Microscopic Identification of the Origin of Generation-Recombination Noise in Hydrogenated Amorphous Silicon with Noise-Detected Magnetic Resonance

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(Received 2 September 1999)

Spin-dependent changes in the noise power of undoped amorphous hydrogenated silicon (a-Si:H) are observed under electron spin resonance conditions. The noise-detected magnetic resonance (NDMR) signal has the g value of holes in the valence band tail of a-Si:H. Both the sign of the NDMR signal and the frequency dependence of its intensity can be quantitatively accounted for by a resonant reduction of the generation-recombination noise time constant τ. This identifies hopping in the valence-band tail as the dominant spin-dependent step governing noise in this material.

PACS numbers: 72.70.+m, 73.61.Jc, 76.70.−r

Although the origin of thermal noise [1,2] is well established, the exact microscopic processes leading to nonthermal noise have proven to be elusive. As an example, generation-recombination noise (gr noise) stems from the statistical nature of charge carrier transitions in semiconductors [3]. Thus, noise measurements can reveal important transport parameters, such as the carrier lifetime τ. In addition, trap densities, capture cross sections, activation energies, etc. may be determined. However, an unambiguous, microscopic identification of the states involved in these noise processes, e.g., the dopants or defects acting as traps, remains difficult [4,5].

Moreover, in many real systems the noise power density S_U as a function of frequency f does not follow the simple Lorentzian expected for gr noise due to a single defect level [6,7]

$$S_U^{\mathrm{gr}}(f) \propto \frac{\tau}{1 + (2\pi f \tau)^2}\,. \tag{1}$$

Rather broad, featureless spectra are observed instead, in many cases exhibiting approximately $1/f$ frequency dependencies. Various models leading to such behavior have been considered for different materials, e.g., a distribution of τ [8], fluctuations of chemical structure [9], spatial inhomogeneities [10], filamentary current transport, or hierarchical transport kinetics [11,12]. Even more so than in the case of gr noise, the experimental identification of the corresponding microscopic processes responsible for $1/f$-like noise remains a challenge [13].

In contrast, spin-selection rules governing hopping and recombination processes allow the microscopic identification of paramagnetic states involved in charge transport by electrically detected magnetic resonance (EDMR) [14,15]. For example, consider a recombination process with two paramagnetic initial states. Since the final state has to be a spin $S = 0$ due to the Pauli principle, only those pairs of initial states with $S = 0$ can recombine, those with $S = 1$ cannot. Inducing electron paramagnetic resonance (EPR) leads to a net enhancement of the relative amount of $S = 0$ pairs and to a resonant reduction of the recombination time.

Using amorphous silicon, where spin-dependent effects have been studied extensively, as an example, we show in this Letter that the same spin dependence also leads to resonant changes in the noise properties. gr noise can be identified and its origin successfully linked to a particular microscopic state in this material, i.e., holes trapped in the valence band tail. We call this new experimental method, combining noise spectroscopy with electron spin resonance, noise-detected magnetic resonance (NDMR).

As samples, intrinsic amorphous hydrogenated silicon (a-Si:H) Cr-n^+-i-n^+-Cr sandwich structures are used. The highly doped n^+ regions are 200 nm thick and serve as Ohmic contacts to the 3 μm thick intrinsic layer. Good Ohmic contacts are of crucial importance to avoid contact noise. In order to decrease the resistance and, thus, the RC-time constant, the sample is illuminated with white light of an intensity of 70 mW/cm^2. We are therefore in fact studying the noise of the photocurrent in a-Si:H. For the NDMR measurements, the sample is introduced into the standard TE$_{102}$ cavity of a commercial X band EPR spectrometer working at 9.35 GHz. A constant microwave power of 2 W is employed in all cases.

