INFLUENCE OF SAMPLE THICKNESS NON-UNIFORMITIES ON THE MODULATED PHOTOCURRENTS IN AMORPHOUS SOLIDS

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INTRODUCTION

One of the experimental techniques being frequently used for determination of the energetic density of states (DOS) in amorphous solids is the method of modulated photocurrents (MPCs). The method consists of photogeneration or photoinjection of excess charge carriers into the sample by sinusoidally modulated light. The phase shift and the amplitude of the resulting photocurrent are measured as functions of modulation frequency. In experimental practice two sample configurations are utilised – with either coplanar or sandwich electrodes. As regards the latter case, the theory of the method has been given in [1-4], and the MPC measurements have been performed in a-Si:H [5-7] and poly-(N-vinylcarbazole) (PVK) [8].
In the case of PVK we have noticed some differences between the courses of measured and calculated MPC phase shifts [8]. These discrepancies occur in the intermediate frequency region, where the carrier-density wavelength is comparable with the sample thickness. For this reason, we have attributed the behaviour to the sample thickness non-uniformities. The aim of the present communication is to give an approximate analysis of this effect.

ANALYTICAL RESULTS

The basic expression, determining the MPC intensity in an ideal sample with smooth, parallel surfaces (cf. figure 1a), has the form of [1-4]

$$\Delta I(\omega) = \Delta I_0 \frac{1-\exp\left[-\mathrm{i}\omega L_0^2\tilde{\Phi}(\omega)/\mu_0 V\right]}{\mathrm{i}\omega L_0^2\tilde{\Phi}(\omega)/\mu_0 V}, \tag{1}$$

where the function

$$\tilde{\Phi}(\omega) = C_t \int_0^\infty \frac{N_t(\varepsilon)\mathrm{d}\varepsilon}{\mathrm{i}\omega + 1/\tau_r(\varepsilon)}. \tag{2}$$

Here, ω is the angular frequency of light modulation, ΔI_0 - the ac-current intensity at illuminated electrode, L_0 - the sample thickness, μ_0 - the microscopic carrier mobility, V - the applied voltage, ε - the energy variable (measured from the edge of conduction band), C_t - the carrier capture coefficient, $N_t(\varepsilon)$ - the trap density per energy unit. $\tau_r(\varepsilon) = \nu_0^{-1}\exp(\varepsilon/kT)$ is the mean carrier dwell-time in the trap (ν_0 denotes

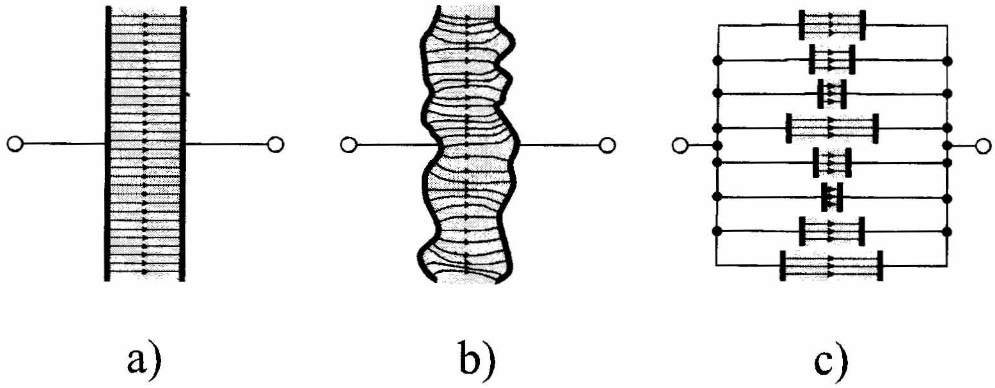

Figure 1. The ideal sample (a), the sample with rough surfaces (b), and its model (c).

