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Citation: Applied Physics Letters **91**, 051917 (2007); doi: 10.1063/1.2764113
View online: http://dx.doi.org/10.1063/1.2764113
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Type-I optical emissions in Ge/Si quantum dots

P. F. Gomes,[a)] F Iikawa, and F. Cerdeira
Intituto de Física "Gleb Wataghin," Unicamp, CP 6165, Campinas, São Paulo 13083-970, Brazil

M. Larsson, A. Elfving, G. V. Hansson, W.-X. Ni, and P.-O. Holtz
Department of Physics, Linköping University, S-581 83 Linköping, Sweden

(Received 4 May 2007; accepted 2 July 2007; published online 2 August 2007)

The authors studied the optical emission of Ge/Si quantum dots under externally applied biaxial stress using samples grown with different temperatures varying from 430 to 700 °C. The optical emission energy of samples grown at low temperatures is rather insensitive to the applied external stress, consistent with the type-II band alignment. However, for samples grown at high temperatures we observed a large blueshift, which suggests type-I alignment. The result implies that recombination strength can be controlled by the growth temperature, which can be useful for optical device applications. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2764113]

Ge/Si self-assembled quantum dots are promising systems for optoelectronic applications because their discrete electronic states can be modulated by dot size and strain, giving transition energies in a spectral range that is widely used for optical communication.[1,2] The band alignment at the Si/Ge interface was the object of a long controversy. It is now generally accepted that Ge/Si quantum dots present type-II band alignment at the interfaces, where holes are confined in the Ge dot, while the electrons are confined in the Si layer surrounding the dot.[3–6] Nevertheless, recent work by various authors[7,9] shows evidence of photoluminescence (PL) spectra in some Ge/Si quantum dots which exhibit either type-I or type-II emission. In this work, we address this question by performing PL measurements under an externally applied biaxial tensile stress. Deformation potential theory predicts that under such a strain the PL peak suffers a large blueshift for type-I recombination and a *small* redshift when the recombination is type II. A similar experiment was performed by Thewalt *et al.*[9] in Si-rich SiGe/Si *quantum wells*. They observed that decreasing the laser intensity by several orders of magnitude the emission peaks switched from type-I behavior to type-II behavior. They concluded that carrier accumulation due to optical excitation can result in emission with type-I characteristics even when the Si/GeSi interface had type-II character. We study a series of samples of Ge/Si self-assembled quantum dots grown by molecular beam epitaxy at different growth temperatures. In samples grown at high temperatures we observe a large blueshift in the PL peak with increasing strain (type-I behavior), while the samples grown at low temperatures present practically no shift, consistent with type-II alignment. Changing the laser intensity by a couple of orders of magnitude does not alter these results.

The pseudomorphic self-assembled Ge quantum dots (QDs) grown on Si substrate are under intrinsic biaxial compressive strain due to the lattice mismatch between Ge and Si.[1,10] For these dots, the band profile of the minimum of the conduction band (Δ points) and maximum of the valence band (VB) (Γ points) is shown in Fig. 1(a). The Δ points in the Ge layer split into doublet higher branch, Δ_2, and a quadruplet lower one, Δ_4.[1] Therefore, the minimum in the Ge dot is Δ_4 and in the Si layer is Δ. Applying an external biaxial tensile strain the Δ_2 component of both materials shifts down, while the Δ_4 component shifts up, as indicated in Fig. 1(a). In the VB the heavy hole level shifts down. In the Ge dot both the electron and heavy hole levels shift in opposite directions under strain, resulting in a large blueshift of the type-I recombination. However, if the electron is localized in the Si layer, its energy shifts under strain in the same direction as the heavy hole in Ge, resulting in a small redhift of the recombination energy.[9]

The Ge/Si QDs were grown on Si (001) substrate by molecular beam epitaxy at different growth temperatures resulting in different dot distributions (Table I). Samples differ in dot size and density: small dots with high dot density for low growth temperatures and large dots with small dot density for high growth temperatures.[8] We used a homemade pressure cell to apply a biaxial stress on the epitaxial film.[11–13] It is based on bending the sample placed between a ring and a sphere [Fig. 1(b)]. One surface is under biaxial compressive strain and the other surface, under biaxial tensile strain. The latter corresponds to the front of our samples. The strain was calibrated through the energy shift of the Si layer zero-order optical emission. The PL measurements were performed using a 442 nm line of a He–Cd laser and detected using a single $\frac{1}{2}$ m monochromator coupled with the nitrogen cooled Ge detector.

In Figs. 2(a) and 2(b) we present the PL spectra, measured at 20 K, for two samples grown at lowest and highest

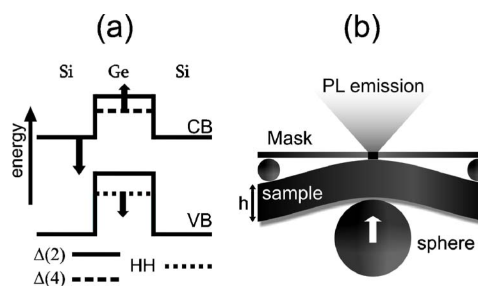

FIG. 1. (a) Potential profile of nonrelaxed Ge/Si QDs. For conduction band only Δ points are shown. The arrows indicate the directions of the energy shift when an external biaxial tensile strain is applied. (b) Lateral view of the pressure cell. A mask with a 1 mm diameter hole was used to select the luminescence at the center of the sample.

