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Depolarization effect in optical absorption measurements of one- and two-dimensional nanostructures

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We measure absolute optical absorption cross-sections of one- (1D) and two-dimensional (2D) nanostructures using a focused laser beam while varying the numerical aperture (NA) of the focusing lens. We find the optical absorption deviates at higher NA. In the high NA regime, absorption by graphene decreases from 2.2% to below 1.8%; for Ge nanowires, it decreases from an expected value by a factor of 1.2. We explain this using the depolarization effect at the focal spot and conclude that these corrections allow for accurate quantitative measurements of optical and optoelectronic processes in 1D or 2D nanostructures. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4752889]

Absorption of light is a key parameter essential for characterizing the intrinsic efficiency of optical processes in materials as well as the performance of optical and optoelectronic devices, including photovoltaic cells, photo detectors, CCD arrays, and lasers.[1–7] Recently, building these devices from nanostructures has attracted wide attention as it makes possible new functionalities as well as high sensitivity. Examples include plasmon enhanced light-emitting devices, optical power limiting devices, optical antennas, and optical wires.[8–11] Accurate measurements of optical absorption in nanostructures, usually quantified with an optical cross section, are critical for the design and optimization of nanostructure based optoelectronic devices.

There are two experimental approaches for measuring optical absorption of nanostructures: by using ensemble or individual measurement. Individual measurements yield more accurate information by revealing the heterogeneity of nanostructures. In order to determine the optical properties of individual structures with sub-micron spatial resolution, localization of laser excitation at the diffraction limit is often required, which is typically achieved by using high numerical aperture (NA) objective lenses. For instance, the use of two focusing and collecting lenses with matching NA, in combination with laser scanning optics allows spatially resolved imaging of light transmission (thus local extinction or absorption) in the diffraction limit (see Fig. 1(a)). While such measurements could yield, in principle, the same results as measurements done with collimated light on macroscopic samples, these two platforms produce different light fields at the sample location. In particular, the focusing optics modifies the light polarization at the focal plane from that of the collimated beam with the degree of modification determined by the NA, or equivalently the semiaperture angle (SA).

This effect, generally referred to as the depolarization effect, is important especially for optical and optoelectronic

characterization of nanostructures since their optical properties are polarization sensitive due to geometric confinement associated with their nanoscale dimensions. Single-layer graphene, a one-atom-thick film, provides an excellent example of two-dimensional (2D) nanostructures. Its absorption of light is strictly limited to light with polarization parallel to the graphene surface with uniform absorption efficiency (∼2.3%) over broad infrared and visible wavelengths.[12] In contrast, graphene does not absorb light polarized perpendicular to its surface.[13] On the other hand, semiconducting nanowires (NWs) as well as carbon nanotubes (CNTs) are
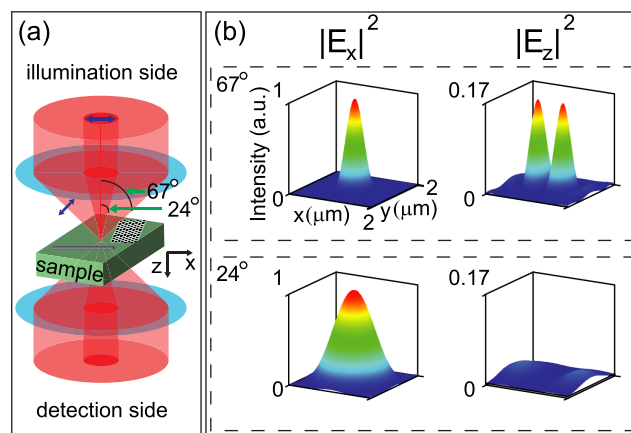

FIG. 1. Measurement scheme and calculated depolarization effect at the focal plane. (a) Linearly polarized light (785 nm) is introduced into an oil-immersion objective lens with an adjustable numerical aperture (upper blue disk), with the polarization direction shown as a blue arrow. The light transmitted through the substrate which has various nanostructures resting on the surface is collected by an oil-immersion lens (lower blue disk) with a fixed numerical aperture (NA = 1.4). The initial polarization before the lens is along the x-direction, and the z-direction is defined as being normal to the sample. Two representative semiaperture angles are shown here. Focusing the light with the larger semiaperture angle tilts the polarization, introducing a measurable z-component. (b) Calculated intensities of the x and z components of the electric field incident upon the sample focal plane are shown for different SAs. All intensities at different SAs are normalized to the x-component.

