

Analysis of photothermal ionization spectra of shallow impurities in silicon

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A general model has been developed for calculating the photoconductive response in a multiply doped semiconductor. With consideration of the effects of sample temperature and thickness, as well as impurity parameters such as concentration and the frequency-dependent photoabsorption cross section and thermal ionization probability as obtained from existing experimental data, rate equations have been developed for the excess photogenerated carrier density $\Delta p(\omega)$ per unit angular frequency ω of incident light. The model is formulated to include illumination by both band-edge light and the modulated far-infrared light used in Fourier-transform spectrometry. Our calculations so far assume no band-edge light. The model has been applied to doubly doped Si(In,Al) and Si(Ga,B), with the effects of compensation taken into account. The temperature and concentration dependence of the discrete lines of the deeper acceptor, which are superimposed on the continuum background of the shallower acceptor, agree well with experimental spectra, indicating that the model holds promise for making photothermal ionization spectroscopy a more quantitative characterization technique.

I. INTRODUCTION

We have developed a general model for calculating the photoconductive response in a multiply doped semiconductor. In particular, the model has been applied to doubly doped Si(In,Al) and Si(Ga,B), under the conditions of low temperature and excitation by modulated far-infrared light, appropriate to Fourier-transform infrared (FTIR) spectroscopy. One motivation for formulating this model was to incorporate the effect of the temperature dependence of the acceptor photothermal ionization probability for those acceptor lines which appear against a continuum background. At temperatures low enough to suppress thermal ionization from an excited level of the deeper acceptor, the net photoresponse is simply a reduction, through photoabsorption, of the number of photons contributing to the continuum background photoresponse of shallower species, resulting in a negative peak in the photocurrent. As the temperature is increased, thermal ionization from the excited levels of the deeper acceptor produces enough excess carriers to result in a positive peak. This effect has been seen by several workers¹⁻³ but has never been incorporated into a general model.

II. THEORY

The formulation of the model may be broken down into five steps: (1) determining the equilibrium neutral acceptor concentrations at a given temperature in the presence of compensation, using standard semiconductor statistics; (2) setting up the rate equations for ionized acceptor and hole generation in the presence of ir radiation, assuming hole capture to be the dominant relaxation process; (3) obtaining an expression for the Fourier component of the excess hole concentration and showing that it is in phase with the modulated ir source; (4) representing the photoabsorption cross sections for the discrete spectrum by

Lorentzian functions and the photothermal ionization probabilities by Boltzmann factors, both based on experimental data; (5) defining a photothermal ionization spectroscopy (PTIS) response function, taking into account the attenuation of the incident light within the sample.

Step 1. Our primary interest is the case of one or more species of shallow acceptors in the presence of compensating shallow donors. We consider the system as being initially in thermal equilibrium at temperature T , in the absence of the exciting light. We assume that the Fermi level $E_F(T)$ is several $k_B T$ below all donor ground-state energies but several $k_B T$ above the top of the valence band. Then standard semiconductor statistics⁴ together with the charge neutrality condition leads to the following equation:

$$\sum_j \frac{N_j}{1 + g_j \eta \exp(E_j/k_B T)} = N_v \eta + N_D, \quad (1)$$

where

N_D = total donor concentration ;

N_v = effective density of holes states in the valence band

$$\approx 2(m_h k_B T / 2\pi \hbar^2)^{3/2} ;$$

N_j = concentration of the j th acceptor species ;

g_j = the degeneracy factor (e.g., 4 for acceptors in Si) ;

E_j = ground-state energy relative to the top of the valence band ;

$$\eta = \exp(-E_F/k_B T) .$$

Equation (1) results in a polynomial equation for η , which can be solved numerically to determine η and, from this, the concentration $N_j^{(0)}$ of neutral acceptors for each species at temperature T at time $t=0$.

Step 2. We now turn on the ir source and consider the photoresponse of the sample. The essential point is that for a modulated source we will be looking at a nonequilibrium situation. We consider the excess photoconductivity at time t :

$$\Delta\sigma(t) \equiv \sigma(t) - \sigma(0) = e[\mu_h \Delta p(t) + \mu_e \Delta n(t)],$$

where Δp and Δn are, respectively, the excess hole and electron concentrations. We shall assume Δn is negligible compared to Δp . (This would not necessarily be the case if, for example, the sample were initially under illumination by band-edge light.) The net acceptor ionization rate is

$$\begin{aligned} \frac{dN_j^{(-)}(t)}{dt} &= -\frac{dN_j^{(0)}(t)}{dt} \\ &= N_j^{(0)}(t) \left[I_{hj}^{(0)} + \int_0^\infty \mathcal{J}(\omega, t) \sigma_{hj}^{(\omega)} I_{hj}^{(\omega)} d\omega \right. \\ &\quad \left. - C_{hj} g_j p(t) N_j^{(-)}(t) \right], \end{aligned} \quad (2)$$

