Ultrafast carrier dynamics around nanoscale Schottky contacts studied by femtosecond far- and near-field optics

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Far- and near-field femtosecond pump–probe spectroscopy has been used to study ultrafast carrier dynamics in matrices of nanometer-scale tungsten disks embedded in GaAs. These studies reveal the dynamics of carrier transfer from a semiconductor into embedded metal clusters in the presence of Schottky contacts and built-in electric fields. The carrier transfer involves transport towards and trapping into the tungsten disks. We find picosecond time constants at higher carrier densities when the built-in field is screened, allowing for efficient carrier transport. Near-field measurements reveal the spatial variation of carrier dynamics. The spatially averaged dynamics can be controlled by the tungsten disk spacing. [S0003-6951(00)04147-4]

Metal-semiconductor composite materials have found many applications in electronics and optoelectronics. For example, annealed low-temperature (LT) grown GaAs, which contains metallic As precipitates,[1] is widely used in device fabrication due to its high resistivity and ultrafast carrier trapping times.[2,3] Another promising metal-semiconductor material consists of lithographically fabricated nanoscale metal disks embedded in GaAs.[4] The resistivity of these structures can be controlled by the disk spacing and semi-insulating behavior can be obtained.[5] Annealed LT-GaAs and metal-disk structures have in common that buried Schottky contacts are formed at the semiconductor-metal interfaces.[1,5] Hence, besides being interesting on their own right, artificial metal-disk structures can serve as a model system for annealed LT-GaAs, e.g., to obtain a deeper understanding of carrier trapping into buried metal clusters[6] which form Schottky contacts. So far, no femtosecond-resolved studies of carrier dynamics in the metal-disk structures have been reported.

In this letter, we report femtosecond pump–probe studies of carrier dynamics in noncontacted nanometer-size tungsten (W) disks embedded in GaAs. Both spatially averaging far-field and spatially resolved optical near-field measurements have been performed. These experiments yield insight into the carrier transfer from the semiconductor into the W disks in the presence of Schottky contacts and built-in electric fields. The transfer involves carrier transport towards and trapping into the metal. We find that these processes occur on picosecond time scales at higher optically excited carrier densities. In this regime, the optically excited carriers can screen the built-in electric field around the Schottky contacts to allow for fast carrier transport towards the metal. Smaller disk spacings result in shorter time constants in far-field experiments. Thus, control of the disk spacing allows one to tailor the structures for ultrafast applications. Temporally and spatially resolved pump–probe measurements with a femtosecond near-field scanning optical microscope (NSOM)[7–10] confirm the conclusions drawn from the far-field experiments.

The investigated structure consists of a 500 nm GaInP layer as mechanical support at the bottom, followed by an absorbing region with (in this order) an 80 nm GaAs layer, W disks with ~80 nm diameter and a thickness of 20 nm, and another 20 nm thick GaAs layer. A 20 nm GaInP protection layer has been grown on top to avoid the formation of surface traps. The structure has been fabricated by overgrowth of the W disks which have been produced by electron beam lithography and subsequent liftoff. The GaAs is *n*-doped (electron density $\sim 10^{16}$ cm^{-3}) and Schottky contacts are formed at the W/GaAs interface.[5] Different rectangular W disk patterns with disk spacings of 0.3, 1, and 2 μm have been defined. The structure described above is obtained after etching off opaque layers to allow for transmission experiments. The thin top and active layers ensure good spatial resolution in NSOM measurements.

Both far-field (FF) and near-field (NF) degenerate pump–probe measurements are performed at room temperature with 100 fs pulses, centered at a photon energy of 1.46 eV, from a 100 MHz mode-locked Ti:sapphire laser. The sample is excited from the bottom side with a pump beam of ~10 μm diameter. The pump induced transmission changes are probed from the top side. The probe pulses are either sent

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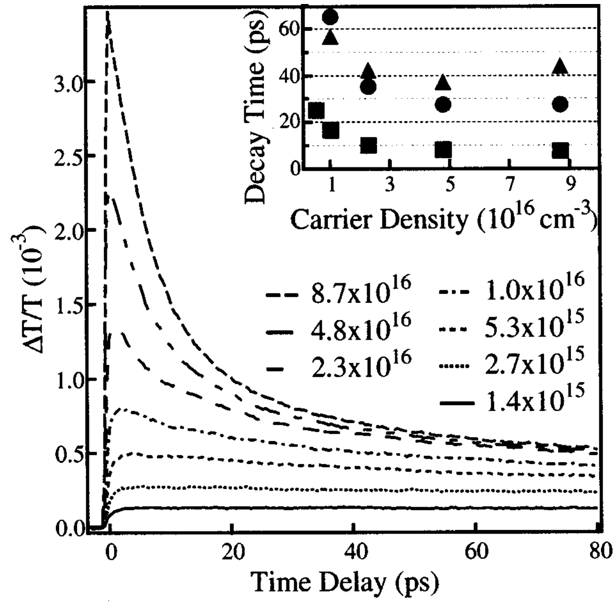

FIG. 1. Far-field data: differential transmission traces from the sample region with 0.3 μm W-disk spacing for different carrier densities (in cm^{-3}). Inset: decay times vs carrier density for different disk spacings: 0.3 μm (squares), 1 μm (circles), 2 μm (triangles).

