Nonexponentiality in photoinduced current transients in undoped semi-insulating gallium arsenide

P. K. Giri and Y. N. Mohapatra[a]
Department of Physics, Indian Institute of Technology, Kanpur 208016, India

An isothermal spectroscopic technique called time-analyzed transient spectroscopy (TATS) has been used to study photoinduced current transients in undoped semi-insulating GaAs. It is demonstrated that this has many advantages over conventionally used photoinduced transient spectroscopy (PITS). Specifically, TATS provides both quantitative measure and qualitative insights to the nonexponentiality of current transients commonly encountered in these materials. Using this spectroscopy, features related to enigmatic negative peaks resulting from rising current transients in these materials are reported. A simple kinetic model has been proposed to explain the essential features of rising transients leading to negative peaks in both TATS and PITS spectra. © *1995 American Institute of Physics.*

I. INTRODUCTION

Semi-insulating gallium arsenide (SI GaAs) is well recognized as a substrate material for application in high-speed integrated circuits and optoelectronic devices; however, characterization of SI materials poses problems not encountered in conducting materials. Photoinduced transient spectroscopy (PITS),[1-12] which is based on the analysis of thermal behavior of photocurrent transients induced by light pulses, has been widely used to study several high-resistivity semiconductors,[10,13-16] especially in detection and characterization of deep traps in SI GaAs substrates.[4,10,16-20]

Apart from the difficulties in trap concentration estimation by this method, oversimplification such as the neglect of carrier recapture and assumptions regarding exponential nature of photocurrent decay transients have posed serious interpretive problems in analyzing PITS results. Moreover, in conventional PITS experiments the choice of rate window is limited at the fast end by the time required to extinguish the light and at the slow end by the decay of the photocurrent at the values where it approaches the dark current level.[18] Extraction of trap parameters by isothermal transient analysis[18,21,22] has not been promising because the base-line determination of the exponential is crucial and it is important to know the dark current value at each temperature.

Normally one expects to observe decaying photocurrent transients leading to positive peaks in PITS spectra. However, most often negative peaks arising out of rising transients have been observed in the case of SI GaAs. Although the occurrence of a negative peak in PITS spectra has been widely discussed in the literature,[1,4,6,8,17,19] it is one of the most poorly understood features of PITS spectra and continues to be an enigma. The defect phenomena responsible for this feature have not been isolated and their relevance, if any, to the compensation mechanism of SI GaAs has not been identified.

In spite of the inherent nonexponential nature of the photoinduced current transients, several workers have attempted to fit multiple exponentials[12,23-25] for the observed

transients basing their analysis on questionable assumptions. Abele and co-workers[18] and Blight and Thomas[19] have pointed out the severity of problems associated with fitting multiple exponentials by successive subtraction method or by the method of moments. Hence, the ability to analyze the nature of the nonexponentiality of transients is a major challenge in both characterization and understanding of phenomena in these materials.

In this work we attempt to critically analyze the experimental photoinduced current transients involved in characterization of SI GaAs material. We demonstrate the advantages of an isothermal spectroscopy, called time-analyzed transient spectroscopy (TATS), over conventional PITS in the analysis of these transients. We focus especially on TATS analysis of rising transients, which normally result in negative peaks in PITS spectra of SI GaAs. We show that TATS analysis is a better guide in understanding phenomena underlying occurrence of negative peaks. We propose a simple kinetic model for the observed nonexponential transients leading to negative peak.