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expected to show behaviors of one-dimensional (1D) structures. Light is most strongly absorbed when its polarization is along the longitudinal (axial) direction of NWs or CNTs, which is often quantified by the absorption anisotropy ratio, $\delta = (Q_\parallel - Q_\perp)/(Q_\parallel + Q_\perp)$, where $Q_\parallel$ ($Q_\perp$) is the absorption efficiency for light having longitudinal (perpendicular) polarization. For NWs, the absorption efficiencies are defined as ratio of the absorption cross-section to the physical cross section.[14] It is, thus, natural that the light absorption in 1D and 2D nanostructures would be sensitive to the illumination geometry in addition to their intrinsic optical properties. However, this effect is often unjustifiably ignored. As we show in this letter, the depolarization of focused light causes significant changes to the measured absorption values, whose effect is larger for high NA optics. We investigate this effect by measuring absorption cross-sections of prototypical 1D (semiconducting NWs) and 2D (graphene) nanostructures using confocal transmission absorption measurements while varying the SA of illumination.

In Fig. 1(b), we first discuss the depolarization of a focused laser at two different SAs. Here, we use a calculation based on conventional electromagnetic theory, first developed in 1959 by Richards and Wolf, in order to understand how polarization changes when a linearly polarized monochromatic laser beam is focused.[15] It clearly shows that a linearly polarized laser beam will produce a polarization component perpendicular to the focal plane. This depolarization effect has experimentally been seen.[16,17] It can also be understood phenomenologically from the vectorial nature of the electromagnetic field and ray optics.[16] Different rays of a light beam have the same polarization prior to a focusing lens, but the polarization of each ray would be tilted differently with the outermost ray having the largest tilt (Fig. 1(a)) and the strongest depolarized field component. For example, the $|E_z|^2$ component could be as large as 20% of $|E_x|^2$ along the original polarization direction for a semiaperture angle $\theta = 67°$. In nanostructures, these two polarization components are absorbed differently, and hence accurate absorption measurements require an appropriate correction when a high NA lens is used.

Fig. 1(a) shows our experimental scheme. A linearly polarized, collimated, CW laser beam (785 nm) was focused by an oil immersion objective lens with variable NA (between 0.6 and 1.35; Olympus 100x) to obtain different SAs from 24° to 67°. Both NW and graphene samples are deposited on transparent fused silica substrates, which have prepatterned alignment marks for locating samples for further characterization, e.g., atomic force microscopy (AFM). Our graphene is grown using chemical vapor deposition (CVD) methods on copper foils and then transferred onto the substrate following a wet transfer technique.[18] Our NWs are CVD grown germanium (Ge) NWs sonicated lightly in methanol and deposited onto the substrate.[19] The transmitted light was collected using another oil immersion lens with a larger collecting SA (73°) at the back of the sample. In order to produce an absorption mapping, the intensity was recorded as a function of the focused laser spot position, which was controlled by custom-built laser scanning optics. A series of such images, normalized to the total laser power, was taken with different SAs. We use this to investigate the depolarization effect in the absorption measurements.
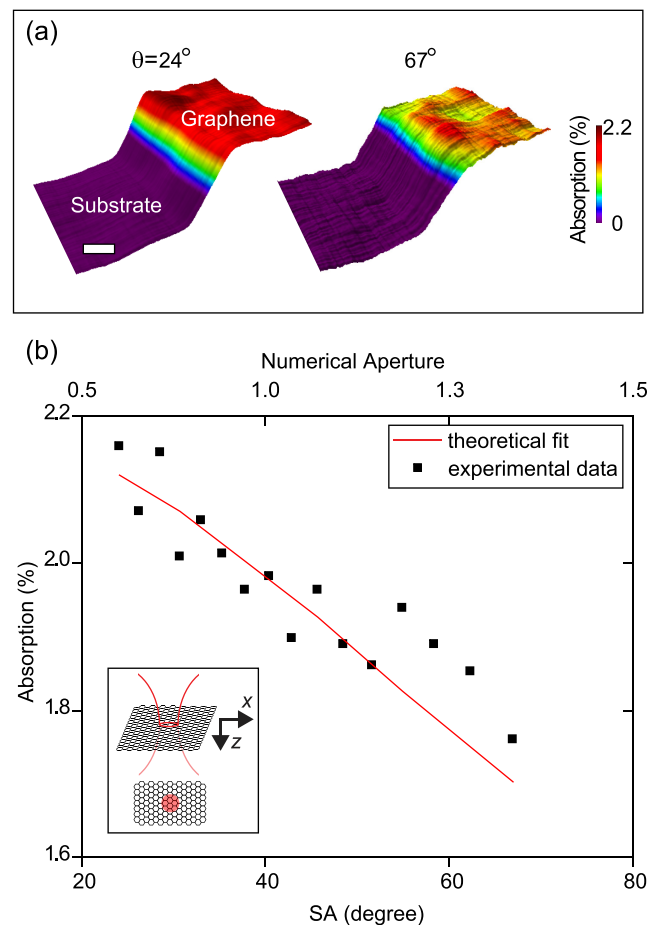

