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Transient measurements with an ultrafast scanning tunneling microscope on semiconductor surfaces

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We demonstrate the use of an ultrafast scanning tunneling microscope on a semiconductor surface. Laser-induced transient signals with 1.8 ps rise time are detected. The investigated sample is a low-temperature grown GaAs layer placed on a sapphire substrate with a thin gold layer that serves as a bias contact. For comparison, the measurements are performed with the tip in contact to the sample as well as in tunneling above the surface. In contact and under bias, the transient signals are identified as a transient photocurrent. An additional signal is generated by a transient voltage induced by the nonuniform carrier density created by the absorption of the light (photoember effect). The transient depends in sign and in shape on the direction of optical excitation. This signal is the dominating transient in tunneling mode. The signals are explained by a capacitive coupling across the tunneling gap. © 1998 American Institute of Physics. [S0003-6951(98)03613-4]

Until now photoconductively gated tunneling microscopes have been used for measuring voltage pulses propagating on transmission lines, implying that all measurements were limited to metallic surfaces.¹⁻³ In this letter we extend the use of an ultrafast scanning tunneling microscope (USTM) to the investigation of laser-induced transients measured directly on a semiconductor. Details of the instrument are described in Ref. 3. In order to ensure a fast transient and to test the temporal resolution of the instrument, we use low-temperature (LT) grown GaAs with a subpicosecond carrier life time. The sample is prepared as follows. A 1.5- μm -thick LT GaAs layer is grown by molecular beam epitaxy at 250 °C on a GaAs substrate on top of a sacrificial AlAs layer. The sample is treated by rapid thermal annealing at 600 °C for 1 min. By dissolving the sacrificial layer the LT GaAs layer is lifted off and placed on a sapphire substrate with a 6 nm Ti and 40 nm Au layer. The sample is then annealed at 360 °C to ensure contact between the Au layer and the LT GaAs. The setup is sketched in Fig. 1. The excitation wavelength is 820 nm and the absorption coefficient is measured to be 1.5 μm^{-1} .⁴ This implies that 90% of the incident light is absorbed and that the initial photoinduced carrier concentration is strongly nonuniform in the light propagation direction. The pump beam (on the sample) and probe beam (on the tunneling probe) are modulated around 600 kHz with a difference frequency of 1.4 kHz. The lock-in amplifier is locked to the difference frequency. The fundamental modulation frequencies are chosen high enough so that thermal effects do not follow the modulation.⁵ For smaller modulation frequencies these effects are detected as a huge offset of the signal and make measurements in the tunneling mode impossible.

In addition to measurements with the tip in the tunneling regime, we performed measurements with the tip in direct contact with the sample. It has to be pointed out that these measurements are performed in air and although it has been

observed that the buildup of oxide is delayed on LT GaAs compared to SI GaAs,⁶ the semiconductor surface is expected to be covered with an oxide layer. By bringing the tip in contact repeatedly and measuring on the same spot the oxide is removed and the photoinduced current increases from an initial 1.5 nA to approximately 8 nA (for a bias voltage $V_b = -7$ V). The current-voltage (I - V)-curve with the average current induced by the mode-locked laser source is shown in Fig. 2. The amplitude of the current strongly depends on the alignment of the laser spot with respect to the tip position. But consistently, the I - V -curve shows the characteristics of two asymmetric back-to-back Schottky barriers. The asymmetry is consistent with three effects: (i) a residual oxide layer between tip and sample, (ii) the smaller contact area of the tip-sample contact, and (iii) the lowering of the Au-LT GaAs contact barrier due to the stronger illumination on this side of the sample.

The top three curves in Fig. 3 show the transient, delay-dependent signals measured with the STM tip in contact. In this case, the setup can also be regarded as two photoconductive switches in series with the first one on the sample excited by the pump beam and the second one on the probe excited by the probe beam. For high bias ($|V_b| > 3$ V) the

FIG. 1. Experimental setup for back side excitation of an LT GaAs layer and detection of transient field changes with a gated tunneling tip from the front side.

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FIG. 2. Average photocurrent for back side illumination with the tip in contact to the LT GaAs layer.

amplitude and sign of the signals follow the photoinduced average current in Fig. 2. The full width at half maximum (FWHM) for the fastest signal is 2.2 ps. The response time is limited by the temporal resolution of our instrument, indicated also by the symmetric rise and fall times of the signal. The signal can be explained as a transient photocurrent through the LT GaAs layer. The sign follows the voltage applied across the LT GaAs layer. The response time is determined by the carrier lifetime in the investigated sample and the photoconductive switch gating the signal in the tunneling circuit. A carrier lifetime of 400 fs has been determined by differential transmission and a photoconductive switch response time of 800 fs by electro-optic sampling

FIG. 3. Transient signals for back side illumination with the tip in contact (top three curves) for different sample bias voltages and with the tip tunneling (bottom black curve). The two bottom curves compare the signal in tunneling mode and contact mode at the same position under the same bias conditions $V_b = -7$ V $I_{dc}^{contact} = 8$ nA, $I_{dc}^{tunnel} = 0.05$ nA. Both curves are normalized, the unnormalized amplitude of the initial contact mode signal is six times higher than the tunneling mode amplitude.

measurements.⁷ Two observations deviate from the simple picture considering a photocurrent driven by the applied field: the signal does not vanish for $V_b = 0$ V and for $V_b > 0$ V the initial positive peak is followed by a small negative component.

