

Analytical description of thermally stimulated luminescence and conductivity without the quasiequilibrium approximation

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A generalized mathematical description of thermally stimulated luminescence (TL) and thermally stimulated conductivity (TSC) is presented in terms of a formulation that replaces the quasiequilibrium (QE) and kinetic-order approximations by two new functions $Q(T)$ and $P(T)$, respectively. These functions are related and can be described in terms of the physically meaningful processes of charge trapping, release, and recombination. With the $Q(T)$ and $P(T)$ functions and their relations to the reaction rates, we show that slow-retrapping (first-order) processes will be dominant in systems for which QE is satisfied, and that QE and non-first-order kinetics are incompatible. We develop general expressions for TL and TSC in terms of the $Q(T)$ and $P(T)$ functions and arrive at analytical solutions to the rate equations for the first-order case without the QE approximation and without the presence of $Q(T)$ in the final equations. These general first-order equations are then parametrized and compared to curve shapes generated both from the Randall-Wilkins expressions and from numerical solutions to the rate equations. We show that the general first-order equations reproduce the numerical solutions over a wider range of parameter cases than do the Randall-Wilkins equations. In addition, we suggest a simple method for the extraction of activation energies from experimental TL and TSC peaks, independent of the QE approximation.

I. INTRODUCTION

Thermally stimulated luminescence (or thermoluminescence, TL) and thermally stimulated conductivity (TSC) have long been used as methods for determining the characteristic parameters of deep, localized states in semiconductors and insulators. The methods involve the establishing of a nonequilibrium, metastable concentration of trapped charges at deep states within the band gap of the material (usually by illuminating the specimen with ionizing radiation of energy greater than the band gap). Heating of the sample then results in the thermally stimulated release of the trapped charges which subsequently recombine. If the recombination is radiative, a transient luminescence signal results (known as a TL "glow peak" or "glow curve"); furthermore, if the transitions involve passage through the delocalized bands a transient conductivity signal is recorded (TSC). The TL and TSC peaks so generated may be used to determine the energy depth of the trap from which the charge was stimulated, along with information concerning other factors such as capture cross sections and relative concentrations.^{1,2}

The usual methods of analysis for TL and TSC curves proceed using the descriptions of these processes devised during the 1940s by Randall and Wilkins³ and Garlick and Gibson.⁴ The model described by these authors consists of a single electron-trapping level and a single hole-trapping level. During excitation electrons and holes become trapped, after which thermal stimulation causes the release of the trapped electrons into the conduction band, giving a TSC signal. Recombination with the trapped holes follows, with the subsequent production of TL. The

concept of free holes recombining with trapped electrons is equally acceptable. Later analysis introduced a third, thermally disconnected (electron) level,^{5,6} but the basic approach remained unaltered. The fundamental challenge has been to use the adopted model to describe the theoretical curve shape for TL and/or TSC, and from this to extract information regarding the trapping parameters (trap depth, capture cross section, etc.), to which end a wide range of analytical methods has been suggested. The usual procedure is to write a set of differential rate equations to describe the flow of charge between the traps and recombination centers during thermal stimulation. Due to their nonlinear coupling, however, these equations become intractable and analytical solutions are not possible even for the simplest of systems. As a result, additional simplifying relationships have to be introduced; these include the "kinetic-order" (KO) assumption regarding the relative rates of recombination and retrapping, and the "quasiequilibrium" (QE) approximation regarding the rate at which free charge accumulates in the delocalized bands versus the rate of recombination. It is this last approximation which results in the decoupling of the rate equations. The resulting expressions for the TL and TSC peaks are then used as the basis for several families of analytical procedures, which may be conveniently listed as initial rise techniques,⁴ heating rate methods,⁷ peak shape methods,^{8,9} and peak fitting methods.¹⁰

The accuracy of each of these methods at determining the trapping parameters has been examined by numerically solving the rate equations, without any simplifying assumptions, and in this way producing a simulated TSC or TL peak. One then uses the above techniques to

determine, from the simulated curves, the various trapping parameters, which are then compared to the input values. The results of such exercises have, in general, been pessimistic. The most comprehensive examination of this type was undertaken by Kelly *et al.*¹¹ who concluded that the approximations used to derive the simple solutions were valid for only a part of the range of physically reasonable parameters. The exact solutions to the rate equations led to a wider range of peak shapes, positions, and magnitudes than could be described by the simplified solutions. They also noted that the exact solutions could sometimes be adequately described by more than one set of parameters using the simplified solutions. These deliberations led Kelly *et al.*¹¹ to the conclusion that analyses of TL and TSC peaks using simplifying assumptions are unlikely to lead to unambiguous solutions in the absence of extensive additional information. In particular, these authors questioned the validity of the QE approximation. Thus the validity of using TL and/or TSC to arrive at estimates of the trapping parameters is potentially undermined by uncertainty regarding the QE approximation in particular, and the simplified descriptions in general.

The purpose of this paper is to present a more generalized description of TL and TSC phenomena which does not rely on the QE approximation. The analysis to be presented follows from an approach to TL and TSC kinetics introduced by Lewandowski and McKeever in an earlier paper.¹² In this proposal, the QE and KO approximations are replaced by two physically meaningful functions, namely, the $Q(T)$ and $P(T)$ functions, respectively. These functions are related and can be described in terms of the meaningful processes of charge trapping, release, and recombination. The abandonment of the QE approximation yields a generalized equation for TL (and one for TSC) which is capable of describing a wide range of cases. With this new equation we are able to gain greater insight into the validity of the QE approximation than was available hitherto and, furthermore, we are able to make some general statements regarding the validity of the various simplified equations to describe TL and TSC. Finally, the analysis allows us to suggest a simple experimental method for the extraction of the trap depth values from experimental TL and TSC peaks, independent of the QE approximation.

II. THE MODEL

The analysis to be presented must begin with a choice of a model for the production of TL and TSC. Following our earlier paper¹² we employ the model shown in Fig. 1, which is closely related to that described by Dussel and Bube⁵ and by Saunders.⁶ The model consists of a set of shallow electron traps (ST) which are thermally unstable over the "active" temperature range. The active temperature range denoted from T_0 to T_f is defined as that over which charge is thermally released from the active traps (AT). The active traps are characterized by a thermal trap depth $E = E_c - E_a$, a concentration N of traps, of which $n(T)$ are filled with electrons, and a capture cross

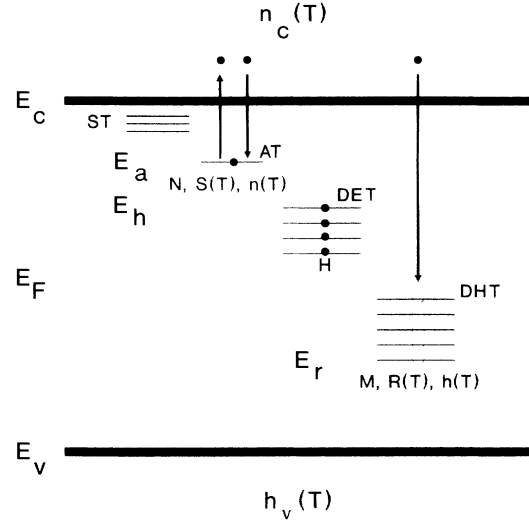


FIG. 1. Energy level diagram of the model under consideration. The model consists of a distribution of shallow levels (ST's), the active level (AT) at energy E_a , a distribution of deep thermally disconnected electron traps (DET's) from energies E_F to E_h , and a distribution of thermally disconnected hole traps (DHT's) from energies E_r to E_F [from Lewandowski and McKeever (Ref. 12)].

section for electrons $S(T)$. In addition, the model contains a concentration H of deep, thermally disconnected electron traps (DET). Thermally disconnected is taken to mean that electrons localized at these traps are thermally stable over the active temperature range. The final category of traps are a set of thermally disconnected hole traps (DHT) existing in concentration M . These are also thermally disconnected in the sense that $E_c - E_a$ is less than $E_r - E_v$ so that little or no simultaneous hole release occurs over the active temperature range. The Fermi energy E_F is located schematically near midgap. H and M are given by,

$$H = \int_{E_F}^{E_h} G(E) dE, \quad (1)$$

and

$$M = \int_{E_r}^{E_F} G(E) dE, \quad (2)$$

where $G(E)$ is the density of states function. The deep hole traps are characterized by an effective capture cross section for electrons $R(T)$ and a concentration of full hole traps $h(T)$. The average recombination cross section, $R(T)$, is given by,

$$R(T) = \frac{1}{M} \sum_i M_i r_i(T), \quad (3)$$

where the sum is over levels with energy less than E_F and greater than E_v . M_i is the concentration of hole traps of defect type i and $r_i(T)$ is the capture cross section for electrons of a hole trap of defect type i . We further

define $n_c(T)$ to be the concentration of free electrons in the conduction band ($E > E_c$) and $h_v(T)$ to be the concentration of free holes in the valence band ($E < E_v$).

We consider the excitation of the system at a low temperature $T < T_0$ by illumination with radiation of energy greater than the band gap. This is continued until equilibrium is achieved, whereupon the excitation is removed and the system is allowed to return to a new equilibrium. The system is then heated at a linear rate $\beta = dT/dt$ in the dark. Initially the trap-emptying process begins as electrons are excited out of the shallow levels. Over the active temperature range (T_0 to T_f), however, these levels no longer represent stable trapping centers so that, as shown in Fig. 1, electrons are excited out of the active level into the conduction band and are removed from the conduction band by retrapping and recombination. The deep electron traps remain full over this temperature range and therefore do not constitute potential trapping sites.

III. THE THEORY

A. The $Q(T)$ and $P(T)$ functions

Following the analysis of Lewandowski and McKeever¹² we write the thermal release probability function per unit time from the active level as,

$$\mathcal{P}(T) = s(T) \exp\left(-\frac{E}{kT}\right), \quad (4)$$

where $s(T)$ is the frequency factor, E is the trap depth, T is the absolute temperature, and k is Boltzmann's constant.

The set of coupled differential equations which describe the traffic of charge between the energy levels during thermal stimulation can now be written, thus,

$$\begin{aligned} \frac{dn_c(T)}{dt} = & -n(T)N_c(T)S(T)v_e(T) \exp\left(-\frac{E}{kT}\right) \\ & + n_c(T)[N - n(T)]S(T)v_e(T), \end{aligned} \quad (5)$$

and

$$\frac{dn_c(T)}{dt} = -\frac{dn(T)}{dt} - \frac{n_c(T)}{\tau(T)}. \quad (6)$$

In these equations, $N_c(T)$ is the effective density of states in the conduction band, given by

$$N_c(T) = 2 \left(\frac{kTm^*}{2\pi\hbar^2} \right)^{3/2}, \quad (7)$$

where \hbar is Planck's constant divided by 2π . The thermal velocity of electrons in the conduction band $v_e(T)$ is given by,

$$v_e(T) = \sqrt{\frac{3kT}{m^*}}, \quad (8)$$

where m^* is the electron effective mass. The recombination lifetime $\tau(T)$ is given by

$$\frac{1}{\tau(T)} = v_e(T)R(T)h(T). \quad (9)$$

With this model the reaction rates for excitation $R_{\text{ex}}(T)$, recapture $R_{\text{recap}}(T)$, and recombination $R_{\text{recom}}(T)$ are thus

$$R_{\text{ex}}(T) = n(T)s(T) \exp\left(-\frac{E}{kT}\right), \quad (10)$$

$$R_{\text{recap}}(T) = n_c(T)[N - n(T)]S(T)v_e(T), \quad (11)$$

and

$$\begin{aligned} R_{\text{recom}}(T) &= n_c(T)/\tau(T) \\ &= n_c(T)v_e(T)R(T)h(T). \end{aligned} \quad (12)$$

It should be noted that these reaction rates are defined to be positive quantities.

