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Dielectric functions and optical parameters of heavily doped and/or highly excited Si:P

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Recently shown photonic and optoelectronic potentialities of Si-based materials and devices require an accurate representation for their optical functions. A predictive model of dielectric function for heavily doped and/or highly excited Si:P is presented. The influence of dopants and of free-carrier population has been calculated independently, allowing the determination of accuracy in usual approximations. The effect of Drude parameters on the heavily doped Si:P optical response is taken into account. All results are supported by experimental data.

Keywords: heavily doped Si, strongly excited Si, dielectric function, Drude–Lorentz approximation, optical model.

1. Introduction

Tabular representation of the dielectric response [1] of semiconductors [2, 3] is often sufficient for analyzing simple materials by optical methods, but it is usually insufficient when the attention is moved towards complex materials. Analytical representations [4, 5] in such cases are much more convenient, allowing rapid predictions and analysis of subtle property changes of a material. They become essential in the fabrication of new materials that exhibit different properties than their classical counterparts [6] and they also find use in quality control of epitaxial growth where constant monitoring of optical properties must take place [7]. Another important application lies in new generation of semiconductor devices [8], where very thin films are stacked together, for example in third generation multi-junction solar cells [9]. Existing solutions and theoretical models allow analyzing either the generic response of a given material for a broad range of energies [10] or accurate response for a very specific case and conditions [11], trading off generality for accuracy and usually [12]

their increased complexity at the same time. Even so, many accurate models and theoretical approaches still working for studying single material fail when the emphasis is turned to a multi-material (composite) structure [13].

Specific optoelectronic properties of heavily doped and/or highly excited semiconductors, which differ in various ways from perfectly known (non-degenerated) materials, lead to new often unpredicted applications [14, 15]. The properties of such materials find use and are appreciated in late generation electronic circuits, and will probably appear in future all-Si optical and photonic applications. Several effects have been discovered [16] in materials [17] to produce often unpredicted properties or enhancement of known properties [18, 19]. Despite extensive studies over the past twenty years, there are several non-elucidated questions about optical features [20, 21]. Results of the systematic investigation into differences among ellipsometrically measured pseudodielectric functions are also found in the literature [13].

Information about the dielectric functions and their critical points (CP) corresponding to interband transitions of such materials is necessary for fabrication and proper control of new materials [3]. Measurements of these interband transitions have been performed by several optical techniques, including reflectance [22, 23], ellipsometry [24, 25] and reflectance modulation techniques such as magnetoreflectance [26], thermoreflectance [27, 28], electroreflectance [29, 30] and wavelength-modulated reflectance [31, 32].

Semiconductors are especially interesting due to an unusual number of physical phenomena and possible applications [33, 34]. Many papers have been published covering the behavior of CP and dielectric functions for Si [35, 36] and other semiconductors [37, 38] as a function of illumination [39, 40], temperature [41, 42], composition of the material [43, 44] or its phase [45, 46], method of preparation [47], or size [48]. Various analytical methods [49, 50] exist that can be used to predict (or estimate) the quantities in a given range of energy [51, 52]. The quantities are, *e.g.*, the dielectric function of a material [53] with a dense free-electron gas [54, 55], composite medium [56] or metamaterial [4]. The methods used are: the density functional theory [57], exchange-correlation potential [58], k.p method [59, 60], tight-binding approximation [61, 62] or parabolic band approximation [63, 64], to name just a few.

Extremely heavily doped Si or highly excited Si, or the Si that is less heavily doped and/or highly excited show optoelectronic properties that differ in various ways from the perfectly known non-degenerate and/or weakly excited material. Such a situation sometimes offers unpredicted applications [6] that find their place in late generation electronic circuits and will appear in the future all-Si optoelectronics and photonics. Despite extensive studies over the past twenty years, there are several non-elucidated questions which for example appear in a systematic investigation into ellipsometrically measured pseudodielectric functions [13].