where

- $I_{hj}^{(0)}$ = temperature-dependent thermal ionization probability per unit time, in the absence of exciting radiation;
- $I_{hj}^{(\omega)}$ = temperature- and frequency-dependent probability that the acceptor will thermally ionize after photoabsorption;
- $\sigma_{hj}^{(\omega)}$ = photoabsorption cross section;
- C_{hj} = capture coefficient for holes;

and

$$\begin{aligned} \mathcal{J}(\omega, t) &= \text{intensity spectrum of the radiation, modulated by the interferometer in the FTIR system according to} \\ \mathcal{J}(\omega, t) &= \mathcal{J}_0^{(\omega)} [1 + \cos \Omega t], \end{aligned} \quad (3)$$

where $\Omega = (2\nu/c)\omega$ is the modulation frequency and ν is the speed of the moving mirror.

The net production rate of holes is

$$\frac{dp(t)}{dt} = \sum_j \frac{dN_j^{(-)}(t)}{dt} - R_{\text{eff}} n(t) p(t), \quad (4)$$

where R_{eff} is the effective electron-hole recombination coefficient, including recombination at the sample surface. We shall assume that $C_{hj} \gg R_{\text{eff}}$, which is equivalent to the condition that the hole capture time τ_c be much less than the recombination time τ_R . This condition appears to be well satisfied for silicon, for which $\tau_R \geq 10^{-7}$ sec for recombination at the surface, while $\tau_c \approx 10^{-11}$ sec for $N_j \approx 10^{16}/\text{cm}^3$. This has the effect of making τ_c the relevant time scale of the experiment; the

term in Eq. (4) proportional to R_{eff} controls only the long-term approach to equilibrium and may be dropped. Then

$$\frac{dp(t)}{dt} \approx \sum_j \frac{dN_j^{(-)}(t)}{dt} = -\sum_j \frac{dN_j^{(0)}(t)}{dt}. \quad (5)$$

Step 3. We neglect the variation of $N_j^{(0)}(t)$ with t in the rate equation, Eq. (2), i.e., assume $N_j^{(0)}(t) = N_j^{(0)}(0)$ on the right-hand side. Then for $t > 0$,

$$\begin{aligned} -\frac{dN_j^{(0)}}{dt} &\approx N_d^{(0)}(0) \int_0^\infty \mathcal{J}(\omega, t) \sigma_{hj}^{(\omega)} I_{hj}^{(\omega)} d\omega \\ &\quad - C_{hj} g_j \Delta p(t) [N_j - N_j^{(0)}(0)], \end{aligned} \quad (6)$$

where we have used the fact that the system was at equilibrium just before the light was turned on, i.e.,

$$\begin{aligned} I_{hj}^{(0)} N_j^{(0)}(0) - C_{hj} g_j p(0) [N_j - N_j^{(0)}(0)] \\ = - \left[\frac{dN_j^{(0)}}{dt} \right]_{t=0} = 0. \end{aligned} \quad (7)$$

Thus for $t > 0$ we can write for the excess hole production rate,

$$\frac{d\Delta p(t)}{dt} = \int_0^\infty G_h(\omega, t) d\omega - \frac{1}{\tau_{hc}} \Delta p(t), \quad (8)$$

where

$$\begin{aligned} G_h(\omega, t) &= \mathcal{J}(\omega, t) \sum_j \sigma_{hj}^{(\omega)} I_{hj}^{(\omega)} N_j^{(0)}(0) \\ &= \text{hole photogeneration rate per unit of } \omega, \end{aligned}$$

and

$$\tau_{hc} = \text{the average hole capture time,}$$

defined by

$$\tau_{hc}^{-1} = \sum_j C_{hj} g_j [N_j - N_j^{(0)}(0)]. \quad (9)$$

Equation (8) is a first-order differential equation with initial condition $\Delta p(0) = 0$.

What we really need is the cosine Fourier transform (FT) of the excess hole concentration, referred to its asymptotic limit taken as a base line, i.e., $\Delta p(t) - \Delta p(\infty)$, because what the Fourier transform spectrometer gives is essentially the corresponding transform of the excess photocurrent, $\Delta i(t) - \Delta i(\infty)$. Transient effects on $\Delta p(t)$ [and $\Delta i(t)$] due to switching on the ir source can be removed by moving the time origin back to $-\infty$. We thus take the time domain for Eq. (8) to be $-\infty < t < \infty$, with initial condition $\Delta p(-\infty) = 0$. To obtain the FT of $\Delta p(t)$ consider the function $\Delta p(\omega, t)$ defined by

$$\frac{d\Delta p(\omega, t)}{dt} = G_h(\omega, t) - \frac{1}{\tau_{hc}} \Delta p(\omega, t), \quad -\infty < t < \infty, \quad (10)$$

with initial condition $\Delta p(\omega, -\infty) = 0$. We can write

$$\Delta p(t) = \int_0^\infty \Delta p(\omega, t) d\omega. \quad (11)$$

TABLE I. Lorentz profile parameters [Eq. (26)] for In $p_{3/2}$ lines in Si(In). Values of ω_0 were converted from values in meV given in Ref. 11. Values of A and Γ were obtained from unpublished data by J. J. Rome.