The usual procedure to solve the rate equations, Eqs. (5) and (6), is to introduce the quasiequilibrium approximation, namely, that $|dn_c(T)/dt| \ll |n_c(T)/\tau(T)|$. This inequality implies that the density of delocalized electrons remains constant over the temperature range of detrapping. If one has $n_c(T_0) \ll n(T_0)$ at equilibrium after excitation, then the QE assumption would mean that, relative to the trapped charge density, free electrons do not accumulate in the conduction band during the release of trapped charge. While this may be a reasonable assumption for the slow-retrapping case, for other cases such as fast retrapping this assumption may break down altogether. As an alternative to the QE approximation, Lewandowski and McKeever¹² introduced the function $q(T)$ such that,

$$q(T) \left(\frac{n_c(T)}{\tau(T)} \right) = \frac{dn_c(T)}{dt}, \quad (13)$$

for all T . From Eq. (6) this becomes

$$Q(T) \left(\frac{n_c(T)}{\tau(T)} \right) = -\frac{dn(T)}{dt}, \quad (14)$$

where $Q(T) = q(T) + 1$. Note that QE means that $q(T) \approx 0$ or $Q(T) \approx 1$ for all T . The value of this function is then a measure of the degree to which the system remains in quasiequilibrium over the course of trap emptying. At this stage we may also define the kinetic-order function $P(T)$, namely,

$$P(T) = \tau(T)[N - n(T)]S(T)v_e(T). \quad (15)$$

This function is a generalization of the concept of kinetic order in that it expresses the degree by which recombination processes dominate over retrapping, or vice versa. The traditional limiting cases considered such as slow retrapping (first order), retrapping, and fast retrapping are achieved by $P(T)$ taking on the constant values $P(T) \ll 1$, $P(T) = 1$, and $P(T) \gg 1$ respectively. The main advantage in defining the $Q(T)$ and $P(T)$ functions in this way is to separate conceptually the QE and KO approximations and further to allow the system's adher-

ence to these restrictions to vary with temperature. In previous formulations the QE approximation is made before any kinetic-order considerations.

$Q(T)$ and $P(T)$ may also be defined in terms of the reaction rates, thus,

$$Q(T) = \frac{R_{\text{ex}}(T) - R_{\text{recap}}(T)}{R_{\text{recom}}(T)}, \quad (16)$$

and

$$P(T) = \frac{R_{\text{recap}}(T)}{R_{\text{recom}}(T)}. \quad (17)$$

We also note that

$$Q(T) + P(T) = \frac{R_{\text{ex}}(T)}{R_{\text{recom}}(T)}, \quad (18)$$

and

$$\frac{Q(T)}{P(T)} + 1 = \frac{R_{\text{ex}}(T)}{R_{\text{recap}}(T)}. \quad (19)$$

Using the framework of the $Q(T)$ and $P(T)$ functions we can now make some general statements with regard to the traditional methods of analysis for TL and TSC curves in which assumptions are employed involving the relative sizes of the various reaction rates defined in Eqs. (10)–(12). The traditional methods use the QE approximation, namely $Q(T) \approx 1$, along with the slow-retrapping, retrapping, or fast-retrapping approximations.

Considering the traditional fast-retrapping case first, we have for this case $Q(T) \approx 1$ and $P(T) \gg 1$, which implies from Eq. (19) that $R_{\text{ex}}(T) \approx R_{\text{recap}}(T)$ over all T . If this were strictly true, the active trap would not empty. Furthermore, if $R_{\text{ex}}(T) \approx R_{\text{recap}}(T)$, then from Eq. (16), $Q(T) \approx 0$, contrary to the initial premise. If, however, $R_{\text{recom}}(T)$ was correspondingly very small in Eq. (16) then it still may be that $Q(T) \approx 1$.¹³ While this may be true in principle, in practice if R_{recom} were very small, it is unlikely that a TL signal would be observed in the first place and the TSC signal would not form a peak but instead would rise to an equilibrium level forming a step rather than a peak. Therefore, for a TL and/or TSC peak to be observed in the active temperature range, the $P(T) \gg 1$ and $Q(T) \approx 1$ conditions cannot both be true for all T . This discussion leads to the realization that *it is extremely unlikely to have a TL (or TSC) peak which is described by both fast-retrapping and quasiequilibrium*. In this way we see that the reservations of Kelly *et al.*¹¹ are justified for this case.

For the retrapping case we have $Q(T) \approx 1$ and $P(T) = 1$. This means from Eqs. (17)–(19) that $R_{\text{recap}}(T) = R_{\text{recom}}(T)$ and $R_{\text{ex}}(T) \approx 2R_{\text{recom}}(T) \approx 2R_{\text{recap}}(T)$ over all T . Although these conditions may be true over certain temperature ranges, it is unlikely that they will be true in general since they rely on the functions $n_c(T)$, $n(T)$, $S(T)$, $R(T)$, $h(T)$, and $v_e(T)$, which all have different temperature dependencies. Thus, as before, we see that the warnings of Kelly *et al.*¹¹ may have some justification in this case also.

The final set of conditions are those imposed by Randall and Wilkins³ in their original analysis of TL glow peaks, namely $Q(T) \approx 1$ and $P(T) \ll 1$. This requires $R_{\text{recom}}(T) \gg R_{\text{recap}}(T)$ and $R_{\text{ex}}(T) \gg R_{\text{recap}}(T)$, or $n_c(T)R(T)h(T) \gg n_c(T)S(T)[N - n(T)]$ and $n(T)s(T)\exp(-E/kT) \gg n_c(T)S(T)[N - n(T)]$. These constitute a legitimate self-consistent set of inequalities and are significantly less restrictive than the equalities of the previous case. Such relationships may exist over wide temperature ranges. Stated differently, the slow-retrapping conditions best satisfy the requirements of a realizable system. It is not obvious in this case if the reservations of Kelly *et al.*¹¹ continue to be justified. To answer this we must proceed further.

B. The generalized first-order TL and TSC equations

The experimentally observed TSC and TL signals, $I_{\text{TSC}}(T)$ and $I_{\text{TL}}(T)$, are related to the free-carrier concentration $n_c(T)$ by $I_{\text{TSC}}(T) = AF e \mu(T) n_c(T)$ and $I_{\text{TL}}(T) = \eta n_c(T) / \tau(T)$, where e is the modulus of the electronic charge, $\mu(T)$ is the electron mobility, A is the effective electrode area, F is the magnitude of the applied electric field, and η is the luminescence efficiency of the recombination process ($0 < \eta \leq 1$).

From the definitions of the $Q(T)$ and $P(T)$ functions [Eqs. (14) and (15)] and the first rate equation [Eq. (5)], one may arrive at the following expression for $n_c(T)$,¹²

$$n_c(T) = \frac{\tau(T)n(T)s(T)}{Q(T) + P(T)} \exp\left(-\frac{E}{kT}\right). \quad (20)$$

Then substituting this expression for $n_c(T)$ into Eq. (14) produces

$$\frac{1}{n(T)} \frac{dn(T)}{dt} = -s(T) \left(\frac{Q(T)}{Q(T) + P(T)} \right) \exp\left(-\frac{E}{kT}\right). \quad (21)$$

Integration of this last equation using $dT/dt = \beta$ produces

$$\begin{aligned} n(T) &= n_0 \exp\left[-\frac{1}{\beta} \int_{T_0}^T \left(\frac{Q(\Theta)}{Q(\Theta) + P(\Theta)} \right) s(\Theta) \right. \\ &\quad \left. \times \exp\left(-\frac{E}{k\Theta}\right) d\Theta \right] \\ &= n_0 \exp[-g(T)], \end{aligned} \quad (22)$$

where n_0 is the initial electron concentration in the active trap, and

$$g(T) = \frac{1}{\beta} \int_{T_0}^T \left(\frac{Q(\Theta)}{Q(\Theta) + P(\Theta)} \right) s(\Theta) \exp\left(-\frac{E}{k\Theta}\right) d\Theta. \quad (23)$$

With Eqs. (20) and (22), the TL expression becomes

$$\begin{aligned}
I_{\text{TL}}(T) &= \eta \left(\frac{n_c(T)}{\tau(T)} \right) \\
&= \eta n_0 \frac{s(T)}{Q(T) + P(T)} \exp\left(-\frac{E}{kT}\right) \exp[-g(T)].
\end{aligned} \tag{24}$$

Since $I_{\text{TSC}}(T)$ and $n_c(T)$ are related by $AFe\mu(T)$, deriving the TSC equation essentially consists of deriving an expression for $n_c(T)$. Since $n_c(T) = \tau(T)I_{\text{TL}}(T)/\eta$,

$$n_c(T) = \tau(T)n_0 \frac{s(T)}{Q(T) + P(T)} \exp\left(-\frac{E}{kT}\right) \exp[-g(T)]. \tag{25}$$

Unlike the TL solution, the TSC equation requires one to specify the form of $\tau(T)$. This is done by imposing the charge neutrality condition. For temperatures greater than T_0 , this is expressed,

$$n_c(T) + n(T) + H = h(T) + h_v(T). \tag{26}$$

For the model considered in this study, $h_v(T) \approx 0$ over the active temperature range. Therefore, from Eq. (9),

$$\tau(T) = \frac{1}{v_e(T)R(T)[n_c(T) + n(T) + H]}, \tag{27}$$

and Eq. (25) becomes

$$\begin{aligned}
n_c(T)^2 + n_c(T)[n(T) + H] \\
= \frac{s(T)}{v_e(T)R(T)} \frac{n(T)}{Q(T) + P(T)} \exp\left(-\frac{E}{kT}\right).
\end{aligned} \tag{28}$$

The solution to this equation is considered in more detail in the Appendix. In the present derivation it will be assumed that the concentration of trapped electrons in both the active level and the deep electron traps is much greater than the concentration of free electrons at any temperature T within the active temperature range, that is, $n(T) + H \gg n_c(T)$. This is essentially the same approximation used by Lewandowski and McKeever in their earlier analysis.¹² The result of this approximation is that one may ignore the $n_c(T)^2$ term compared to $n_c(T)[n(T) + H]$ in the above equation resulting in

$$\begin{aligned}
n_c(T) &= \frac{s(T)}{v_e(T)R(T)} \frac{1}{Q(T) + P(T)} \\
&\times \left(\frac{n(T)}{n(T) + H} \right) \exp\left(-\frac{E}{kT}\right).
\end{aligned} \tag{29}$$

At this stage one may employ the expansion,

$$\begin{aligned}
\frac{n(T)}{n(T) + H} &= \sum_{j=1}^{\infty} (-1)^{j+1} \left(\frac{n(T)}{H} \right)^j \\
&= \sum_{j=1}^{\infty} (-1)^{j+1} \left(\frac{n_0}{H} \right)^j \exp[-jg(T)],
\end{aligned} \tag{30}$$

provided $H > n(T)$. With $s(T) = N_c(T)S(T)v_e(T)$, the

TSC equation becomes

$$\begin{aligned}
n_c(T) &= \frac{S(T)}{R(T)} \frac{N_c(T)}{Q(T) + P(T)} \exp\left(-\frac{E}{kT}\right) \\
&\times \sum_{j=1}^{\infty} (-1)^{j+1} \left(\frac{n_0}{H} \right)^j \exp[-jg(T)].
\end{aligned} \tag{31}$$

At this point we introduce the first-order, slow-retrapping approximation [$P(T) \ll 1$] realizing that the vast majority of physically observable cases fall into this category. Under the condition $P(T) \ll Q(T)$ where $Q(T)$ is on the order of 1, except at the very end of the peak, the TSC and TL equations become

$$\begin{aligned}
n_c(T) &= \frac{S(T)N_c(T)}{R(T)Q(T)} \exp\left(-\frac{E}{kT}\right) \sum_{j=1}^{\infty} (-1)^{j+1} \left(\frac{n_0}{H} \right)^j \\
&\times \exp\left(-\frac{j}{\beta} \int_{T_0}^T s(\Theta) e^{-E/k\Theta} d\Theta\right),
\end{aligned} \tag{32}$$

and

$$\begin{aligned}
I_{\text{TL}}(T) &= \frac{\eta n_0 s(T)}{Q(T)} \exp\left(-\frac{E}{kT}\right) \\
&\times \exp\left(-\frac{1}{\beta} \int_{T_0}^T s(\Theta) e^{-E/k\Theta} d\Theta\right).
\end{aligned} \tag{33}$$

If the QE approximation is made by setting $Q(T) = 1$, the TL equation takes on the form determined by Randall and Wilkins.³ To obtain the Randall-Wilkins form of the TSC equation one must in addition require that $H \gg n_0$. This requirement is a consequence of inclusion of deep thermally disconnected traps in the present model and their absence in the model proposed by Randall and Wilkins¹² and is also in accordance with the conclusion of "mixed-order" kinetic analysis by Chen *et al.*¹⁴

By comparing Eqs. (32) and (33) with and without the QE approximation, it is clear that,

$$I_{\text{TSC}}^{\text{QE}}(T) = Q(T)I_{\text{TSC}}(T), \tag{34}$$

and

$$I_{\text{TL}}^{\text{QE}}(T) = Q(T)I_{\text{TL}}(T), \tag{35}$$

where $I_{\text{TSC}}^{\text{QE}}(T)$ and $I_{\text{TL}}^{\text{QE}}(T)$ represent the observed TSC and TL profiles under conditions of quasiequilibrium and first order. It should also be noted that the above relationship for TSC is only valid within the $n(T) + H \gg n_c(T)$ approximation, while the TL relationship is perfectly general for first order for this model.

