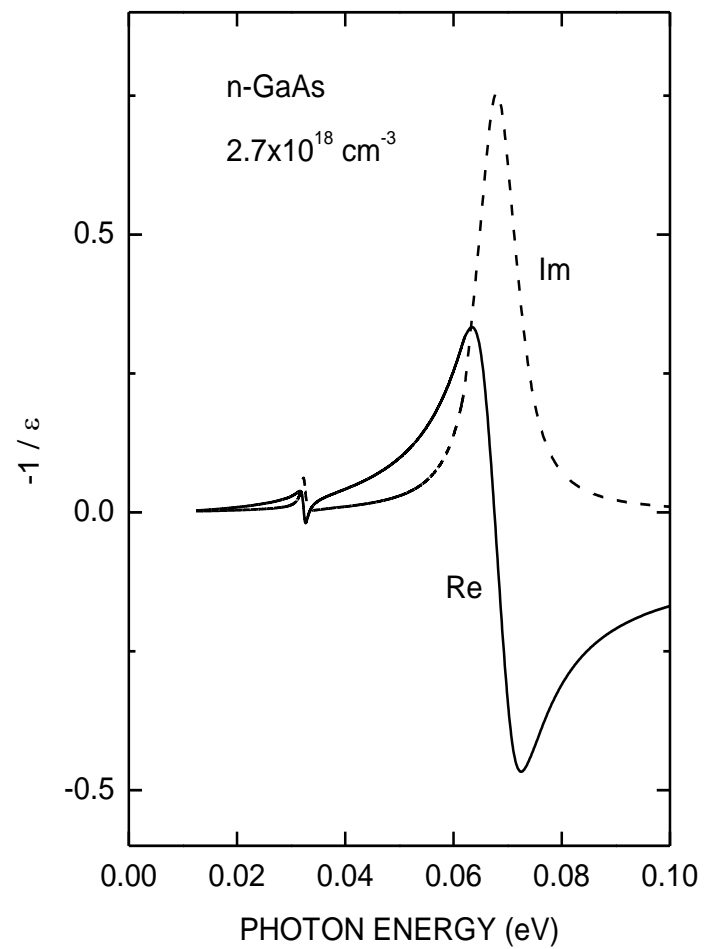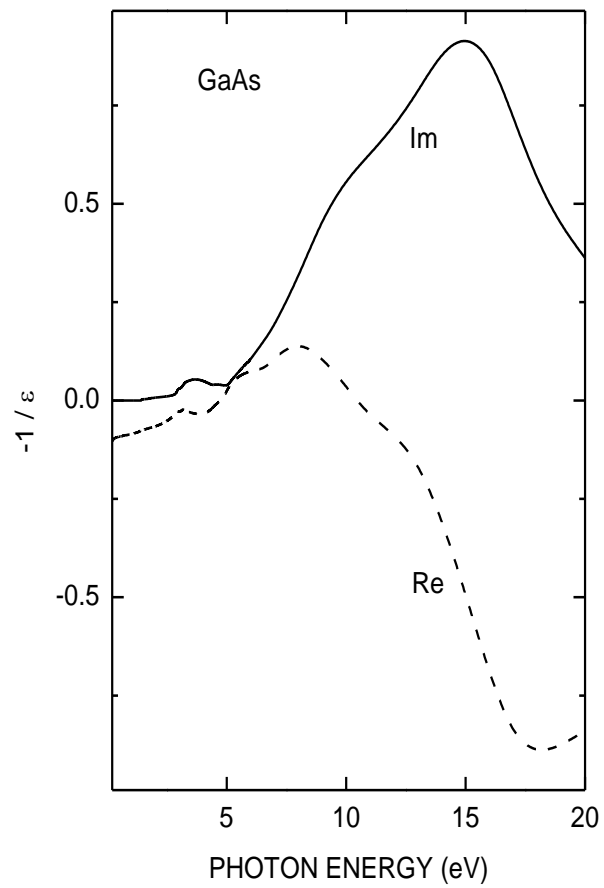
PENETRATION DEPTH (μm)

PHOTON ENERGY (eV)
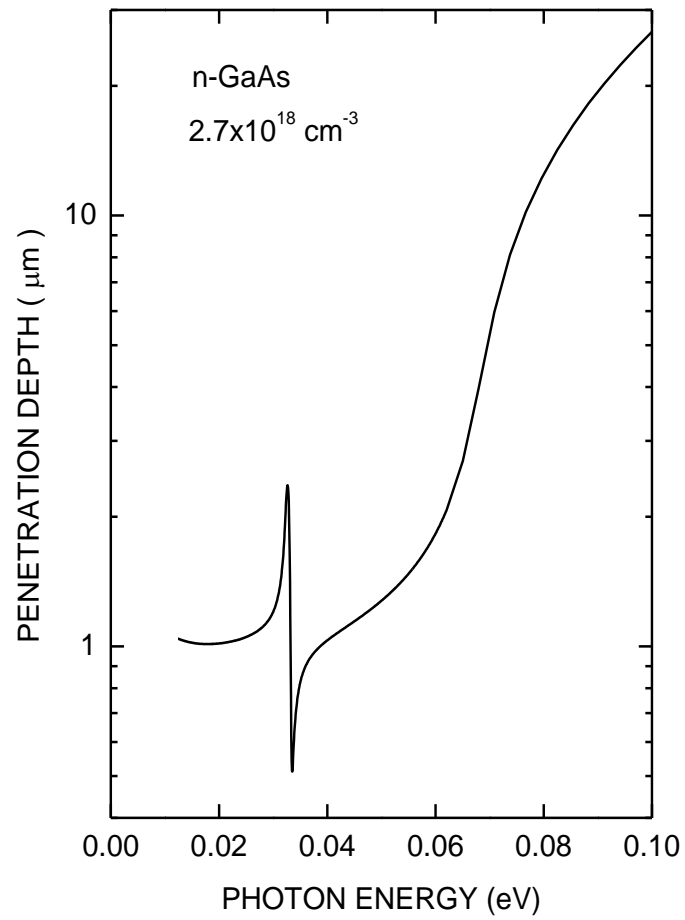