The noise is measured as voltage fluctuations while a constant current is drawn through the sample [16]. The fluctuations are amplified and fed through a bandpass filter of width BW. A root mean square (rms) device then yields $U_{\mathrm{rms}} = \sqrt{S_U BW}$. Thus, we obtain a time continuous measure of the noise power within BW [17]. Resonant changes ΔU_{rms} of this signal can now be taken to detect the magnetic resonance (NDMR). Since the expected resonant changes of the noise are small, we use magnetic field modulation and lock-in detection. The sensitivity of this new method is inherently limited by the accuracy with which U_{rms} can be measured, given the statistical nature of noise. For an averaging time t, this accuracy is given by [18]

$$\frac{\sigma(U_{\mathrm{rms}})}{\overline{U_{\mathrm{rms}}}} = \frac{1}{\sqrt{2BWt}}\,, \tag{2}$$

where $\sigma(U_{\mathrm{rms}})$ is the standard deviation, and $\overline{U_{\mathrm{rms}}}$ is the average value. The setup used in our experiments operates at the statistical limit (2) within a factor of 2. Equation (2) represents a fundamental restriction for the applicability of NDMR. To stress this point, consider the thermal noise power density [1,2]:

$$S_U^{\text{thermal}}(f) = 4k_B TR\,, \tag{3}$$

with the Boltzmann constant k_B and the temperature T. Resonant changes ΔR in the sample resistance R (as detected in conventional EDMR) will result in resonant changes $\Delta S_U^{\text{thermal}}$ (detected in NDMR) according to

$$\frac{\Delta U_{\text{rms}}}{U_{\text{rms}}} = \frac{1}{2}\,\frac{\Delta S_U^{\text{thermal}}}{S_U^{\text{thermal}}} = \frac{1}{2}\,\frac{\Delta R}{R}\,. \tag{4}$$

The relative signal amplitude, $\Delta R/R$, measured in EDMR, thus determines the minimum relative signal amplitude in NDMR. In a-Si:H, a comparatively large EDMR signal amplitude of $10^{-5} < \Delta R/R < 10^{-4}$ is usually observed at room temperature. Still, in NDMR, at a typical bandwidth $BW = 2$ kHz, measurement times above 10^4 s would be necessary for each value of the external magnetic field to obtain a reasonable signal-to-noise ratio.

A characteristic NDMR spectrum obtained at room temperature is shown in Fig. 1. We find a single resonance at $g = 2.0097$ with $\Delta H_{pp} = 18$ G and a maximum $\Delta U_{\text{rms}}/U_{\text{rms}} = 7 \times 10^{-4}$ or $\Delta S_U/S_U = 1.5 \times 10^{-3}$. The signal is quenching; i.e., the noise is reduced upon resonance. No differences in the line shape were found as a function of filter frequency. For comparison, an EDMR spectrum taken under the same conditions is also shown in Fig. 1 (dotted line). Even taking into account the large fluctuations, a cross talk of EDMR into NDMR can be excluded due to the different line shape of the two signals. More importantly, the relative amplitude of the EDMR signal $\Delta R/R \approx 8 \times 10^{-5}$ is approximately an order of magnitude smaller than the one of NDMR. According to the discussion above, this means that the NDMR is not simply due to changes in the thermal noise [cf. (4)], but that additional processes leading to nonthermal noise are spin dependent.

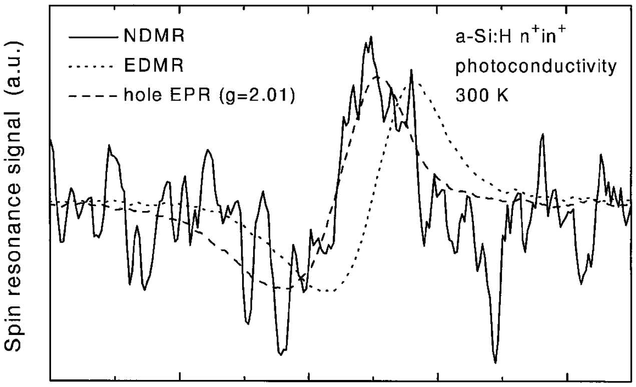

FIG. 1. X band noise-detected magnetic resonance spectrum of an a-Si:H Cr-n^+-i-n^+-Cr sandwich structure (full line) in comparison to the EDMR spectrum under the same experimental conditions (dotted line). The dashed line represents the EPR resonance attributed to holes ($g = 2.01$, $\Delta H_{pp} \approx 17$ G). Both the EDMR and the hole EPR are normalized to the NDMR signal intensity. The good agreement between the EPR hole spectrum and the NDMR spectrum indicates that the resonant changes in noise power density result from transitions involving holes.