II. EXPERIMENTAL DETAILS

The samples used in this work are commercial undoped semi-insulating GaAs cut from a 400-μm-thick wafer. The SI property is due to the presence of a complex center, known as EL2, which compensates the shallow accepters.[26] We used the planar structure with electrical ohmic contacts in the form of stripes on front surface by evaporating Au/Ge alloy in vacuum. The metal contacts are annealed at 450 °C in flowing forming gas. The samples were mounted on a liquid-nitrogen-cooled cold-finger-type cryostat with temperature controlled to within 0.1 K in the range 90–400 K. A copper-Constantan thermocouple was mounted on the sample holder close to the device. Light pulses were provided from the He-Ne laser source (6328 Å) with the help of a mechanical chopper with cutoff time less than 0.2 ms. The light beam was tightly focused to avoid the effect of carrier injection through contacts. Typically a dc voltage of 30 V was applied across 2–3 mm separation between the contacts. The *I-V* characteristics in this range was linear. The current transient

[a]Electronic mail: ynm@iitk.ernet.in

is digitized at each temperature with the help of Keithley 236 source measure unit. The whole setup is personal-computer (PC) controlled except for temperature control. Each acquired transient is then used for further analysis.

III. METHOD OF ANALYZING DATA

The decay of photocurrent in semi-insulating III-V materials, after removal of an intrinsic light, carries information regarding emission of carriers from deep traps in the material. The form of current transient due to a single trap is most often assumed to be of the form

$$I(t) = I_0 e_n \exp(-e_n t), \tag{1}$$

where I_0 is a constant and e_n is the characteristic emission rate from the trap at any temperature. This rate is given by

$$e_n = \gamma_n \sigma_{Tn} T^2 \exp(-E_T/k_B T) = \tau_T^{-1}, \tag{2}$$

where $\gamma_n = (G_{T0}/G_{T1})(N_c v_n/T^2)$ is a temperature-independent constant, σ_{Tn} is the trap capture cross section of electrons, G_{T0} and G_{T1} are respectively, the degeneracy of the occupied and the unoccupied trap, N_c is the conduction-band effective density of states, and v_n is the mean electron speed.

Among many methods of analysis of such transients, PITS is the most common. In this method, the transients are analyzed by plotting the difference $I(t_1) - I(t_2)$ as a function of temperature [as in deep-level transient spectroscopy (DLTS)] for a specific choice of sample times t_1 and t_2.

The need for a similar technique that is isothermal without losing the advantages of spectroscopy has been recognized in the analysis of capacitance transients in semiconductors.[27,28] In the TATS method the spectroscopic signal is defined as

$$S(t) = I[t,T] - I[t(1+\gamma),T], \tag{3}$$

where I represents isothermal current transient at temperature T and γ is an experimentally chosen constant. This is equivalent to defining a moving window in time at a fixed temperature instead of using a fixed window and scanning the temperature as is commonly done in the case of DLTS and PITS. It can be easily shown that $S(t)$ has a maxima when plotted against $\ln(t)$ and the maxima occur at time t_m give by the relation

$$e_n = \frac{\ln(1+\gamma)}{\gamma t_m}, \tag{4}$$

assuming the exponential form of the current transient as given in Eq. (1). The peak value of the TATS signal is given by

$$S_{\max}(t_m) = \frac{I_0}{t_m} f(\gamma), \tag{5}$$

where

$$f(\gamma) = (1+\gamma)^{-(1+\gamma)/\gamma} \ln(1+\gamma).$$

There are several advantages of TATS analysis over PITS. TATS is a spectroscopy in the time domain alone[27] and therefore distortion in line shape due to possible temperature dependence of the strength of the transient (i.e., pre-exponential factor containing occupancy, etc.) does not occur. In addition, in methods involving temperature scanning such as PITS and DLTS the line shape is dependent on the trap parameters and the range of time scales involved. The width of the TATS peaks depends only on the parameter γ, which is chosen to optimize resolvability and signal-to-noise ratio. For example, the full width half-maximum (FWHM) of the line shape is a constant given by

$$\text{FWHM} = \ln(t_+) - \ln(t_-) = 2.496, \tag{6}$$

for $\gamma = 1$, t_+ and t_- being the times corresponding to the half-maximum of the TATS peak. Because of this, it is very easy to detect and estimate the degree of nonexponentiality from TATS line shape. A convenient way of characterizing nonexponentiality is to represent it by a stretched exponential transient of the form