the frequency factor, k the Boltzmann constant and T the sample temperature). The above equations correspond to the simplest possible case, when the effects of carrier diffusion, space charge and trap filling are negligible. Moreover, it is assumed that the carriers are photogenerated in a very thin sample layer or photoinjected from the adjacent electrode.
The formula (1) can be rewritten as

$$\Delta I(\omega) = \Delta I_m(\omega)\exp[-\mathrm{i}\varphi_I(\omega)], \tag{3}$$

where the real functions $\Delta I_m(\omega)$ and $\varphi_I(\omega)$ are the MPC amplitude and phase shift, measured in the experiment. Thus, comparing the equations (1) and (3), one can calculate $\Delta I_m(\omega)$ and $\varphi_I(\omega)$ for a given trap distribution.
The rigorous calculation of MPCs in the sample having rough surfaces is difficult because of complicated field distribution (figure 1b). In the present analysis, we treat the real sample as a population of connected in parallel 'subsamples', having different thicknesses (figure 1c). It is assumed that the surfaces of each 'subsample' are flat and parallel as well as that the electric field inside it is uniform.
We shall denote by h(L) the probability distribution of the 'subsample' thicknesses L. Since the MPC is equal to the sum of currents, flowing through separate 'subsamples', the extension of the equation (1) to the considered case has the form of

$$\Delta I(\omega) = \Delta I_0 \int_0^\infty h(L) \frac{1-\exp\left[-\mathrm{i}\omega L^2\widetilde{\Phi}(\omega)/\mu_0 V\right]}{\mathrm{i}\omega L^2\widetilde{\Phi}(\omega)/\mu_0 V}\mathrm{d}L. \tag{4}$$

In general, the above integral can be calculated only numerically. One can, however, obtain the approximate formulae for MPCs in the low- and high-frequency regions.

It is easy to prove that the functions $\omega \operatorname{Re}\tilde{\Phi}(\omega)$ and $-\omega \operatorname{Im}\tilde{\Phi}(\omega)$ increase monotonically with the modulation frequency. Let us denote by

$$\tau_0 = \langle L\rangle^2 / \mu_0 V \tag{5}$$

the free carrier time-of-flight through the sample of average thickness $\langle L\rangle$ (the angle brackets stand for averaging over the distribution h(L)). For low modulation frequencies, when $\omega\tau_0 \operatorname{Re}\tilde{\Phi}(\omega) << 1$ and $-\omega\tau_0 \operatorname{Im}\tilde{\Phi}(\omega) << 1$, the exponential function in the equation (4) may be expanded into a power series. Then, making use of the equation (3), one gets the formulae

$$\varphi_l(\omega) \approx \frac{1}{2} c_1 \omega\tau_0 \operatorname{Re}\tilde{\Phi}(\omega), \tag{6}$$

$$c_1 = \langle L^2\rangle / \langle L\rangle^2,$$

$$\Delta I_m(\omega) \approx \Delta I_0 . \tag{7}$$

Since $\langle L^2\rangle = \langle L\rangle^2 + \sigma^2$, where σ is the standard deviation of distribution h(L), the coefficient $c_1 = 1 + \sigma^2/\langle L\rangle^2$.

For high modulation frequencies, when $-\omega\tau_0 \operatorname{Im}\tilde{\Phi}(\omega) >> 1$, the exponential function in the equation (4) may be omitted. Using the equation (3), one obtains then the formulae

$$\tan\varphi_l(\omega) \approx -\frac{\operatorname{Re}\tilde{\Phi}(\omega)}{\operatorname{Im}\tilde{\Phi}(\omega)}, \tag{8}$$

$$\Delta I_m(\omega) \approx \frac{c_2 \Delta I_0}{\omega\tau_0 \left|\tilde{\Phi}(\omega)\right|}, \tag{9}$$

$$c_2 = \langle L\rangle^2 \langle L^{-2}\rangle .$$

The equations (6)–(9) are almost identical with those referring to the ideal sample (in this case $c_1 = c_2 = 1$). One can conclude that for limiting cases of low and high frequencies the phase shift and amplitude of the MPC are almost independent of the form of h(L), except for multiplicative factors. Therefore, in both frequency regions the sample thickness non-uniformities do not influence significantly the MPCs. This implies that the methods of DOS determination, utilising the MPC course in these frequency ranges (cf. [1-4]), are applicable to the samples with rough surfaces.