Fig. 8a. Penetration depth of light in IR.
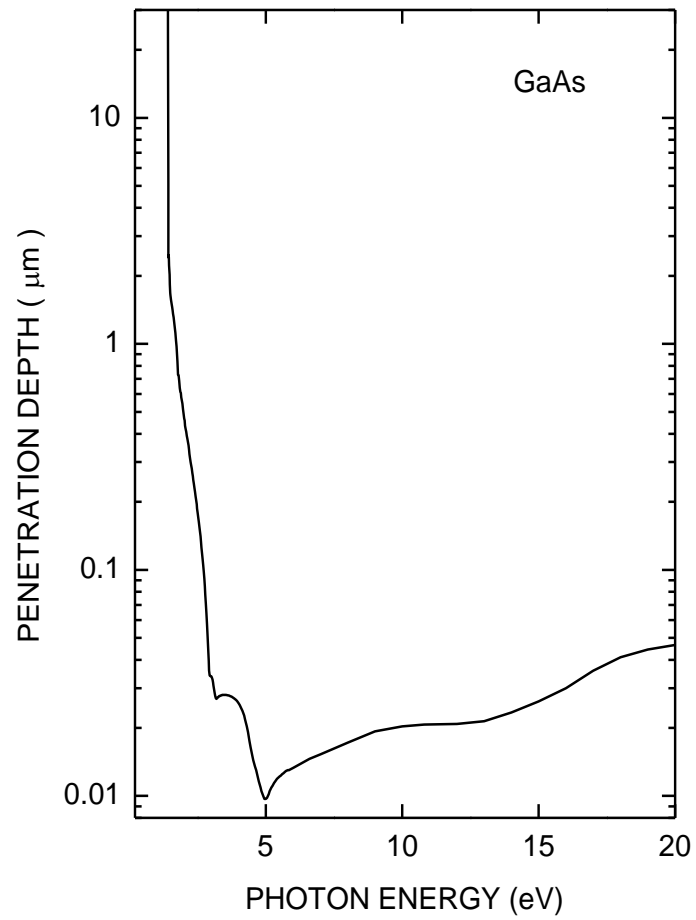
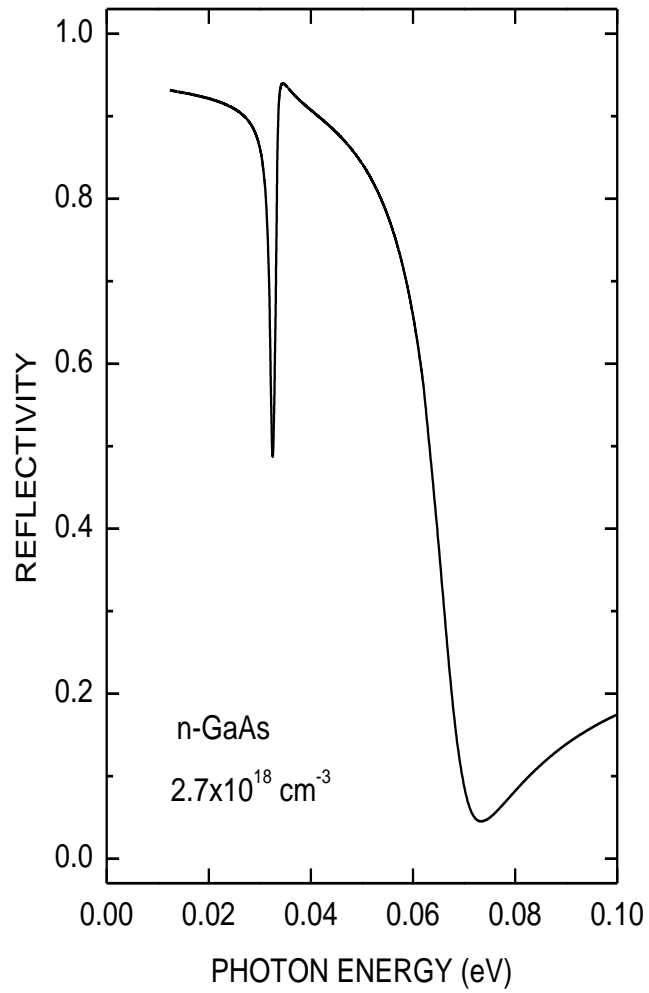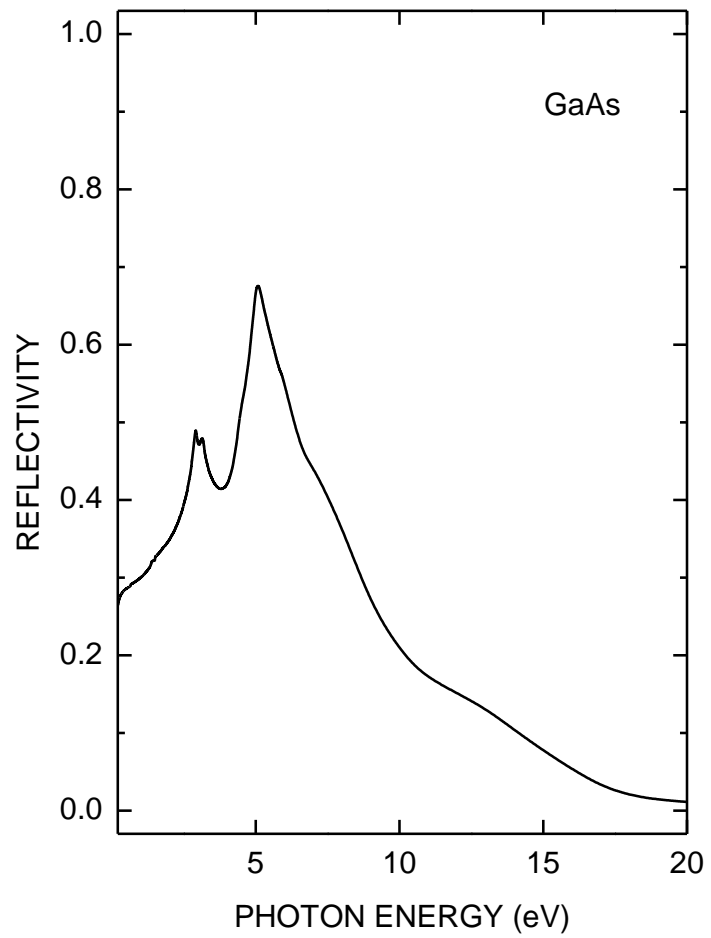Fig. 8b. Penetration depth in NIR-VIS-UV.

Fig. 9a. Normal-incidence reflectivity in IR.

Fig. 9b. Normal-incidence reflectivity in NIR-VIS-UV.