

Letter to the Editor

ON THE RESOLUTION ABILITY OF CHARGE TRANSIENT SPECTROSCOPY A COMPARISON WITH THERMALLY STIMULATED DISCHARGE (TSD) MEASUREMENTS

О РАЗРЕШАЮЩЕЙ СПОСОБНОСТИ СПЕКТРОСКОПИИ ПЕРЕХОДА ЗАРЯДА В СРАВНЕНИИ С ИЗМЕРЕНИЯМИ ТЕРМАЛЬНОГО ВЫЗВАНОГО РАЗРЯДА

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The deep-level transient spectroscopy (DLTS) developed originally by Lang [1] occurs now in many versions. One of them, referred to as charge transient spectroscopy (QTS), was suggested in [2, 3]. All the modifications of the DLTS are widely used for the investigation of deep levels in semiconductors. Possibilities to expand the DLTS principle utilization to other materials or to relaxation processes of different origins were pointed out in [4, 5]. The paper [6] deals with the QTS as an alternative of the thermally stimulated depolarization of a dielectric, the comparison of the respective sensitivities of both methods is in favour of QTS. The dielectrics contain usually several kinds of dipoles, even distributed spectra of relaxation times are expected in some cases. The question arises about the QTS ability to resolve contributions of different relaxation mechanisms to the overall response, mainly in comparison with the TSD, the latter being considered to possess high resolution power.

The following brief analysis concerns a concrete QRS apparatus described in more detail in [7]. The apparatus utilized the correlation method suggested in [8], while using a multichannel signal-to-noise ratio degradation. Yet there is a simultaneous decrease of the resulting signal amplitude [8, 9]. With an increasing number of channels ($n > 3$) the selectivity exhibits saturation and we therefore confine our considerations to a standard case, with $n = 3$. If we suppose a single dipole species to contribute to the polarization of the dielectric, the process is governed by its relaxation time

$$\tau = \tau_0 \exp\left(\frac{E}{kT}\right) \tag{1}$$

(where τ_0 is preexponential factor). The formula for the specified QTS mode [7] can be derived:

$$S = K \left\{ \frac{2\tau(x)}{\Delta t} \left[\exp\left(-\frac{\Delta t}{\tau(x)}\right) - \exp\left(-\frac{\Delta t}{2\tau(x)}\right) \right] - \frac{3\tau(x)}{2\Delta t} \left[\exp\left(-\frac{2\Delta t}{\tau(x)}\right) - \exp\left(-\frac{\Delta t}{\tau(x)}\right) \right] + \frac{\tau(x)}{4\Delta t} \left[\exp\left(-\frac{4\Delta t}{\tau(x)}\right) - \exp\left(-\frac{2\Delta t}{\tau(x)}\right) \right] \right\}, \tag{2}$$

where $x = E/kT$, Δt denoting in our case the "time window" related to the relaxation time τ_{max} at the maximum response [1], K is a constant involving the equilibrium amount of polarization. The assumption of noninteracting dipoles with different time constants is required to conserve the superposition principle.

Now let us concentrate on Fig. 1 which shows a QTS signal computed for the case of a single polarization (depolarization) according to (2). As a measure of the selectivity we take the FWHM (full width-at-half maximum), see Δ in Fig. 1. It may be evident from the figure that the selectivity improves with increasing time window Δt , while keeping the remaining parameters constant. When reducing the Δt at the QTS peaks are shifted towards higher temperatures and broadened. Note the amplitude of the signal does depend on Δt .

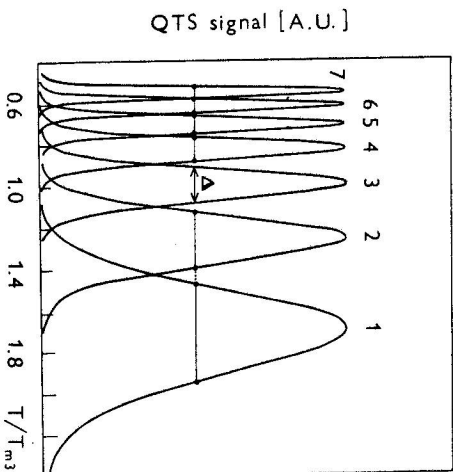


Fig. 1. QTS signal calculated from (2), the variable x being defined $x = \frac{E}{kT_{m3}} \cdot \frac{T_{m3}}{T}$ (T_{m3} - temperature of the peak maximum (3)), where $\frac{E}{kT_{m3}} = 10$. The peaks correspond to various widths of the time window: (1) - $\Delta t = 0.01$, (2) - 0.1 , (3) - 1 , (4) - 10 , (5) - 100 ms, (6) - 1 and (7) - 10 s. FWHM is labelled as Δ .

An exact criterion for the resolution ability cannot be derived. One can obtain an illustrative picture from looking at the fine structure of the resulting QTS signal, composed of two peaks which stem from two dipole species differing in the respective activation energies E_a and E_b . The evolution of such a signal with increasing time window Δt is envisaged in Fig. 2. Two counteracting tendencies are evident; separation between the peaks (a) and (b), respectively, reduces for enhanced Δt , while FWHM at the time becomes smaller; thus one can deduce an increasing resolution ability for Δt becoming large. Curve (3) demonstrates the case with a valley lying 5% below the link of the two maxima (see the insert in Fig. 2 for the "5%" criterion). The ratio of the respective activation energies E_b/E_a is nearly equal to 1.18 in this case.

