Dispersive Low-Temperature Transport in *a*-Selenium

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Transient hole transport in *a*-Se below 180 K is shown to be a stochastic process. The transition from the well-defined high-temperature transport with Gaussian dispersion to the low-temperature stochastic transport is not paralleled by a change in the activation energy of the hole drift velocity.

The thermally activated carrier drift mobilities in the prototype elemental chalcogenide glass *a*-Se are commonly interpreted to indicate carrier motion in extended states with frequent interruption by shallow trapping.[1] There is, however, no direct experimental evidence for the proposed transport mechanism. The experimental situation appears more satisfying for the model substance of a tetrahedrally bonded elemental glass *a*-Si. Here, LeComber and Spear[2] argued that the distinct lowering of the activation energy for electron drift, which they observed on cooling the sample to below ~250 K, evidences the transition from shallow-trap–controlled transport to hopping among localized band tail states. Hence, the high-temperature (HT) and low-temperature (LT) activation energies were associated with shallow-trap depth and hopping energy, respectively. No similar experiments were reported for *a*-Se. Unfortunately, however, time-of-flight experiments in *a*-Se were restricted to temperatures above ~180 K where the carrier transit exhibits little dispersion and the transit time is well defined. At LT the dispersion of the propagating carrier packet increases rapidly and the associated current signal becomes so featureless that a characteristic transit time t_T cannot be established without ambiguity.[3] The question, therefore, remained open whether the activation energies of the drift mobilities in *a*-Se would eventually decrease at lower temperatures in a manner similar to that reported for electron drift in *a*-Si. Interest into this problem was recently revived when Scher and Montroll[4] (SM) provided a theoretical framework for the analysis of featureless transient current signals which enables us now to extend our knowledge of electronic transport in *a*-Se to much lower temperatures. This Letter highlights such a study for hole transport in *a*-Se. This is the first instance where the transition from the well-defined transport at HT to the extremely dispersive transport at LT has been studied. It is found, unexpectedly, that the transition is *not* accompanied by a change in the activation energy.

SM[4] propose that the dispersive transport, common to a large number of disordered solids, has to be treated as a stochastic succession of events with broad event-time distribution which can extend beyond the time range of the experimental observation ($\sim t_T$). An example of such a process is hopping among localized states with the hopping distance as stochastic variable. This theoretical approach has the important consequence that the dispersion of the propagating carrier packet is non-Gaussian which leads to transport characteristics distinctly different from transport in ordered solids. The essential features of stochastic transport have been verified in *a*-As_2Se_3 and molecularly doped polymers.[5,6] It will be shown below that the LT hole transport in *a*-Se conforms to the stochastic transport mechanism also and, therefore, one has the unique possibility to study the transition from the well-defined transport at HT with Gaussian dispersion to the stochastic dispersive transport at LT.

Eleven samples in the thickness range 10–125 μm were prepared by open-boat evaporation onto a substrate (polished Al or Au coated Kel-F) held at 55°C. To ensure blocking contacts the Se surface was coated with a <1-μm layer of Lexan polycarbonate before the semitransparent Au top electrode (0.19 cm^2, 200 Å) was applied by evaporation. The time-of-flight measurements were standard and details are described elsewhere.[5] The free holes were generated by a 5-nsec 3371-Å laser pulse of low intensity which lagged the bias field by ~0.5 sec.

Figure 1 shows the Arrhenius plot of the hole velocity $v_h = L/t_T$ for a 75-μm sample for various applied electric fields. Included are several current signals recorded at 10 V/μm at the temperatures indicated. The traces clearly demonstrate the dramatic change of the current shape as T is lowered. At HT t_T (arrow) is clearly defined by the sharp drop in the current level which marks