FIG. 2. The experimentally measured absorption in graphene depends on semiaperture illumination angle. (a) Absorption of light at the same area of the sample for two different semiaperture angles. The absorbance of the substrate was set to zero following uniform background correction. Scale bar, 5 μm. (b) Experimentally measured (black squares) and theoretical curve (red line) of absorption of graphene as a function of semiaperture angle. The theoretical curve takes into account the depolarization effect at the focal plane obtaining the fitted tangential absorption of light to be 2.2% for graphene. Inset: Laser beam diameter is smaller than the area of the graphene sheet, which is assumed in the calculation.

Two representative absorption images for a graphene sample are shown in Figure 2(a). A uniform background signal is subtracted from both images. These images are taken for two different SAs. For a 24° semiaperture angle (NA = 0.6), the (area averaged) absorption of light for graphene is measured 2.16±0.07%, close to the well known 2.3%;[12,20] however, for 67° (NA = 1.35), it is only 1.76±0.06%. In both cases, the intensity of focused laser (approximately 80 kW/cm^2) is well below the saturation intensity.[21] This behavior is consistent with the depolarization effect. At a small SA of 24°, the total intensity has little contribution from the normal polarization component (Fig. 1(b) bottom, $|E_z|^2$), while at a large SA of 67°, the normal component's global maximum intensity is almost 20% that of $|E_x|^2$ (Fig. 1(b) top). As all dipole transitions are constrained within the π bands of graphene at this wavelength,[13] any normal component of the light will not be absorbed and thus increases (decreases) the total transmission (absorption) of light. Therefore, as we increase the semiaperture angle of the illumination, the $|E_z|^2$ component increases, decreasing the measured absorption. Indeed, our data are fully consistent
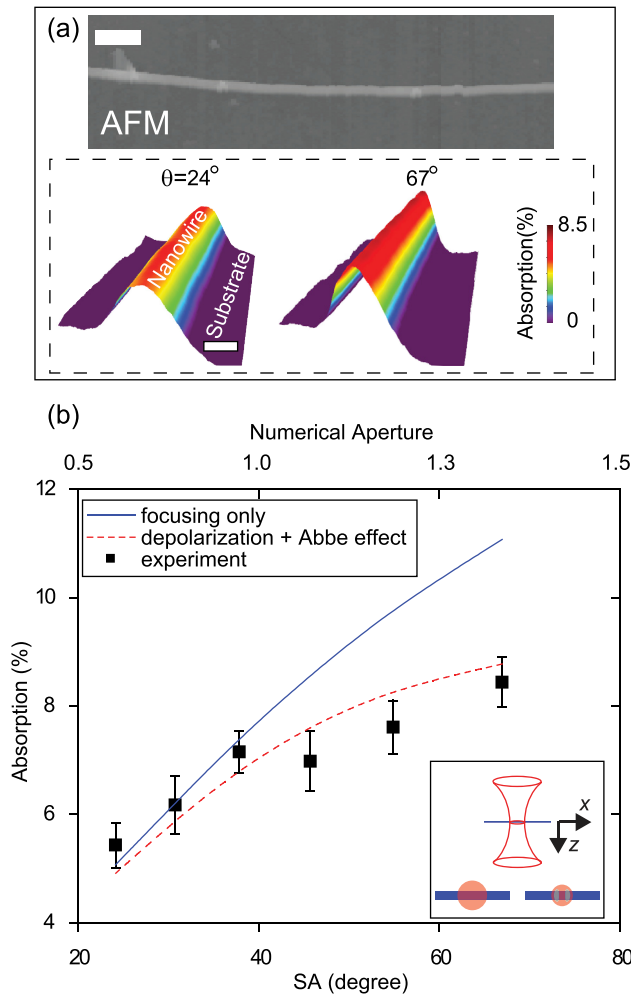
FIG. 3. Dependence of measured absorption versus illumination semiaperture angle in Ge nanowires. (a) Upper: AFM image of a representative Ge nanowire (d ∼ 65 nm). Scale bar, 1 μm. Lower: Measured absorption of light of the same nanowire for two different semiaperture angles. Scale bar, 0.5 μm. (b) Experimentally measured (black squares) and calculated (blue and red curves) absorption for various semiaperture angles of illumination. The blue curve considers only the Abbe diffraction effect in the absence of depolarization. The red curve, however, includes the depolarization effects. Both calculations for this nanowire use $Q_\parallel = 85\%$ in the portion of the nanowire being illuminated. The latter uses $\delta = 70\%$. Inset: Schematic illustrating that the nanowire diameter is smaller than the beam diameter. Smaller semiaperture angles lead to larger laser spot diameters (left) and vice versa (right), with the z component (shaded area) only being significant in the latter.