$$I \sim \exp[-(e_n t)^\beta], \quad \beta \leqslant 1, \tag{7}$$

where β is the stretching factor. This form of transient, also known as William–Watts decay,[29] has been invoked in many physical systems including defect analysis.[30,31] The TATS signal of stretched exponential has a maximum at a t_m given by

$$(e_n t_m)^\beta = \frac{\beta \ln(1+\gamma)}{(1+\gamma)^\beta - 1}, \tag{8}$$

which reduces to the expression in Eq. (4) for $\beta = 1$. Simple calculation shows that the peak position in time t_m does not change appreciably on lowering β. Specifically, for $\gamma = 1$, the peak position (t_m/τ) is 0.990 times the perfect exponential case even for a β as low as 0.5. Hence, estimate of the time constant is independent of the stretching factor β in most cases. The experimental inaccuracies are much larger than is expected due to nonexponentiality factor β.

However, nonexponentiality shows up as broadening of the line shape. If we use Eq. (4) for peak position t_m, then the FWHM of the TATS signal is proportional to $(1/\beta)$ for the case of $\gamma = 1$; hence, a quantitative measure of nonexponentiality is obtained simply by inspection of TATS spectral line shape. For more precise determination of the β factor, one can use precomputed values of FWHM as a function of β. For example, for the case of $\gamma = 1$, we have simulated the FWHM of the TATS line shape for various values of β, which fits into the following straight-line formula:

$$\frac{1}{\beta} = \frac{\text{FWHM} - 0.078}{2.417}, \tag{9}$$

for determination of β correct up to three decimal places.

We have taken advantage of TATS as a guide to nonexponentiality in analyzing our data on current transients.

IV. RESULTS AND DISCUSSION

A. Typical current transients and PITS

Typical photocurrent decay curves are shown in Fig. 1 for two different temperatures. In SI GaAs both these types of transients are commonly observed. In our discussion we refer to transients labeled (a) in Fig. 1 as decaying transients
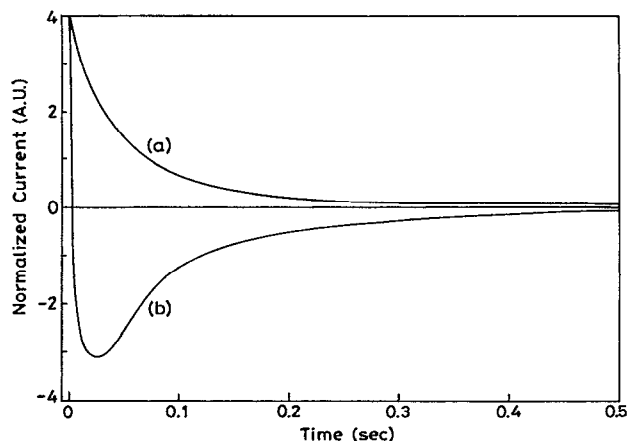
FIG. 1. Typical photocurrent transients in SI-GaAs sample: (a) decaying current at 174.9 K; (b) rising current at 293.2 K.

FIG. 2. Typical PITS spectrum of SI-GaAs sample with $t_1 = 0.141$ s, $t_2 = 0.279$ s, showing negative peak along with broad positive peak.

and to curves of type (b) as rising transients. Decaying photocurrent transients in these materials are interpreted to be due to slow thermal release of carriers from traps filled during the light pulse. Most of the excess carriers would have recombined at a much faster rate normally due to band-to-band recombination; hence, the slow decaying transient carries information regarding the emission rate of carriers from the defect center. However a "clean" interpretation of rising transients such as curve (b) has not been possible and many models have been advanced mainly through PITS studies.[1,4,6,12,19]

