ナノメカニクスによる半導体量子ドットの発光特性制御
Control of photoluminescence of quantum dots by nanomechanics

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1. **Intruduction** In the report, we describe the simultaneous measurement of the indentation force and PL of In0.5Ga0.5As/GaAs QDs, together with experimental improvements on the precise fabrication of the nanoprobe. The simultaneous measurement of the indentation force and PL of QDs is achieved by introducing a sensitive loadcell into our previous nanoprobe indentation system [1, 2]. The nanoprobe is milled by FIB to obtain a flat apex aperture. Based on the quantitative relation between the nanoprobe indentation force and the discrete energy levels of the QDs, which is determined experimentally, a theoretical analysis is performed based on a three-dimensional finite element calculation and six-band strain-dependent Hamiltonian. The distribution of biaxial strain under the central part of the nanoprobe and the contributions of the strain components to the energy shifts of the QDs are discussed from the analytical results.
2. **Experimental procesures** The In0.5Ga0.5As QDs studied in the present work were prepared on GaAs(001) by chemical beam epitaxy [3, 4] and embedded in a 50 nm thick capping layer of GaAs. The configuration used for the nanoprobe PL measurement (Unisoku, USM-100R) was described previously, where a nanoprobe (Al-coated optical fiber) with a spherical apex rather than a flat apex was adopted [1, 2]. In the present experiment, however, the nanoprobe was milled by FIB to obtain a flat apex aperture with a radius of 425 nm (shown in the insert of Fig. 1). Then, the nanoprobe was installed to apply indentation force on the QD sample, and the PL of the sample was recorded through its aperture. A small cylindrical loadcell (aluminum alloy, Tokyo Sokki Kenkyujo) of the resistance-bridge type was specially designed for high sensitivity and for installation in the small sample holder of the nanoprobe PL system. With this loadcell below the QD sample, an indentation force as low as 200 μN can be measured with good reproducibility. The experiments were carried out at a low temperature (10 K) and in an ultra-high vacuum ($<1.4 \times 10^{-9}$ Pa). After the abovementioned indentation experiments, the following unloading indentation experiments were performed in order to check for reversibility in our experiments. The symmetry of the PL spectrum shifts of the QDs due to loading and unloading in our experiments indicated elastic deformation.
3. **Results** Fig. 1 shows the dependence of the indentation force on the nominal distance of the piezo-driven nanoprobe, as measured by the loadcell in a direction perpendicular to the sample surface. The nominal distance is estimated from the voltage applied to the probe-driving piezo-unit. The negative values of the nominal distance in Fig. 1 indicate the gap between the top surface of the QD sample and the flat apex of the nanoprobe, while positive values indicate the further downward displacement of the piezo-driven nanoprobe after the contact of the nanoprobe apex with the sample surface. The positive values comprise the deformation/displacement of the nanoprobe, the QD sample, and the loadcell. The zero point corresponds to the onset of the contact between the nanoprobe and the sample surface, determined by the emergence of the first sharp peak in the spectrum. From the linear fit in Fig. 1, we obtain the dependence of the PL spectrum shifts on the indentation force, as shown in

Fig. 2(a). A vertical slice of this figure contains the PL spectrum for a given indentation force [5], and 58 spectra of this type comprise Fig. 2(a). The step-wise discontinuity in the diagram is a result of the enlargement of the indentation force axis, which is done to improve the visualization. The brightness in Fig. 2(a) corresponds to the PL peak intensity. The bright streaks show the shift of the emission energy of a single QD due to the nanoprobe indentation-induced strain. By tracing the typical bright streaks in Fig. 2(a), the shifts of the emission energy of the representative QDs are obtained as shown in Fig. 2(b). An energy shift as large as 90 meV and its linear dependence on the indentation force are observed in Fig. 2(b). In order to present the difference in the shifts quantitatively, we define the shift rate of the emission energy as $R_s = \frac{\Delta E}{\Delta F}$, where ΔE is the shift of the emission energy of the QD and ΔF is the change of the indentation force. The value of R_s in Fig. 2(b) varies from 99 to 146 meV/mN. The relatively large variation of Rs obtained can be clarified in a repeated indentation with the horizontal step-wise movement of the nanoprobe. In this repeated indentation experiment, the nanoprobe is gradually pressed downward onto the QD sample, and then lifted up and moved horizontally by 6.6 nm. This sequence is repeated eight times. The shifts of the emission energy of the QDs obtained in this experiment are shown in Fig. 3. Some of the bright streaks in Fig. 3 gradually change in shift rate according to the horizontal position of the nanoprobe. One streak of the emission energy, which is selected because of its strong dependence on the horizontal position of the nanoprobe, is traced by the dashed white lines in Fig. 3. As compared with this streak, the other bright streaks in Fig. 3 exhibit a weak dependence on the horizontal position of the nanoprobe. This difference in the dependence can be attributed to the variation in the local strain distribution, i.e., the relative position of the QDs and the nanoprobe.