with this expected behavior (Fig. 2(b)). Furthermore, our data are in excellent agreement with our theoretical calculation (solid line in Fig. 2(b)). We obtained our theoretical fit assuming 2.2% absorption in the tangential polarization direction and zero absorption in the normal direction.

Our results shown in Fig. 2 clearly demonstrate that graphene's optical absorption requires correction (as much as 20%) at large semiaperture angle illumination and that the decrease in the apparent optical absorption is proportional to the amount of depolarization component (in this case $|E_z|^2$). It also experimentally confirms the magnitude of $|E_z|^2$ predicted by theory, which has never been done before.

In Fig. 3, we explore the depolarization effect in 1D linear structures. GeNWs are chosen as they are an ideal material for this study. First, GeNWs have large absorption cross-sections as Ge's direct gap is similar in energy to its indirect gap, producing a better signal-to-noise ratio in our measurement as compared to other indirect band gap materials (such as Si). In addition, their polarization anisotropy was shown to be strongly diameter dependent, increasing from 50% to 98% as the diameter changes from 70 nm to 20 nm.[22]

Both optical microscopy and AFM are used to locate and characterize the GeNWs first. An example AFM image is shown in Fig. 3(a) (upper) for a GeNW with diameter ∼65 nm. In Fig. 3(a) (lower), after aligning the light polarization (before focusing) to the NW's axis, we compare the absorption of the NW at two different SAs as in graphene case. The data at 67° show a higher and narrower absorption peak compared to the case of 24°. This behavior is expected. As the semiaperture angle increases, the focused spot size becomes smaller (the Abbe diffraction effect) and allows more light to be localized around the NWs (Fig. 3(b), inset), especially when the NW diameter is smaller than the beam spot. In Fig. 3(b), we plot the peak absorption values (Fig. 3(b)) measured while varying the SAs of illumination. They monotonically increase for larger SAs closely following the prediction of the Abbe effect (blue curve). However, at large SA, the depolarization effect introduces a significant amount of field with polarization along z direction and thus lowers the absorption.

In order to obtain the theoretical fits for absorption with and without the depolarization effect for NWs, we assume the effective diameter of this NW to be 60 nm after considering the native oxide of GeNWs.[23] The anisotropic ratio, δ, is predicted to be 70% for this diameter.[22] We further divide the focused laser beam spot into two regions. One region is the overlapping area between the NW and laser spot. In this region, the field intensity of each polarization is denoted as I_{NW}^x, I_{NW}^y, and I_{NW}^z with x being along the longitudinal direction of the NW. Similarly, for the region excluding the overlapping region, the intensity of each polarization is denoted as I_E^x, I_E^y, and I_E^z. The experimentally measured absorption is expressed as $(Q_\parallel, Q_\perp, Q_\perp) \cdot (I_{NW}^x, I_{NW}^y, I_{NW}^z)/I_{total}$, where $I_{total} = (I_{NW}^x + I_{NW}^y + I_{NW}^z) + (I_E^x + I_E^y + I_E^z)$ and $Q_\parallel$ and $Q_\perp$ are defined previously as the absorption efficiency for different polarizations. We obtained our theoretical fit assuming $Q_\parallel = 85\%$ (red curve). This value is consistent with respect to the cylindrical NW geometry calculation combined with the previously measured absorption efficiency for the assumed diameter.[19,22]

In conclusion, we demonstrate that the depolarization effect strongly modifies the optical absorption of one- and two-dimensional nanostructures when high NA lenses are used to illuminate the nanostructures. Depending on the dimensionality of nanostructures, different corrections are required. For quantitative optical studies of individual nanostructures using high NA objective lens, these corrections become increasingly important. Furthermore, the variable semiaperture angle measurement presented here is not only limited to the examples mentioned above but also could be useful for understanding the optical coupling in nanostructures under different polarization field configurations.

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