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Nanometer-scale dielectric constant of Ge quantum dots using apertureless near-field scanning optical microscopy

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Tip-enhanced near-field scattering images of Ge quantum dots (QDs) with 20–40 nm height and 220–270 nm diameter grown on a Si substrate have been observed with a spatial resolution of 15 nm. Changing the wavelength of the incident light, the contrast of the images is reversed. It is found that the scattering intensity is caused by the dielectric constants of the materials under the probe. By changing the wavelength of the incident light, we have obtained information about the dielectric constant dispersion of single Ge QDs. © 2010 American Institute of Physics.

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Conventional far-field optical microscopy cannot provide spatial resolution better than half of the optical wavelength (about 300 nm) because of the diffraction of light. The recently developed near-field scanning optical microscopy (NSOM) aperture probe technique offers a resolution below the diffraction limit but the practical resolution is limited to 60–100 nm.

An alternative form of NSOM that utilizes the local field enhancement at the end of an externally illuminated metallic probe (an “apertureless” probe) has also been demonstrated, and various aspects of this new imaging technique, for example, tip-enhanced Raman scattering, photoluminescence, near-field Rayleigh scattering, etc., are being explored. The resolution of the so-called tip-enhanced or apertureless NSOM (ANSOM) is only limited by the radius of curvature of the probe, and therefore it promises unprecedented optical resolution (<15 nm) compared with conventional fiber-based NSOM.

In recent years, a considerable amount of work has been devoted to the study of semiconductor self-assembled quantum dots (QDs) that can be successfully grown from III-V, II-VI, and group-IV lattice-mismatched semiconductors using a Stranski–Kratsanov growth technique. A great deal of research has been focused on Ge/Si self-assembled QDs with an interest in being able to fabricate integrated optoelectronic devices upon existing silicon technology infrastructure. The growth of high-quality, Ge/Si self-assembled QDs can be achieved by molecular beam epitaxy or CVD.

Electrical and optical properties of Ge/Si QDs are strongly modified as their sizes shrink below the nanometer scale. The interband transition at the Γ point (the center of the Brillouin zone) and the E1 transition, which is attributed to a three-dimensional critical point with M1 type of Van Hove singularities have been observed in some types of Ge/Si QDs. To the best of our knowledge, these experiments are based on ensemble averages of QDs.

In this paper, we introduce nanometer scale dielectric constant mapping of Ge QDs grown on a Si substrate without a capping layer (just the thin oxide layer produced in air) by using apertureless NSOM. Contrasts of the scattered light between single Ge QDs and the Si substrate are dependent on the dielectric constants, which are mainly determined by the direct transition of Si in the UV region and by the E1 transition at the M1 saddle point of Ge in the visible region. By changing the wavelength of the incident light, we have obtained information about the dielectric constant dispersion of single Ge QDs. The spectral position of the E1 transition in single Ge QDs is found to shift to higher energy as compared with that in bulk Ge.

Ge dots were grown on a Si substrate without a capping layer using an ultrahigh-vacuum chemical vapor deposition system. The temperature of the Ge layer deposition was 800 °C. Two kinds of dots are obtained on a Si substrate depending on the growth temperature as follows: one is dome shaped and the other is pyramid shaped.

The experimental setup for near-field scattering measurements is a combination of a tapping mode atomic force microscope (AFM) and a confocal optical microscope. The AFM microcantilevers are commercially available, standard etched Si tips, coated with platinum and titanium, with a tetrahedral shape. The radius and resonant frequency Ω of the tips are 15 nm and 70 kHz, respectively.

The CW light sources are a violet diode laser (3.06 eV), an Ar ion laser (2.50 and 2.60 eV), a frequency-doubled Nd:YAG laser (2.33 eV), a Nd:YAG pumped tunable dye laser employing Rhodamine 6G (2.10 eV), a HeNe laser (1.96 eV), and a Ti:Sapphire laser (1.55 eV). The incident light is focused by means of a long working distance objective lens (NA=0.42) onto the end of the AFM cantilever at approximately 40° incidence [see Fig. 1(a)]. Back scattered light returns through the same objective lens and is focused onto the optical fiber. Therefore the detection is confined to the area within a few micrometers around the tip-sample junction. The scattered light through the optical fiber is detected by the 200 kHz photoreceiver.

The signal contains the modulated scattered light at the frequency of the cantilever oscillations and its harmonics (ac component) due to the nonlinearity of scattered signal dependence on the tip-sample distance. The ac component is demodulated by a lock-in amplifier at the second harmonic 2Ω of the resonant frequency of the cantilever. Figure 1(b) shows a plot of the scattered intensity as a function of the
tip-sample distance. The near field scattering signal decreases completely within a distance of 50 nm from the sample surface, reflecting the extremely short distance scale of the tip-sample interaction. Therefore we safely conclude that the observed scattered signal does not contain an artifact background signal.

Figure 2(a) is a typical result of AFM topography of Ge QDs grown on a Si substrate. The Ge QDs have two kinds of shapes as follows: dome and pyramid.12 Typical base and height sizes of Ge QDs are 270 and 40 nm for dome shape and 220 and 21 nm for pyramid shape, respectively. Simultaneously recorded scattering images are shown in Fig. 2(b) 3.1 eV and Fig. 2(c) 2.11 eV, respectively. Arrows in the images refer to the same QD. The contrast of the scattered light images is reversed in the Figs. 2(b) and 2(c). The spatial resolution is approximately 15 nm, which is determined by the near-field scattering measurements of following two samples: gold nanoparticles with 80 nm in diameter and InAs self assembled QDs with 3 nm in height and 20–30 nm in diameter (not shown here).

