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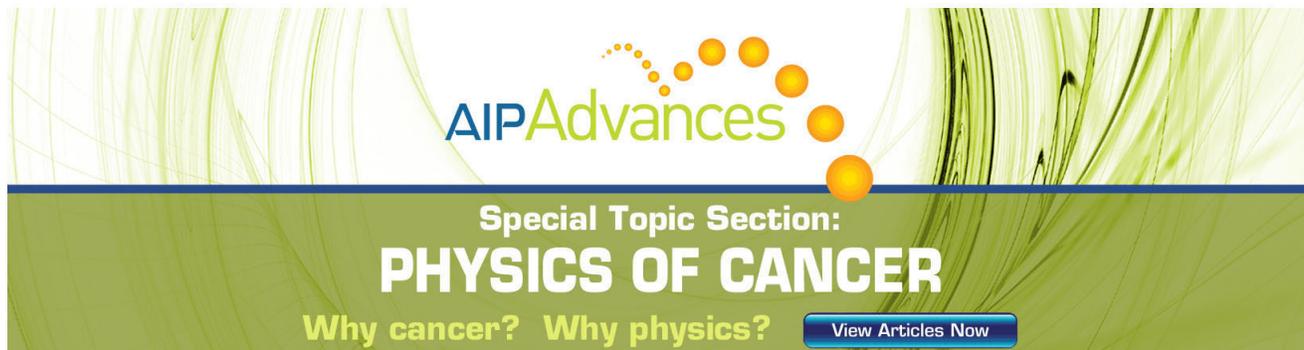
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Field emission measure of the time response of individual semiconducting nanowires to laser excitation

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A simple technique is explored to determine the temporal photo-response, τ , of individual semiconducting SiC and Si nanowires (NWs), with a high time resolution. Laser-assisted field emission (LAFE) from the NWs is first shown to be highly sensitive to continuous laser illumination. Pulsed illumination is then combined with measurements of the total energy distributions to determine τ which were rather large, 4–200 μ s. The time response scaled roughly with the square of the NWs length and could be attributed to laser-induced heating. LAFE is thus a new tool for quantifying rapid thermo-optical effects in such nano-objects. © 2011 American Institute of Physics. [doi:10.1063/1.3627168]

Photo-field emission or laser-assisted field emission (LAFE) has been studied for both fundamental physics^{1,2} and within the perspective of developing new photocathodes.^{3–5} The potential for optically controlled rapid field emission (FE) sources⁴ is particularly attractive for applications such as microwave tubes⁶ and time-resolved electron microscopy.⁷ In the interesting case of properly prepared semiconducting (SC) emitters, FE can be highly sensitive to light for both p-type and high resistivity n-type samples.^{8,9} This high sensitivity is associated with a current saturation in the FE I/V curves and voltage drops within the SC. Though the emission at low current follows the Fowler-Nordheim (FN) behavior for metallic emitters and is insensitive to light, the supply of mobile carriers at higher current can be insufficient for full screening and the electric field then penetrates into the SC creating a band bending and a high resistance depletion region to considerable depths associated with strong voltage drops. The dependence of FE with light (and temperature T) is because the depletion zone is highly sensitive to the generation of free carriers.^{8,9}

These effects have been demonstrated for manually-fabricated tips^{2,9} and for Si tip arrays fabricated from doped Si wafers by photolithography.^{3,4} It is timely to explore these effects in SC nanowires (NWs) because they are attracting enormous attention these last years. Understanding the optical response of such NWs is thus important for a wider community interested in other photonic devices such as photodetectors, solar cells, etc.¹⁰ Our previous studies on FE from nominally undoped SiC NWs,¹¹ and more recent studies on Si NWs,¹² revealed strong nonlinearities in the FN plots with T -dependent saturation currents as predicted by theory.⁸ The drops were determined by the total energy distributions (TEDs) of the emitted electrons which, when combined with the measured total current, becomes a type of two point transport measurement.¹³ In the saturation region, measured voltage drops along the NWs reached up to several

hundreds of volts^{11,12} and were T -dependent. These measurements allowed us to identify the carrier transport mechanisms¹⁴ in our SiC NWs, which is difficult using only FN plots as measured by most authors (e.g. Refs. 3 and 4). An important point in our work is that we showed a clear SC FE effects for mass-produced NWs.

Here, we extend these measurements to determine the time-resolved photo-response of SiC and Si NWs with LAFE. The scientific question was to identify the mechanism for the photo-response which combines the complexity of photoconductivity with FE. An experimental problem was that the emitted currents were very small (pA to sub nA) and they had to be measured at high frequency. The TEDs permit to solve this problem because the voltage drops vary as a function of laser illumination (Figure 1(a)), similar as with T .^{11,14} It turns out that this differentiation has no limitation in time resolution, being similar to synchronous measurements. Note that the emitted electrons originate from the conduction band in Figure 1(a). It is possible that they come from the surface states which would modify somewhat the schematic. However, this modification would be a minor perturbation to the band bending and position of the TEDs. It would have no bearing on the thermal response time, which does not depend on the absolute position of the spectra.