Line	ω_0 (cm ⁻¹)	$A = \int \sigma(\omega) d\omega$ (10 ⁻¹⁶ cm)	Γ (cm ⁻¹)	$\sigma_0 = \frac{A}{\pi\Gamma}$ (10 ⁻¹⁶ cm ²)
1	1145.41	0.643	1.5	0.1365
2	1175.82	5.20	1.1	1.505
3	1208.00	0.2165	1.05	0.0656
4	1216.31	1.44	0.7	0.656
4(A,B)	1218.57 (4A)	2.06	1.0	0.656
6	1236.56	0.245	0.95	0.0822
8	1241.96	0.174	0.75	0.0738
∞ (In)	1254.79			
∞ (Al)	556.78			

The solution to Eq. (10), in the limit

$$\Omega\tau_{hc} \ll 1, \quad (12)$$

is

$$\Delta p(\omega, t) \approx G_{h0}^{(\omega)} \tau_{hc} [1 + \cos(\Omega t)], \quad (13)$$

where

$$G_{h0}^{(\omega)} = \mathcal{F}_0^{(\omega)} \sum_j \sigma_{hj}^{(\omega)} I_{hj}^{(\omega)} N_j^{(0)}(0). \quad (14)$$

For Si, $\Omega\tau_c \leq 10^{-6}$, so the condition (12) is well satisfied. As pointed out by Darken,^{5,6} the significance of the resulting Eq. (13) is that the FT of the excess photocurrent $\Delta i(t) - \Delta i(\infty)$ will be in phase with the modulated ir light. Thus

$$\Delta p(t) = \int_0^\infty G_{h0}^{(\omega)} \tau_{hc} [1 + \cos(\Omega t)] d\omega, \quad (15)$$

$$\Delta p(\infty) = \int_0^\infty G_{h0}^{(\omega)} \tau_{hc} d\omega, \quad (16)$$

$$\begin{aligned} \Delta p(t) - \Delta p(\infty) &= \int_0^\infty G_{h0}^{(\omega)} \tau_{hc} \cos(\Omega t) d\omega \\ &= \frac{c\tau_{hc}}{2\nu} \int_0^\infty G_{h0}^{(\omega)} \cos(\Omega t) d\Omega. \end{aligned} \quad (17)$$

Denoting the cosine FT of $\Delta p(t) - \Delta p(\infty)$ by $\Delta p(\Omega)$, we have

$$\Delta p(\Omega) = \frac{\pi c \tau_{hc}}{4\nu} G_{h0}^{(\omega)}. \quad (18)$$

Step 4. Up to this point, no assumptions have been made regarding the form of $G_{h0}^{(\omega)}$. The simplest assumptions to make are (1) the ir source has appreciable intensity only over the discrete portion of the impurity spectrum for each impurity species which may be present, and (2) the photoabsorption cross section $\sigma^{(n)}$ to the n th excited level of species j is so sharply peaked that it can be represented by a δ function centered at the absorption frequency $\omega_{jn} = (E_{jn} - E_0)/\hbar$.

The consequence of these assumptions is that

$$G_{h0}^{(\omega)} \approx \mathcal{F}_0^{(\omega)} \sum_{j,n} \sigma_{hjn} I_{hj}^{(\omega)} N_j^{(0)}(0) \delta(\omega - \omega_{jn}), \quad (19)$$

where σ_{hjn} is the integrated cross section for the n th line of species j , proportional to the oscillator strength of the line. Then the Fourier-transformed photoresponse should consist of discrete peaks, each with area proportional to $\mathcal{F}_0(\omega_{jn}) \sigma_{hjn} I_{hj}(\omega_{jn}) N_j(0)$. This result has been used to obtain relative acceptor concentrations in Ge (Ref. 5), assuming they are in the same ratio as the areas under the corresponding peaks (in) and (jn). This is equivalent to assuming

$$\mathcal{F}_0(\omega_{in}) \sigma_{hin} I_{hi}^{(\omega_{in})} = \mathcal{F}_0(\omega_{jn}) \sigma_{hjn} I_{hj}^{(\omega_{jn})}. \quad (20)$$

TABLE II. Lorentz profile parameters [Eq. (26)] for Ga $p_{3/2}$ lines in Si(Ga). Values of ω_0 were converted from values in meV given in Ref. 11. Values of A and Γ were obtained from unpublished data by J. J. Rome.