We have focused in particular on Si:P (moderately and heavily P-doped crystalline Si) and highly excited Si (containing an extremely dense free-carrier gas) looking for general tendencies of the dielectric functions of a system. The results are directly

useful in the design, manufacturing and control of newly fabricated materials and devices [3, 36] by allowing a prediction of functioning and yield [4, 58, 65]. New fabrication techniques permit extremely heavy Si:P doping, more than $N_d = 2.3\times10^{21}$ cm^{-3}, which exceeds the solid state solubility of P in Si at $T = 300$ K.

The general behavior of the dielectric function under small doping variation was studied in [1]. But any consistent model for describing the whole doping range, from intrinsic Si to extremely heavily doped Si (at the limit of the solubility), does not exist to date. In our investigations we used as a reference the experimental data for undoped Si [2] and for heavily doped Si (Si:P) [66].

Usually behavior variations resulting from doping or from generated/injected carriers are treated in the same way. A femtosecond excitation of intrinsic Si (by injection of a free-carrier population) has shown that the corresponding dielectric functions change differently [67]. Thus, the problem becomes essential in design and manufacturing of devices containing a dense free-carrier population.

Our study, which is based on the theoretical approach of [11] and on the data of [1–3, 68], confirms that the changes of dielectric function caused by injected carriers or by heavily doping are not the same. Questions about the sensitivity of the nonlinear dielectric functions to the carrier–carrier interactions are still open.

2. Theory and computational methodology

2.1. Harmonic oscillator approximation and Drude approach

We propose a method of predicting dielectric functions of heavily doped Si (Si:P) and heavily doped Si under high excitation (*e.g.*, by incident light) that both lead to a dense additional/extrinsic free-carrier population. The well-known harmonic oscillator (HMO) approximation has been extensively studied over the years and some classical problems and disagreements between experimental data for Si and theory have been identified, like an inaccurate representation of the absorption edge associated with the direct bandgap [4, 5, 7–13, 65–67]. Simple extensions to the classical model might solve the problem, but they lead to the loss of generality of the model itself [10–12]. For analyzing dielectric functions in materials that vary little, one might need a tool that is suited for a particular application and spectral range, rather than more generic but less accurate one. Such extensions have been developed for intrinsic Si and Si-derived materials in the past [11, 12], and they work well when parameterization of a given dielectric function is performed, but they are not capable of predicting optical properties when structural changes occur. For structural changes, a model equation introduced by Aspnes group [11] was expanded by the addition of a possible functional dependence of its parameters in regard to changing doping density, and later combined with a Drude extension that leads to the following set of equations:

$$\varepsilon(\omega, N_d) = \varepsilon_\infty + \sum_j \frac{C_{0j}(N_d)}{\omega^2}(A + B - C - D) - E \qquad (1)$$

where:

$$A = \exp\left[i\beta_j(N_d)\right]\left[\omega_{gj}(N_d) - \omega - i\Gamma_j(N_d)\right]^{\mu_j(N_d)}$$

$$B = \exp\left[-i\beta_j(N_d)\right]\left[\omega_{gj}(N_d) + \omega + i\Gamma_j(N_d)\right]^{\mu_j(N_d)}$$

$$C = 2\mathrm{Re}\left\{\exp\left[-i\beta_j(N_d)\right]\left[\omega_{gj}(N_d) + i\Gamma_j(N_d)\right]^{\mu_j(N_d)}\right\}$$

$$D = 2i\mu_j(N_d)\,\omega\,\mathrm{Im}\left\{\exp\left[-i\beta_j(N_d)\right]\left[\omega_{gj}(N_d) + i\Gamma_j(N_d)\right]^{\mu_j(N_d) - 1}\right\}$$

$$E = \frac{N_{\mathrm{e-h}}\, e^2}{\varepsilon_0 m_{\mathrm{opt}} m_e \omega^2} \frac{1}{1 + i\dfrac{1}{\tau_D\,\omega}}$$

while C_0 and g are, respectively, the amplitude and the critical frequency of the Lorentz oscillator, Γ is the broadening coefficient, N_d is the dopant density, $N_{\mathrm{e-h}}$ is the carrier density, τ_D is the Drude damping time, m_{opt} is the optical mass of carriers, μ is the pole order, and β is the phase factor of the pole. The rest of the parameters have their usual meaning.

