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Charge diffusion noise in monocrystalline PbS nanoparticle films

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Charge transport in monocrystalline lead sulfide (PbS) nanoparticle films is investigated by current noise measurements. Monocrystalline and single-sized PbS nanoparticles are synthesized via the gas phase and deposited electrostatically onto semiconducting (GaAs) and on isolating (SiN_x) substrates with planar electrode contacts. The particles form inhomogeneous films. Low-frequency current noise of 1-ML-thick films are measured at various voltages, exhibiting diffusion noise characteristics, which indicates a random-walk (diffusion) phenomenon of electrons between the particles. © 2000 American Institute of Physics. [S0003-6951(00)03747-5]

Gas-phase generated PbS nanoparticles may be potential candidates for future electronic devices.¹ In this work the electronic transport properties of thin films of monocrystalline PbS nanoparticles are investigated by low-frequency current noise measurements. Earlier investigation showed a huge increase of photocurrent on metal–semiconductor–metal structures after deposition of a monolayer of nanocrystalline PbS particles.¹ Nevertheless, an explanation of the charge transport mechanism in these structures is missing.

Monocrystalline PbS nanoparticles with a size of 20 nm are synthesized by an evaporation-nucleation method using a differential mobility analyzer.² The particles are deposited statistically onto substrates until a nonhomogeneous film with a mean thickness of one monolayer is reached. The substrates are made of (a) GaAs or (b) GaAs topped with a 200-nm-thick SiN_x layer for insulation. Onto the substrates lateral, interdigitated gold electrodes are fabricated to create eight pairs of finger electrodes with 50 μm fingers width and a spacing of 2.5 μm (Fig. 1).

Low-frequency current noise measurements (with frequency f from 5 Hz to 10 kHz) are performed on wafer level. A current–voltage amplifier (gain 10^{-8} A/V), a voltage amplifier (gain 100) and an analog-to-digital sigma-delta converter with fast Fourier transform conversion by a personal computer are used in order to investigate the current noise spectra. Direct current (dc) voltages up to 6.3 V are applied to the samples.

At voltages higher than 1.5 V diffusion noise, where the

current noise S_i is proportional to $(1/f^m)$ with two characteristic slopes $m=1/2$ and $3/2$ [Ref. 3], can be recognized on GaAs-type samples. Hence, the weighted current noise spectrum fS_i is proportional to $(1/f^{m-1})$ (slopes $1/2$ and $-1/2$) as depicted in Fig. 2. Several of our GaAs-type samples showed this behavior with shifting characteristic frequencies $f_{c,\text{GaAs}}$, defined as the frequency at the maximum of $fS_i(f)$ (Fig. 2). The fS_i characteristic of SiN_x -type samples is depicted in Fig. 3. For voltages higher than 2 V two characteristic frequencies f_{c1,SiN_x} and f_{c2,SiN_x} can be recognized. Initial SiN_x substrates, prior to PbS deposition, show white noise only.

FIG. 1. Sketch of 1-ML-thick film composed of 20 nm PbS nanoparticles deposited onto interdigitated Au electrodes on (a) GaAs substrate and (b) GaAs substrate topped with a 200-nm-thick layer of SiN_x .

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FIG. 2. PbS film on GaAs: Weighted current noise spectra of a film composed of a single monolayer of 20 nm monocrystalline PbS nanoparticles. The substrate is a GaAs wafer with Au electrodes. The inset shows the double-logarithmic dc current–voltage characteristic.

Diffusion noise occurs if there are areas with either different carrier concentration or different carrier mobility. According to the equation given by Shur⁴ we calculate that the Schottky contact for the initial samples would deplete the whole substrate homogeneously. Hence, the substrate does not have regions with different carrier concentration and mobility. Thus, no diffusion noise can occur. PbS nanoparticles between the electrodes create areas with different conductiv-

FIG. 3. PbS film on SiNx: Weighted current noise spectra of a film composed of a single monolayer of 20 nm monocrystalline PbS nanoparticles. The substrate is a GaAs wafer topped with 200-nm-thick SiNx and with Au electrodes. The inset shows the double-logarithmic dc current–voltage characteristic.

ity resulting in different carrier concentration or different carrier mobility. They modulate the charge transport due to the interparticle potential barrier and the particle-substrate interface. Charge carriers are diffusing with a diffusion coefficient D_n between the PbS particles due to thermal agitation. This process is controlled by the interparticle potential barrier which is influenced by the voltage bias. The diffusion coefficient of this random electron walk between the grains can be calculated from the characteristic frequency shift³ of the samples, see Figs. 2 and 3:

$$f_{c,x} = c_{1,x} D_{n,x} = c'_{1,x} \exp[(E_{0,x} + k_{1,x} V)/kT], \quad (1)$$

$$f_{c,x} = c_{2,x} \exp[(k_{1,x} V)/kT], \quad (2)$$

where $f_{c,x}$ is the characteristic frequency for x -type samples (GaAs or SiNx), $c_{1,x}$ and $c'_{1,x}$ are geometry constants (in cm^{-2}), $c_{2,x} = c'_{1,x} \exp(E_{0,x}/kT)$ is a constant (in cm^{-2}) including the geometry and the activation energy $E_{0,x}$ (in eV) at zero bias, $D_{n,x}$ the interparticle diffusion coefficient of the charge carriers (in $\text{cm}^2 \text{s}^{-1}$), kT the thermal energy (in eV), and V the applied voltage (in V). $k_{1,x}$ is the voltage coupling constant (in e) expressing the influence of the voltage on the diffusion coefficient.

We do not have an explanation for the characteristic frequency $f_{c1,\text{SiNx}}$ and further investigation have to be carried out. Interestingly, the fits of $f_{c,\text{GaAs}}$ and $f_{c2,\text{SiNx}}$ according to Eq. (2) show comparable results. The voltage coupling constants $k_{1,\text{GaAs}} = 0.003\,659\, \text{e}$ (correlation coefficient $r^2 = 0.952$) and $k_{1,\text{SiNx}} = 0.003\,693\, \text{e}$ ($r^2 = 0.985$) are almost the same. This we can only explain with the interparticle potential barrier, which is dominantly responsible for determining the diffusion coefficient. The fits of the factors $c_{2,x}$ are $c_{2,\text{GaAs}} = 1677\, \text{Hz}$ and $c_{2,\text{SiNx}} = 1425\, \text{Hz}$. Due to the similarity of the particle films and the geometry, the differences can only be explained by different activation energies $E_{0,x}$ depending on the particle-substrate interface. The effect of higher dc current characteristic by a factor of 1000 in the GaAs-type samples in comparison to SiNx-type samples (insets Figs. 2 and 3) indicates that the main charge transport takes place in the substrate and the surface. If the charge transport would be dominated by the PbS particles film, the dc current would be comparable.

We have studied the charge transport in monocrystalline PbS nanoparticle films. The current noise indicates a random walk (diffusion) phenomenon of electrons between the particles. This means that the electrons are localized on the particles and the random walk is controlled by the interparticle potential barrier which is influenced by the voltage bias. Nevertheless, the main charge transport takes place in the substrate.

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