Line	ω_0 (cm ⁻¹)	$A = \int \sigma(\omega) d\omega$ (10 ⁻¹⁴ cm)	Γ (cm ⁻¹)	$\sigma_0 = \frac{A}{\pi\Gamma}$ (10 ⁻¹⁴ cm ²)
1	469.75	0.460	0.65	0.225
3	541.45	0.0944	0.65	0.04624
4	548.23	0.567	0.325	0.555
4(A,B)	551.42	0.590	0.777	0.242
6	568.71	0.131	0.45	0.0930
8	573.72	0.139	0.35	0.126
10	581.30	0.0472	0.425	0.0354
∞ (Ga)	586.62			
∞ (B)	358.04			

Equation (20) has some validity in Ge because of the closeness of the ground-state energies of different acceptor species. We would not expect it to be valid in Si, essentially because the temperature dependence of the photothermal ionization probability $I_{hj}(\omega_{jn})$ (Refs. 7-9) introduces large differences between $I_{hi}(\omega_{in})$ and $I_{hj}(\omega_{jn})$ at the low temperatures of measurement. We have therefore gone beyond the above two assumptions by using experimental data to represent the cross sections by a superposition of Lorentzian peaks with a continuum contribution above the absorption edge, and the photothermal ionization probabilities by Boltzmann factors.

Step 5. One consequence of treating the cross sections and ionization probabilities more realistically is that the discrete portion of one impurity spectrum may overlap with the continuum of a shallower impurity, with the form of the discrete spectrum depending strongly on temperature and on the attenuation of the incident light within the sample. In order to examine this situation more closely we shall first define a PTIS response function.

For a spectral intensity $\mathcal{I}(\omega, t)$ incident on a slab of thickness d , the intensity at depth z is¹⁰

$$\mathcal{I}_z(\omega, t) = \mathcal{I}(\omega, t) \frac{(1-R)(e^{-\alpha z} + R e^{-2\alpha d} e^{\alpha z})}{1 - R^2 e^{-2\alpha d}}, \quad (21)$$

where R is the reflectivity and α is the linear absorption coefficient. The spectral component of the excess photoconductivity at depth z is proportional to $\Delta p_z(\Omega)$ and therefore $\mathcal{I}_z(\omega, t)$, so the spectrum of the excess photo-current is

$$\Delta i(\Omega) \sim \int_0^d \Delta \sigma_z(\Omega) dz \sim G_{h0}^{(\omega)} \left[\frac{(1-R)(1-e^{-\alpha d})}{\alpha(1-R e^{-\alpha d})} \right]. \quad (22)$$

The absorption coefficient may be written

$$\alpha(\omega) = \sum_j \sigma_{hj} N_j^{(0)} + \alpha_0(\omega), \quad (23)$$

where $\alpha_0(\omega)$ is the contribution from all other sources, including Breit-Wigner-Fano (BWF) excitations, localized vibrational excitations of electrically inactive impurities such as C and O and of impurity or impurity-vacancy complexes, and optical phonon excitation of the host lattice.