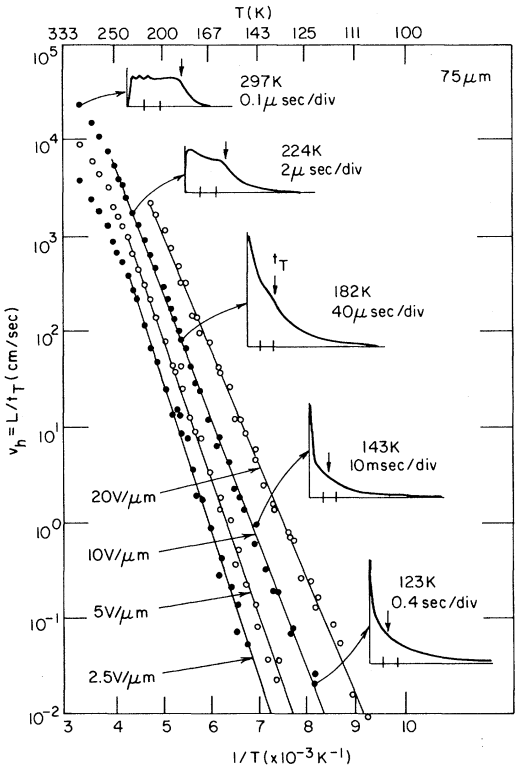

FIG. 1. Semilog plot of hole velocity versus $1/T$ for various fields, and transient current traces at several temperatures. Arrows indicate transit times.

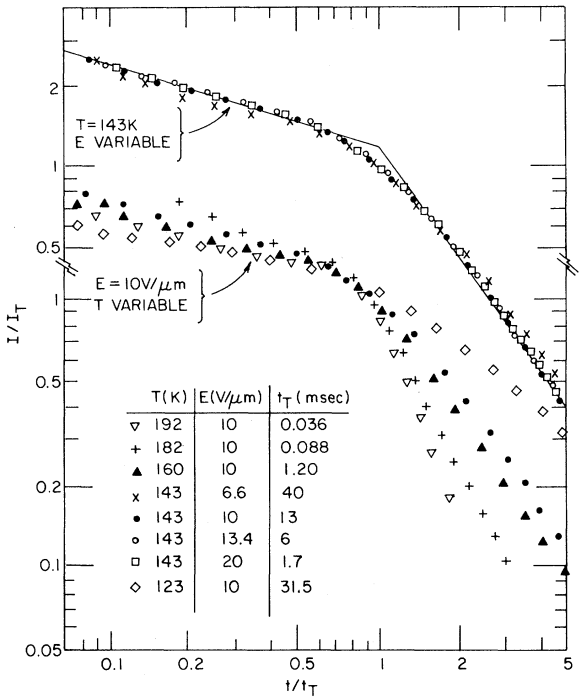

FIG. 2. Log-log plots of current transients. Upper part: universality with respect to E at fixed T. Lower part: T dependence of relative dispersion at fixed E.

the arrival of the injected charge sheet at the opposite electrode. The procedure used to determine t_T at LT when there is no such fiduciary will be described in the next paragraph. The essential results of Fig. 1 are as follows: (i) Below ~250 K and down to the lowest experimental temperature of ~120 K, the activation energy Δ_h is well defined; (ii) no change of Δ_h is observed on lowering T into the range of strong dispersion at ~180 K; (iii) Δ_h is field dependent, but there is no specific temperature at which the field dependence sets in; (iv) in agreement with earlier studies the temperature dependence of v_h weakens above ~250 K.[1,3]

It remains to be established now that the dispersive current signal is indeed due to a stochastic process. The evidence will be derived from the current shape $i(t)$ and the L dependence of t_T. SM predict that $i(t)$ decays with time as $t^{-(1-\alpha)}$ for $t<t_T$ and $t^{-(1+\alpha)}$ for $t>t_T$, and therefore, $\ln i$ versus $\ln t$ has tangents of slope $-(1-\alpha)$ and $-(1+\alpha)$, respectively. The "transit time" t_T defined by the intersection of the tangents has been used to calculate the hole velocity in Fig. 1. $0<\alpha<1$ is a parameter describing the microscopic details of the transport process and therefore is a measure of the local disorder. α decreases as disorder becomes more dominant. Changing experimental conditions at fixed T should merely result in a parallel shift of $i(t)$ along the logarithmic axes, since the parameter α is largely field independent; i.e., the relative dispersion of $i(t)$ is independent of t_T.[4] This "universality" of $i(t)$ is distinctly different from the familiar Gaussian dispersion which changes relative to t_T as $t_T^{-1/2}$. At HT the transient hole response in a-Se conforms with the Gaussian description[7] while, as demonstrated in Fig. 2, at LT it exhibits the features of a stochastic process, i.e., algebraic time dependence and universality. To obtain the upper graph in Fig. 2, current traces recorded at the indicated fields at 143 K were plotted in logarithmic units and then normalized with respect to time and current. Within the experimental error the various traces generate a t_T-independent universal curve which on both sides of $t/t_T=1$ can be approximated by an algebraic time dependence. The lower part of Fig. 2 shows similar traces, but now recorded at a constant field of 10 V/μm for various temperatures. Although the algebraic