As far as transients of type (b) are concerned, note that in most earlier studies only the rising part of the transients have been analyzed. However our data clearly show that all such transient also have a slow decaying component reaching a minimum and then rising to saturation. We have taken care to confirm that the decaying component is not related to any artifact of experiment such as time constant of chopper or $R\text{-}C$ circuit response. This is further borne out by the fact that the slowly decaying component is highly temperature dependent and is always observed along with negative transient. Hence, we believe that the current dipping to a minimum before rising is an essential part of the whole transient and this major deviation from exponentiality needs to be explained. Attempts to fit such transients with multiexponentials incorporating both rising and decaying transients poses severe problems. It normally involves fitting 3–4 exponentials with many independent parameters and is extremely sensitive to the subtraction of base current level. Many different combinations of parameters lead to similar quality of fitting. For these precise reasons isothermal analysis of transients has not been popular. Any analysis of this phenomena that focuses only on the later rising part, as has been done in the literature, can be misleading.

The most popular way of characterizing traps in these materials has been PITS. The decaying transients give rise to positive peaks in PITS spectra. Transients such as given in curve (b) of Fig. 1 result in a negative peak feature in such spectra and have been widely discussed in the literature. Figure 2 shows conventional PITS spectra in the higher
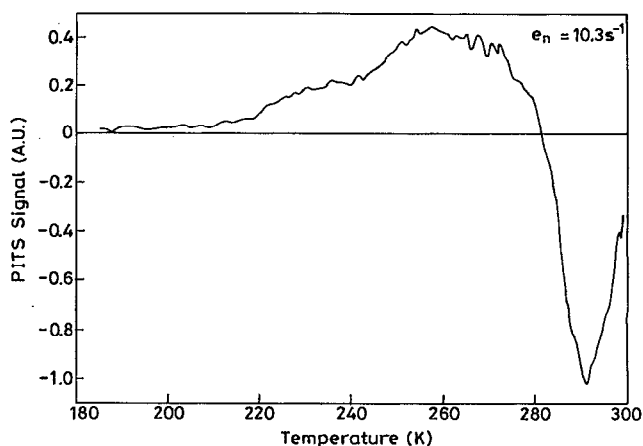
temperature regime where the negative peak feature is clearly observed. This figure also shows a broad positive peak prior to the negative peak and has been seen in most samples. The shape of the negative peak in PITS spectra is highly distorted owing to the presence of this broad positive peak; however, the significance of this feature in relation to the negative peak has not been discussed.[4,19] We also see a well-formed positive peak at lower temperature in the PITS spectra which is discussed later in this section.

In our experiments, as in other reports, we have seen that the strength of the transients is highly dependent on temperature for a constant filling time leading to distortion of PITS spectra. Moreover, most transients are nonexponentials to varying degrees making PITS analysis questionable. Hence, PITS is not well suited for detailed studies involving questions of phenomena, although it can be used as a general purpose survey technique. Specifically, it fails miserably in trying to understand negative peaks or rising transients in SI GaAs.

B. TATS of positive peak

As has already been pointed out in Sec. III, there are several advantages associated with TATS analysis, it being a completely isothermal technique. To demonstrate its efficacy we choose at first to analyze the decaying transients which lead to positive peak in PITS spectra in the temperature range 165–200 K. In Fig. 3(a) we show TATS signals for these transients at several temperatures. The emission time constant is obtained from the peak position using Eq. (4). The corresponding Arrhenius plot is shown in Fig. 3(b) which leads to an activation energy of 0.41 eV with corresponding capture cross section of 6.1×10^{-13} cm^2. This can be attributed to the level called EL6 reported in the literature.[4,32]