Comparison with TSD. The thermally stimulated depolarization method (for details see [10]) relies on polarizing the dielectric at an elevated temperature T_p with subsequent cooling to a temperature T_0 . At the latter temperature the polarization voltage is removed and the sample of the dielectric shortcircuited by a current meter. Then, when heating the sample (mostly at constant heating rate $b = dT/dt$) a depolarization current is flowing in the outer circuit. In the

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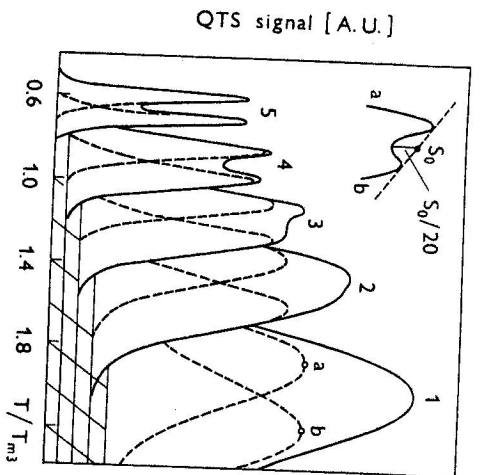


Fig. 2. QTS signal as a sum of two peaks (a) and (b), respectively, resulting from two types of dipoles with closely spaced activation energies providing the ratio $E_b/E_a = 1.18$. The curves correspond to various widths of the time window: (1) - $\Delta t = 0.01$, (2) - 0.1, (3) - 1.0, (4) - 10, and (5) - 1000 ms. Insert shows the "5%" criterion.

case of an ideal dielectric containing only a single species of noninteracting dipoles one can derive the following equation for the TSD:

$$\frac{i(T)}{P_0} = \left[\tau_0 \exp\left(\frac{E}{kT}\right) \right]^{-1} \exp \left\{ - \int_{\tau_0}^T \left[b \tau_0 \exp\left(\frac{E}{kT'}\right) \right]^{-1} dT' \right\} \quad (3)$$

(P_0 stands for the amount of polarization of the dielectric prior to heating). Characteristic TSD spectra which depend upon the heating rate b are well known [10]: when decreasing b , the resolution ability of the TSD increases, but the signal amplitude is lowered, as well. Contrary to the increasing b , however, the resolution becomes lower, in addition there is a danger of temperature gradients within the sample. Currently used experimental values for b vary about $b \sim 0.15 \text{ K s}^{-1}$. Fig. 3 shows a numerical calculation (according to (3)) for the case of two types of dipoles with the respective activation energies E_a and E_b , the value $E_a = 0.5 \text{ eV}$ being taken for the former one. The energy E_b was determined according to the "5%" criterion. This requirement is satisfied for $E_b = 0.54 \text{ eV}$, i.e. the ratio E_b/E_a is close to 1.08. For the same E_a and E_b values a calculation was performed for a QTS response, while choosing $\Delta t = 1 \text{ ms}$ for the time window as usually found in practice. The two dipole relaxations present are not discernible from the shape of the QTS curve (see Fig. 3). This demonstrates the generally valid fact that under standard conditions (i.e. $b \sim 0.15 \text{ K s}^{-1}$ and $\Delta t \sim 1 \text{ ms}$) the resolution power of the TSD is evidently higher than that of QTS. As it could be deduced from the preceding analysis (Fig. 1 and 2) there is a possibility to improve the QTS resolution without any sensitivity loss by a remarkable widening e.g. for the large time window ($\Delta t = 840 \text{ ms}$), is reproduced in Fig. 3. Evidently, we get the same resolution as with the TSD.

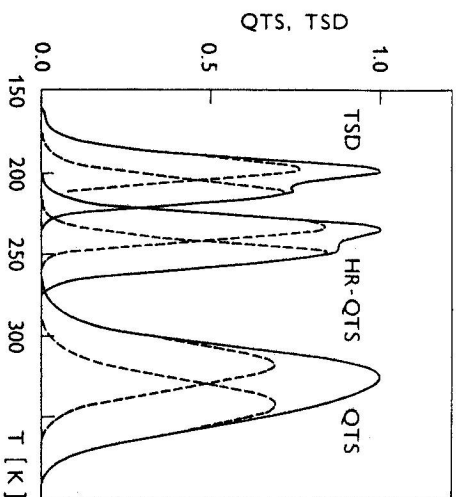


Fig. 3. The calculated TSD signal (left) for the case of two dipole kinds with activation energies $E_a = 0.5 \text{ eV}$ and $E_b = 0.54 \text{ eV}$ ($\tau_0 = 10^{-11} \text{ s}$, $b = 0.15 \text{ K s}^{-1}$). The QTS signal (right) was computed for the same energies taking the widely used time window $\Delta t = 1 \text{ ms}$. In the middle of the figure there is the QTS signal in high resolution (HR-QTS) regime ($\Delta t = 840 \text{ ms}$).

The main advantages of the QTS when compared with the TSD (e.g. external sensitivity, arbitrary heating rate, simple analytical expression for the signal, etc.) were mentioned in [6]. This list can be complemented by the possibility to obtain all the parameters during a single temperature scan (in multichannel mode) mainly for the distributed polarizations, the determination of which is not consistent with the single TSD temperature scan. The basic conclusion of the present analysis is the requirement to apply large time windows to QTS experiments to achieve a resolution capability comparable with that of the TSD. Nevertheless, the TSD is the only technique which makes it possible to operate in the partial-heating mode (multistage TSD - [10]), which mode is particularly useful when studying composite dielectrics like polymers.

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