Assume $\Delta i(\Omega)$ is referenced to a standard detector

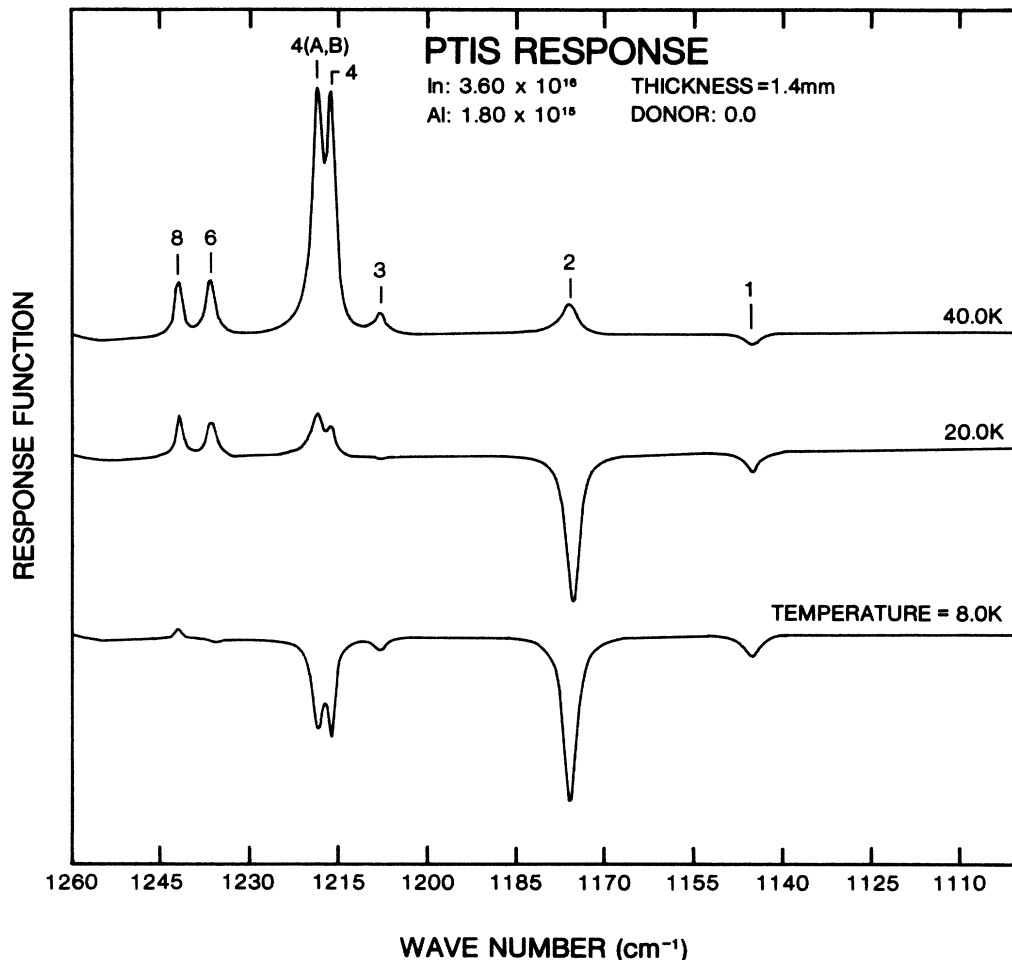


FIG. 1. Model calculation of the PTIS response function [Eq. (24)] for Si(In,Al) in the region of overlap of the In $p_{3/2}$ spectrum with the Al continuum. In concentration $3.6 \times 10^{16}/\text{cm}^3$, Al concentration $1.8 \times 10^{15}/\text{cm}^3$, donor concentration zero and sample thickness 1.4 mm. Other sources of absorption are ignored.

whose response $\Delta i_0(\Omega)$ is flat in the frequency range of interest, and that R is independent of frequency in this range. Then the relative PTIS response of the sample, $\Delta i(\Omega)/\Delta i_0(\Omega)$, is proportional to the response function $f(\omega, T)$, defined by

$$f(\omega, T) = \left(\frac{g_h^{(\omega)}}{\alpha(\omega)} \right) \left(\frac{1 - e^{-\alpha(\omega)d}}{1 - R e^{-\alpha(\omega)d}} \right), \quad (24)$$

where

$$g_h^{(\omega)} = \sum_j \sigma_{hj}^{(\omega)} I_{hj}^{(\omega)} N_j^{(0)}(0) \quad (25)$$

= number of holes generated by a photon

of frequency ω , per unit length of sample,

via photothermal ionization.

III. CALCULATIONS

We have applied the theory described in the preceding section to the calculation of model PTIS spectra for

Si(In,Al) and Si(Ga,B). Experimental values of photoabsorption line positions, line strengths (i.e., integrated peak areas) and linewidths, taken from measurements on singly doped float-zone samples Si(In) and Si(Ga) of known dopant concentration,¹¹ were used to obtain the parameters σ_0 (peak height), ω_0 (peak position), and 2Γ (full width at half-maximum) in the expression

$$\sigma(\omega) = \frac{\sigma_0}{1 + \left[\frac{\omega - \omega_0}{\Gamma} \right]^2}, \quad (26)$$

for several $P_{3/2}$ lines of the deeper acceptor. The parameter values are given in Tables I and II. The ionization limits are designated by ∞ . The corresponding limits for Al and B are included for comparison.

The continuum contribution for each species j was approximated by the expression, due to Lucovsky,¹²

$$\sigma_{j^{(\omega)}}^{(\text{cont})} = \frac{1}{n} \left(\frac{E_{\text{eff}}}{E_0} \right)^2 \frac{16\pi e^2 \omega_{jl}^{1/2} (\omega - \omega_{jl})^{3/2}}{3m^* c \omega^3}, \quad (27)$$

where n is the index of refraction of the medium and E_{eff}/E_0 is the ratio of the effective electric field at the