As discussed in Sec. III, inspection of the TATS spectra directly shows that transients are nonexponential. This is clearly shown in Fig. 4 where experimental TATS points are shown by symbols and a perfect exponential of the time constant corresponding to the maximum t_m is shown as a
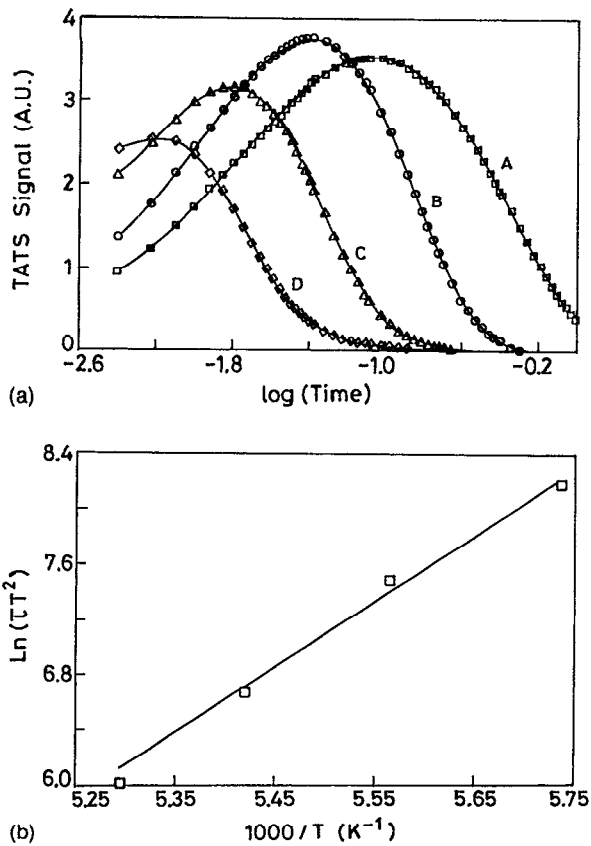
FIG. 3. (a) A series of TATS spectra in a SI-GaAs sample at different temperatures: (A) 174.3 K; (B) 179.7 K; (C) 184.5 K; and (D) 188.8 K. (b) Arrhenius plot for deep trap corresponding to positive TATS peaks shown in (a).
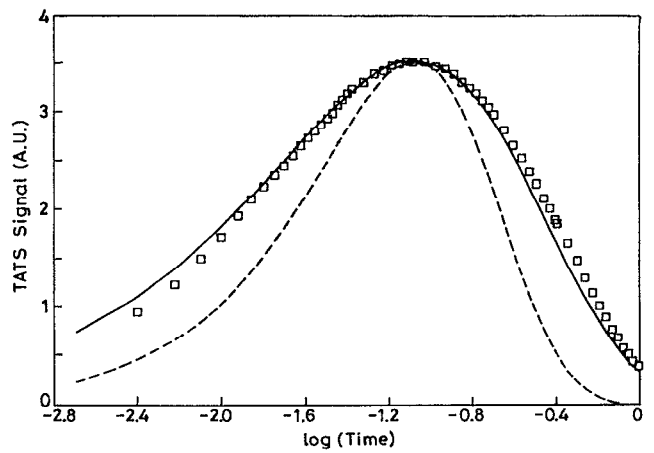

FIG. 4. Comparison of experimental spectra with calculated ones to demonstrate the degree of nonexponentiality. The experimental spectra (with symbols) are broader than the spectrum corresponding to a perfect exponential (dotted) line. The bold line corresponds to a stretched exponential with $\beta = 0.668$ determined from the FWHM of the experimental curve.

dashed curve. From the experimental FWHM the stretching factor β is obtained using Eq. (9) and the corresponding TATS spectra are shown by the bold line in the figure. This clearly shows the power of TATS in determining the degree of the nonexponentiality of transients in a straightforward way. Similarly the stretching factor determined at different temperatures ranges between 0.67 and 0.9, the lower value being obtained at lower temperature. The significance of this systematic deviation from exponentiality with temperature is not clear to us but it could be related to recapture of carriers. The height of the TATS peak at different temperatures does not scale with $(1/t_m)$ as would be expected from Eq. (5). This is mainly due to the change in the degree of occupancy achieved for a fixed exposure time at different temperatures. Normally this would contribute to line-shape distortion in PITS spectra.