As shown in Fig. 2, the average photocurrent at $V_b = -7$ V in contact is around 8 nA. In order to obtain a reasonably stable tunneling current for operating the instrument in constant current mode, we have to select an approximately 100 times smaller tunneling current. We choose a tunneling current of 0.05 nA.

In the lower part of Fig. 3 the signal in tunneling mode is shown and compared in the contact mode for $V_b = -7$ V. In tunneling the amplitude drops by a factor of 6 while the signal shape does not change significantly; the FWHM is marginally longer for the tunneling measurement. We find that the signal sign and amplitude is independent on V_b . In addition, the sign is the same for the $V_b = 0$ V measurement. The high tunneling gap resistance ($R_t \sim 100$ G Ω) means that there is only a negligible voltage applied across the LT GaAs layer with a resistance of $R_{LT} \sim 1$ G Ω . In this sense, the measurements with $V_b = 0$ V and the tip in contact are comparable to the measurements with the tip in tunneling.

If we retract the tip out of the tunneling regime, the transient signal amplitudes drop but the signal does not disappear. As for the transmission line measurements,⁸ this is a strong indication that the observed signal is due to capacitive coupling. However, the resemblance of the contact measurement in the tunneling measurement is completely contrary to the signal shapes of voltage transients on transmission lines measured with the same tip.⁹ In that case the transient signal in tunneling mode represented the derivative of the contact measurement. Nevertheless, both observations can be explained by capacitive coupling. The main difference is that the time constant of the transmission line impedance ($Z \sim 100$ Ω) and gap capacitance ($C \sim 0.5$ fF) $Z \cdot C$ is on the order of 50 fs, whereas the time constant of the LT GaAs resistance (minimum 10 k Ω) and gap capacitance is on the order of a few 100 ps. In this case the signal is first integrated by the tunneling junction and then differentiated at the transition from the tunneling junction to the impedance formed by the tunneling wire and the transmission line on the probe.

In this specific, highly insulating sample we do not expect to observe a contribution from the surface field. The sign of the signal is consistent with an explanation that is independent of the doping type and the contact characteristics, i.e., the nonuniform carrier concentration induced by the laser excitation. In this case the initial concentration gradient (high concentration on the Au side, low concentration on the tip side) and the higher mobilities of the electrons (compared to the holes) lead to a net electron current towards the tip interface (Dember effect).¹⁰ The sign of the current is equivalent to a negative bias voltage. In other words, for high bias contact mode we observe a drift determined current and for tunneling mode and zero bias contact mode the transient current is driven by diffusion.

We test this model by illuminating the LT GaAs layer from the front (or tip) side via a mirror. Accordingly, the injected carrier density gradient changes sign and the observed signal is expected to change sign as well. Figure 4

FIG. 4. Transient signals for front (tip) side illumination with the tip in contact (top three curves) for different sample bias voltages and with the tip tunneling (bottom curve). The bias voltages are indicated in the graph, $I_{dc}^{tunnel} = 0.05$ nA.

shows a comparison of the contact measurements (at the top of the graph) with the tunneling measurement. Again, the biased contact measurements can be interpreted as a transient photocurrent and serve as a reference for the tunneling measurement. As for the back side illumination, the zero bias contact measurement and the tunneling measurement are similar in shape. The main contribution of the signal in tunneling mode now has a positive sign, as opposed to the signal measured in the back side illumination (Fig. 3). The signal shape, however, is more complicated with a delayed negative contribution in the tunneling mode as well as in the contact mode with zero or positive bias. The transient in tunneling mode has a 1.8 ps (10%–90%) rise time and a 1.2 ps FWHM. Certainly, the front side illumination is more versatile and does not require a transparent substrate, but the excitation geometry leads to a more complicated initial carrier density profile. The light is incident under an angle of

approximately 50° and the tip casts a shadow. For the front side illumination we still expect the main carrier gradient to point away from the tip leading to an electron diffusion away from the tip. Due to the shadow of the tip and the angle of incidence there are carrier gradients which can lead to a current in the opposite direction, possibly with a delay. In contrast to the tunneling mode measurements, the measurements with an applied voltage in contact are similar for both excitation geometries. The transient signals are dominated by drift of the carriers in the applied field and are less dependent on the carrier profile than in the diffusion dominated measurements without bias.

In summary, the transient signals in contact with a voltage applied across the LT GaAs layer are identified as a conductance increase due to laser generated carriers. Without an applied voltage the signal can be explained by nonuniform carrier generation and the buildup of a Dember field. The same mechanism causes the signal in tunneling mode. In tunneling the signals resemble the unbiased contact case as in this case the tunneling resistance is too high to apply a noticeable voltage to the sample. In contrast to the transmission line measurements the tunneling measurements do not show a derivative of the tunneling measurements. In both cases the signals can be explained by a signal picked up by the gap capacitance. The spatial resolution should in this case be determined by the tip radius with an ultimate resolution limit on the order of 10 nm. The tip-side illumination opens the possibility of time-resolved transient field or current measurements of small and closely spaced semiconductor structures.

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