$$H_\varepsilon^V = -\begin{bmatrix} P+Q & -S & R & 0 & \sqrt{\frac{1}{2}}S & -\sqrt{2}R \\ -S^* & P-Q & 0 & R & \sqrt{2}Q & -\sqrt{\frac{3}{2}}S \\ R^* & 0 & P-Q & S & -\sqrt{\frac{3}{2}}S^* & -\sqrt{2}Q \\ 0 & R^* & S^* & P+Q & \sqrt{2}R^* & \sqrt{\frac{1}{2}}S^* \\ \sqrt{\frac{1}{2}}S^* & \sqrt{2}Q & -\sqrt{\frac{3}{2}}S & \sqrt{2}R & P+\Delta_0 & 0 \\ -\sqrt{2}R^* & -\sqrt{\frac{3}{2}}S^* & -\sqrt{2}Q & \sqrt{\frac{1}{2}}S & 0 & P+\Delta_0 \end{bmatrix} \begin{matrix} \left|\frac{3}{2},\frac{3}{2}\right\rangle \\ \left|\frac{3}{2},\frac{1}{2}\right\rangle \\ \left|\frac{3}{2},-\frac{1}{2}\right\rangle \\ \left|\frac{3}{2},-\frac{3}{2}\right\rangle \\ \left|\frac{1}{2},\frac{1}{2}\right\rangle \\ \left|\frac{1}{2},-\frac{1}{2}\right\rangle \end{matrix},$$

$$P = a_v(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}), \quad Q = b[\varepsilon_{zz} - \tfrac{1}{2}(\varepsilon_{xx} + \varepsilon_{yy})],$$

$$R = \tfrac{\sqrt{3}}{2} b(\varepsilon_{xx} - \varepsilon_{yy}) - id\varepsilon_{xy}, \quad S = -d(\varepsilon_{xz} - i\varepsilon_{yz}).$$