C. TATS of negative peak

We use TATS to analyze the nature of the nonexponentiality of rising transients observed at higher temperature. As in PITS spectra, rising transients lead to negative peaks in TATS spectra for several temperatures. Note that the sample shows two negative peaks as opposed to what is usually observed in PITS spectra. This is possibly due to the fact that the smaller negative peak gets submerged in already highly distorted PITS line shape.
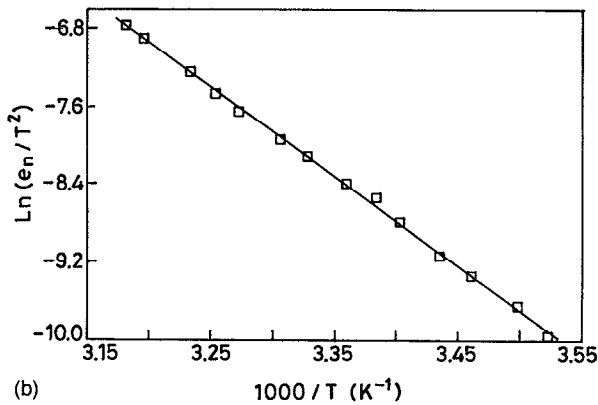
The dominant negative peak in TATS spectra is due to the rising part of the current transient as shown in curve (b) of Fig. 1. Note that the TATS signal falls to zero sharply on the faster side of the peak. In fact it is initially positive corresponding to the decaying part of the transient. The TATS signal goes through zero at a time approximately corresponding to the minimum of the transient. Hence, it is clear that by no stretch of imagination can the transient corresponding to the negative peak be considered exponential or mildly nonexponential by choosing to focus only on the later rising part of such transients.

However, for thermally stimulated relaxation processes, use of Arrhenius plots to estimate the approximate energy involved is a robust procedure, not withstanding nonexponentiality. Hence, we treat the TATS peak shifts with temperature as an indicator of shift in time constant and, hence, plot an Arrhenius plot given in Fig. 5(b). The activation energy so obtained is 0.79 eV with an estimate of capture cross section from the slope as 2.3×10^{-11} cm^2. The activation energy of negative peaks found in earlier works[4,7,19,33] normally ranges between 0.65 and 1.2 eV. Therefore, it has always been attributed to midgap centers, although the definite identification has remained controversial.

Figure 6 shows TATS of transients at a particular temperature (290 K) for various values of filling time, i.e., the time for which the laser light is on. The change in height of the TATS peak of the main negative peak is an indicator of the slow increase in the corresponding filled trap concentration. However, it is seen that the second negative peak height remains constant indicating that saturation of the peak occurs in time scale faster than the shortest filling time scale used in these experiments. The extent of trap filling is found to be highly temperature dependent and studying filling behavior is rewarding using an isothermal spectroscopy such as TATS.

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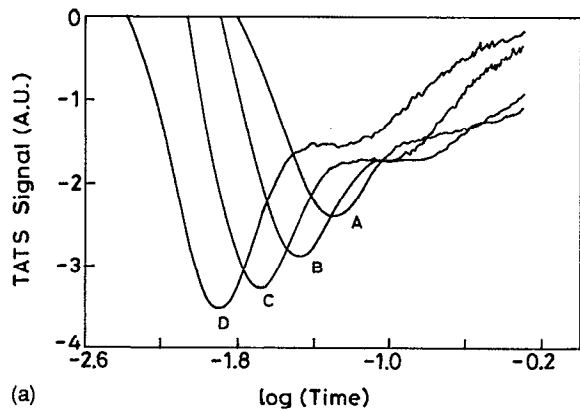
FIG. 5. (a) TATS spectra with $\gamma = 1$ in SI-GaAs samples at different temperatures: (A) 293.9 K; (B) 297.7 K; (C) 302.5 K; and (D) 307.4 K showing two negative peaks for each temperature. (b) Arrhenius plot for the dominant negative peak of (a).