We now discuss a simple model for the spin dependence of the widely encountered gr noise processes. The model consistently accounts for both sign (quenching) and frequency dependence of the NDMR signal. It is motivated by the results of Verleg and Dijkhuis [16], who have extensively studied a-Si:H n^+-i-n^+ structures with conventional noise spectroscopy. They propose that gr processes are the origin of the nonthermal noise in this material. For simplicity, we shall, for the present, approximate the experimentally observed, rather broad frequency dependence of the noise power density (Fig. 2, upper part) by an ideal Lorentzian (1) with a well-defined carrier lifetime τ. An estimate for τ can be experimentally obtained from the so-called characteristic frequency $f_{1/f}$, i.e., the frequency where $\partial S_U/\partial f = -1$ [16]. For the illumination intensities applied, $f_{1/f} \approx 8.5$ kHz, or $\tau = \tau_{1/f} \approx 1.9 \times 10^{-5}$ s [19].

To describe NDMR, we shall treat the effect of paramagnetic resonance on the noise power density as a reduction

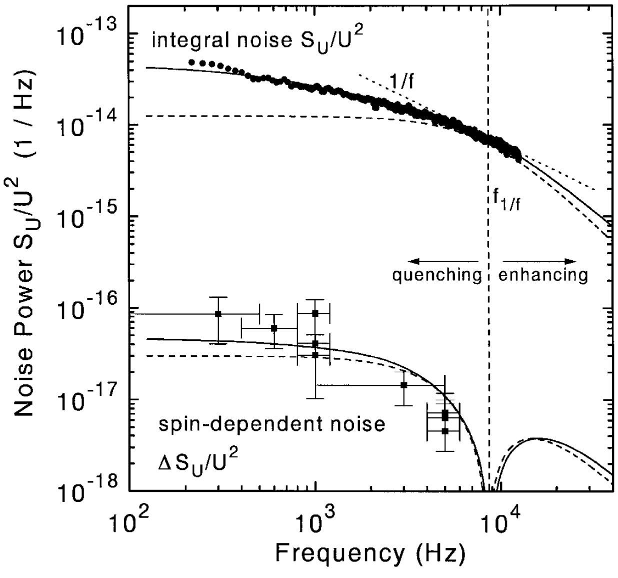

FIG. 2. Upper part: nonthermal noise power density in Cr-n^+-i-n^+-Cr a-Si:H sandwich structures at room temperature. Lower part: resonant change of the noise power density under spin resonance conditions. The horizontal error bars correspond to the 3 dB bandwidth of the filters. The dashed lines represent the ideal S_U^{gr} for a single carrier lifetime $\tau = 1.9 \times 10^{-5}$ s and the corresponding expected ΔS_U^{gr} for a resonant reduction of τ by $\Delta\tau = 4 \times 10^{-8}$. The solid lines give a fit to the integral noise power density and the corresponding resonant change if a distribution of lifetimes is assumed.

