Ellipsometry study of SiGe nanocrystals embedded in SiO_2

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Silicon-germanium alloys, especially in form of small nanocrystals (NCs), are promising materials for future nanotechnology applications [1-5]. For example, replacement of the charge trapping layer in FLASH memory with an array of NCs embedded in a dielectric matrix provides the possibility to increase the data storage density and improve the data retention [3, 4]. However, recent works [6, 7] demonstrated that in small size (less then 5 nm in diameter) silicon NCs embedded in Gd_2O_3 quantum confinement results in an up-shift of bottom of their conduction band towards the conduction band of the surrounding insulator leading to a decrease in depth of electron traps and impairing the charge trapping and storage. Potentially, the problem may be solved using SiGe NCs with bandgap tuned by changing Si/Ge ratio [2]. At the same time, possibility of NCs band energy manipulation can be used in resonant tunneling devices [8] and for opto-electronic applications [9]. To provide fundamental insight into SiGe electron state spectrum, in this work we investigated optical properties of several types SiGe-NCs embedded in SiO_2 films on Si substrates.

The films containing NCs were grown by RF magnetron co-sputtering of Ge and Si targets mounted on a pure SiO_2 target on Si(100) substrates using the Alcatel SCM650 system. The growth conditions were varied, in particular the substrate temperature (room temperature, 400°C and 500°C), argon gas pressure (1×10^{-3} and 1×10^{-2} mbar), the discharge power (30, 80 and 100 W), and surface ratio of Si and Ge targets (Ge/Si = 1/1 and 1/2). The latter parameter affects the ratio of the fluxes of Si and Ge atoms forwarded to the substrate surface. After the deposition, the samples were annealed at 900-1000°C in nitrogen atmosphere for 1-6 hours. Investigation of NCs size distribution and spatial homogeneity of our samples was reported [10]. Thereby, we used samples with following labels, mean radii and Ge fraction: G=2.8±1.8 nm (Ge = 0.97), J=2.4±0.3 nm (Ge = 0.68), K=1.9±0.2 nm (Ge = 0.68), M=2.2±0.5 nm (Ge = 0.81), and Q=11.5±4.2 nm (Ge = 1.0). Note, that deviation given for the NCs radii is the root-mean-square deviation. Optical properties of the Si-nc samples were characterized using spectroscopic ellipsometry (SE) measurements in the photon energy range 1.5-5 eV (Sopra GES-5 Optical Platform). Though, SE does not directly give optical constants, such as refractive index (n) and extinction coefficient (k), they can be extracted by applying an adequate mathematical model [11]. In order to account for the spherical shape of NCs, we used Maxwell-Garnett model [12] for representation of NCs containing film as mixture of SiO_2 matrix with the dispersion law composed of Lorenz peaks [11].

The observed spectra of n and k shown in Fig. 1 (a) and (b) are consist of Lorenz peaks and related to presence of NCs. It clear from Fig. 1 (b), that for photon energy above 4 eV, k-coefficient strongly increases with increasing photon energy. Taking in account that k is related to the optical absorption coefficient α by simple equation $k = (\alpha\lambda)/4\pi$, the observed behavior of k can be explained by presence of a non-stoichiometric germanium oxide [13-15]. Latter indicates that relatively large part of Ge was oxidized during the film growth or the post-deposition annealing. Portion of the spectra in Fig. 1 (a) and (b) for phonons energy less then 4 eV corresponds to the optical response of non-oxidized SiGe in NCs because the refractive index is higher than that one of silicon oxide (1.45-1.5 for SiO_2 and 1.9-2.1 for SiO [16]) or germanium oxide (1.6-1.95 for GeO_x [13, 14]). In photons energy range below 4 eV all samples except of sample Q exhibit broad range of k-values (cf. Fig. 1 (b)) without, however, strong characteristic peaks related to excitation of direct optical transitions in crystalline SiGe [16]. Therefore, we suggest that in our samples SiGe is in amorphous phase. The sample Q (Fig. 1) contains large-size NCs, probably including the crystalline phase of SiGe. Using analysis of Lorenz peaks position during extraction of n and k from the ellipsometry datasets (not shown here) we found that the samples G and Q with similar Ge fraction demonstrate similar peaks with positioned at around 2.5 eV, while the samples J and K, also with similar Ge content, but lower than that in samples G and Q exhibit the peak shifted to a higher energy (3.5-3.7 eV). Sample M with the lowest Ge content shows only a broad peak at ~4 eV. In contrast to our earlier work on Si NCs [6], it appears impossible to resolve bandgap width of Ge-rich NCs from spectra shown in Fig. 1, probably because the sizes of NCs in our samples are not small enough to cause the quantum confinement bandgap shift [17].

In summary, optical analysis of SiGe nanocrystals embedded in SiO_2 reveals significant contribution of Ge-oxide to the spectra of refractive index and extinction coefficient. The obtained data demonstrate that in small size nanocrystals a non-oxidized SiGe is predominantly in amorphous phase. The energy position of the corresponding optical absorption peak is significantly influenced by the Si/Ge ratio.

References

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Figures

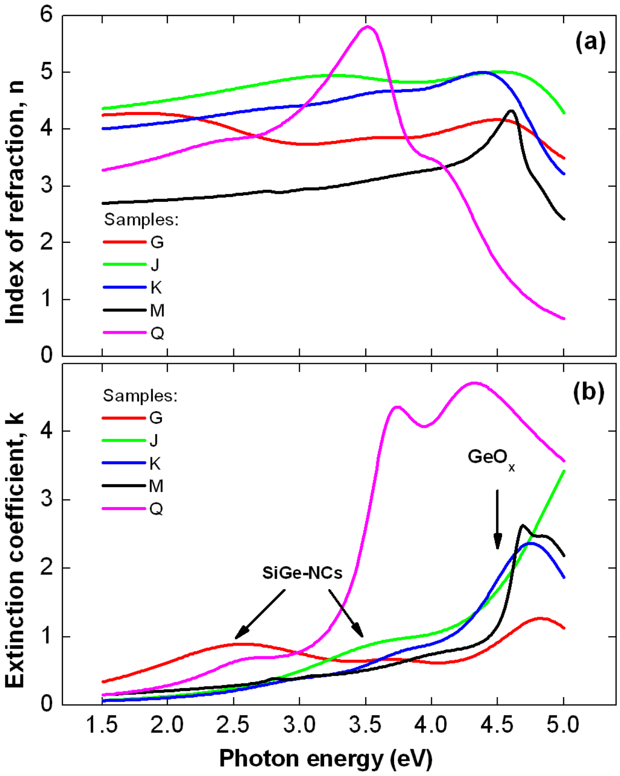

Fig. 1. Spectral dependences of refractive index (n) and extinction coefficient (k) extracted from ellipsometry measurements on samples with SiGe nanocrystals embedded in SiO_2. Samples description is given in text.