It also demonstrates that the height of conventional PITS peaks cannot be considered as a healthy indicator of the extent of filling.

Both the functional form of the transient and features in the TATS line shape open up a new set of questions regarding the mechanism behind these transients. It is clear that such transients should by no means be considered as a de-
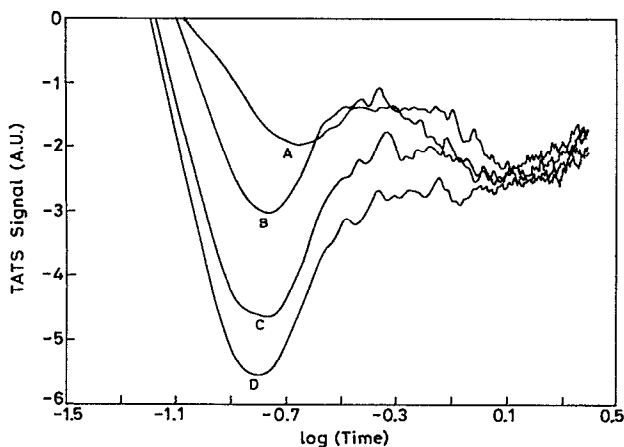

FIG. 6. TATS spectra for SI GaAs at temperature 290 K for different filling times: (A) 20 ms; (B) 70 ms; (C) 100 ms; and (D) 200 ms.

viation from the exponential. It would be inappropriate to characterize the nonexponentiality by stretched exponential parameter β, or distribution in time constants either discrete or continuous.

Explicit inclusion of capture processes in the rate equations, as has been attempted in recent models,[12] cannot lead to transient solutions with a definite cusplike behavior in the functional form: They would be still either rising or decaying depending on initial conditions. In order to isolate the essential features that any mechanism must possess, we propose a simple kinetic model without attempting to pinpoint any physical mechanism at this stage. The most significant feature of our model is to introduce a new rate process which acts as a delay between capture of a carrier and its emission. For simplicity, we assume that only one dominant electron trap and one band, say, the conduction band, is involved in the process. An electron gets captured at an empty trap taking the defect to a metastable state from which it must relax to the ground state. It can then finally emit the carriers to the band. We can write down the rate equations as follows:

$$\frac{dA^+}{dt} = -A^+A^+C + e_n A^0, \tag{10}$$

$$\frac{dA^*}{dt} = A^+A^+C - rA^*, \tag{11}$$

with the constraint that $A^0 + A^* + A^+ = 1$, where A^+, A^*, and A^0 are the concentrations of empty, occupied metastable state, and occupied ground state, respectively, r is the rate of conversion from A^* to A^0, and C is the product of capture cross section σ, average thermal velocity v_{th}, and the total trap concentration. It is also assumed that the concentration of electrons in the band is equal to the concentration of empty traps, an assumption mostly valid in these materials in the dark. The rate equations are written with all concentrations normalized with respect to the total trap concentration. The desired delay between capture and emission is introduced by the negative term in Eq. (11) where it is treated like an emission term insofar as it depends only on the concentration of the metastable state. The rate equations can conveniently be summarized in a state transition diagram as is given in inset of Fig. 7.