time dependence is preserved the different traces now do not generate a universal curve. The relative dispersion clearly broadens as T is reduced, which indicates that α decreases with decreasing temperature and disorder becomes increasingly important. Between ~140 and 170 K the sum of the exponents characterizing $i(t)$ is close to the theoretical value of -2. No theoretical framework is available to analyze $i(t)$ at higher temperatures where the transition from the Gaussian to the universal dispersion occurs. Below ~140 K the sum is smaller than theoretically predicted. It is likely that in this temperature range pulse distortions due to trapped charge become noticeable which tend to flatten the initial part of $i(t)$. Similar observations were made and analyzed along these lines for hole transport in a-As_2Se_3.[5]

In the Gaussian regime the mean displacement $\langle l \rangle$ of the carrier packet from the illuminated electrode increases in proportion to time, $\langle l \rangle \propto t$, and therefore, $t_T \propto L$. In the stochastic regime $\langle l \rangle \propto t^{\alpha}$; hence $t_T \propto L^{1/\alpha}$, where α is the same parameter introduced to describe $i(t)$. Consequently, $i(t)$ and $t_T(L)$ are correlated and with increasing dispersion, i.e., decreasing α, the L dependence of t_T should become stronger. The proportionality between t_T and L at HT has been verified extensively.[1,3] Figure 3 shows the result at LT in a plot of $\ln t_T$ versus $\ln L$, measured at 10 V/μm at different temperatures. In agreement with the stochastic transport model one notes that (i) the relationship between t_T and L is superlinear, (ii) the deviation from linearity begins at around 180 K where the dispersion of $i(t)$ becomes significant, and (iii) the exponent describing the L dependence increases towards lower temperatures in qualitative correlation with the increase of the relative dispersion of $i(t)$ (Fig. 2, lower part).

On the basis of the evidence shown in Figs. 2 and 3 it is concluded that at LT hole transport occurs via a stochastic process of the form described by SM.[4] That the transition from the Gaussian to the stochastic process is not paralleled by a change in the activation energy of the hole velocity suggests that the same basic mechanism of carrier propagation prevails in the entire temperature range accessed in these experiments. While one mechanism is the previously proposed multiple-trap model,[1] a hopping process is suggested as a simpler alternative. In either case disorder becomes dominant below ~180 K and causes the distribution of event times (trap release time or hopping time) to broaden into the time range of t_T. Both transport mechanisms are

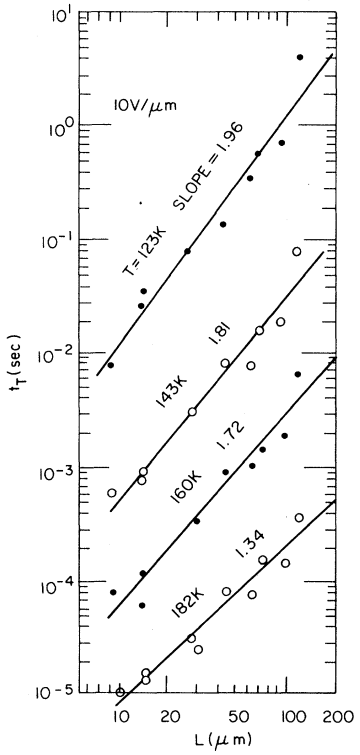

FIG. 3. Log-log plots of t_T versus L at various temperatures.

compatible with the deviation from the well-defined thermally activated behavior which is observed above ~250 K (Fig. 1). For the trap-controlled process a microscopic mobility proportional to T^{-n}, where $n \sim \frac{3}{2}$, has been proposed.[1] For the hopping mechanism, one would expect that at temperatures sufficiently above ~180 K disorder effects are largely overcome, so that the temperature dependence weakens and eventually approaches a nonactivated behavior characteristic of particles executing diffusive Brownian motion.[8]

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[8]See, for instance, M. H. Cohen, J. Non-Cryst. Solids 2, 432 (1970).