References

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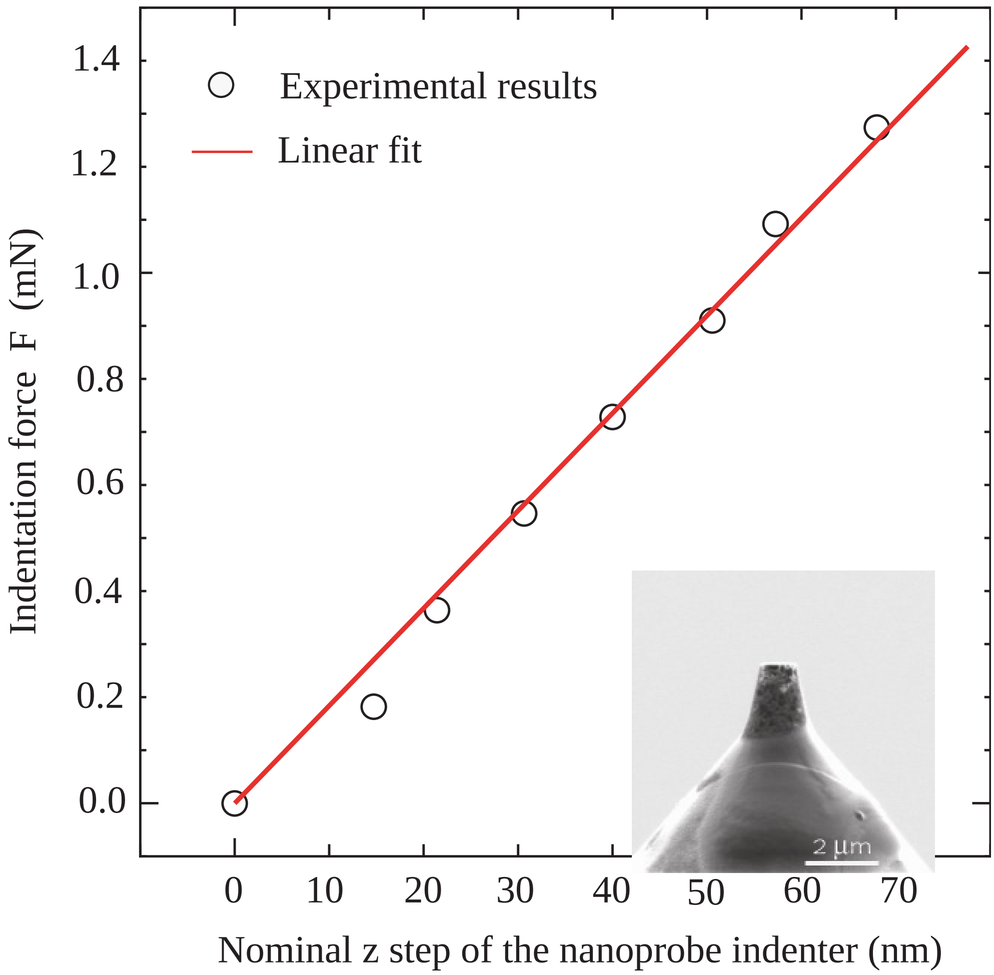
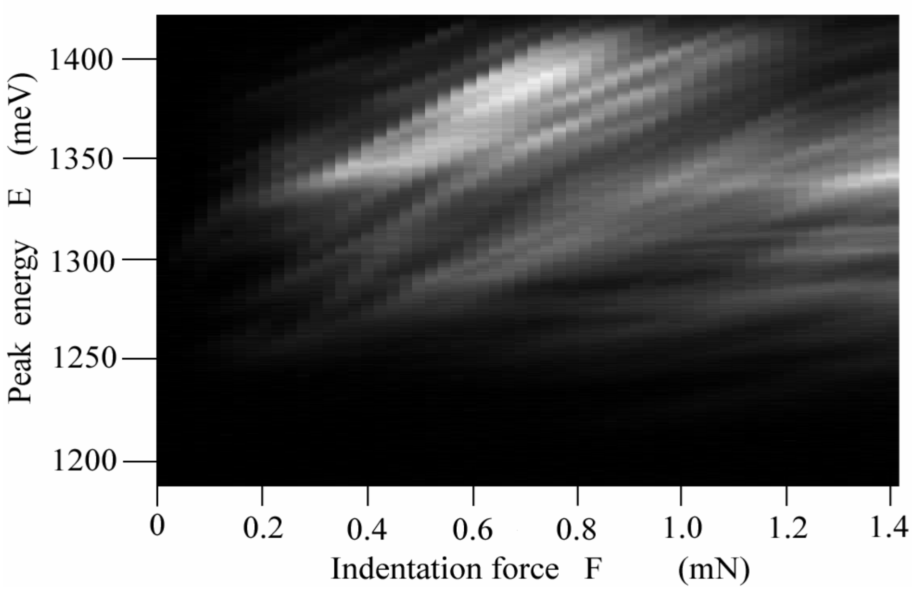
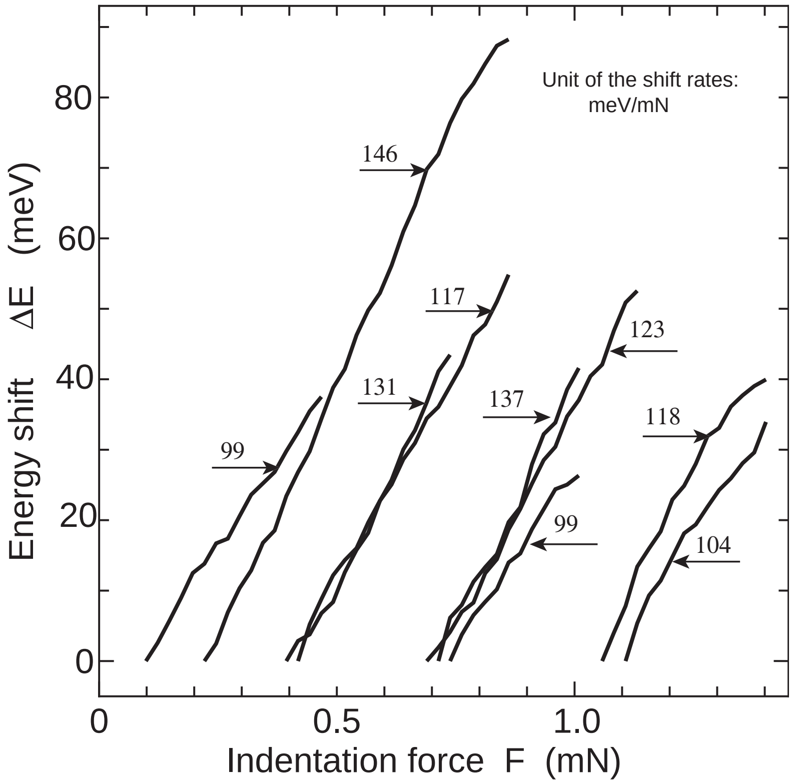