Introducing the dependence of the model parameters on doping density and on the free electron gas density allows to predict the optical properties of Si for any doping density. In the 1.5–5.5 eV spectral range, we found three characteristic oscillator energies sufficient, at energies E_{g1} = 3.4 eV, E_{g2} = 3.62 eV and E_{g3} = 4.27 eV, where E_{g1} and E_{g3} correspond to the first and second direct transitions in Si, while E_{g2} is the established energy for e–h pair creation in Si [68]. This is also coherent with literature data [11–13]. Also, the integration of the Drude part with the HMO approximation allows discrimination between the changes in dielectric function for lower energies, since the Drude-dependent influence area (hence connected with free carriers) is localized mainly on the left-hand side of the spectrum and an increase in e–h pair density always influences the lower energies first. On the other hand, the lattice disturbance caused by doping itself is present mainly at the characteristic transition energies E_g and neighbouring area.

2.2. Evolution of parameters with variation of doping concentration

The experimental results of JELLISON *et al*. [1 and private communications] for heavily doped Si:P were used to determine model parameters versus the doping density. The dielectric function was predicted on the basis of [12, 69] and on the well-known effects originating from doping. In general, the oscillator amplitude C_0 decreases with

increasing doping concentration, because of the additional defects and lattice disturbances caused by doping. The broadening parameter Γ tends to increase for the same reasons. The critical point energy E_g (or frequency ω_g) shows a slight shift towards lower energies due to the renormalization of the band structure caused by additional free-carrier population originated from doping and state-and-band filling (see Fig. 1).