FE studies were carried out on individual SC NWs attached by conductive carbon glue to tungsten tips (Figure 1(b)). Our experimental setup for the FE measurements was described previously.^{11,14} It includes a hemispherical energy analyzer to measure the TEDs. We performed optical measurements with both static and dynamic illumination using an Ar laser to investigate the response time. The laser pulses for dynamic measurement were carried out using an electro-optical modulator (Pockels cell) at frequency from DC up to 20 MHz. Laser light was focused onto the field emitter with a standard setup of spatial filter, beam expander, and focusing lens mounted outside the vacuum chamber in front of a viewport. The laser beam was perpendicular to the W tip/NW axis and polarized parallel to this axis. The diameter of the

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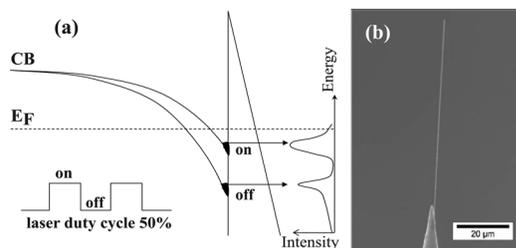


FIG. 1. (a) Simplified schematic of the band and potential profile showing the effect of laser modulation where the emitted electrons originate from the conduction band (see Refs. 11 and 14 for more details). Only the bottom of the conduction band is shown. E_F represents the Fermi level of the support tip and the position of the TEDs at very low currents. Two typical TEDs are depicted for both laser on and laser off in these experiments. (b) SEM image of a SiC nanowire attached to a tungsten tip with diameter $\Phi = 100$ nm and length $L = 85$ μm .

focal spot was typically $\simeq 15$ μm . Measurements when focusing on the bare W tip showed negligible current changes or temperature rises even at full laser intensity (250 mW), which was largely above those used for the NW measurements. The absolute light intensity falling on the NW could only be approximately estimated and most measurements were relative. This is because of the uncertainty in the focus and the positioning along the length (L) of the longest NWs and the necessity to estimate diffraction effects. As well much of the light is transmitted. We estimate our minimum to maximum radiating power densities to be 0.5–50 kW/cm^2 .

The effect of light on the FN plots (i.e. $\log(I/V^2)$ vs $1/V$) for SiC NWs ($E_{\text{gap},300\text{K}}(\text{SiC}) = 2.33$ eV) is shown in Figure 2. Continuous illumination ($\lambda = 514$ nm, $E = 2.41$ eV) and different light intensities were used. The metallic-like FN and the saturation regions are clearly visible. However, the slopes of the I-V curves measured here for the saturation regime are quite large in comparison to the theory. We have reported previously¹⁴ that this can be attributed to the Poole-Frenkel effect assured by energy-trap levels in the band gap in the depletion region. This reduces the response to the laser. In Figure 2, the current increased up to 11 times under illumination in the saturation regime which is orders of magnitude more than we measured for the bare W tip.

In Figure 3, we show series of TED spectra for a SiC NW ($L = 85$ μm) (a) and a Si NW ($L = 20$ μm) (b). The spectra measures the Fermi level at the end of the NW with respect to

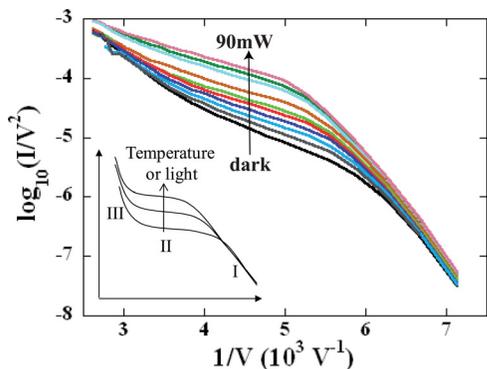


FIG. 2. (Color online) Typical Fowler-Nordheim plots at room temperature for various laser powers showing three regions (I, II, and III). In saturation region (II), the current increases with optical power as predicted by theory (see inset) for field emission from semiconductors.

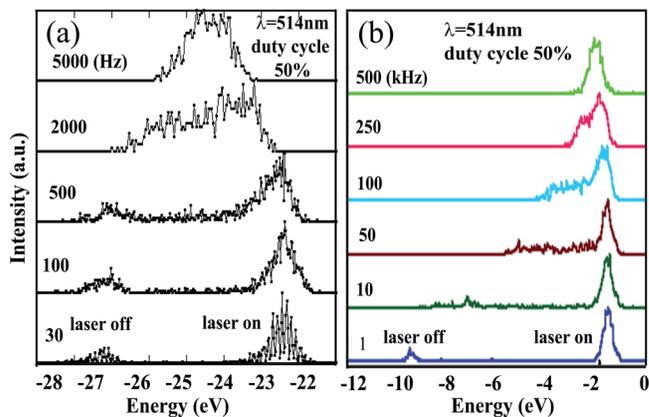


FIG. 3. (Color online) The energy distributions of emitted electrons with different frequency modulation for SiC (a) and Si (b) nanowires. The peak of low intensity corresponding to laser off and the second one corresponding to laser on. The two energy peaks shift are distinguishable above 5 kHz (a) and 250 kHz (b).

the voltage applied to W support tip. Zero electron volt corresponds to no voltage drop along the NW. The laser had a 50% duty cycle at frequencies up to 500 kHz. In this figure, one sees that the peaks, and hence the voltage drops, undergo large optically-induced shifts. The peak at low energy and intensity corresponds to laser off and the second corresponds to laser on. The voltage drops along NWs were smaller and the emission currents were larger with illumination due to the generation of additional carriers. The key new point is that the separation between the peaks diminished as the frequency was raised until the 2 peaks could no longer be distinguished. For the SiC and Si samples, this occurred at 5 and 250 kHz, respectively, giving time constants of 0.2 ms and 4 μs .