From the above expressions we can see that a first-order TL or TSC peak is described by the approximate solutions $I_{\text{TL}}^{\text{QE}}(T)$ and $I_{\text{TSC}}^{\text{QE}}(T)$, modified by the $Q(T)$ function. From a practical standpoint we are still faced with the problem of knowing what the shape of this function is. As they stand, the above first-order equations are not practical as experimental tools since they cannot be described solely in terms of macroscopically measurable parameters, unlike the original Randall-Wilkins equation. In an attempt to overcome this one may proceed in two different ways.

As a first approach one may recognize that the defini-

tion of $Q(T)$ results in useful relations between $Q(T)$ and the observed TSC and TL spectra. For instance, $Q(T)$ can be represented in terms of TSC thus,

$$Q(T) = \frac{\beta\mu(T)\tau(T)}{I_{\text{TSC}}(T)} \frac{d}{dT} \left(\frac{I_{\text{TSC}}(T)}{\mu(T)} \right) + 1 \approx \frac{\beta\tau(T)}{I_{\text{TSC}}(T)} \frac{dI_{\text{TSC}}(T)}{dT} + 1, \quad (36)$$

where typically the temperature dependence of $\mu(T)$ is weak and can usually be ignored. In terms of TL, $Q(T)$ may be expressed as

$$Q(T) = \frac{\beta}{I_{\text{TL}}(T)} \frac{d[\tau(T)I_{\text{TL}}(T)]}{dT} + 1. \quad (37)$$

Both the above expressions require knowledge of $\tau(T)$; however, this requirement can be removed if one has access to simultaneous TL and TSC since $Q(T)$ can also be written [assuming $\mu(T) = \text{constant}$] as

$$Q(T) = \frac{\eta\beta}{AF e \mu I_{\text{TL}}(T)} \frac{dI_{\text{TSC}}(T)}{dT} + 1. \quad (38)$$

As discussed in a previous paper¹² these expressions establish that the QE approximation is only valid at the temperature of the TSC peak maximum. In principle, the

shape of $Q(T)$ can be determined experimentally from a measurement of either TSC alone [Eq. (36)], or TL alone [Eq. (37)], or from a simultaneous measurement of both TL and TSC [Eq. (38)]. However, in practice this line of attack is fraught with difficulties since, experimentally, the scaling constants are unknown, and the function $\tau(T)$ is often unknown also.

An alternative approach is to use Eqs. (34) and (35); and Eqs. (36) and (37) to produce the relation,

$$I^{\text{QE}}(T) = \beta\tau(T) \frac{dI(T)}{dT} + I(T), \quad (39)$$

where $I(T) = I_{\text{TSC}}(T)$ for TSC and $I(T) = \tau(T)I_{\text{TL}}(T)$ for TL. Equation (39) is simply a first-order, linear, differential equation with the solution

$$I(T) = \frac{1}{\beta} \exp\left(-\frac{1}{\beta} \int \frac{dT}{\tau(T)}\right) \times \int_{T_0}^T \exp\left(\frac{1}{\beta} \int \frac{d\Theta}{\tau(\Theta)}\right) \frac{I^{\text{QE}}(\Theta)}{\tau(\Theta)} d\Theta. \quad (40)$$

The complete relation for TL is obtained by setting $I(T) = \tau(T)I_{\text{TL}}(T)$, $I^{\text{QE}}(T) = \tau(T)I_{\text{TL}}^{\text{QE}}(T)$, and by substituting Eq. (33) with $Q(T)$ set to unity into Eq. (40). This done, the generalized first-order TL equation becomes

$$I_{\text{TL}}(T) = \frac{\eta n_0}{\tau(T)\beta} \exp\left(-\frac{1}{\beta} \int \frac{dT}{\tau(T)}\right) \int_{T_0}^T \exp\left(\frac{1}{\beta} \int \frac{d\Theta}{\tau(\Theta)}\right) s(\Theta) \exp\left(-\frac{E}{k\Theta}\right) \exp\left[-\frac{1}{\beta} \int_{T_0}^{\Theta} s(\Omega) \exp\left(-\frac{E}{k\Omega}\right) d\Omega\right] d\Theta. \quad (41)$$

It should be pointed out that the only approximation used in the derivation of the above equation for TL is that $P(T) \ll Q(T)$ and it is in all other ways a perfectly general solution for this model.

The complete expression for TSC obtained from the above TL equation via $I_{\text{TSC}}(T) = AF e \mu \tau(T) I_{\text{TL}}(T) / \eta$ is

$$I_{\text{TSC}}(T) = \frac{AF e \mu n_0}{\beta} \exp\left(-\frac{1}{\beta} \int \frac{dT}{\tau(T)}\right) \int_{T_0}^T \exp\left(\frac{1}{\beta} \int \frac{d\Theta}{\tau(\Theta)}\right) s(\Theta) \exp\left(-\frac{E}{k\Theta}\right) \times \exp\left[-\frac{1}{\beta} \int_{T_0}^{\Theta} s(\Omega) \exp\left(-\frac{E}{k\Omega}\right) d\Omega\right] d\Theta. \quad (42)$$

The approximations involved in the above TSC solution are: $n(T) + H \gg n_c(T)$, $\mu(T) = \text{constant}$, and, $P(T) \ll Q(T)$.

Although Eqs. (41) and (42) are more complex than their quasiequilibrium counterparts, they represent more accurately the true shapes of the TL and TSC curves than do the simple Randall-Wilkins forms. It should be noted that the above expressions still contain the unknown term $\tau(T)$. However, $\tau(T)$ can either be considered a constant [where its temperature dependence is ignored compared with the $\exp(-E/kT)$ terms] or it may simply be parametrized thus, $\tau(T) = C_\tau T^\alpha$, where C_τ and α are constants. If either of these simplifications are introduced we have achieved the desired aim of producing full expressions for TL and TSC written entirely in terms of macroscopic parameters only.

IV. CALCULATIONS

A. Equation parametrization

We now consider curve shapes obtained from the Randall-Wilkins (RW) equation, the generalized first-order equations derived in the previous section, and those obtained by numerically solving the rate equations directly using an adaptive fourth-order, Runge-Kutta-Fehlberg (RKF) routine. Our goal is to determine if the generalized equations give superior descriptions compared to the Randall-Wilkins equations and to determine under what conditions the Randall-Wilkins equations may be safely applied. Before this is done, however, some comments regarding the parametrization of $R(T)$ and $S(T)$ and the relationship between TL and TSC are

in order.

Lewandowski and McKeever¹² show that the relationship between TL and TSC is given by the following expression,

$$I_{\text{TSC}}(T) = I_{\text{TL}}(T) \frac{AFe}{\eta H} \frac{\mu(T)}{R(T)v_e(T)} \times \sum_{j=0}^{\infty} \left(-\frac{n_0}{H}\right)^j \exp[-jg(T)]. \quad (43)$$

From this expression it was observed that if $H \gg n_0$ (or N) then one may approximate the series by the first term, and also, if $\mu(T)/[R(T)v_e(T)] \approx \text{constant}$, then the TL and TSC have the same shape and peak temperature. For many of the curve shapes presented in this section these conditions will indeed hold. In the curves that follow, we have assumed that the following set of conditions hold true:

(1) $\eta = 1$. This, in fact, is a consequence of the model we have chosen in Fig. 1 wherein all recombination events are radiative.

(2) $\mu(T) = \mu = \text{constant}$. This allows us to monitor $n_c(T)$ instead of $I_{\text{TSC}}(T)$ since, under this assumption, they differ by a constant.

(3) In order to use the expressions derived in the previous section some knowledge regarding the functional form of the unknown functions $R(T)$, $S(T)$, $\tau(T)$, or $s(T)$ must be inferred or assumed. Because of the relationships between these functions, it is only necessary to assume a functional form for any two. In this paper we will assume that $R(T)$ and $S(T)$ may be approximated by power-law temperature dependencies of the form DT^{-d} and

CT^{-b} respectively. These assumptions regarding $R(T)$ and $S(T)$ are necessary to begin the parametrization of the generalized first-order TL and TSC equations. As noted earlier, $v_e(T) = (3kT/m^*)^{1/2}$, and while different values of d were used for different calculations, typically we used $d = 1.5$. Thus $R(T)v_e(T)$ will have a weak temperature dependency compared with the exponential dependencies of the trapped charge concentrations.

(4) No *a priori* assumptions are made concerning $n_c(T)$ or $n(T)$. These functions take on values dictated by the numerical solutions to the rate equations. Often, however, it is found that $n_c(T)$ or $n(T)$ or both can be considered to be very much less than H . When this happens, this will be noted. However, we should take note that when this *does* occur, and when taken together with points (1)–(3) above, we see from Eq. (43) that the TL and TSC peaks will indeed have maxima at approximately the same temperature.

As previously noted, we assume:

$$R(T) = DT^{-d}, \quad \text{and} \quad S(T) = CT^{-b}. \quad (44)$$

With $v_e = (3kT/m^*)^{1/2}$ and $N_c(T) = 2[kTm^*/(2\pi\hbar^2)]^{3/2}$, we have

$$s(T) = N_c(T)S(T)v_e(T) = BT^{2-b}, \quad (45)$$

and using Eq. (27) with $H \gg n_c(T)$ and n_0 , the recombination lifetime could be parametrized thus,

$$\frac{1}{\tau(T)} \approx v_e(T)R(T)H = \Lambda T^{1/2-d}. \quad (46)$$

These choices produce the following parametrized first-order equations. For $d \neq 3/2$,

$$\begin{pmatrix} I_{\text{TL}}(T) \\ I_{\text{TSC}}(T) \end{pmatrix} = \begin{pmatrix} K_{\text{TL}}T^{1/2-d} \\ K_{\text{TSC}} \end{pmatrix} \exp\left(-\frac{\Lambda T^{3/2-d}}{\beta(3/2-d)}\right) \int_{T_0}^T \exp\left(\frac{\Lambda \Theta^{3/2-d}}{\beta(3/2-d)}\right) \Theta^{2-b} \exp\left(-\frac{E}{k\Theta}\right) \times \exp\left[-\frac{B}{\beta} \int_{T_0}^{\Theta} \Omega^{2-b} \exp\left(-\frac{E}{k\Omega}\right) d\Omega\right] d\Theta. \quad (47)$$

For the case where $d = 3/2$ the equations are

$$\begin{pmatrix} I_{\text{TL}}(T) \\ I_{\text{TSC}}(T) \end{pmatrix} = \begin{pmatrix} K_{\text{TL}}T^{-1} \\ K_{\text{TSC}} \end{pmatrix} T^{\Lambda/\beta} \int_{T_0}^T \Theta^{2-b+(\Lambda/\beta)} \exp\left(-\frac{E}{k\Theta}\right) \times \exp\left[-\frac{B}{\beta} \int_{T_0}^{\Theta} \Omega^{2-b} \exp\left(-\frac{E}{k\Omega}\right) d\Omega\right] d\Theta. \quad (48)$$

In these equations $K_{\text{TL}} = \eta n_0 B \Lambda / \beta$ and $K_{\text{TSC}} = AF e \mu n_0 B / \beta$. The necessity of presenting two forms of the parametrized equations, one for $d \neq 3/2$ and one $d = 3/2$, follows from our choice of parametrization and does not represent a fundamental requirement of the theory. There are six unknown parameters, K_{TL} (K_{TSC}), Λ , d , b , E , and B . With regard to fitting, it would be necessary for any algorithm to use Eq. (47) and then switch to Eq. (48) if d becomes equal to $3/2$. As they stand, Eqs. (47) and (48) require the following approximations: $H \gg n_c(T)$ and n_0 , $P(T) \ll Q(T)$ (first order),

and $\mu(T) = \text{constant}$ for TSC. In principle, one need not make the $H \gg n_0$ approximation. Instead of Eq. (46), one could construct $1/\tau(T)$ as $v_e(T)R(T)[n(T)+H]$ using only the $n(T)+H \gg n_c(T)$ approximation and insert Eq. (22) for $n(T)$. This, however, leads to more complicated equations with a larger number of unknown parameters.