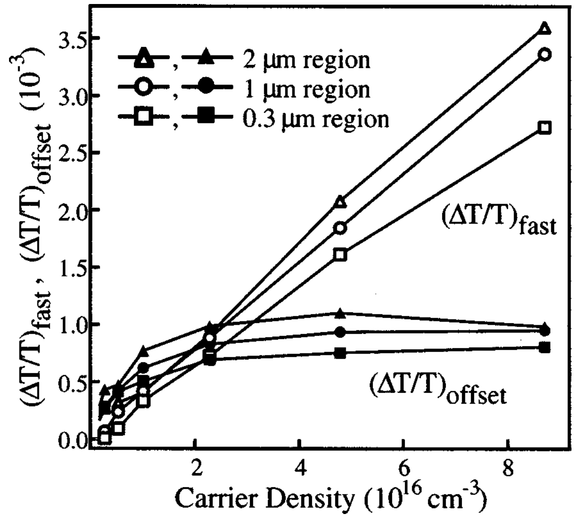

FIG. 2. Amplitude (open symbols) and offset (filled symbols) of far-field pump–probe traces obtained from exponential fits vs carrier density for different disk spacings.

through a cleaved fiber (spot diameter on the sample 4 μm: far-field) or through a metal-coated NSOM fiber tip with an aperture <250 nm (near-field). To ensure high temporal resolution, the probe pulses are precompensated with a prism pair setup in front of the fiber. A detailed description of the femtosecond NSOM can be found in Ref. 7.

Figure 1 shows FF pump–probe traces obtained from the pattern with 0.3 μm disk spacing at different optically excited carrier densities. At low carrier densities, the differential transmission signal $\Delta T/T$ is essentially constant over 80 ps. In contrast, above a threshold carrier density, we observe a fast decay within the first 20 ps, followed by a contribution that decays very slowly. The transition from the quasiconstant signal to the decaying one occurs at a carrier density of approximately 10^{16} cm^{-3}, equal to the doping level. Decay times τ are extracted fitting the pump–probe traces to $(\Delta T/T)_{\text{offset}}+(\Delta T/T)_{\text{fast}}\, e^{-\Delta t/\tau}$ (Δt time delay). The offset $(\Delta T/T)_{\text{offset}}$ describes the long time behavior and is assumed to be time independent in the time window of observation; $(\Delta T/T)_{\text{fast}}$ is the amplitude of the time dependent signal contribution. The decay times τ are shown in the inset of Fig. 1 versus carrier density for the three different disk spacings. We observe a faster decay for smaller disk spacings. This result indicates that the disks are responsible for the fast decay but not intraband relaxation processes, which should not depend on the disk spacing. Moreover, the inset demonstrates that the decay time can be controlled by the disk spacing and can be as short as 10 ps.

Figure 2 plots $(\Delta T/T)_{\text{fast}}$ and $(\Delta T/T)_{\text{offset}}$ versus carrier density for the three different disk spacings. One can clearly see that the amplitude $(\Delta T/T)_{\text{fast}}$ substantially contributes to the pump–probe signal only above the threshold density of $\sim 10^{16}$ cm^{-3}. Above this density, the amplitude $(\Delta T/T)_{\text{fast}}$ increases linearly with carrier density. In contrast, the offset is constant at higher carrier densities.

In the following, we assume that the pump–probe traces directly reflect the carrier dynamics in the GaAs[11] in the presence of the Schottky contacts and built-in electric fields. Before optical excitation, the built-in fields in the depletion regions of the Schottky contacts point to the W disks, i.e., the bands are bent upwards.[5] At optically excited carrier densities smaller than the doping density, any changes in the space-charge region can be neglected and the built-in fields and the band bending persist. Thus, the transport of optically excited electrons towards the W disks is suppressed since diffusion towards the W disks and drift away from the disks cancel each other. As a consequence, most of the electrons remain in the GaAs where they recombine on a nanosecond time scale.[12] This slow process is observed in the pump–probe traces since electrons yield the major contribution to the signal for detection close to the band edge.[13]

We note that the holes in the depletion region move towards the W disks driven by the built-in field. If the optically excited carrier density exceeds the doping density, a substantial screening field builds up as the electrons and holes are separated. The total field is decreased and the band bending is diminished.[14] Both electrons and holes can then diffuse towards the W disks where they are trapped and recombine. We attribute the fast decay seen for initial optically excited carrier density $>10^{16}$ cm^{-3} to this effect. Once the carrier density has decreased to $\sim 10^{16}$ cm^{-3}, the built-in field is restored. Then electron transport towards the disks is again suppressed, reflected by the offset $(\Delta T/T)_{\text{offset}}$ of the pump–probe data. These conclusions are strongly supported by the density dependence of the data in Figs. 1 and 2. We like to emphasize that the decay time τ includes the dynamics of diffusion towards the disks as well as trapping and recombination at the disks.