One notices that the peak shifting is somewhat different in the two cases, with the peaks finishing closer to the high energy peak for SiC. This is not because the basic physics is different but because the shifting depends on the slopes of the saturations in the $I(V, T)$ curves and the chosen operating parameters.

The response time is obviously too large for the direct creation of electron-hole pairs by photo-absorption over the band gap. Slow response times for LAPE have often been observed and several explanations have been advanced. It has been speculated as being due to diffusion and RC effects^{4,15} without much quantification. Thermal effects were discounted.

The Fourier equation for the thermal response can be solved with a simple characteristic time constant τ if one assumes that the heat capacity C , thermal conductivity κ , and density ρ vary little with temperature and that the current-induced Joule heating and energy loss due to the black-body radiation are negligible for the low currents and $T (< 1000$ K) of these experiments. τ is then given by

$$\tau \sim 4\rho CL^2/\pi^2\kappa. \quad (1)$$

Note particularly τ is proportional to L^2 and is independent of the NWs diameter d when C , κ , and ρ are assumed not to depend on d or T . τ is estimated in Table I using the bulk values for C and ρ (Ref. 16) and experimental values for κ found for SiC (Ref. 17) and Si (Ref. 18) NWs. It shows a

TABLE I. Parameters used in calculation and the estimated photo-response $\tau(\text{est})$ from Eq. (1) compared to the measured $\tau(\text{meas})$.

| | L (μm) | ρ (g/cm^3) | C ($\text{J}/\text{g K}$) | κ ($\text{W}/\text{cm K}$) | $\tau(\text{est})$ (μs) | $\tau(\text{meas})$ (μs) |
|-----|-----------------------|-----------------------------------|-------------------------------|-------------------------------------|--------------------------------------|---------------------------------------|
| SiC | 85 | 3.2^{16} | 0.69^{16} | 0.7^{17} | 90 | 200 |
| Si | 20 | 2.3^{16} | 0.7^{16} | 0.4^{18} | 6 | 4 |

rather good agreement between the estimated and measured response times for both NWs. This is a strong proof that the increases in FE current are due to the laser heating the NW, followed by equilibrium carrier generation. At faster time-scales, the NW does not have the time to heat or cool, but establishes at an intermediary temperature. Heating is enhanced by the linear form of the NWs which reduces heat loss to the support.

There are several consequences of these experiments which are worth underlining. First, without the use of the TEDs to show that the time response was due to temperature, one could not be certain of the origin of the current increases or TED shifts. One can now use the measured $I(V,T)$ curves or shifts of the TED vs T (Refs. 11 and 14) to calibrate the increase in temperature of the NWs with laser light and, thus, have a calibrated nanometric, μs bolometer. The maximum optical excitation of the SiC NW at 90 mW corresponds to an increase in T of $\sim 250^\circ$. Second, one would generally search for much more rapid responses for these nanometric objects. However, the scaling of τ with L^2 means, for example, that a $0.5 \mu\text{m}$ Si NW would have $\tau = 4 \times (0.5/20)^2 = 2.5 \text{ ns}$, which is reasonably fast. Third, there is no limitation in the time resolution for states with different energy shifts (or spectra) and it should thus be possible to measure extremely fast processes. We expect to pass the timescales attained by fast electronics,^{4,19} but for very low currents, and even approach ultrafast pump-probe experiments,²⁰ but for individual NWs.

The actual absorption mechanism is outside the scope of this article, but several comments can be made. The photon energy was slightly larger than the gap for SiC and quite a bit larger for Si. However, the same response time was found for the SiC NW with a photon energy of 1.96 eV, considerably less than the gap. Absorption may have been due to the Frank-Keldish effect²¹ in the depletion zone or due to excitation to gap default states or mediated through surface states. The response time does not depend on the absorbed power and careful calibration will be necessary to quantify absorption.

In this letter, it has been shown that FE from bottom-up SC NWs can be strongly modulated by laser light. This

opens perspectives for the fabrication of new types of photocathodes. Also an original and simple technique for temporal photo-response measurements from individual semiconducting NWs has been demonstrated with potentially a very fast time resolution. The slow response of the studied NWs has been attributed to optically induced thermal excitation. We have experimentally demonstrated that the response time scales roughly with L^2 . Even for thermally activated emission, this can give rather fast response times for short NWs. Measurements on faster responding systems with rapid pulsed lasers are being considered.

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