of carrier lifetimes as discussed in the introduction. The spin-dependent changes of the noise can then be analyzed as the change in Lorentzian noise power density upon a change of τ by $\Delta\tau$. To illustrate this, we have plotted S_U^{gr} in Fig. 3 as a function of τ for various values of frequency f_{BP} determined by the bandpass center frequency in the experiment. $\tau_{1/f}$ is indicated as a vertical line. The values of the curves for different f_{BP} at $\tau_{1/f}$ constitute the well-known frequency dependence of gr noise power density (cf. Fig. 2, upper part). From the differential change in noise power density with lifetime, $\partial S_U^{\mathrm{gr}}/\partial\tau$, at $\tau = \tau_{1/f}$, as given by the thick lines, the sign as well as the size of the NDMR signal can be obtained. Thus, we are able to deduce the NDMR signal originating from one single gr process for different bandpass filters, i.e., as a function of frequency. For $f_{BP} < f_{1/f}$, a decrease in lifetime will obviously lead to a decrease in noise power density (quenching), whereas, for $f_{BP} > f_{1/f}$, it will result in an increase in S_U^{gr} (enhancing). Quantitatively,

$$\Delta S_U^{\mathrm{gr}} = \frac{\partial S_U^{\mathrm{gr}}}{\partial\tau}\,\Delta\tau \propto \frac{1 - (2\pi f)^2\tau^2}{[1 + (2\pi f)^2\tau^2]^2}\,\Delta\tau\,. \qquad (5)$$

This dependence of the NDMR signal intensity on $f = f_{BP}$ is fitted to the experimental data in the lower part of Fig. 2 (dashed line). The horizontal error bars indicate the bandwidth of the different bandpass filters used. The only free parameter of the fit is the resonantly induced change $\Delta\tau$, which is determined to be $\Delta\tau = (4 \pm 1) \times 10^{-8}$ s. The predicted change in sign of the NDMR signal for $f_{BP} > 7$ kHz could not be verified due to the dominating thermal noise at frequencies above $\approx$10 kHz and the small signal amplitude. Taking into account the rather crude description of the experimentally observed noise by gr noise with a single $\tau_{1/f}$, the model describes the NDMR signal intensity and sign very well. In particular, while S_U changes by about a factor of 3 over the frequency range studied, ΔS_U changes by an order of magnitude, both experimentally and in the theoretical description.

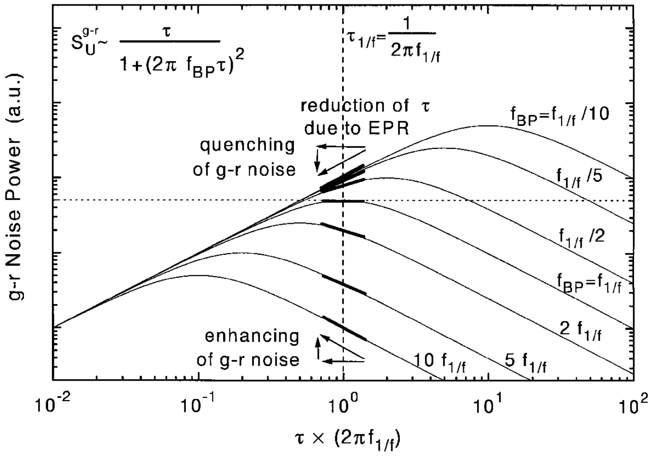

FIG. 3. Generation-recombination noise power density S_U^{gr} plotted as a function of carrier lifetime τ for various bandpass center frequencies f_{BP} used for the measurements. For f_{BP} smaller than the characteristic frequency of the gr noise process $f_{1/f}$, a resonant decrease of the carrier lifetime due to spin resonance leads to a reduction of S_U^{gr}. For $f_{BP} > f_{1/f}$, a small resonant increase is expected.

The full conventional noise spectrum can be accounted for only by a distribution of time constants τ. The solid line in the upper part of Fig. 2 shows the results of a simulation based on a distribution of time constants between 5×10^{-6} and 1×10^{-4} s. This distribution can be incorporated into the derivation of the expected NDMR signal as a superposition of contributions (5) with different τ and relative weight. Using a constant $\Delta\tau = 4 \times 10^{-8}$ s, the frequency dependence of the NDMR intensity, as shown by the solid line in the lower part of Fig. 2, is obtained. It is important to note that while the inclusion of a distribution of τ significantly affects the conventional noise spectrum, the corresponding frequency dependence of the NDMR signal intensity does not change drastically.