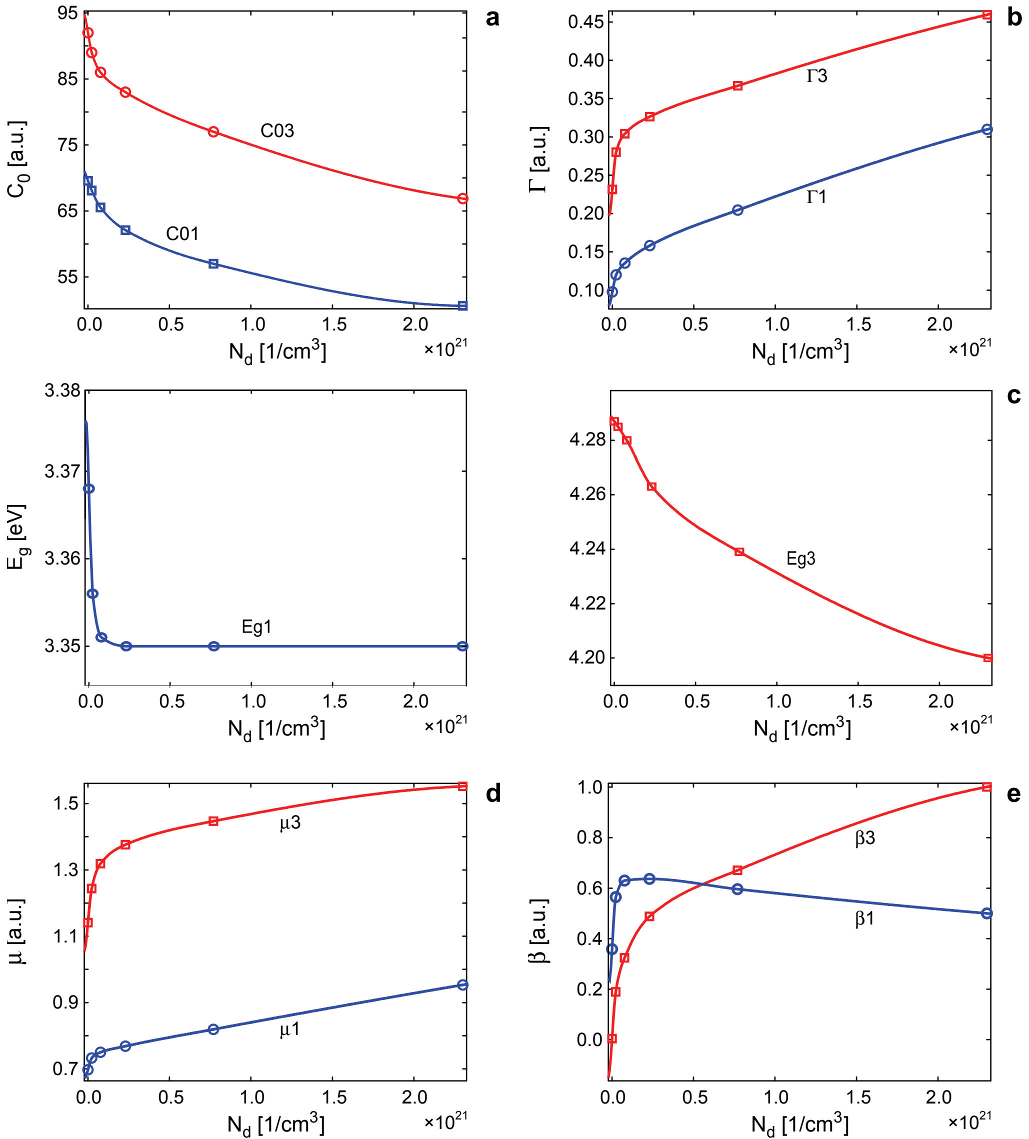

Fig. 1. Evolutions of oscillator amplitude C_0, broadening parameter Γ and critical point energy E_g versus doping density are presented on graphs (**a**), (**b**) and (**c**), respectively. Below the changes of the pole order $|\mu|$ (**d**) and the pole phase factor β (**e**) versus doping concentration are presented. Tendency lines were obtained by calculating a piece-wise polynomial for each pair of points.

Table 1. Parameters used in the three-oscillator model, for five different doping concentrations N_d. Also goodness of fit (R) and sum of squared errors (SSE) are presented for each doping density.

	Doping concentration N_d [cm^{-3}]					
Parameters	1×10^{14}	2.30×10^{19}	7.70×10^{19}	2.30×10^{20}	7.70×10^{20}	2.30×10^{21}
C_1	69.56	68.1	65.53	62.1	57	50.62
C_2	239.3	316.5	343	335.7	325.3	311.9
C_3	92	89	86	83	77	66.88
γ_1	0.09775	0.12	0.1353	0.1582	0.2045	0.3098
γ_2	0.361	0.345	0.3408	0.3396	0.346	0.381
γ_3	0.2316	0.2801	0.3042	0.3264	0.3668	0.4594
β_1	0.3582	0.5648	0.63	0.6465	0.596	0.5
β_2	0.307	0.089	0.03906	0.03642	0.06536	0.1675
β_3	0.004174	0.1891	0.3238	0.4878	0.6708	1.001
$\lvert\mu_1\rvert$	0.6976	0.7328	0.7501	0.7685	0.8193	0.9533
$\lvert\mu_2\rvert$	0.4398	0.3149	0.2966	0.3108	0.3331	0.3743
$\lvert\mu_3\rvert$	1.141	1.244	1.319	1.376	1.447	1.552
E_{g1}	3.368	3.356	3.351	3.35	3.35	3.35
E_{g2}	3.654	3.621	3.604	3.592	3.606	3.636
E_{g3}	4.287	4.285	4.28	4.263	4.239	4.2
R-square	0.9998	0.9999	0.9999	0.9999	0.9999	0.9997
SSE	1.0909	2.2556	1.4831	0.9643	0.8062	0.6568

It has been stated before that the condition $\mu < 1$ is necessary to satisfy Kramers–Kronig relations [11] and we expect the pole order μ to decrease as the doping density increases since the associated oscillator strength becomes relatively weaker while the phase factor $|\beta|$ should increase with doping concentration due to the increased fraction of Brillouin zone over which band transitions take place. As can be seen in Fig. 1, the parameter $|\mu|$ varies similarly with the corresponding broadening coefficient Γ.

Parameters corresponding to an oscillator with critical energy $E_g \approx 3.62$ eV have a slightly different behavior than predicted. This might be due to additional mechanisms that occur at this transition that are not present for transitions at $E_g = 3.38$ and 4.29 eV. Parameters of the three-oscillator model (for five different doping densities) are listed in Table 1.