Additional insight into the carrier dynamics is obtained from near-field measurements, which support our reasoning. Figure 3(a) shows a two-dimensional image of the differential transmission $\Delta T/T$ taken at $\Delta t = 15$ ps in the 1 μm disk region at a carrier density $\sim 9\times 10^{16}$ cm^{-3}. The disk pattern is clearly visible. The contrast is only due to variations in the differential transmission but not due to topographical effects

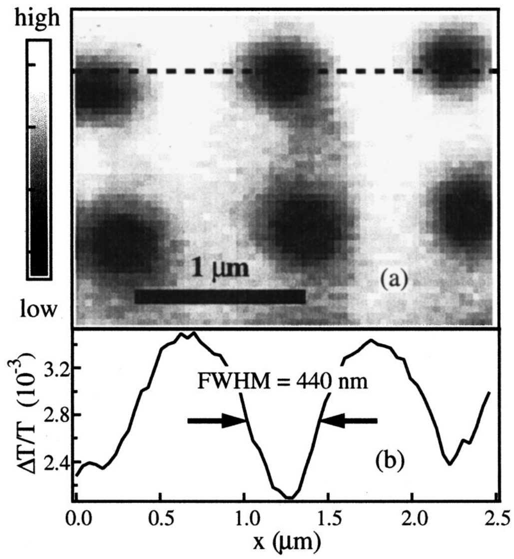

FIG. 3. (a) Two-dimensional optical near-field image of the differential transmission at time delay $\Delta t = 15$ ps and carrier density $\sim 9 \times 10^{16}$ cm^{-3}; (b) line scan along the dashed line in (a), FWHM: full width at half maximum.

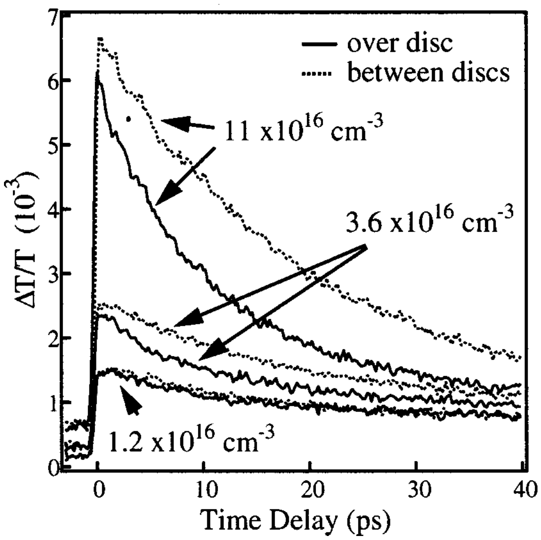

FIG. 4. Near-field differential transmission traces taken over (solid lines) and between the disks (dashed lines) at different carrier densities.

since the simultaneously measured topography does not show any pattern. In the line scan of Fig. 3(b), a reduced signal is observed in a region around a W disk with a full width at half maximum of 440 nm. Given the spatial resolution of 250 nm, we conclude that the carrier density is reduced in a region with a diameter of ~360 nm around the disks (here Gaussian functions have been assumed). Such a carrier density profile is characteristic of diffusive transport towards a trapping center.[8]

The image is complemented by the NF pump–probe traces of Fig. 4, which have been taken either over or between the W disks. No significant difference is observed between these two positions at low optically excited carrier densities. The signals mainly show a long living contribution reflecting recombination, as already seen in the FF data. Faster transients are only observed at higher carrier densities. In this regime, the differential transmission decays faster over the disk (time constant 11 ps) than between the disks (time constant 20 ps). The different decay times are in line with the spatial variation of the carrier density in Fig. 3 and confirm that diffusive transport towards a trapping center occurs[8] at higher carrier densities when the built-in electric field is screened. Thus, the NF pump–probe data strongly support our interpretation that the W disks act as fast trapping centers at higher carrier densities.

The faster decay closer to the W disks is also the reason for the dependence of the decay time on the disk spacing in FF measurements, as shown in the inset of Fig. 1. For larger disk spacing, the spatial average measured in FF experiments contains a larger contribution from regions with a slow decay, giving rise to a longer decay time.

In conclusion, we have presented far-field and near-field pump–probe studies of ultrafast carrier dynamics in GaAs with embedded nanometer-size W disks. These studies shed new light on the transfer of carriers from semiconductors to embedded metal particles in the presence of buried Schottky contacts. The results should be applicable to other metal-semiconductor composite materials.

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