It should also be noted that, given the above parametrizations, we can write

$$\frac{I_{\text{TSC}}(T)}{I_{\text{TL}}(T)} = \frac{K_{\text{TSC}}}{K_{\text{TL}}} T^{d-1/2}, \quad (49)$$

where we observe that if $d = 1/2$, the TL and TSC peaks will have the same shape and peak temperature. This is nothing more than the parametrized version of the statement $R(T)v_e(T) = \text{constant}$.

The rate equations (5) and (6) were parametrized in the same way as Eqs. (47) and (48). To allow for a direct comparison with the results obtained from the Randall-Wilkins solution, it too must be parametrized in the same fashion. The RW equation is given by³

$$n_c(T) = n_0 \tau s(T) \exp\left(-\frac{E}{kT}\right) \times \exp\left[-\frac{1}{\beta} \int_{T_0}^T s(\Theta) \exp\left(-\frac{E}{k\Theta}\right) d\Theta\right], \quad (50)$$

where τ is taken to be constant. Parametrizing the RW equation according to the above prescription and making use of an approximation by Keating¹⁵ produces the parametrized RW fitting equation,

$$n_c(T) = n_0 \tau B T^{2-b} \exp\left(-\frac{E}{kT}\right) \exp\left[\frac{-BkT^{4-b}}{\beta E}\right] \times \left(1 + \frac{(b-4)kT}{E}\right) \exp\left(-\frac{E}{kT}\right), \quad (51)$$

to be used in fitting to the RKF solutions.

B. Curve shapes

Table I summarizes the range of parameters used for the calculations. In addition to the N , H , C , and D parameters, Table I also shows the temperatures of the TSC and TL peak maxima, T_{mc} and T_{ml} respectively. All

calculations were performed with b and d set to $3/2$, with $\beta = 4.0$ K/min, $E = 0.300$ eV, and $n_0 = N$.

From Eq. (49), $d = 3/2$ and therefore $I_{TSC}(T) = \text{const} \times T I_{TL}(T)$. This means that, for these parameters, $T_{ml} < T_{mc}$. We see from Table I this is indeed the case with T_{ml} less than T_{mc} by ~ 0.1 K except for cases 15 and 18 where they differ by 0.226 K and 0.965 K respectively. This arises from the fact that, for case 15, H and N differ by only an order of magnitude while for case 18 they are the same. Since Eqs. (47) and (48) assume that $H \gg n_0$, the prediction from Eq. (49) will not be perfectly valid for these cases.

1. Fitting the RW equation to the RKF solutions

After generating data using the RKF analysis, the RW expression from Eq. (51) was fitted to the resulting TSC and TL curves in order to determine parameter values. In the following discussion a sample of these data sets, which emphasize the more significant trends observed, is examined in greater detail.

The graphs in Fig. 2 show six TSC data sets generated using RKF. These data sets (a)–(f) correspond, respectively, to cases 1, 3, 4, 14, 17, and 15 from Table I. The parametrized RW expression was fitted to these data sets with the results presented in Table II. Note that the leading scale factor is not listed and the parameter C is tabulated instead of B since C is more directly related to the value of the capture cross section. From this table we see that the results of fitting to TSC (actually n_c) yielded a range of activation energies from 0.291 to 0.318 eV. The “poorest” fits (largest value for χ^2) were

TABLE I. Summary of the system parameters used for the calculation of the TSC and TL curves. Power-law temperature dependencies of the form $R(T) = DT^{-d}$; $d = 3/2$ and $S(T) = CT^{-b}$; $b = 3/2$ were assumed for the recombination and capture cross sections. T_{mc} and T_{ml} are the temperatures of the TSC and TL peak maxima respectively. For all calculations, the activation energy was 0.300 eV, the heating rate was 4.0 K/min, and $N = n_0$.

Case	N (m^{-3})	H (m^{-3})	C ($\text{K}^b \text{m}^2$)	D ($\text{K}^d \text{m}^2$)	T_{mc} (K)	T_{ml} (K)
1	10^9	10^{12}	1.6×10^{-17}	10^{-15}	131.285	131.114
2	10^9	10^{12}	1.6×10^{-16}	10^{-15}	121.645	121.507
3	10^9	10^{12}	1.6×10^{-15}	10^{-15}	113.279	113.165
4	10^9	10^{12}	1.6×10^{-14}	10^{-15}	105.959	105.864
5	10^9	10^{12}	1.6×10^{-17}	10^{-14}	130.312	130.149
6	10^9	10^{12}	1.6×10^{-17}	10^{-13}	130.197	130.035
7	10^9	10^{12}	1.6×10^{-17}	10^{-12}	130.185	130.024
8	10^9	10^{12}	1.6×10^{-16}	10^{-14}	120.756	120.625
9	10^9	10^{12}	1.6×10^{-15}	10^{-13}	112.363	112.258
10	10^9	10^{12}	1.6×10^{-14}	10^{-12}	105.100	105.011
11	10^9	10^{12}	1.6×10^{-15}	10^{-13}	112.364	112.258
12	10^{10}	10^{12}	1.6×10^{-15}	10^{-13}	112.375	112.258
13	10^9	10^{13}	1.6×10^{-15}	10^{-13}	112.353	112.248
14	10^{10}	10^{13}	1.6×10^{-15}	10^{-13}	112.354	112.248
15	10^{11}	10^{12}	1.6×10^{-15}	10^{-13}	112.484	112.258
16	10^9	10^{14}	1.6×10^{-15}	10^{-13}	112.352	112.246
17	10^{11}	10^{14}	1.6×10^{-15}	10^{-13}	112.353	112.246
18	10^{12}	10^{12}	1.6×10^{-15}	10^{-13}	113.219	112.254
19	10^9	10^{15}	1.6×10^{-15}	10^{-13}	112.352	112.246
20	10^{12}	10^{15}	1.6×10^{-15}	10^{-13}	112.352	112.246

obtained for data sets (a), (b), and (c), in which both N and H are small (10^9 and 10^{12} m^{-3} respectively). When N and H were increased (10^{10} or 10^{11} m^{-3} and 10^{12} , 10^{13} , or 10^{14} m^{-3}) as in sets (d), (e), and (f), the fits were markedly improved (see Table II). For set (f), the

values of N and H are large but the condition $H \gg N$ is only weakly satisfied since $H/N = 10$. As a result, set (f) is not as good a fit as sets (d) and (e).

When fitted to RKF TL data, a similar trend is observed (see Fig. 3). In most cases, however, the param-

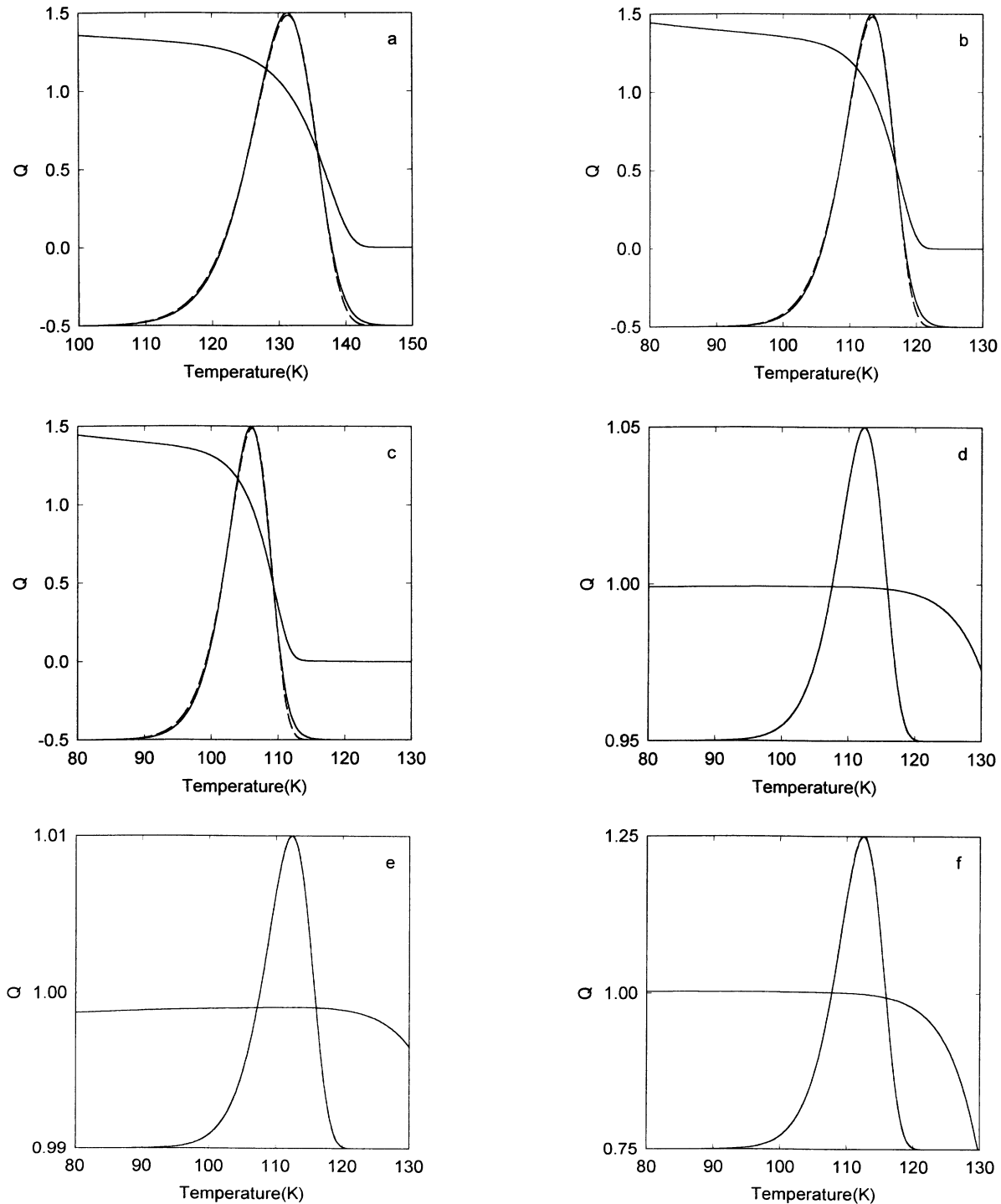


FIG. 2. TSC curves generated using the Runge-Kutta-Fehlberg method. The solid lines represent the RKF data; the dashed lines represent the best fit to the RW expression. Solid lines are also used to represent the $Q(T)$ function. The RKF data are normalized and thus no scale is shown; the scale shown is for the $Q(T)$ function. The parameter values used in each data set are given in Table I: set (a) case 1; (b) case 3; (c) case 4; (d) case 14; (e) case 17; (f) case 15. See Table II for fitted parameters.