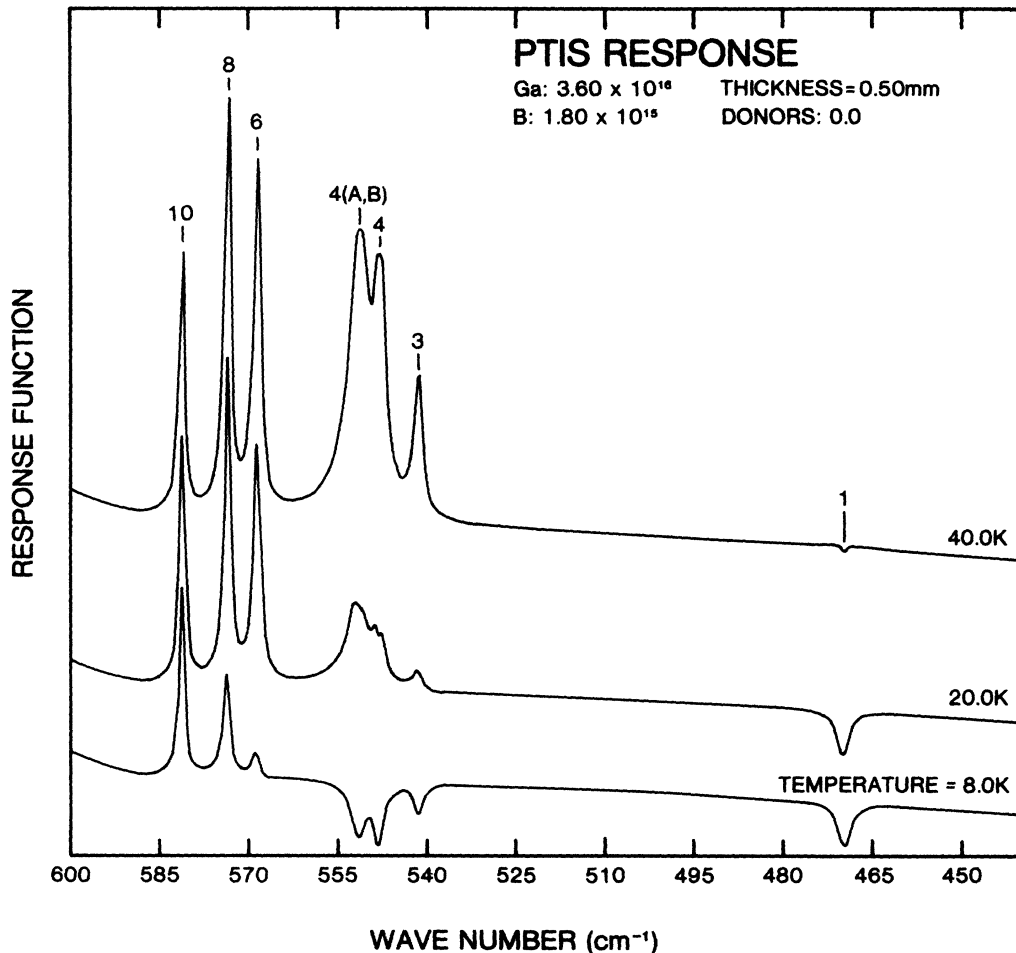


FIG. 2. Model calculation of the PTIS response function [Eq. (24)] for Si(Ga,B) in the region of overlap of the Ga $p_{3/2}$ spectrum with the B continuum. Ga concentration $3.6 \times 10^{16}/\text{cm}^3$, B concentration $1.8 \times 10^{15}/\text{cm}^3$, donor concentration zero, and sample thickness 0.50 mm. Other sources of absorption are ignored.