3. Results

3.1. Heavily P-doped Si

The model reproduces the experimental results nearly perfectly for doped Si in the energy range $E = 2.5$–5.0 eV. The real and imaginary parts of ε reproduced by our model and the experimental data of [1] are presented in Figs. 2**a** and 2**b**,

respectively. For E = 1.2–2.5 eV, the imaginary part of the ε function is very small, except for extremely high P-doping densities, leading to relatively significant differences between the experimental and simulated data. The residual plot of the imaginary component of the dielectric function, shown in Fig. 2**c**, confirms the fact that the greatest relative differences occur for low energies, but in any case the errors are one order smaller than systematic differences observed on experimentally measured dielectric functions of Si [13]. Figure 2**c** confirms the quality of our model. Parameterization of Eq. (1) allowed us to simulate the dielectric functions for any possible P-doping density measured so far for Si, which is presented in Fig. 2**d**. With the help of Eq. (1), the dielectric function can be calculated for any doping density between $N_d = 10^{14}$ to 2.3×10^{21} cm^{-3}, where the first corresponds to intrinsic, float-zone Si and the last is the solubility level of P in Si.

The absolute values of ε around the two main energies E = 3.38 eV and E = 4.29 eV decrease with doping concentration. This effect is due to the lattice disturbance caused by defects.

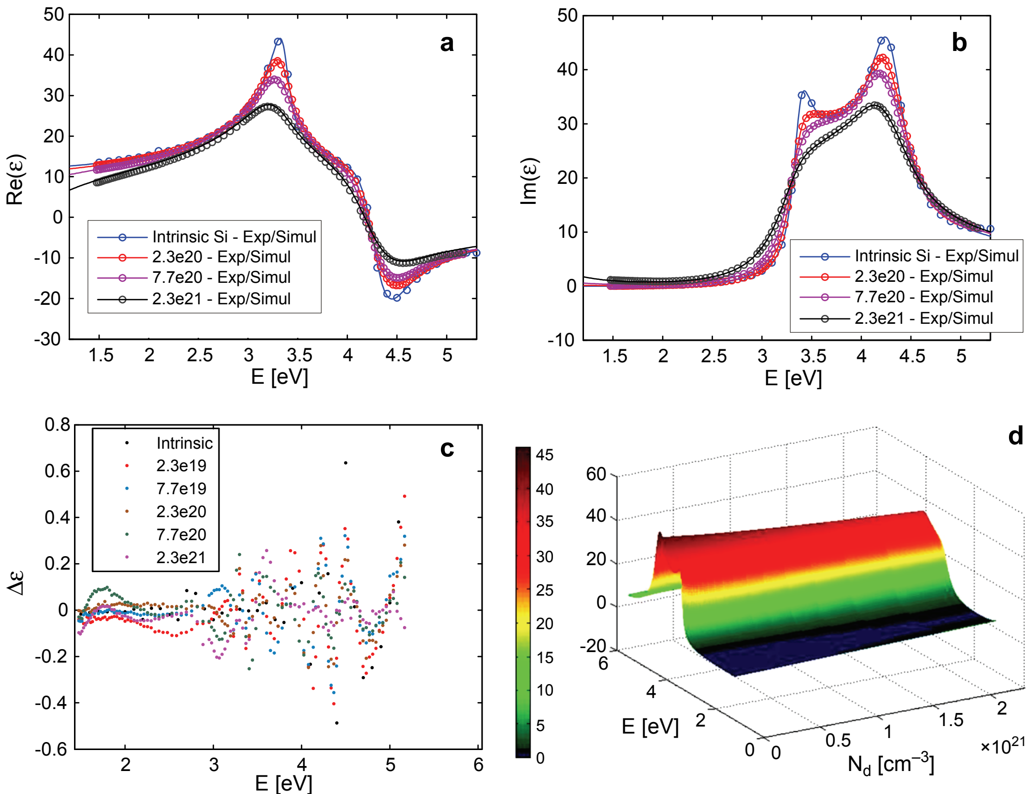

Fig. 2. Simulated (solid line) and experimental (points) data for different P-doping concentrations are shown: Re(ε) (**a**), Im(ε) (**b**), residual plot for the imaginary part of the dielectric function $\Delta\varepsilon$ (**c**) and Im(ε) of Si for all possible P-doping densities (**d**).