TABLE II. Parameters obtained from fitting the Randall-Wilkins TSC and TL equations to Runge-Kutta-Fehlberg data.

Set(Case) ^a	Source	χ^2	C ($K^b m^2$)	E (eV)	b
(a)(1)	actual		1.60×10^{-17}	0.300	1.50
	fit-(TSC)	0.0156	6.31×10^{-16}	0.305	2.22
	fit-(TL)	0.0120	3.21×10^{-16}	0.300	2.18
(b)(3)	actual		1.60×10^{-15}	0.300	1.50
	fit-(TSC)	0.0193	6.14×10^{-14}	0.302	2.31
	fit-(TL)	0.0156	3.46×10^{-14}	0.297	2.29
(c)(4)	actual		1.60×10^{-14}	0.300	1.50
	fit-(TSC)	0.0224	6.92×10^{-13}	0.299	2.41
	fit-(TL)	0.0187	4.00×10^{-13}	0.295	2.39
(d)(14)	actual		1.60×10^{-15}	0.300	1.50
	fit-(TSC)	8.85×10^{-5}	4.78×10^{-14}	0.310	2.00
	fit-(TL)	8.33×10^{-9}	9.76×10^{-16}	0.299	1.41
(e)(17)	actual		1.60×10^{-15}	0.300	1.50
	fit-(TSC)	8.85×10^{-5}	4.78×10^{-14}	0.310	2.00
	fit-(TL)	1.24×10^{-5}	1.40×10^{-16}	0.300	1.47
(f)(15)	actual		1.60×10^{-15}	0.300	1.50
	fit-(TSC)	2.24×10^{-5}	5.41×10^{-17}	0.302	0.74
	fit-(TL)	0.0080	2.53×10^{-14}	0.291	2.25

^aParameter case from Table I.

eter values obtained from TL fitting are closer to the actual values than those obtained from TSC fitting. In the case of sets (d) and (e), the parameters obtained from TL fitting are nearly perfect. Also shown in the graphs of Figs. 2 and 3 are the $Q(T)$ functions which were evaluated from the RKF data.

We notice, from Figs. 2 and 3, that in most cases the $Q(T)$ function is close to unity (i.e., the system is near QE) for temperatures below the peak maximum. Beyond the peak maximum; however, a sharp decrease in the $Q(T)$ function is seen. Note that in sets (a), (b), and (c) the quality of the fit is poorest on either side of the peak maximum; this is a direct result of deviations from quasiequilibrium which render the Randall-Wilkins equation invalid. The departure from QE is not as severe in sets (d), (e), and (f); thus the validity of the Randall-Wilkins equation is better in these cases.

Table II shows fitting values obtained for the data shown in Figs. 2 and 3. The values given in Table II reveal, as mentioned earlier, that in most cases fitting to TL data yields parameter values which are closer to the actual values. The primary reason for this improvement is that the Randall-Wilkins equations assumes that τ is a constant. Since there is only one recombination pathway, $\tau = \tau_r$, thus $\tau/\tau_r = 1$ and the RW expression for TL [i.e., Eq. (51) divided by τ] is completely independent of the temperature dependence of τ . The RW expression for TSC [Eq. (51)], on the other hand, is directly proportional to τ , thus any temperature dependence in τ will directly affect the quality of the fit. As mentioned in other reports,^{16,17} large concentrations of thermally disconnected traps reduce the temperature dependence of τ , thus increasing the validity of the Randall-Wilkins expression for TSC. In most cases, however, one expects better results when fitting to TL rather than TSC.

2. Comparison of the generalized expressions to the RKF results

In order to compare the results discussed above to the generalized expression it was necessary to solve numerically the integrals in Eqs. (47) and (48). The inner integral was evaluated by using Keating's approximation.¹⁵ For the outer integral it was necessary to use an adaptive Simpson's rule algorithm.¹⁸ In order to save computation time when calculating the value at some temperature T_i , the integral was only evaluated from T_{i-1} to T_i and added to the previously saved value of the integral from T_0 to T_{i-1} .

Since computation times from several minutes to a few hours are common when calculating the generalized expression using a 486/33 MHz personal computer, it was not practical to use the generalized expression as a fitting equation. In order to perform comparisons, both the RW expression and the generalized expression were calculated using exactly the same parameter values which were used in the RKF calculations. Figure 4 shows overlay plots of TSC evaluated by each of the three methods. The RKF plots are the same as those shown in Fig. 2. Notice that the RW curves for data sets (a), (b), (c), and (f) do not match the RKF curves whereas for sets (d) and (e) they do match quite well (compare to the fitting results of Fig. 2). The curves for the generalized solution, however, are in good agreement with every one of the RKF curves except for set (f) in which the requirement $H \gg N$ is not well satisfied. It was found that the generalized solution was in excellent agreement with the RKF solution for all parameter ranges tested, provided that the stated assumptions were not violated.

Similar observations to those stated above were made for the TL curves shown in Fig. 5. For sets (a) through

(e), the results are much the same as for TSC. For set (f), however, a significant improvement in the agreement between the RKF data and the generalized solution is observed. Once again, this is attributed to the fact that TL does not depend on the temperature dependence of τ .

Having confidence in the integrity of the generalized solution, the validity of the RW expression may be investigated over a wider range of parameter space. By comparing the RW expression to the solutions of the generalized expression the lengthy computation time of RKF is avoided. The testing involved calculating TSC curves

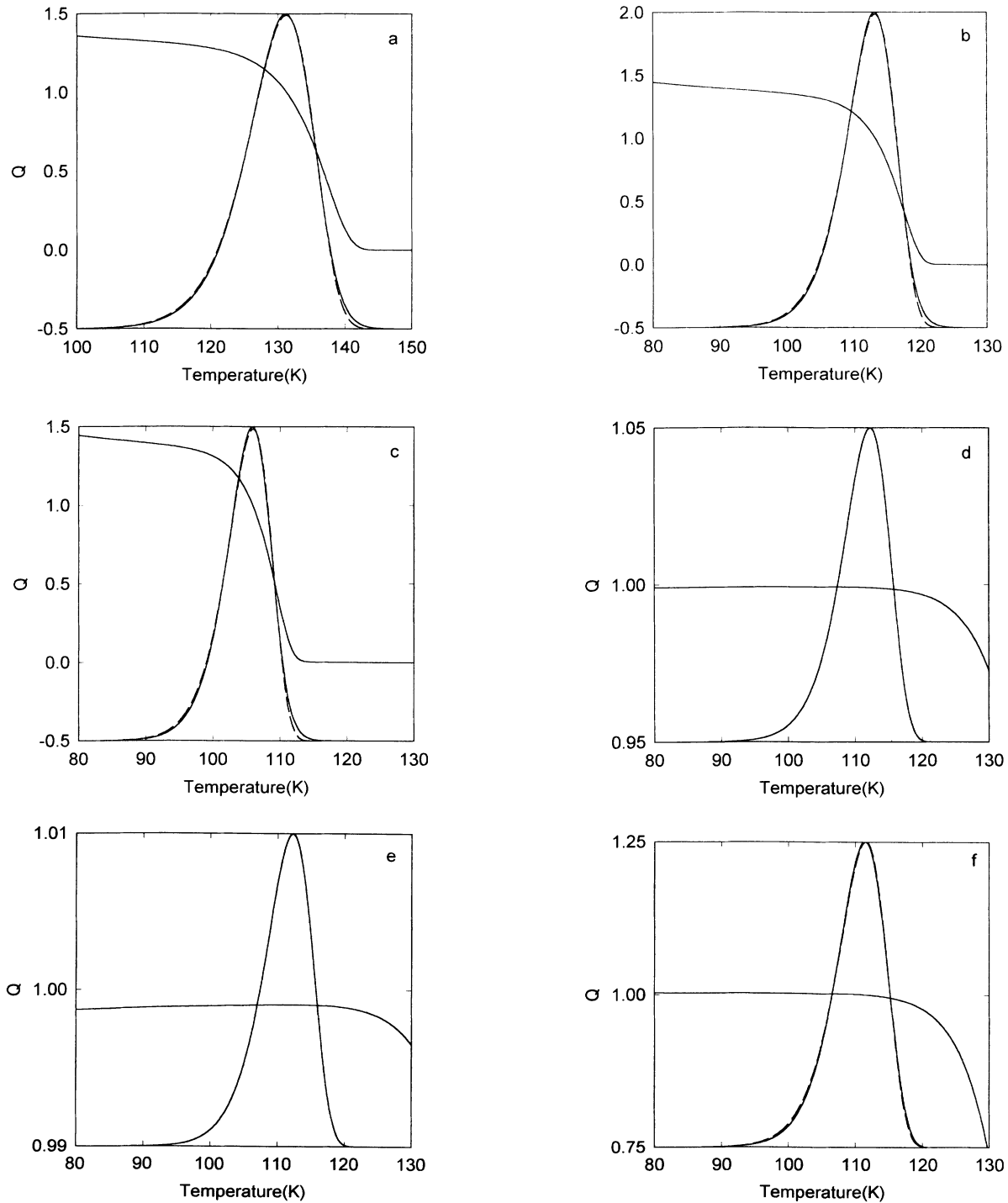


FIG. 3. TL curves generated using the Runge-Kutta-Fehlberg method. The solid lines represent the RKF data; the dashed lines represent the best fits to the RW expression. Solid lines are also used to represent the $Q(T)$ function. The RKF data are normalized and thus no scale is shown; the scale shown is for the $Q(T)$ function. The parameter values used in each data set are given in Table I: set (a) case 1; (b) case 3; (c) case 4; (d) case 14; (e) case 17; (f) case 15. See Table II for fitted parameters.

using both the RW and the generalized expressions over a wide range of parameters. After normalizing each data set the χ^2 value between the two curves was calculated and a three-dimensional (3D) plot was constructed in order to easily determine ranges of “good” and “poor”

agreement. The 3D plots are shown in Figs. 6 and 7. In each of these figures we observe parameter ranges for which χ^2 is independent of parameter changes and regions in which χ^2 changes rapidly. Notice that the χ^2 value corresponding to Fig. 2(a) (case 1) is indicated on