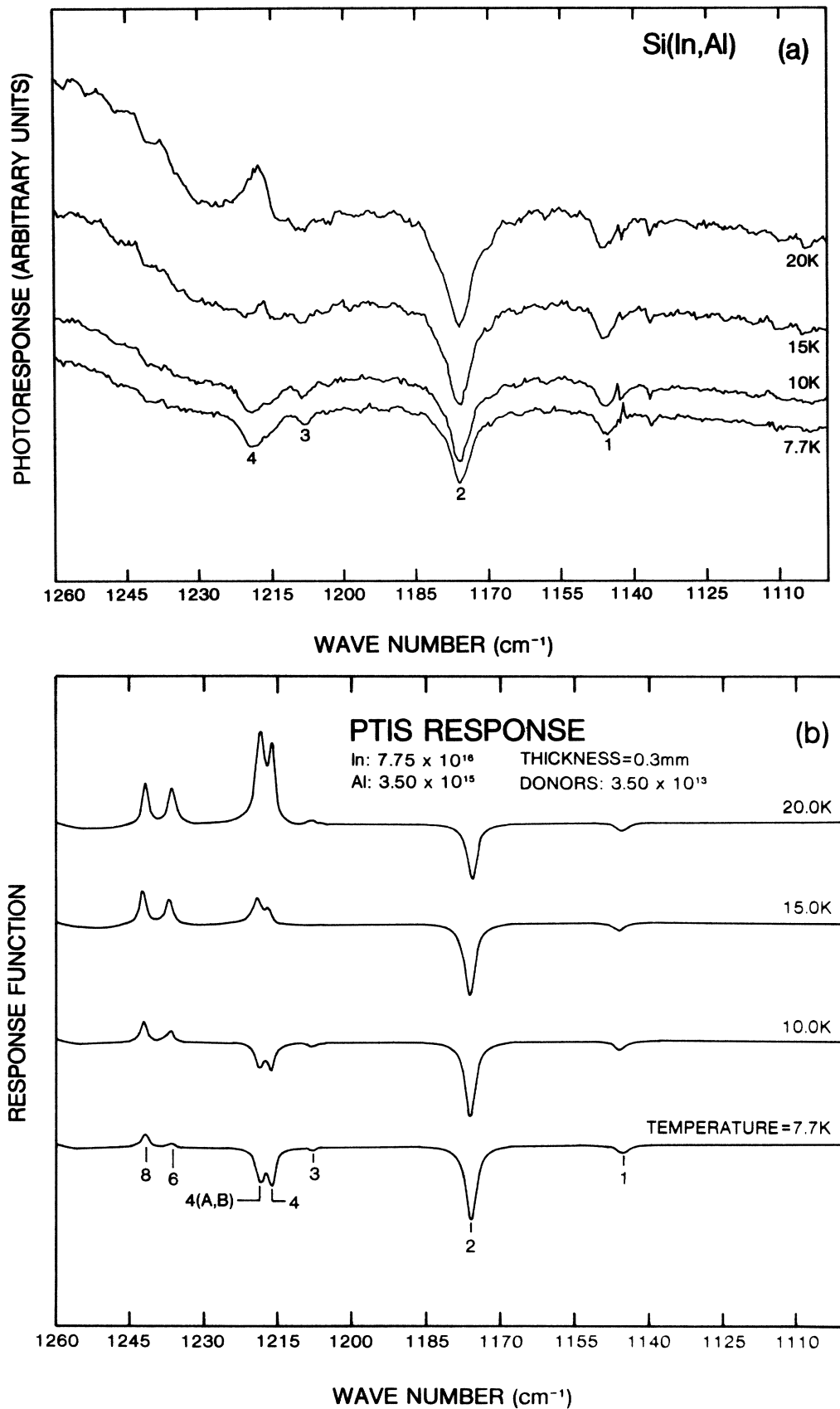


FIG. 3. (a) Portion of an experimental PTIS spectrum of a float-grown Si(In,Al) sample of thickness 0.3 mm, having In concentration $7.75 \times 10^{16}/\text{cm}^3$, Al concentration $3.5 \times 10^{15}/\text{cm}^3$, and donor concentration $3.5 \times 10^{13}/\text{cm}^3$. (b) Model response function for the sample in (a), calculated from Eq. (24).

