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Space Charge Effect on the Transient Current Induced by Pulsed Laser Irradiation

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The laser power dependence of the transient current induced by the pulsed laser irradiation on pyrene in n-hexane was measured. The peak time of the current decreased with increasing the produced charge. This result was inconsistent with a theoretical prediction on the space charge effect, and showed the electric field inhomogeneity was important.

The time profile of the transient current produced by the pulsed irradiation is known to be affected by the space charge effect(SCE).[1,2] In this work, we have measured the laser power dependence of time profiles of the transient current produced by the pulsed laser irradiation of pyrene dissolved in hexane, and compared with the theoretical prediction on SCE.

The apparatus is similar with the previous one.[3] The third harmonics(355nm) of a Nd-YAG laser was introduced to the photoionization cell without focusing in order to avoid inhomogeneity in the ionization region. The size of the beam was restricted to 5 x1 mm with a mask in front of the cell; the electrode spacing was 8 mm. The applied voltage between the electrodes was 1kV.

The laser power dependence of time profiles of the transient current produced by the 355 nm irradiation on the 10^{-5}mol/dm^3 pyrene solution in hexane are shown in Fig 1; time resolution was set to 0.3 ms. At an laser pulse power of 0.045 mJ/pulse, the time profiles were described by the time of flight model,[5] and thus SCE was negligible. At higher laser power, the transient current increased remarkably with time after the initial decrease[1,4] and showed a maximum at about 0.3-0.5 s. The peak time(t_p) of the current, which is defined as the time between the laser irradiation and the second peak of the time profile, became smaller with charge as shown in Fig. 2.

According to the model calculation of SCE[1] assuming a very thin layer for photoionization region, the average electric field increases with separation of + and - charge layers, and the current increases with time and has a maxima, which corresponds to the arrival of charges to the electrodes. When the charge layer is produced at the center between two electrodes, the arrival time of the charge(t_a) can be described according to the thin layer model as[1]

$$t_a = (L/2\mu\Delta E)\ln\{(1+\Delta E/2E_0)(1-\Delta E/E_0)\} \quad , \quad (1)$$

where E_0 is the applied external field(1.25 kV/cm), L the electrode spacing, and ΔE is the discrepancy of the electric field owing to SCE and is related with the charge density(σ) of the thin layer and the dielectric constant of the solution(ε) as $\Delta E=\sigma/\varepsilon$. μ is the summation of the mobilities of anion and cation. According to the thin layer model, t_a=t_p. Equation 1 indicates that calculated value of t_a is an increasing function of ΔE; e.g., t_a increases about 5% when σ=2000 nC/m^2 in this experiment. This prediction was in contrast with our result that the peak time of current was shortened with increase of the charge.

In the thin layer model,[1,2] it is assumed that the charge layer keeps its thickness during the motion of charges under the external and the averaged SCE fields. However, there exists inhomogeneity of the electric field within the charge layer; the charges in the front (close to the electrode) of the charge layer feels larger field than that in the back(far from the electrode). Since the velocity of a charge is proportional to the field, the former moves faster than the latter does. This difference of velocities within the charge layer should widen the thickness of the layer with time. Therefore, the front of the layer arrives at the electrode earlier than that predicted by the thin layer model. After the arrival of the front side, the charge gradually disappears at the electrode and the current decreases with time. Width of the charge layer becomes larger as the total amount of the charge increases. The peak of the transient current should correspond to the arrival of the front side and should be shortened with the increase of the charge. This is inconsistent with the thin layer model, where tp is assumed to be identical with t_a.[1,2] The present result indicates that the inhomogeneous SCE in the charge layer is another key factor to govern the time profile of the transient current.

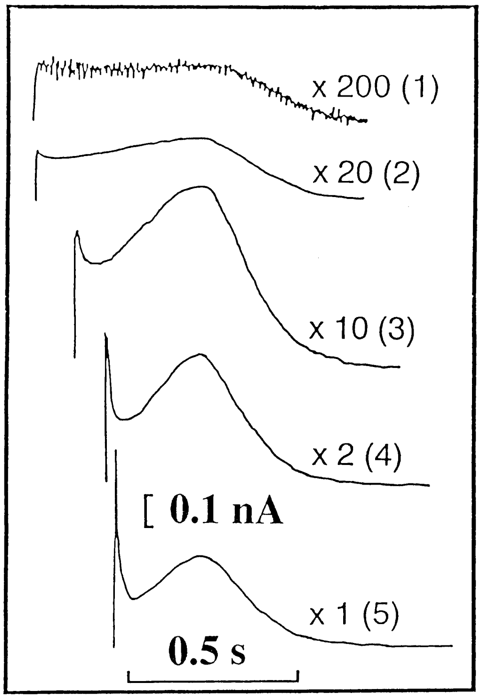

Fig. 1. Laser power dependence of the transient current from pyrene solution in hexane. Laser powers were (1) 0.045, (2)0.15, (3)0.32, (4)0.43, (5)0.82 mJ/pulse.

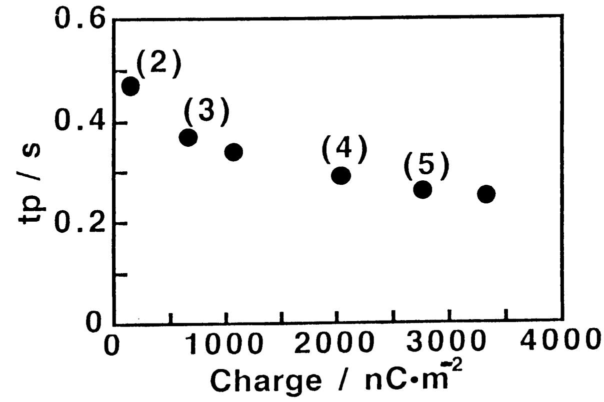

Fig. 2. Dependence of t_p on the produced charge. The numbers on the circles correspond to the numbers in Fig. 1.

References

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