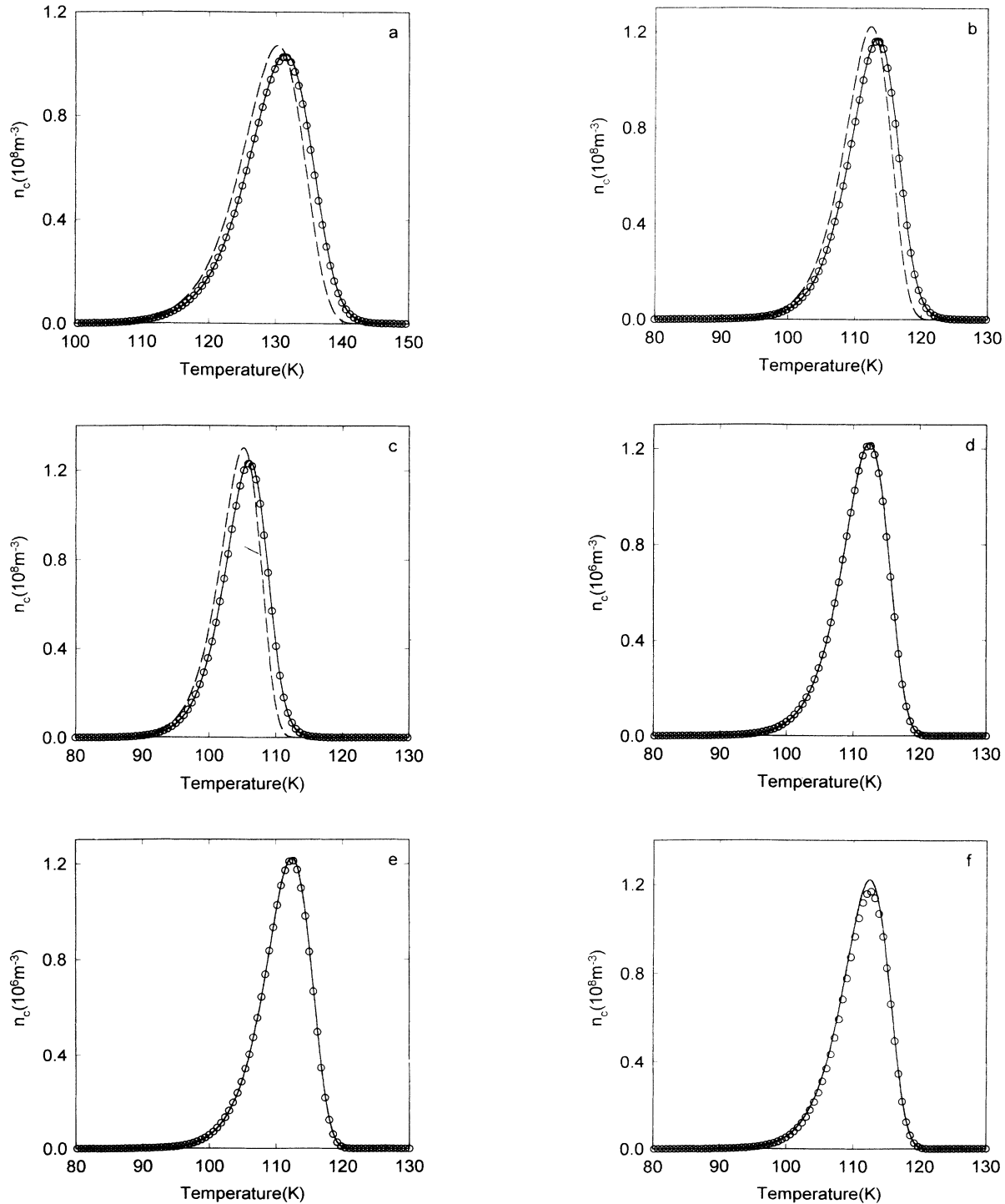


FIG. 4. Comparison of the TSC curves generated using the Runge-Kutta-Fehlberg method with those generated using the generalized [Eq. (48)] and Randall Wilkins [Eq. (51)] expressions. Open circles represent RKF data (every third data point is shown); solid and dashed lines represent the generalized and the RW expressions, respectively. The parameter values used in each data set are given in Table I: set (a) case 1; (b) case 3; (c) case 4; (d) case 14; (e) case 17; (f) case 15.

the surface plot in Fig. 6. This indicator is intended to provide a reference point so that the reader might visualize the quality of fit based on the value of χ^2 .

The inference from the above data is that, while the generalized equations for TL and TSC, namely Eqs. (47)

and (48), are accurate descriptions of the TL and TSC process for all first-order cases, the RW equation only serves as an accurate description for some parameter ranges. In this sense the reservation of Kelly *et al.*¹¹ may have some validity, but clearly the description of TL and

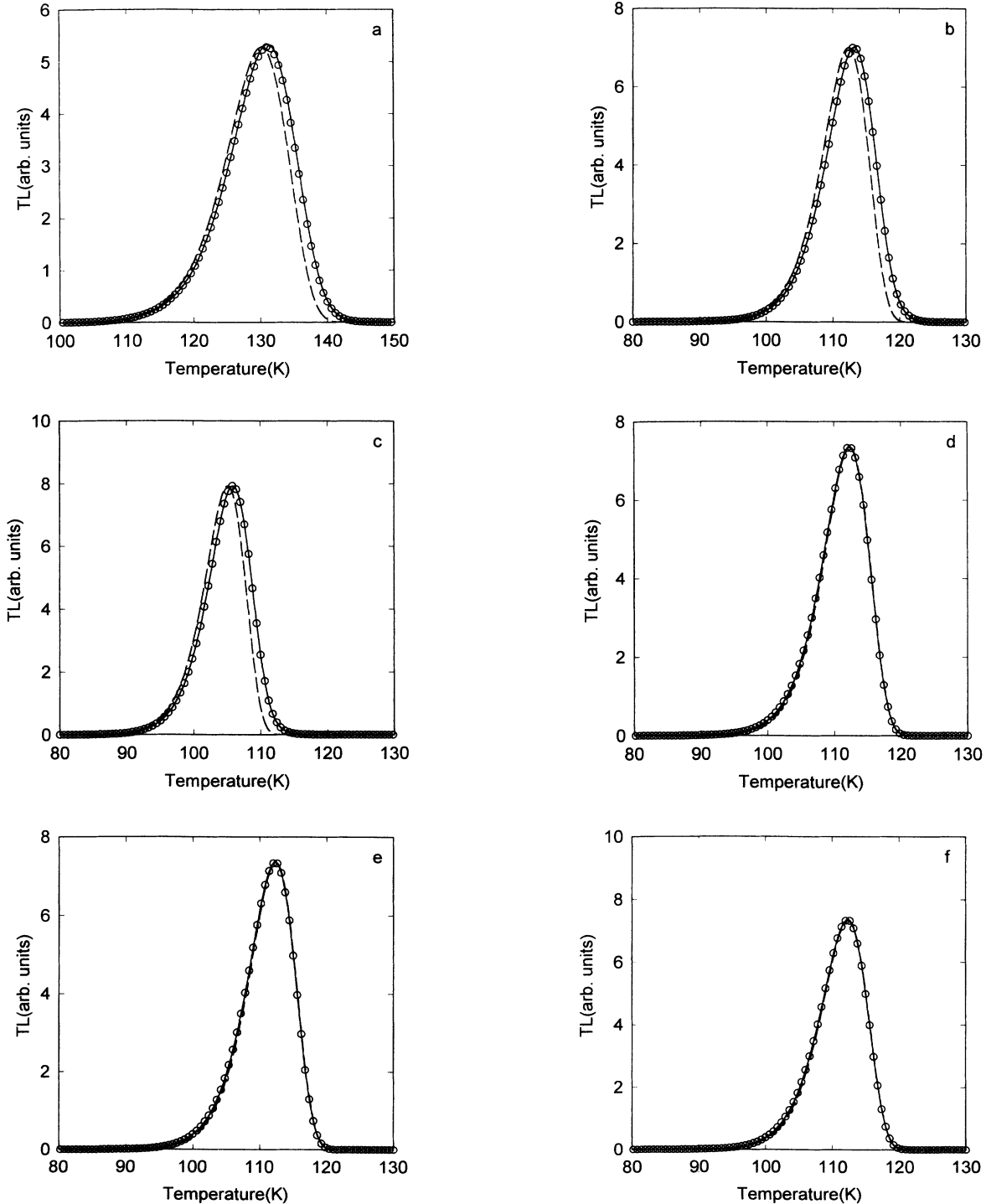


FIG. 5. Comparison of TL curves generated using the Runge-Kutta-Fehlberg method with those generated by the generalized and Randall-Wilkins expressions. Open circles represent RKF data (every third data point is shown); solid and dashed lines represent the generalized and RW expressions, respectively. The parameter values used in each data set are given in Table I: set (a) case 1; (b) case 3; (c) case 4; (d) case 14; (e) case 17; (f) case 15.

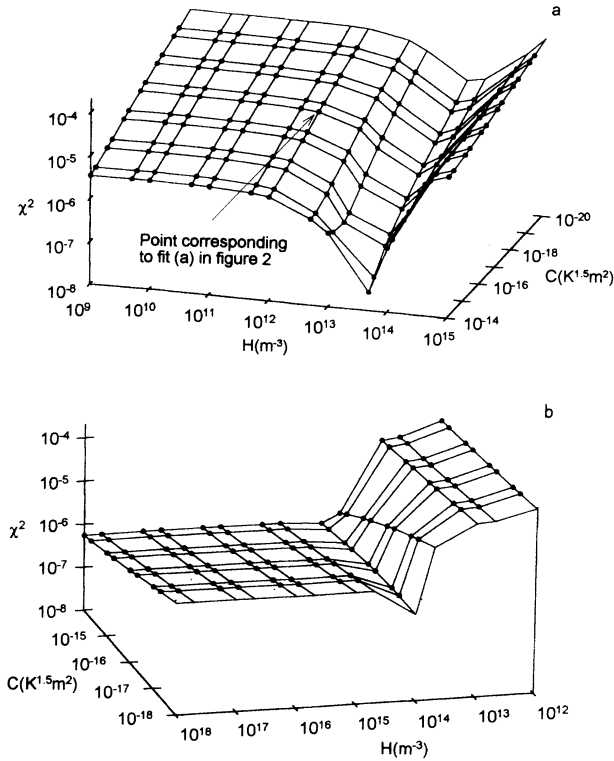


FIG. 6. Variation in χ^2 (from the difference between the Randall-Wilkins and generalized expressions) as a function of C and H . Parameter values: $E = 0.300$ eV and $\beta = 4.0$ K/min. Additional parameters: (a) $b = d = 3/2$, $D = 10^{-15}$ $\text{K}^{3/2}\text{m}^2$; (b) $b = d = 2$, $D = 10^{-13}$ K^2m^2 .

TSC using the RW equation for first-order processes is much more hopeful than for the equivalent non-first-order cases.

V. A SIMPLE METHOD TO DETERMINE THE ACTIVATION ENERGY

In the previous sections we have found that we were able to derive more general first-order expressions which

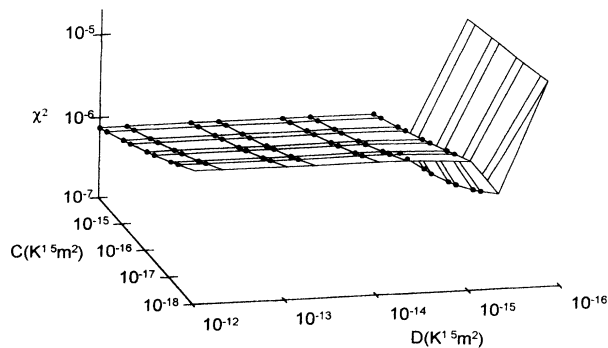


FIG. 7. Variation in χ^2 (from the difference between the Randall-Wilkins and generalized expressions) as a function of C and D . Parameter values: $E = 0.300$ eV, $b = d = 3/2$, $H = 10^{15}$ m^{-3} , and $\beta = 4.0$ K/min.

describe the expected TL and TSC profiles from the assumed model of Fig. 1 [Eqs. (41) and (42)]. After parametrizing the general first-order equations we arrived at expressions containing up to six parameters [Eqs. (47) and (48)]. While these expressions may have the benefit of being more general, in a typical experimental setting where one might wish to engage in peak fitting, six parameters is usually considered too many. The goal then is to arrive at one or more of the parameters through an independent technique and thereby reduce the order of the fit. The approach taken in devising such a method is to exploit the knowledge that for a TSC peak $Q(T) = 1$ at the peak maximum [i.e., $Q(T_{mc}) = 1$]. Since for first-order kinetics $I_{\text{TSC}}^{\text{QE}}(T) = Q(T)I_{\text{TSC}}(T)$, we find that at the TSC peak maximum temperature the system satisfies the QE approximation and therefore the QE solution and the actual solution are identical.

