

Simultaneous Measurement of Thermally Stimulated Conductivity and Thermoluminescence

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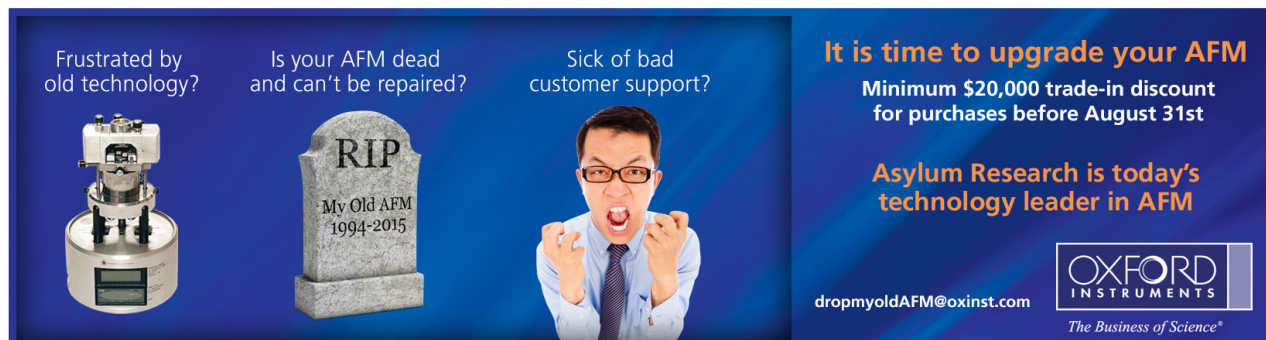
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Simultaneous Measurement of Thermally Stimulated Conductivity and Thermoluminescence

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The relation between thermally stimulated conductivity (TSC) and thermoluminescence (TL) is discussed assuming that the transition of electrons (holes) from traps to recombination centers is through the conduction (valence) band. It is shown that under this general assumption, the TL maxima shift to lower temperatures compared to the corresponding TSC ones. A method for calculating the recombination probabilities by a simultaneous measurement of TL and TSC is given.

Thermoluminescence (TL) and thermally stimulated conductivity (TSC) peaks have usually been analyzed by the same methods,¹ which is justified when one assumes a constant lifetime of conduction carriers. This approach implies that the two curves have the same shape, and more specifically, that the maxima appear at the same temperatures. Experimentally, however, simultaneous measurements of TL and TSC revealed in many cases a shift between the corresponding peaks, in most cases the TL peak appeared at lower temperatures than the TSC one.²⁻⁹ For a certain case of a second-order TL peak,¹⁰ the occurrence of this shift was proved theoretically. The purpose of the present paper is to show generally that under the assumption that the electrons are excited into the conduction band before the luminescent recombination, the TL peaks should always appear at lower temperatures than the TSC ones. Furthermore, a method for evaluating the recombination probability by the use of the simultaneous measurement of these two phenomena is given.

Three simultaneous differential equations have been written¹¹ to account for the transitions of electrons between a single trap, the conduction band, and a recombination center. One of these equations is

$$I(t) = -\frac{dm}{dt} = Amn_c, \quad (1)$$

where I is the TL intensity, t is the time (sec), m is the concentration of carriers in the recombination center (cm^{-3}), and A is the so-called "recombination probability" ($\text{cm}^3 \text{sec}^{-1}$). A is known to be equal to the product of the cross section for recombination and the thermal velocity of the free carriers.¹¹

Equation (1) merely states that the rate of recombination is proportional to the concentrations of free electrons (n_c) and holes in centers (m) and that the TL intensity is equal (in suitable units) to this rate. As such, this equation has much more general bearing than just describing the situation when only one trap and one center are involved. As long as only transitions into one recombination center are measured, one does not care whether there are transitions into other centers. Such transitions may change the number of electrons in the conduction band, but Eq. (1) holds true for the net concentration n_c . Moreover, no information about the trap of traps contributing to n_c is needed as long as $n_c = n_c(t)$ can be measured by conductivity measurements.

Assuming that $n_c(t)$ is known independently, Eq. (1) can be solved to give

$$m = m_0 \exp[-A \int_0^t n_c(t') dt'] , \quad (2)$$

where m_0 is the initial concentration of holes in centers. On inserting (2) into (1) we obtain

$$I = Am_0 n_c(t) \exp[-A \int_0^t n_c(t') dt'] . \quad (3)$$

It should be noted that since we have not assumed any specific heating function [$T = T(t)$], Eqs. (2) and (3) are also valid for the case of $T = \text{const}$ (phosphorescence). Writing Eq. (3) for two arbitrary points t_1 and t_2 and dividing one by the other, we have

$$I(t_2)/I(t_1) = n_c(t_2)/n_c(t_1) \exp[-A \int_{t_1}^{t_2} n_c(t) dt] . \quad (4)$$

All the quantities in Eq. (4) except for A are measurable and thus A can be evaluated from the equation. For the determination of $n_c(t)$, we have to measure the conductivity glow curve. The conductivity is given by $\sigma = e\mu n_c$, where e is the electronic charge and μ is the mobility. In many cases the mobility and its dependence on temperature are known and $n_c(t)$ follows immediately. An alternative method for evaluating $n_c(t)$ is by thermally stimulated electron emission (TSE).⁷ This method has the advantage that the knowledge of the mobility is not needed, however, experimental complications due to surface effects sometimes occur.