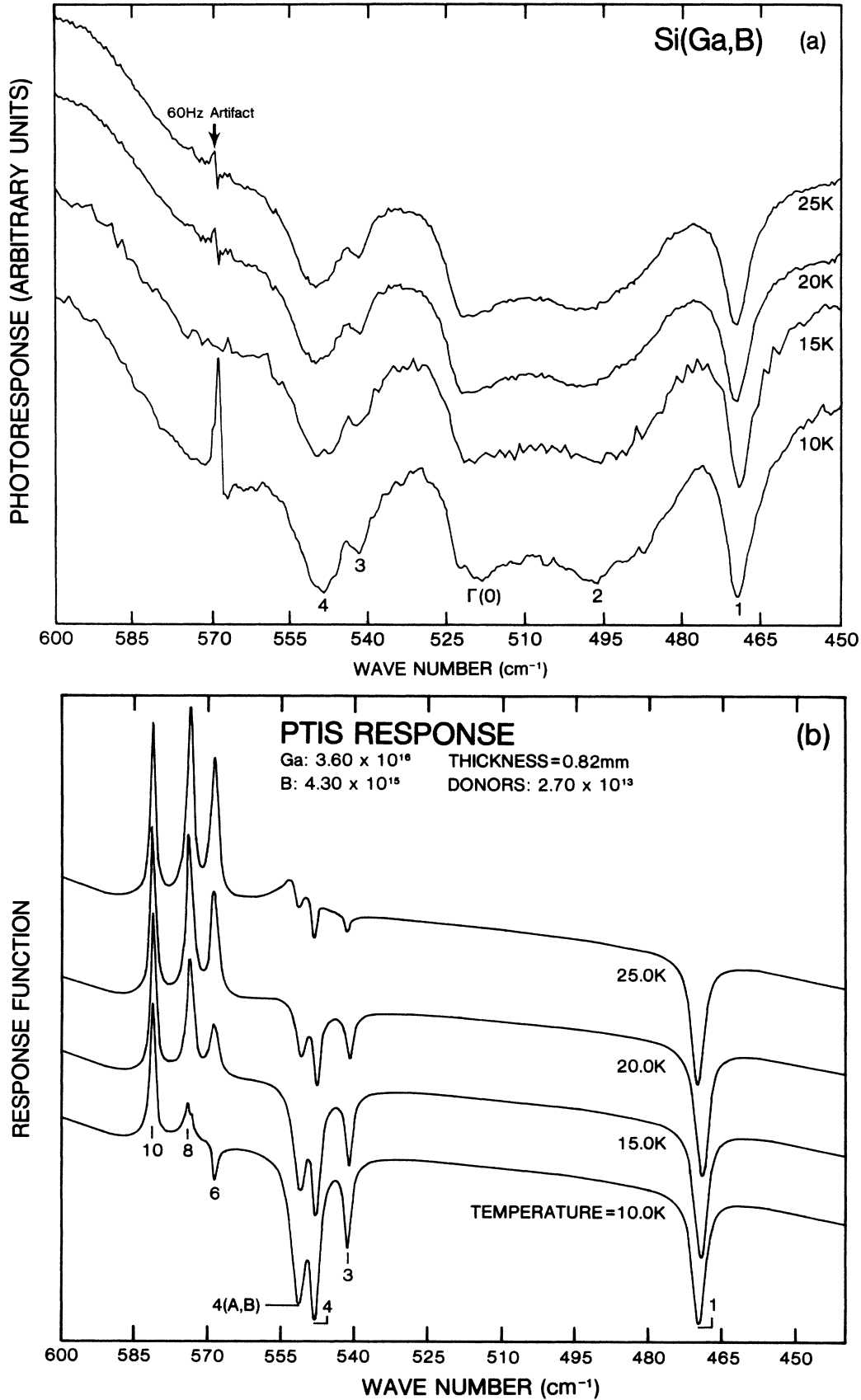


FIG. 4. (a) Portion of an experimental PTIS spectrum of a float-grown Si(Ga,B) sample of thickness 0.82 mm, having Ga concentration $3.6 \times 10^{16}/\text{cm}^3$, B concentration $4.3 \times 10^{15}/\text{cm}^3$, and donor concentration $2.7 \times 10^{13}/\text{cm}^3$. (b) Model response function for the sample in (a), calculated from Eq. (24).

impurity to the average field in the medium. Following Ref. 12, we take this ratio to be 2.5 for all acceptors. Equation (27), though approximate, will be sufficient for our purposes.

For the photothermal ionization probability we have used the expression derived by Abakumov and Yassievich,⁸

$$I_{hj}^{(\omega_{jn})} = \left[1 + \frac{U_{jn}}{k_B T} \right] \exp \left[- \frac{U_{jn}}{k_B T} \right], \quad (28)$$

where $U_{jn} = \hbar\omega_{jI} - \hbar\omega_{jn}$ is the binding energy of a hole in level n of species j , and ω_{jI} is the ionization limit of species j . An improved expression for this probability has been obtained by Jongbloets *et al.*,⁹ but numerical comparison of the two expressions indicates that they do not differ very much over the temperature range of interest.⁷

Figures 1 and 2 show calculations of the $f(\omega, T)$ for model samples of Si(In,Al) and Si(Ga,B) at various temperatures. These figures clearly show the effect of the increase in photothermal ionization probability with temperature, especially in transforming negative In and Ga peaks into positive ones. By comparison, the actual reduction in the concentration of neutrals N_{In} and N_{Ga} through photoionization, though taken into account in the calculations, was quite small in this temperature range. It should be noted that existence of a background is necessary for negative peaks to occur; in the absence of background, Eq. (24) gives positive peaks only.

Figures 3 and 4 compare actual PTIS spectra with model calculations. The samples used here were float-zone,¹³ but similar effects have been seen in Czochralski-grown Si(In,Al) (Ref. 14). The model calculations compare well with the observed spectra with respect to the relative intensities of the various lines in a given spectrum, as a function of temperature, especially the temperature at which the signature of a given line changes from negative to positive. The experimental linewidths are broader than those occurring in the model since the latter were fit to data on singly doped samples, which generally show narrower lines. As indicated in Fig. 4(a), the gallium line 2 is

suppressed by the presence of the $\Gamma(0)$ optical phonon, and therefore was omitted in Fig. 4(b). Phonon and other possible contributions to the absorption were omitted in these calculations, i.e., $\alpha_0(\omega)$ was taken to be zero in Eq. (23). The donor concentration in these samples was small, of order 1% of the shallower acceptor, and had negligible effect on the calculated spectra. More generally, in the absence of band-edge light Eq. (1) shows that even in a heavily compensated sample the only effect of the donors is essentially to reduce the concentration of neutrals of the shallower acceptor by an amount equal to the donor concentration. In the PTIS spectrum this will tend to make the positive peaks of the deeper acceptor appear at lower temperatures (in addition to reducing the intensity of the shallow-acceptor lines).

IV. SUMMARY

In this paper we have derived a response function for photothermal ionization in multiply doped p -type silicon, which incorporates experimental line strengths, positions, and widths obtained from photoabsorption measurements on singly doped samples. The model has been applied to doubly doped Si(In,Al) and Si(Ga,B) and comparison with actual spectra shows good agreement for the relative strengths of the In lines and of the Ga lines, and validates the physical mechanism underlying the effect of temperature on the signatures of these lines. The model provides a general framework which can be extended to include other effects, such as donor contribution to the PTIS response in the presence of band-edge light.

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