NUMERICAL RESULTS

In order to investigate the MPC features in the full frequency domain, we have calculated numerically the photocurrent phase shift and amplitude from the equations (3)–(4). The calculations have been performed for the exponential DOS,

$$N_t(\varepsilon)=\frac{N_{tot}}{kT_c}\exp\left(-\frac{\varepsilon}{kT_c}\right), \qquad (10)$$

with N_{tot} - the total trap density and T_c – the characteristic temperature, determining the DOS decay rate. In this case the functions $\mathrm{Re}\,\widetilde{\Phi}(\omega)$ and $\mathrm{Im}\,\widetilde{\Phi}(\omega)$ can be expressed by simple formulae (see Appendix). Three distribution functions h(L) have been chosen:

the uniform distribution,

$$h(L)=\frac{1}{\Delta L}, \quad L_0-\frac{\Delta L}{2}\le L\le L_0+\frac{\Delta L}{2}, \qquad (11)$$

$\sigma=\Delta L/2\sqrt{3}$,

the Gaussian distribution,

$$h(L)=\frac{1}{\sigma\sqrt{2\pi}}\exp\left[-\frac{(L-L_0)^2}{2\sigma^2}\right], \qquad (12)$$

and the gamma distribution,

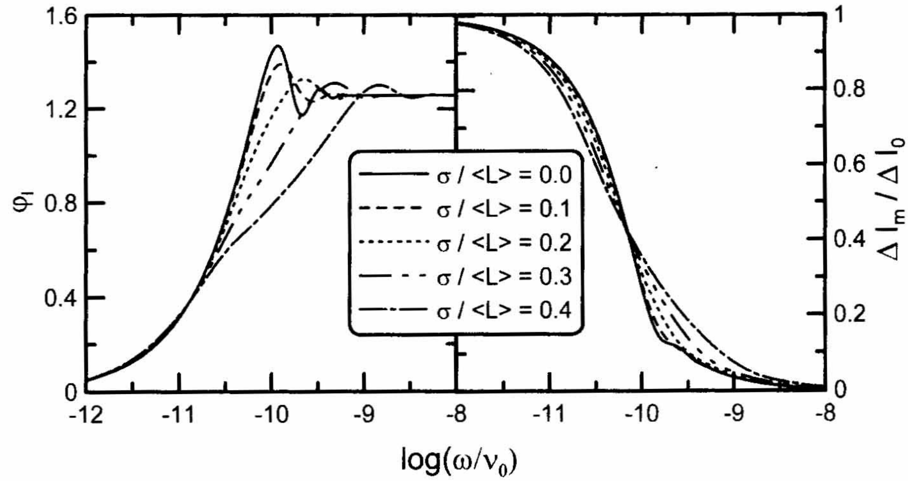

Figure 2. MPC phase shift and amplitude for uniform distribution (11) of 'subsample' thicknesses and exponential DOS (10). The calculations have been carried out for $T/T_c = 0.8$, $\tau_0\nu_0 = 10^{-5}$ and $C_tN_{tot}/\nu_0 = 10^{13}$.