3.2. Highly excited Si

The model presented is able to determine changes in the dielectric function independently of the lattice disturbance, *i.e.*, in the case of injection or excitation of the dense free-carrier gas. As can be seen in Fig. 3**a**, the influence of free-carriers on the dielectric function appears mainly at low energies, being negligible in the range around the direct transition peak at $E = 3.4$ eV and above, where the most important role is played by lattice disturbances and effects introduced by doping. In Figure 3**a**, one can clearly see that the free-carrier related Drude part of the dielectric function, that is present for energies $E = 1$ to 3 eV for the densities studied, is mainly responsible

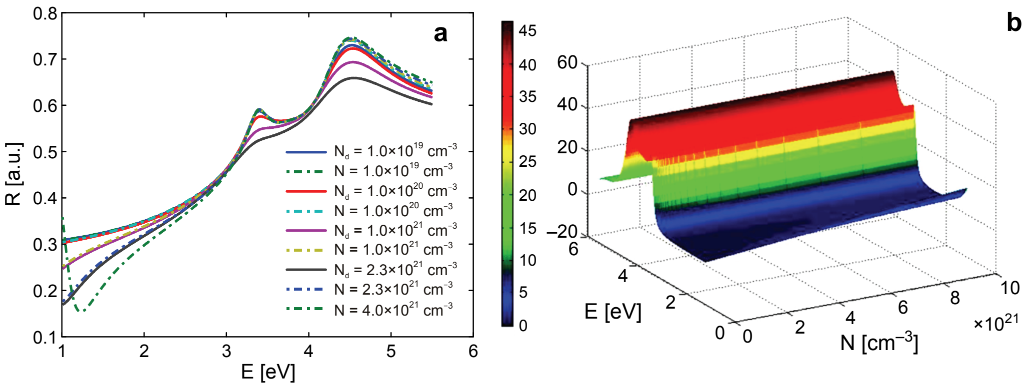

Fig. 3. Simulated reflectivity R for different P-doping concentrations (solid curves) and corresponding density of excited free carriers (dot–dashed curves) (**a**). Simulated response of Im(ε) for free-carrier densities covering the range from intrinsic to near-metallic (**b**).

for the changes of reflectivity, but it has little or no meaning around the main transition peaks. For the highest presented density $N = 4\times10^{21}$ cm^{-3}, one can observe the appearance of the so-called plasma minimum around $E = 1.1$ eV. This minimum in reflectivity depends on carrier density N, carrier optical mass m_{opt} and carrier damping time τ and tends to move towards higher energies when N increases and/or τ and m_{opt} decrease. For higher free-carrier densities, ultimately near-metallic values, the reflectivity reaches values close to unity. One has to keep in mind that the existence and measurement of such high carrier densities in Si is possible only on a femtosecond-time scale [70, 71]. That poses additional very complex problems, that we have addressed elsewhere [72].

4. Conclusions

The main objective of this work has been successfully met with an excellent precision. The model determines the dielectric functions over the complete P-doping range, from intrinsic to extremely highly doped Si (up to 2.3×10^{21} cm^{-3}), thanks to a parameterization of tendency lines for variables of Eq. (1), which were obtained by

analysis of experimental results of ellipsometrically measured dielectric functions of Si:P. The Drude–Lorentz parameters describe the optical functions of lightly or moderately doped Si in the corresponding full range of free-carrier densities up to a metal-like density. The results are consistent with physical predictions and previous works [1–4]. A clear distinction between optical functions of Si with an injected free-carrier population and one originating from doping could be established. Future work shall include the effects of the temperature on the dielectric function of Si and extension to other semiconductors and dielectrics.

Simulated optical functions (reflectivity, absorption coefficient) agree with experimental results published previously [73, 74] and obtained by us on MINDs with extremely high steady-state free-carrier densities.

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