Fig.1 Dependence of indentation force on z step of the piezo-driven nanoprobe in nanoprobe indentation experiment. The milled flat apex of nanoprobe is shown in the insert.



(a)



(b)

Fig.2 Dependence of energy band gap shifts of QDs on indentation force. (a) Indentation force dependence of nanoprobe PL spectrum. The brightness in the figure shows the intensity of peak. (b) Traces of some emission lines in (a). Values are the energy shift rates of each peak.

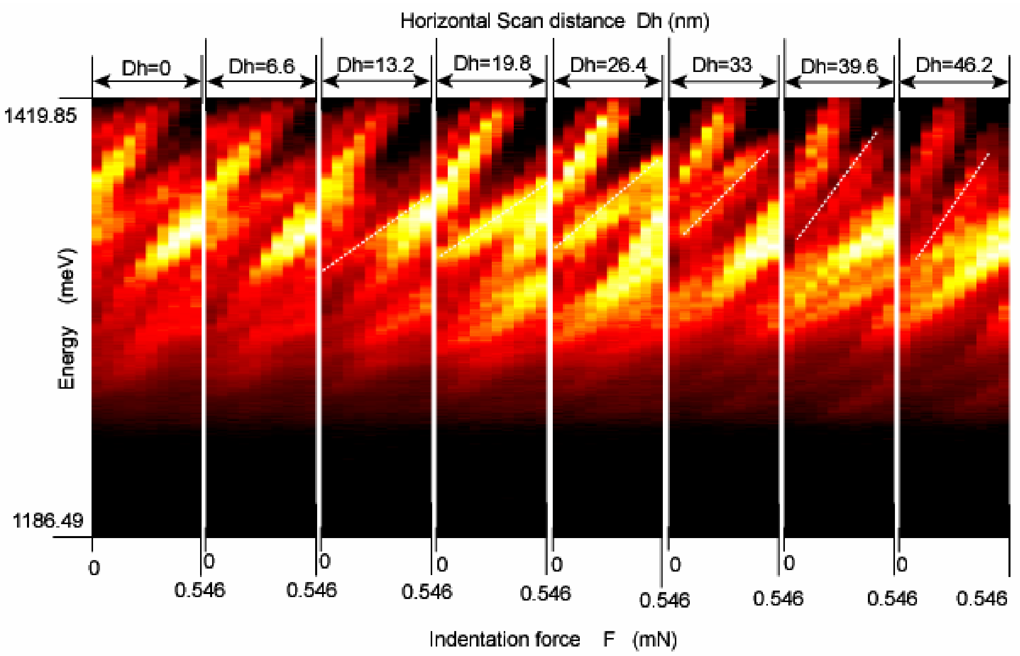


Fig.3 Shifts of emission energy from QDs in the experiment of repeated indentation with horizontal step-wise movement of nanoprobe. The dashed white lines in the figure are corresponding to the shift of emission energy of one QD at different position relative to the nanoprobe.