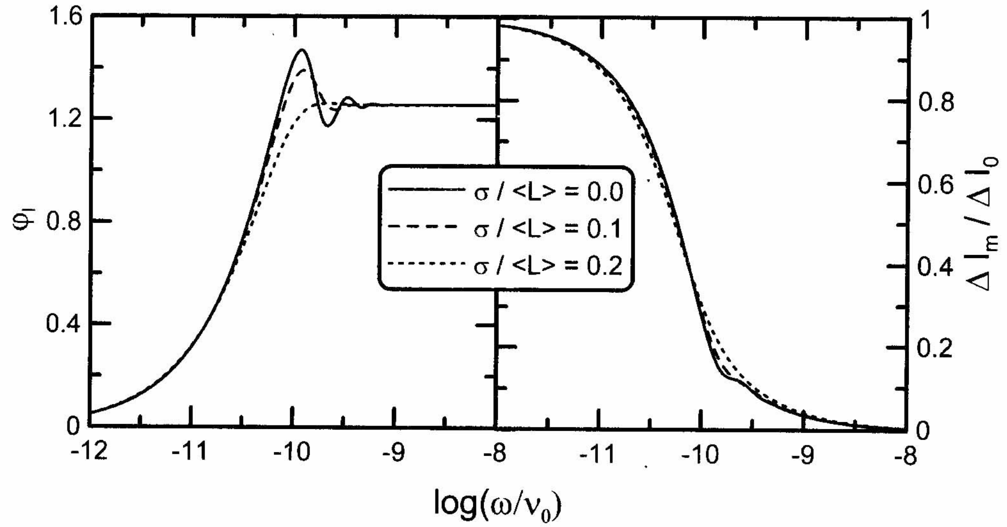

Figure 3. MPC phase shift and amplitude calculated for Gaussian distribution (12). The parameters are as for figure 2.

$$h(L) = \frac{L^{a-1}}{\Gamma(a)(\Delta L)^a} \exp\left(-\frac{L}{\Delta L}\right), \quad a > 0, \tag{13}$$

$$\langle L \rangle = a\Delta L, \quad \sigma = \sqrt{a}\Delta L,$$

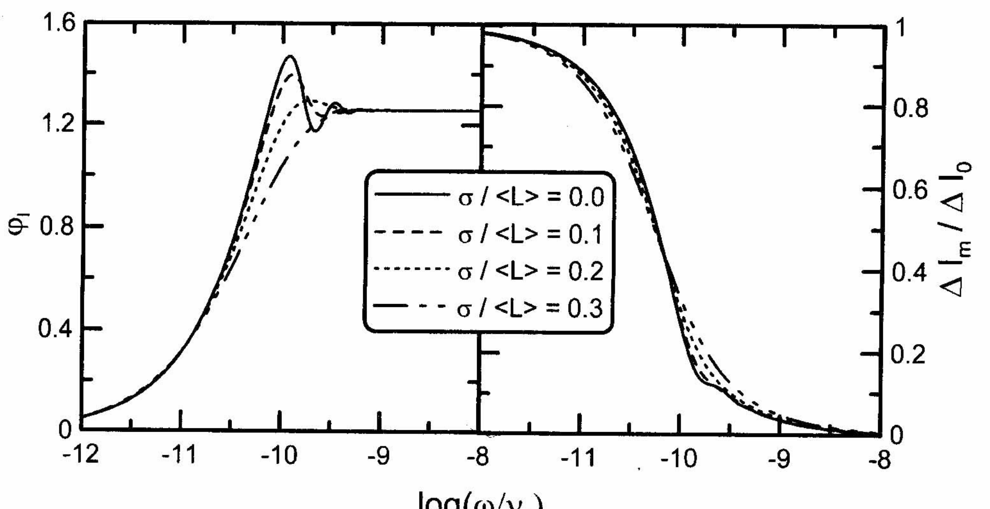

Figure 4. MPC phase shift and amplitude calculated for gamma distribution (13). The parameters are as for figure 2.

where $\Gamma(\ldots)$ is the gamma function. For the distributions (11) – (12) $\langle L \rangle = L_0$. These distributions are symmetrical, contrary to the gamma distribution (13). One can notice that the uniform distribution (11) has a simple interpretation. It corresponds to the sample with flat, but non-parallel surfaces.