The NDMR spectra contain one more important piece of information: the g value and linewidth of the observed resonance, which allow for the microscopic identification of the paramagnetic state involved in the noise process. As already mentioned, such a direct identification is not possible in conventional noise experiments. The EDMR signal of a-Si:H has been extensively studied. It is well established that only two resonances can be observed at room temperature: a dominant resonance attributed to a transition of electrons to dangling bond states (e-db line) and a weak resonance due to the hopping of holes between localized states in the valence band tail (h line) [20,21]. An EDMR spectrum of the sample, taken under the same conditions as in the NDMR measurements, is shown as a dotted line in Fig. 1, normalized to the NDMR signal intensity. As expected, it consists mainly of the e-db line ($g = 2.0059$). Obviously, the agreement between NDMR and EDMR signal is very poor, as mentioned above. In contrast, the somewhat broader h line ($g \approx 2.01$) as obtained from conventional EPR, also normalized to NDMR and shown as the dashed line in Fig. 1, fits the NDMR signal much better. Figure 1 thus clearly shows that the resonant changes of the noise must dominantly be due to the EPR of holes and not of electrons. Therefore, with the help of the microscopic labeling provided by EPR, we have now identified gr processes involving hole states as the dominant spin-dependent process in nonthermal noise of intrinsic a-Si:H.

The relative NDMR signal amplitude $\Delta S_U/S_U = 1.5 \times 10^{-3}$, about 2 orders of magnitude larger than the EDMR hole signal intensity of 3×10^{-5}, points to comparatively strong spin-dependent contributions to the noise. Kaplan, Solomon, and Mott have estimated the maximum EDMR signal intensity to 10^{-1} in the case of weakly interacting initial states [22]. It is interesting to note that the NDMR signal intensity is much nearer to this value than typical EDMR intensities, supporting the notion that in EDMR spin-independent parallel conductivity processes reduce the signal intensity, while spin-independent parallel noise processes seem much less important. This is readily understandable, as the hole mobility in intrinsic a-Si:H is small as compared to the electron mobility ($\mu_e > 10 \times \mu_h$) [23], indicating that holes contribute only weakly to conductivity. Therefore, the resonant change of conductivity (EDMR) due to the hole resonance is small; the EDMR signal is dominated by the e-db transition involving electrons. In contrast, the NDMR experiment indicates that holes localized in the valence band tail do play an appreciable role in the noise properties, whereas resonant changes of the noise power due to electronic transitions are not observed under the current conditions. We have to conclude that gr processes involving electrons have a different, most likely significantly shorter, characteristic lifetime τ, which reduces the noise power (1) and the NDMR signal intensity (5) so that they cannot be detected here. We are therefore left with hopping in the valence band tail both as the dominant gr noise process as well as the spin-dependent transition influencing noise. These findings confirm the conclusions drawn in Ref. [16]. Finally, it can be expected that the NDMR signal intensity should further increase at temperatures below 200 K, since at these temperatures the thermal energy is not sufficient to excite holes above the valence band mobility edge and holes can be transported only via hopping in the band tail [20]. However, flow cryostats are conventionally used to change the measurement temperature in X-band EPR spectrometers. The gas flow leads to a strong increase of the spurious electronic noise for frequencies below 10 kHz, i.e., in the frequency range investigated in NDMR experiments, thus seriously reducing the sensitivity of the setup. Therefore, while NDMR is, in principle, not limited to room temperature, dedicated cryostats contributing a negligible amount of additional noise would be required for such experiments, e.g., cold finger cryostats [24].

In conclusion, we have shown that electron paramagnetic resonance leads to resonant changes in the electronic noise of semiconductors. This allows the direct identification of a defect state dominating nonthermal noise for the first time. A phenomenological model, based on the power density of ideal generation-recombination noise, can well account for the measured sign and amplitude of the NDMR signal. It can be hoped that an extension of the experiments reported here to other semiconductor materials and actual devices will provide new insight into the microscopic origin of electronic noise processes.

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