Figure 1 shows schematically a peak of $n_c = n_c(t)$, the corresponding TL peak, and the quantities $I(t_1)$, $I(t_2)$, $n_c(t_1)$, $n_c(t_2)$, and $\int_{t_1}^{t_2} n_c(t) dt$, which are necessary for evaluating the recombination probability A . It is to be noted that only relative values of $I(t)$ are needed, whereas absolute values of $n_c(t)$ are necessary (for the integral). Some simplification can be achieved by choosing t_1 and t_2 so that $I(t_1) = I(t_2)$ or $n_c(t_1) = n_c(t_2)$. The significance of the method is that the integration can be done in a limited range, this in comparison to the "straight-forward" method for finding A ² by writing Eq. (1) as $A = I(t)/m(t)n_c(t)$ and finding $m(t)$ from $m(t) = \int_t^\infty I(t') dt'$, namely, from the area under the TL curve from t to infinity. The complication is that integration of the experimental curve to infinity is usually inaccurate. Moreover, the possible variations of A with temperature can be neglected only in limited ranges.

Preliminary measurements of the recombination probabilities have been done for a peak at $\sim 350^\circ\text{K}$ in a type II_a insulating diamond¹² and for a peak at $\sim 260^\circ\text{K}$ in a semiconducting diamond.¹³ The mobility has been taken to be $2000 \text{ cm}^2 \text{ V}^{-1}$ which is accurate within the temperature range of interest to a factor of 2. The value of A is found to be of the order of $10^{-5} \text{ cm}^3 \text{ sec}^{-1}$ for the insulating diamond and the cross section for recombination is

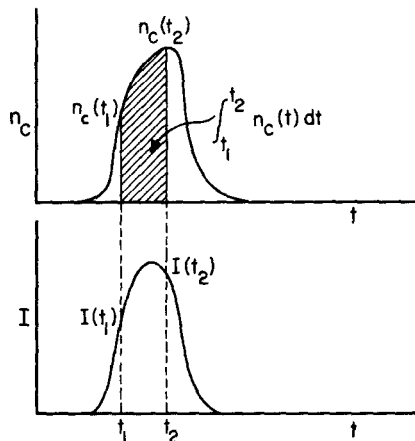


FIG. 1. Quantities measured to find the recombination probability A (schematic).

evaluated therefrom to be about 10^{-12} cm^2 . In the semiconducting diamond, the recombination probability is found to be $10^{-12} \text{ cm}^3 \text{ sec}^{-1}$ and the cross section for recombination 10^{-19} cm^2 . The values are estimated in this case only to an order of magnitude mainly because of experimental difficulties related to the geometrical shape of the samples and the uncertainty about the effective area of the electric contacts. Writing Eq. (1) as $A = I/mn_c$ one can see that if the values of I/m in two samples differ by only a few orders of magnitude, as is the case here, the cross sections are roughly inversely proportional to the conductivities. Thus, since the conductivity in semiconducting diamond is more than ten orders of magnitude higher than in the insulator, the large difference between the cross sections is to be expected.

By equating the derivative of Eq. (3) to zero, one has

$$\left(\frac{dn_c}{dt}\right)_{\text{max}} = A [n_c(t_{\text{max}})]^2 , \quad (5)$$

where the subscript "max" indicates that the values are taken at the maximum of the luminescence peak. The right-hand side of Eq. (5) is always positive, and therefore, the left-hand side should also be positive. This means that when the TL peak reaches the maximum point, the curve of $n_c(t)$ is still increasing, or in other words, that $n_c(t)$ maximizes later (at a higher temperature) than $I(t)$. Thus, the TSE peak would appear at a lower temperature than the TL peak. Since the dependence of the mobility on temperature is weak this would usually be so for the TSC peak as well. This is in agreement with the references mentioned above.²⁻¹⁰

In some cases recombinations into more than one center contribute to the measured TL. Let the concentrations of the empty centers be m_1, m_2, \dots, m_k and the respective recombination probabilities A_1, A_2, \dots, A_k . We now obtain

$$I_i(t) = -\frac{dm_i}{dt} = A_i m_i n_c, \quad i = 1, 2, \dots, k. \quad (6)$$

Each equation can be solved independently using the same $n_c(t)$. The total measured emission would be the sum over i of the partial intensities $I_i(t)$. If the detector has different sensitivities for various i 's (corresponding usually to various centers and therefore to various wavelengths), the I_i 's are to be multiplied by constants α_i

$$I(t) = \sum_{i=1}^k \alpha_i I_i(t) = \sum_{i=1}^k \alpha_i A_i m_{i0} n_c(t) \times \exp(-A_i \int_0^t n_c(t) dt). \quad (7)$$

The condition for maximum would be now

$$\sum_{i=1}^k \alpha_i A_i m_{i0} \exp\left(-A_i \int_0^{t_{\max}} n_c(t) dt\right) \times \left[\left(\frac{dn_c}{dt} \right)_{\max} - A_i [n_c(t_{\max})]^2 \right] = 0, \quad (8)$$

or

$$\alpha \left(\frac{dn_c}{dt} \right)_{\max} = \beta [n_c(t_{\max})]^2,$$

where

$$\alpha = \sum_{i=1}^k \alpha_i A_i m_{i0} \exp\left(-A_i \int_0^{t_{\max}} n_c(t) dt\right)$$

and

$$\beta = \sum_{i=1}^k \alpha_i A_i^2 m_{i0} \exp\left[-A_i \int_0^{t_{\max}} n_c(t) dt\right].$$

α and β are sums of positive factors and thus they are positive. The condition is thus similar to that

obtained in Eq. (5) and bears similar implications, namely, that even under these general conditions the TL peak should appear at a lower temperature than the corresponding TSC or TSE ones.

In conclusion, a method is proposed for evaluating probabilities and cross sections for recombination by simultaneous measurement of TL and either TSC or TSE. The experimental evidence for the shift of TL peaks toward a lower temperature compared to the corresponding TSC and TSE peaks, found for many samples, is explained under rather general conditions. The only limitation is that the measured emission would not include substantial components due to transitions from excited states within the forbidden band.

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