The obtained plots of $\varphi_l(\omega)$ and $\Delta I_m(\omega)$, corresponding to several values of the ratio $\sigma/\langle L \rangle$, are shown in the figures 2–4. The value $\sigma/\langle L \rangle = 0$ refers to the case of ideal sample. It can be seen that the curves $\varphi_l(\omega)$ and $\Delta I_m(\omega)$, computed for different distributions with the same relative standard deviation $\sigma/\langle L \rangle$, somewhat differ. Nevertheless, one can state that $\sigma/\langle L \rangle$ is the main parameter determining the influence of the form of distribution on the MPC. With the increase of $\sigma/\langle L \rangle$, the MPC phase shift maxima in the intermediate frequency region move towards higher frequencies and gradually vanish. This indicates that the DOS determination method, utilising the position of phase shift maxima [3], applies only to the high-quality samples, for which $\sigma/\langle L \rangle < 0.1$. The changes of the MPC amplitude with increasing value of $\sigma/\langle L \rangle$ are less distinct. In the low-frequency domain the amplitude slightly decreases and in the high-frequency domain somewhat increases.

CONCLUSIONS

In the communication we have investigated the influence of the sample thickness non-uniformities on the MPCs at sandwich electrode configuration. In the both low- and high-frequency regions the sample surface roughness influences only slightly the MPCs. This can be explained by the fact, that in these frequency ranges the carrier-density wavelength is, respectively, much larger and much smaller than the average sample thickness. In the intermediate frequency domain the MPC phase shift depends significantly on the sample surface quality, whereas the dependence of the MPC amplitude is much weaker. These results are obtained for the oversimplified model of the sample with uneven surfaces. We believe, however, that they apply also, without significant changes, to the real samples.

APPENDIX

According to the equation (2), the functions $\mathrm{Re}\,\tilde{\Phi}(\omega)$ and $\mathrm{Im}\,\tilde{\Phi}(\omega)$ are given by the integrals:

$$\mathrm{Re}\,\tilde{\Phi}(\omega) = C_t \int_0^\infty \frac{N_t(\varepsilon)\,\tau_r(\varepsilon)}{1+\omega^2\tau_r^2(\varepsilon)}\mathrm{d}\varepsilon\,, \tag{A1}$$

$$\mathrm{Im}\,\tilde{\Phi}(\omega) = -\omega C_t \int_0^\infty \frac{N_t(\varepsilon)\tau_r^2(\varepsilon)}{1+\omega^2\tau_r^2(\varepsilon)}\mathrm{d}\varepsilon\,. \tag{A2}$$

In the case of exponential DOS distribution (10) these integrals can be transformed to the form of:

$$\mathrm{Re}\,\tilde{\Phi}(\omega)=\frac{\alpha}{\tau_t\nu_0}\left(\frac{\omega}{\nu_0}\right)^{\alpha-1}\int_{\omega/\nu_0}^{\infty}\frac{x^{-\alpha}\mathrm{d}x}{1+x^2}, \tag{A3}$$

$$\mathrm{Im}\,\tilde{\Phi}(\omega)=-\frac{\alpha}{\tau_t\nu_0}\left(\frac{\omega}{\nu_0}\right)^{\alpha-1}\int_{\omega/\nu_0}^{\infty}\frac{x^{1-\alpha}\mathrm{d}x}{1+x^2}. \tag{A4}$$

Here, $\tau_t = 1/C_tN_{tot}$ denotes the mean free carrier time-of-life in the conduction band and $\alpha = T/T_C$. Since usually $\omega \ll \nu_0$, in the above equations the lower integration limits may be replaced by zero. In order to ensure the convergence of the first integral, the condition $\alpha < 1$ must be then fulfilled. The resulting integrals can be calculated with the aid of the formula [9]

$$\int_0^{\infty}\frac{x^{b-1}\mathrm{d}x}{1+x^c}=\frac{\pi}{c\sin(b\pi/c)},\quad 0<b<c,$$

which gives the equations

$$\mathrm{Re}\,\tilde{\Phi}(\omega)=\frac{\alpha\pi}{2\cos(\alpha\pi/2)\tau_t\nu_0}\left(\frac{\omega}{\nu_0}\right)^{\alpha-1}, \tag{A5}$$

$$\mathrm{Im}\,\tilde{\Phi}(\omega)=-\frac{\alpha\pi}{2\sin(\alpha\pi/2)\tau_t\nu_0}\left(\frac{\omega}{\nu_0}\right)^{\alpha-1}. \tag{A6}$$

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