These rate equations are solved numerically on a PC. The choice of parameter e_n is taken from the experimental Arrhenius plot, and normalized capture coefficients and initial concentrations are chosen to correspond approximately to EL2 parameters in SI GaAs. The rate r is chosen so as to produce qualitative features of our data. Figure 7 shows the TATS of a particular solution obtained numerically and compares it with a typical experimental TATS curve for negative peak. The ability of any model to mimic cusplike behavior of the transient or positive-to-negative crossover in the TATS spectrum puts a severe restriction on the class of models that need be considered. The ability of our model to mimic these essential features of the data opens up new possibilities as regards study of underlying mechanisms. We have assumed that e_n is the dominant thermally controlled parameter; how-
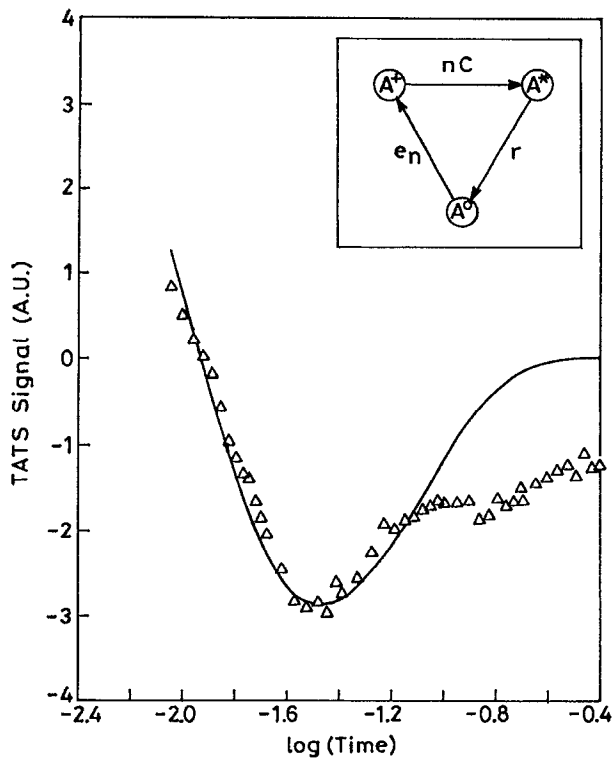
FIG. 7. TATS spectra for the experimental data (symbols) and calculated data (bold line) from the proposed model with EL2 parameters. The chosen parameters as defined in Eqs. (10) and (11) are: $e_n = 19.5$ s^{-1}; $C = 1 \times 10^4$ s^{-1}; $r_c = 1$ s^{-1}; $A^+(0) = 0.011$; $A^*(0) = 0.96$.

the M center in InP. At low temperatures, EL2 centers in GaAs show optically induced metastability without a change in charge state. We believe that the occurrence of complex transient forms should be taken as possible indicator of involvement of metastable states even at such relatively high temperatures in these materials.

V. CONCLUSIONS

We have used isothermal spectroscopy to analyze photo-induced current transients in SI GaAs. We demonstrate the efficacy of such a method and show its advantages over conventional PITS. We further use this method to study the enigmatic negative peak and propose a qualitative model that successfully mimics the new essential features of the negative peak identified in this work. The possible role of metastable-to-stable relaxation processes in giving rise to a negative peak in PITS or TATS has also been proposed.

ACKNOWLEDGMENTS

ever, it is possible that even the rate of conversion r is temperature dependent and, hence, the energy measured through the Arrhenius plot is an effective energy associated with this combined process.

Most earlier models[1,4,6,12] to explain rising transients predict exponential transients hoping to attribute nonexponentiality to second-order effects such as the simultaneous presence of capture processes; however, the negative nature of the transient is attributed to special initial conditions on parameters such that occupation of traps in the dark is higher than under illumination.[1,6,12] Our experimental results and the phenomenological model discussed above suggest that a new term in the rate equation is required instead of tailoring the initial conditions alone. It has been suggested earlier that presence of a large dark current plays a crucial role in giving rise to the negative peak.[12] In contrast, note that we have observed negative peaks even at temperature well below room temperature where the dark currents are negligibly small. There has been a qualitative explanation of negative peak based on surface states;[19] however, recent experimental results[12,34] show that the negative peak is not merely a property of surface-related effects.

The identification of the nature and origin of physical processes giving rise to metastability, as suggested in our kinetic model, would require more detailed experimental investigations; however, the existence of metastable states with involvement of slow relaxation processes has often been invoked in relation to deep levels.[35] For example, charge-state-driven bistability of states is known to occur in the case of

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