Figure 2(d) shows the cross-sectional height and normalized scattering intensity of a dome shaped Ge QD indicated by the arrows in the AFM and scattering images. The normalized scattering intensity from Ge QDs is dependent on the wavelength of the incident light. The scattering intensity from the Ge QDs is 2.86 or 0.29 times as large as that from the Si substrate at 2.11 or 3.10 eV, respectively. Note that we could not determine the absolute values of the scattering intensities at the different wavelengths in this experiment because the optical alignment is changed when we change the light sources. As shown in Fig. 1(b), the penetration depth of the optical near field is 30–50 nm, which is comparable to the height of the QDs. Hence it is found that the scattering intensity is almost proportional to the height of the QDs.

To make clear the spectral response, we observed the near-field scattering intensity of single Ge QDs, normalized to the Si substrate, for various wavelengths of the incident light. The spectrum is obtained by dividing the average value of the scattering intensity of 10–20 individual QDs by that of the Si substrate. Figure 3(b) shows the spectral dispersions of dome (solid circles) and pyramidal (open circles) shaped QDs. Each spectrum has a broad peak around 2.0–2.5 eV.

As shown in Fig. 3(a), dielectric constants of bulk Si and Ge are mainly determined by the direct transition of Si in the UV region and by the E1 transition at the M1 saddle point of Ge in the visible region.14 The near-field scattering intensity is sometimes calculated by using the point dipole model of the oscillating sphere, which relates to the dielectric constant of the sample.4 However our experimental data [Fig. 3(b)] could not be quantitatively reproduced by a calculation based on that model, since the calculated contrast is almost unity as compared with experimental data. Here we used the finite-dipole model in which the tip is approximated by a
spheroid. In the finite-dipole model, the scattered field is written as, $E_s \propto (1 + r_p)^2 \alpha_{eff} E$, with $r_p$ being the Fresnel reflection coefficient for $p$-polarized light, $\alpha_{eff}$ the effective polarizability, and $E$ the incident field. The value of $\alpha_{eff}$ is described by geometrical parameters of the spheroid: the radius of curvature at the apex $R$, the half major axis $L$, and the distance between the spheroid and the sample surface $H$. The details of the equation are described in Ref. 15. The finite-dipole model is valid in the quasistatic regime, i.e., $L \ll$ wavelength. Although it is not clear whether this requirement is fulfilled in our experiment, the calculation based on the finite-dipole model will be made, and applicability of this model will be checked.

We introduce an expression of the effective polarizability in the case of an elongated spheroid, i.e., $R$ and $H \ll L$, $\alpha_{eff} \propto g \beta/(1 - g \beta)$, where $\beta = (e - 1)/(e + 1)$, $e$ the dielectric constant of sample. Here $g$ is the fraction of effectively induced charge at the focus point of the spheroid and totally induced charge by mirror charge induced in the sample. In the case of $g = 1$, i.e., the mirror charge in the spheroid is concentrated on the focus point, $\alpha_{eff} \propto (e - 1)/2$.\(^\text{17}\)

The observed signal is proportional to the cross-term between the scattered near-field light $E_s$ and the background light $E_{br}$ because $E_s \ll E_{br}$. The interference generally induces a difficulty in measuring the scattering intensity due to the relative phase change among $E_s$ and $E_{br}$.\(^\text{16}\) In this experiment, the relative phase $\phi_s - \phi_b$ is adjusted by tuning the incident light position to obtain constructive interference on the Si substrate, i.e., $\phi_s - \phi_b = 0$. Therefore the observed signals for the Si substrate and the Ge QDs are proportional to $|E_s|^2$ and $|E_s^{QDs}| \cos(\phi_s^{QDs} - \phi_b)$, respectively.

The calculated scattering intensity of bulk Ge normalized to the Si substrate at the second harmonic of the modulation frequency is plotted as a solid line in Fig. 3(b). The experimental data can be well reproduced by the calculation based on the finite-dipole model. It is expected that the presence of a wetting layer and intermixing of Ge and Si lead to the slight difference. From Raman scattering measurements, the intermixing of Ge and Si is observed.\(^\text{12}\) Since the dielectric constant of Si around 2.0–2.3 eV is smaller than that of Ge, the intermixing effect reduces the contrast of the scattering. And the thin Ge wetting layer on the Si substrate also reduces the contrast.

The contrast of dome shaped QDs is 1.3 times as large as that of pyramidal shaped ones. A possible reason is a coupling between the tip and the Si substrate through the Ge QDs, because the thickness of the Ge QDs is in the range of the near-field interaction depth as shown in Fig. 1(b).\(^\text{18,19}\) Another possibility is the difference of the shape and Ge content.

The experimentally obtained spectral weight of the E1 transition in a single Ge QD is found to shift to higher energy of about 0.1 eV with decreasing the height. There are two following possible reasons for the blueshift: (1) a quantum confinement effect\(^\text{20-23}\) and (2) a compressive strain due to the lattice mismatch between the Ge QDs and the Si substrate by the Stranski–Krasnov growth.\(^\text{12}\) The base sizes of our QDs are too large to expect a quantum confinement effect; the height sizes of our QDs are permissible to expect it.\(^\text{5}\) Therefore the observed blueshift is determined by the interplay between the above two effects. A band calculation of Ge QDs grown on a Si substrate will be needed for further understanding of the blueshift, which remains for future work.

In conclusion, we have observed the near-field scattering images of Ge QDs fabricated on a Si substrate. By changing the wavelength of the incidence light, we have obtained information about the spectral dispersion of single Ge QDs, which indicates a blueshift.

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\(^5\)Dalberg, M. Grundmann, and N. N. Ledentsov, Quantum Dot Heterostructure (Wiley, Chichester, 1999).
\(^17\)The contrast $|\epsilon_{Ge} - 1|/|\epsilon_{Si} - 1|$ based on the elongated spheroid approximation is consistent with the experimental data.