[a)]Electronic mail: pgomes@ifi.unicamp.br

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TABLE I. Description of the samples.

	Growth temp. (°C)	Density (cm^{-2})	Diameter (nm)	Height (nm)
Sample 1	700	1.5×10^9	200	20–25
Sample 2	580	7×10^9	60–70	~11
Sample 3	480	1.4×10^{11}	~30	<2
Sample 4	430	1.5×10^{11}	20–25	<2

growth temperatures, respectively, taken at progressively higher values of biaxial strain. All samples exhibit broad optical emission bands in the absence of an external strain. Under biaxial tensile strain, the emission bands practically remain at the same energy for samples grown at 480 and 530 °C and present a large blueshift for samples grown at 580 and 700 °C. The energy shift of the PL peak position versus biaxial tensile strain of all samples is plotted in Fig. 2(c). We also varied the laser intensity, from ~8 to 160 W/cm^2; however, we observed the same behavior shown in Fig. 2(c). This strained-induced energy shift can be estimated using a linear deformation potential theory.[10] We used the values of the parameters given in Ref 1 The type-I optical emission corresponds to the recombination of electron (in Δ_4) and heavy hole both in the Ge dot and we can easily calculate it using the values of the parameters for the Ge. On the other hand, for the type-II emissions the electron (in Δ_2 in Si) and heavy hole (in Ge) involve different materials. Since the hydrostatic deformation potential is obtained experimentally only for its difference, it cannot be described by the simple model used here for type-II transition, and thus it is an opened parameter. However, we used in this work the values of parameters for the Si (for Ge, the energy shift is very similar) in order to analyze the qualitative behavior. The theoretical curves are plotted in Fig. 2(c), where the solid and dashed lines indicate the type-I and type-II energy shifts, respectively. The figure shows that the two samples grown at higher temperatures (solid symbols) exhibit shifts compatible with type-I behavior, while the two samples grown at 430 and 480 °C (represented by open symbols) exhibit type-II behavior. Better quantitative agreement for type-II shift could be attained by adjusting the values of the deformation potentials or using a more complex model. However, the trend indicated in Fig. 2(c) clearly defines which sample shows either type-I or a type-II behavior.

Changing the growth temperature, as shown in Table I, basically changes the dot sizes and densities. However, it should also affect intermixing effects, which result in a different Ge distribution in the dot and, consequently, different built-in strain. Strain relaxation in the dot can also depend on the dot size. Since the estimated value for the conduction band offset is quite small (~40 meV for unrelaxed pure Ge/Si interfaces[1]) it may be sensitive to the distribution of the Ge content and the strain in the dot. All these factors can result in either type-I or type-II recombination.

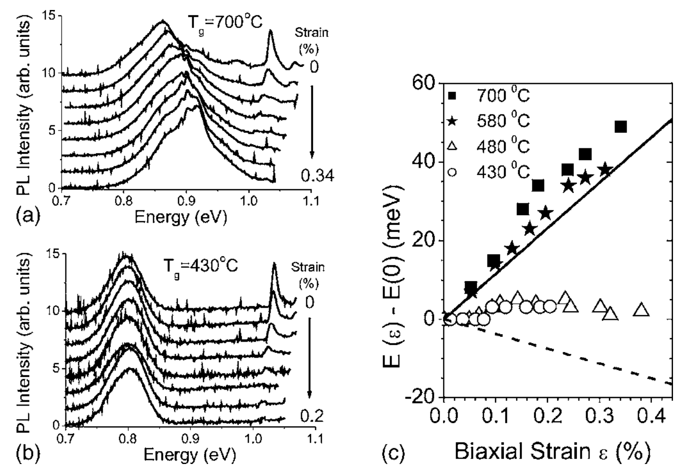

FIG. 2. Strain dependence of PL spectra measured at 20 K for samples grown at (a) 700 and (b) 430 °C. The spectra are shifted vertically for ease of viewing. The fine structures in the spectra of (b) are due to the water absorption. (c) The energy shift of the PL peak position vs strain.

In summary, we observed that the optical emissions in Ge/Si QDs are quite sensitive to the external strain when the quantum dot is grown at low or high temperature. The result suggests that the growth temperature plays an important role on the carrier distribution, resulting in a type-I or type-II recombination. Thus, growth temperature can be used as a parameter to manipulate the transition oscillator strength in optical devices.

The authors acknowledge financial support from FAPESP, CAPES, and CNPq in Brazil and SSF in Sweden, and thank Milton Tanabe and Claudecir Biazoli for technical support.

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