In formulating this approach, we will start with the first-order TSC equation from Eq. (32) and then produce the QE form by requiring that $Q(T) = 1$ and $H \gg n_0$, to yield

$$I_{\text{TSC}}^{\text{QE}}(T) = K s(T) \exp\left(-\frac{E}{kT}\right) \times \exp\left[-\frac{1}{\beta} \int_{T_0}^T s(\Theta) \exp\left(-\frac{E}{k\Theta}\right) d\Theta\right], \quad (52)$$

where $K = AF e \mu \tau n_0$ since for $H \gg n_0$, $1/\tau(T) \approx v_e(T)R(T)H$. Next, the Randall-Wilkins fitting equation, $I_{\text{TSC}}^{\text{RW}}(T)$, is obtained by requiring that $s(T) = \text{constant}$, thus

$$I_{\text{TSC}}^{\text{RW}}(T) = K s \exp\left(-\frac{E}{kT}\right) \times \exp\left[-\frac{1}{\beta} \int_{T_0}^T s \exp\left(-\frac{E}{k\Theta}\right) d\Theta\right], \quad (53)$$

where $I_{\text{TSC}}^{\text{RW}}$ is used instead of $I_{\text{TSC}}^{\text{QE}}$ to denote that in the RW development the frequency factor is assumed constant in addition to making the QE approximation. Now, while the $H \gg n_0$ approximation is very likely to be satisfied in systems exhibiting first-order behavior and therefore the relation $I_{\text{TSC}}^{\text{QE}}(T) = Q(T)I_{\text{TSC}}(T)$ will most certainly hold [where $I_{\text{TSC}}^{\text{QE}}(T)$ is Eq. (52) above], the approximations needed to produce $I_{\text{TSC}}^{\text{RW}}(T)$ [namely that $s(T)$ is constant] may not hold for many cases even when the peak is first order. Therefore, for a great many systems it may be that $I_{\text{TSC}}^{\text{RW}}(T) \neq Q(T)I_{\text{TSC}}(T)$. This is an important detail in regard to this analysis of which more will be said shortly.

Now assume that one has available an experimental TSC curve, $I_{\text{TSC}}(T)$, and say that the parameters, K, s, E in the above RW equation are adjusted to take on the values K', s', E' so that a very good fit is achieved with the data. To a very high accuracy we may then write

$$I_{\text{TSC}}(T) = I_{\text{TSC}}^{\text{RW}}(K', s', E'; T); \quad (54)$$

then it follows that

$$\frac{dI_{\text{TSC}}(T)}{dT} = \frac{dI_{\text{TSC}}^{\text{RW}}(K', s', E'; T)}{dT} \quad (55)$$

$$= -\frac{s'}{\beta} \exp\left(-\frac{E'}{kT}\right) I_{\text{TSC}}^{\text{RW}}(K', s', E'; T) + I_{\text{TSC}}^{\text{RW}}(K', s', E'; T) \frac{E'}{kT^2}. \quad (56)$$

However, since $I_{\text{TSC}}(T) = I_{\text{TSC}}^{\text{RW}}(K', s', E'; T)$, we have

$$\frac{dI_{\text{TSC}}(T)}{dT} = -\frac{s'}{\beta} \exp\left(-\frac{E'}{kT}\right) I_{\text{TSC}}(T) + I_{\text{TSC}}(T) \frac{E'}{kT^2}, \quad (57)$$

which may be written as

$$\frac{E'}{kT^2} - \frac{s'}{\beta} \exp\left(-\frac{E'}{kT}\right) = \frac{1}{I_{\text{TSC}}(T)} \frac{dI_{\text{TSC}}(T)}{dT}. \quad (58)$$

For $T = T_{\text{mc}}$ we may solve for s' as

$$s' = \exp\left(\frac{E'}{kT_{\text{mc}}}\right) \frac{\beta E'}{kT_{\text{mc}}^2}, \quad (59)$$

from which we observe that s' is not an independent variable. Substituting s' into Eq. (58) we find

$$\begin{aligned} I_{\text{TSC}}^{\text{QE}}(T_{\text{mc}}) &= K s(T_{\text{mc}}) \exp\left(-\frac{E(T_{\text{mc}})}{kT_{\text{mc}}}\right) \exp\left[-\frac{1}{\beta} \int_{T_0}^{T_{\text{mc}}} s(\Theta) \exp\left(-\frac{E(\Theta)}{k\Theta}\right) d\Theta\right] \\ &= K s' \exp\left(-\frac{E'(T_{\text{mc}})}{kT_{\text{mc}}}\right) \exp\left[-\frac{1}{\beta} \int_{T_0}^{T_{\text{mc}}} s' \exp\left(-\frac{E'(\Theta)}{k\Theta}\right) d\Theta\right]. \end{aligned} \quad (61)$$

In this expression, $E(T_{\text{mc}}) = E_{\text{corr}}$ since $Q(T_{\text{mc}}) = 1$, $E'(T_{\text{mc}})$ is obtained from Eq. (60) above, and s' is the RW constant-frequency factor given by Eq. (59). Equation (61) is satisfied if

$$s(T_{\text{mc}}) \exp\left(-\frac{E_{\text{corr}}}{kT_{\text{mc}}}\right) = s' \exp\left(-\frac{E'(T_{\text{mc}})}{kT_{\text{mc}}}\right). \quad (62)$$

With s' given by Eq. (59) we find that the solution for E_{corr} is,

$$E_{\text{corr}} = kT_{\text{mc}} \ln\left(\frac{s(T_{\text{mc}})kT_{\text{mc}}^2}{\beta E'(T_{\text{mc}})}\right). \quad (63)$$

Indeed, if $s(T)$ is a constant and therefore is equal to s' which is given by Eq. (59), then Eq. (62) shows that $E_{\text{corr}} = E'(T_{\text{mc}})$.

In general, when considering the suitability of a particular method for use as an analytical technique one must address several concerns, namely, generality, applicability, and ease of use. The method just described is quite general in that it does not use the QE approximation in the derivation of its equations (more accurately,

$$\frac{E'}{k} \left\{ \frac{1}{T^2} - \frac{1}{T_{\text{mc}}^2} \exp\left[\frac{E'}{k} \left(\frac{1}{T_{\text{mc}}} - \frac{1}{T}\right)\right] \right\} = \frac{d \ln I_{\text{TSC}}(T)}{dT}. \quad (60)$$

The form of the left side of Eq. (60) is such that one cannot solve analytically for E' ; however, this may be done numerically quite easily. Another observation is that E' must be temperature dependent. Therefore, the interpretation is that the function $E'(T)$ is that temperature-dependent energy function that one must use in the Randall-Wilkins expression so that it will exactly fit the experimental TSC curve. Furthermore, since $I_{\text{TSC}}^{\text{QE}}(T) = Q(T)I_{\text{TSC}}(T)$, and since $Q(T_{\text{mc}}) = 1$, then $I_{\text{TSC}}^{\text{QE}}(T_{\text{mc}}) = I_{\text{TSC}}(T_{\text{mc}})$ and, therefore, $E'(T_{\text{mc}}) = E$, where E is the real energy.

While the above may be true if $I_{\text{TSC}}^{\text{RW}}(T) \approx I_{\text{TSC}}^{\text{QE}}(T)$, that is, if $s(T) \approx \text{constant}$, $E'(T_{\text{mc}})$ may deviate from the real energy if the frequency factor has a sizable temperature dependence. Indeed, one of the benefits of this formulation of TL and TSC kinetics is that, since one starts with generalized expressions and then subsequently introduces approximations in the development of analytical techniques, one is able to identify clearly the errors involved and derive the relevant correction terms. To produce the corrected energy E_{corr} we must take into account the frequency-factor temperature dependence. To do this we equate $I_{\text{TSC}}^{\text{QE}}(T_{\text{mc}})$ and $I_{\text{TSC}}^{\text{RW}}(T_{\text{mc}})$, thus,

it makes use of the fact that at T_{mc} the QE assumption is valid). Furthermore, the correction terms are known and if additional information is available, these can be accounted for. The method is derived only for first-order kinetics; however, in the vast majority of cases likely to be encountered, this criterion will be satisfied. A major advantage in applying this technique to real data is that it only requires quality data near the peak temperature and furthermore one need only have these data for one heating rate. Typically it is the data near the peak that are the highest quality. This method is in contrast to the initial rise⁴ and the heating rate method of Hoogenstraaten.⁷ While the initial rise method is quite general,¹² it requires that the peak be somewhat isolated and noise free. The method of Hoogenstraaten requires that the peak be known for several heating rates, and, furthermore, Hoogenstraaten's method is derived from the traditional TL and TSC equations which employ the QE approximation.

While in principle the method presented here is only valid for TSC data, all of our calculations show that except for cases when $H \sim N$, $T_{\text{mc}} \approx T_{\text{ml}}$ and therefore this method could safely be applied to TL data as well.

As far as ease of use is concerned, this method requires the numerical solution of Eq. (60) for $E'(T_{mc})$ which can easily be accomplished given knowledge of T_{mc} and the data $I_{TSC}(T)$. One detail to be considered is that since this method requires the calculation of the derivative of the log of data, any noise would be amplified. One solution is to fit the data to a linear combination of smooth functions (such as Gaussians) first and then perform the analysis. Another detail is that Eq. (60) cannot be actually solved at $T = T_{mc}$. In practice, this does not present a problem since numerically one may solve this equation for values very close above and below T_{mc} and then interpolate between the solutions to get $E'(T_{mc})$.

Table III shows the results of this method applied to the data generated from directly solving the rate equations using the parameters of Table I. In all cases the actual value of the energy is 0.300 eV. The results for cases 1 through 4 are shown graphically in Fig. 8. For these cases, $E'(T_{mc})$ agrees with the actual energy to within at most 0.006 eV. Cases 5 through 7 are shown in Fig. 9 and agree to within 0.012 eV. The largest error occurs for cases 15 and 18 for which H and N are within an order of magnitude of each other. It should be pointed out that even in the extreme case when $H = N$ this method produced an energy that was to within 0.048 eV of the actual energy. Correcting for the temperature dependence of the frequency factor using Eq. (63), the energy was found to agree with the actual energy to within at most 0.003 eV for all cases. This clearly shows that the frequency-factor temperature dependence is the dominant source of error.

Figures 8 and 9 show the function $E'(T)$ one must use as the energy so that the RW equation will exactly fit the TSC (or in most cases TL) data. If one were to fit the TSC data using the RW expression with a constant energy parameter, the resulting value would be the average

TABLE III. Summary of results from the effective Randall-Wilkins method applied to the TSC data using the parameters in Table I.

Case	T_{mc} (K)	$E'(T_{mc})$ (eV)	E_{corr} (eV)
1	131.285	0.306	0.303
2	121.645	0.304	0.303
3	113.279	0.302	0.303
4	105.959	0.301	0.303
5	130.312	0.312	0.300
6	130.197	0.311	0.300
7	130.185	0.312	0.300
8	120.756	0.311	0.300
9	112.363	0.310	0.300
10	105.100	0.309	0.300
11	112.364	0.310	0.300
12	112.375	0.311	0.300
13	112.353	0.310	0.300
14	112.354	0.310	0.300
15	112.484	0.314	0.300
16	112.352	0.310	0.300
17	112.353	0.310	0.300
18	113.219	0.348	0.301
19	112.352	0.310	0.300
20	112.352	0.310	0.300

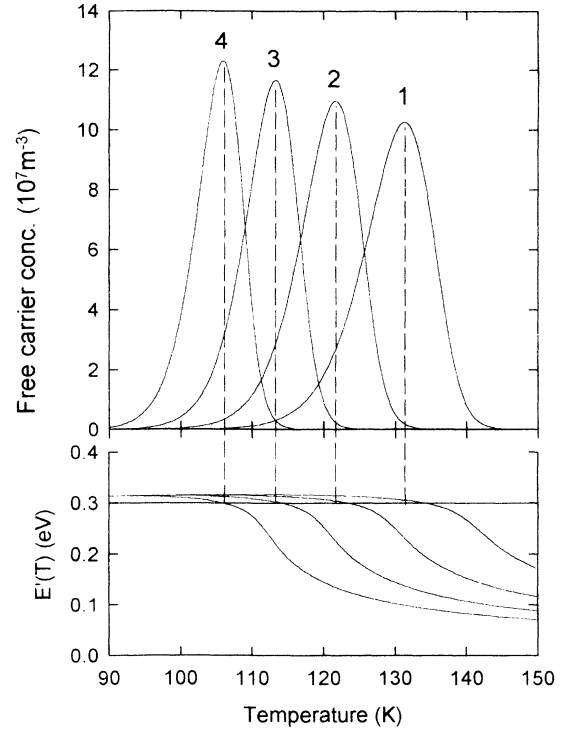


FIG. 8. TSC obtained from numerically solving the rate equations using the parameters from cases 1-4 of Table I and the associated $E'(T)$ curves. For these cases, $E'(T_{mc})$ agrees with the actual energy (0.300 eV) to within at most 0.006 eV.

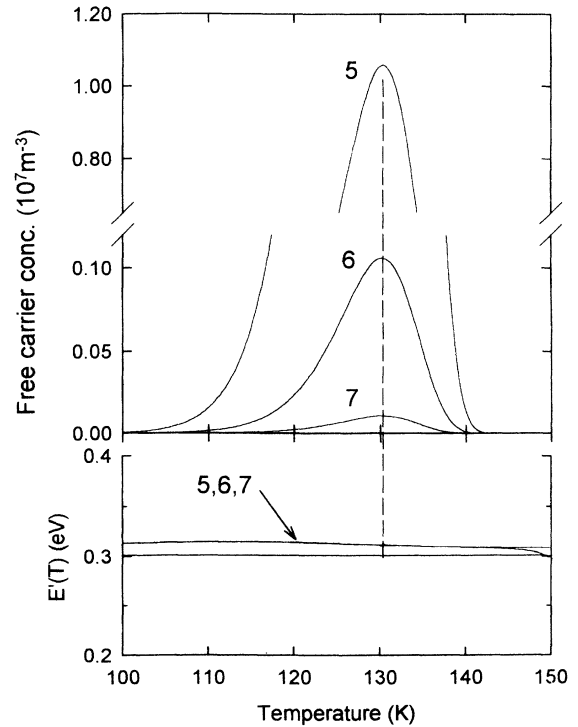


FIG. 9. TSC obtained from numerically solving the rate equations using the parameters from cases 5-7 of Table I and the associated $E'(T)$ curves. For these cases, $E'(T_{mc})$ agrees with the actual energy (0.300 eV) to within at most 0.012 eV.

of $E'(T)$ over the temperature range being fitted. For example, for case 4 in Fig. 8 the $E'(T)$ function is slightly higher than 0.300 eV until just after the peak temperature, when it drops sharply below. Fitting the entire peak with the RW expression would result in the average of $E'(T)$ over this range and would therefore likely produce an energy lower than the actual value. In practice, the quality of experimental data (either from noise or overlap with neighboring peaks) is such that only parts of the peak are actually fitted. If the temperature range fitted is dominated by data past the peak temperature, then the resulting energy will likely be slightly less than the actual value, whereas if most of the fitted portion is below the peak temperature the energy will likely be greater than the actual value.

VI. FINAL COMMENTS AND PRACTICAL IMPLICATIONS

In this paper we have presented a more complete theory of TL and TSC kinetics. By the introduction of the $Q(T)$ and $P(T)$ functions and their relations to the reaction rates, we have shown that slow-retrapping (first-order) kinetics will be the dominant process in systems in which QE is satisfied (i.e., QE and non-first-order kinetics are incompatible). The main advantage arising from the $Q(T)$ and $P(T)$ formulation of TL and TSC kinetics is the explicit separation in the general solutions of the QE and KO approximations. This is in contrast to previous developments where the QE approximation is made before questions of KO are considered. Furthermore, we have developed general expressions for TL and TSC in terms of the $Q(T)$ and $P(T)$ functions and arrived at solutions to the rate equations for the first-order case without the QE approximation and without the presence of $Q(T)$ in the final equations. By parametrizing these general first-order solutions and comparing their curve shapes with both the Randall-Wilkins and numerical RKF solutions to the rate equations, we have shown that the general first-order equations reproduce the numerical solutions over a wider range of parameter cases than do the Randall-Wilkins equations.

The parametrized, general, first-order equations [Eqs. (47) and (48)] could be used in fitting algorithms. However, these equations contain, in general, six parameters which is usually too many for an unambiguous fit. Therefore, it became necessary to develop an independent analytical technique to arrive at one or more of the parameters and thereby reduce the order of a subsequent fit. Furthermore, any analytical method would have to be valid to the same level of theory as the fitting equations themselves, namely, the method could not rely on the QE approximation. In Sec. V we present a simple method for obtaining the activation energy. This method makes use of the knowledge that the QE approximation is only valid at the TSC peak maximum and therefore does not rely on the QE approximation as such. It was found that the main error entering into this analysis arises from the temperature dependence of the frequency factor, which could lead to errors as high as 0.048 eV for extreme cases (i.e.,

when $H = N$). Correcting for this [Eq. (63)] reduced the errors to at most 0.003 eV for all cases. The correction term, however, requires knowledge of $s(T_{mc})$ which in general is unknown. Given experimental TSC data, one method to resolve this is to solve Eq. (60) for $E'(T_{mc})$. Since $E'(T_{mc})$ depends only upon the data $I_{TSC}(T)$ and the value of T_{mc} , $E'(T_{mc})$ is merely a constant. Next, using the parametrization of Eq. (45), Eq. (63) becomes

$$E_{\text{corr}} = kT_{mc} \ln \left(\frac{BT_{mc}^{2-b} kT_{mc}^2}{\beta E'(T_{mc})} \right). \quad (64)$$

Then by substituting Eq. (64) into Eqs. (47) and (48) for E , the general first-order equations could be used for fitting with at most five parameters instead of six. By this method, one has replaced the unknown parameter E with the known parameters T_{mc} and $E'(T_{mc})$. While this method may be more complicated than a three- or four-parameter fit using the RW equations, as we have shown for some cases, the QE approximation can introduce sizable errors into the analysis.

The direction of future work involves an extension of the $Q(T)$, $P(T)$ theory to include systems containing more than one active level.

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APPENDIX: TSC WITHOUT THE CONDITION

$$n(T) + H \gg n_c(T)$$

In this Appendix we will briefly present some of the TSC equations without the $n(T) + H \gg n_c(T)$ approximation. Starting with Eq. (26) as the complete charge neutrality relation, the recombination lifetime [Eq. (9)] becomes

$$\begin{aligned} \frac{1}{\tau(T)} &= v_e(T)h(T)R(T) \\ &= v_e(T)R(T)[n_c(T) - h_v(T) + n(T) + H]. \end{aligned} \quad (A1)$$

If the free carriers are electrons and no holes are in the valence band over the active temperature range [i.e., $h_v(T) \approx 0$], then the recombination lifetime is:

$$\frac{1}{\tau(T)} = v_e(T)R(T)[n_c(T) + n(T) + H]. \quad (A2)$$

The $Q(T)$ and $P(T)$ functions are defined as before in Eqs. (14) and (15). As shown in Sec. IIIB, using these definitions of $Q(T)$ and $P(T)$, the rate equations [Eqs. (5) and (6)], and Eq. (A2), we arrive at the following expressions:

$$n(T) = n_0 \exp \left[-\frac{1}{\beta} \int_{T_0}^T \left(\frac{Q(\Theta)}{Q(\Theta) + P(\Theta)} \right) \times s(\Theta) \exp \left(-\frac{E}{k\Theta} \right) d\Theta \right], \quad (\text{A3})$$

and

$$n_c^2(T) + [n(T) + H]n_c(T) - \frac{S(T)N_c(T)}{R(T)[Q(T) + P(T)]} n(T) \exp \left(-\frac{E}{kT} \right) = 0, \quad (\text{A4})$$

where we see that this last equation is quadratic in $n_c(T)$. The solution is

$$n_c(T) = -\frac{n(T) + H}{2} \pm \frac{1}{2} \left[[n(T) + H]^2 + \frac{4S(T)N_c(T)n(T)}{R(T)[Q(T) + P(T)]} \exp \left(-\frac{E}{kT} \right) \right]^{1/2}. \quad (\text{A5})$$

Since the first term is always negative and the second term is always positive, we must choose the “+” solution so that $n_c(T)$ is always positive. Next, substituting Eq. (A3) into this equation produces the following result:

$$n_c(T) = \frac{1}{2} \left(\left\{ H + n_0 \exp \left[-\frac{1}{\beta} \int_{T_0}^T \left(\frac{Q(\Theta)}{Q(\Theta) + P(\Theta)} \right) s(\Theta) \exp \left(-\frac{E}{k\Theta} \right) d\Theta \right] \right\}^2 + \frac{4S(T)N_c(T)n_0}{R(T)[Q(T) + P(T)]} \exp \left(-\frac{E}{kT} \right) \exp \left[-\frac{1}{\beta} \int_{T_0}^T \left(\frac{Q(\Theta)}{Q(\Theta) + P(\Theta)} \right) s(\Theta) \exp \left(-\frac{E}{k\Theta} \right) d\Theta \right] \right)^{1/2} - \frac{n_0}{2} \exp \left[-\frac{1}{\beta} \int_{T_0}^T \left(\frac{Q(\Theta)}{Q(\Theta) + P(\Theta)} \right) s(\Theta) \exp \left(-\frac{E}{k\Theta} \right) d\Theta \right] - \frac{H}{2}. \quad (\text{A6})$$

Considering the case where $Q(T) \gg P(T)$, i.e., slow retrapping, Eq. (A3) becomes

$$n(T) = n_0 \exp \left[-\frac{1}{\beta} \int_{T_0}^T s(\Theta) \exp \left(-\frac{E}{k\Theta} \right) d\Theta \right], \quad (\text{A7})$$

and Eq. (A4) becomes

$$n_c(T)[n_c(T) + n(T) + H] = \frac{S(T)N_c(T)}{R(T)Q(T)} n(T) \exp \left(-\frac{E}{kT} \right). \quad (\text{A8})$$

Since under conditions of quasiequilibrium $Q(T) = 1$, we find that

$$n_c^{\text{QE}}(T)[n_c^{\text{QE}}(T) + n(T) + H] = Q(T)n_c(T)[n_c(T) + n(T) + H]. \quad (\text{A9})$$

This is the real relation between the free-carrier concentration with and without the QE approximation under first-order kinetics [i.e., the equivalent to Eq. (34)]. We note that, from Eq. (A7), it is unnecessary to label $n(T)$ as $n^{\text{QE}}(T)$ under conditions of first order, since this concentration is not affected by the QE approximation. From Eq. (A2) we see that Eq. (A9) is nothing more than the statement, $I_{\text{TL}}^{\text{QE}}(T) = Q(T)I_{\text{TL}}$, as expected since the general TL expression does not depend on the $n(T) + H \gg n_c(T)$ approximation. If this approximation is made, Eq. (A9) then becomes $n_c^{\text{QE}}(T) = Q(T)n_c(T)$ as expected.

Using Eq. (36) for $Q(T)$ expressed entirely in terms of TSC and Eq. (A2) for the recombination lifetime, Eq. (A9) becomes

$$\begin{aligned} & n_c^{\text{QE}}(T)[n_c^{\text{QE}}(T) + n(T) + H] \\ &= \frac{\beta}{v_e(T)R(T)} \frac{dn_c(T)}{dT} + n_c^2(T) + n_c(T)[n(T) + H], \end{aligned} \quad (\text{A10})$$

where, in a remarkable occurrence, *all traces of $\tau(T)$ vanish*. This is in contrast to the earlier generalized first-order solutions that explicitly contain $\tau(T)$. We rewrite Eq. (A10) in standard form as

$$\frac{dn_c(T)}{dT} + A(T)n_c(T) + J(T)n_c^2(T) = M(T), \quad (\text{A11})$$

where

$$A(T) = \frac{v_e(T)R(T)}{\beta} [n(T) + H],$$

$$J(T) = \frac{v_e(T)R(T)}{\beta},$$

$$M(T) = \frac{v_e(T)R(T)}{\beta} n_c^{\text{QE}}(T)[n_c^{\text{QE}}(T) + n(T) + H].$$

The form of Eq. (A11) is that of the Riccati equation.¹⁹ The method now would be to solve Eq. (A11) for $n_c(T)$ as a function of $n(T)$, $n_c^{\text{QE}}(T)$, and T . Equation (A6) for $Q(T) \gg P(T)$ and with $Q(T) = 1$ would produce $n_c^{\text{QE}}(T)$, and Eq. (A7) for $n(T)$ would complete the solution. This solution would not contain $\tau(T)$ and H would enter in as a new parameter. However, due to the difficulty in solving the Riccati equation for variable coefficients, and due to the complexity of Eq. (A6), it is likely that the resulting solution would be unwieldy for practical applications.

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