Spin dynamics in two-dimensional arrays of quantum dots with different shapes

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Abstract. Electron paramagnetic resonance and spin echo methods are used to probe the spin dynamics in two-dimensional quantum dot (QD) arrays with different shape of nanoclusters. Two types of QD structures were investigated: 1) with single shaped QDs (hut-clusters, having the ratio of height $h$ to lateral size $l$, $h/l = 0.1$), and 2) with two groups of QDs, hut- and dome-clusters ($h/l = 0.2$). Both types of structures demonstrate the EPR signals from electrons localized in QD layers. The orientation dependence of EPR line width for first type structures is well described by model of spin relaxation through precession in the effective magnetic field, arising during tunneling between QDs due to structure-inversion-asymmetry. In the experiments on structures with dome-clusters the additional peculiarity, which cannot be explained in the framework of precession model, is observed. The different orientation dependencies can be explained by different localization degree of electrons in investigated structures. Spin echo measurements give the longest spin decoherence time for structures with single shaped QDs.

1. Introduction

A dimensionality reduction leads to the appearance of new effects, many of them are governed by symmetry of nanostructures. In particular, the symmetry of nanostructure has a crucial impact on the spin dynamics. The low symmetry of nanostructures can lead to appearance of additional spin relaxation mechanisms, which takes place in the arrays of tunnel-coupled quantum dots (QDs) with structure-inversion-asymmetry. At high density of QDs the overlapping between localized states is sufficient for coming in force the most efficient Dyakonov-Perel mechanism of spin relaxation [1]. In this case the spin relaxes during series of random tunneling events (in hopping transport) through precession in the effective magnetic field whose direction can be changed after each tunneling event. There are two ways to increase the spin relaxation time. The first follows from origin of the effective magnetic field. It relates to spin-orbit interaction and can vanish in more symmetrical structures. The second way is related to suppression of probability of tunneling between QDs, that can be realized by the creation of well-separated QD array.

The present paper is aimed to realize the increase of spin relaxation time in Si/Ge structures with quantum dots. To reach this goal we examine the electron spin coherence in QD structures with different shapes of nanoclusters.
2. Samples and Experiment

Samples were grown by molecular-beam epitaxy on n-Si(001) substrates with a resistivity of 1000 Ω cm. The QD structure with single shaped QDs (hut-clusters) was optimized with the aim to enlarge the electron binding energy in strain-induced potential well in Si near Ge QD. The vertical stack of four Ge QDs layers was inserted into the 0.6 µm epitaxial n-Si layer (Sb concentration 4·10^{16} cm^{-3}) at the distance of 0.3 µm from the substrate. The space charge spectroscopy confirms the localization of electrons in Si near QDs with binding energy ~ 50 meV [2]. The test structures uncovered by Si capping layer was investigated by scanning tunneling microscopy (STM). It was shown that the Ge islands have the shape of hut-clusters with the average lateral size l = 20 nm and height h = 2 nm. The density of QDs is ~ 10^{11} cm^{-2}.

The QD structure with dome-clusters contains 6 layers of QDs separated by 30 nm Si spacer layers. Each QD layer are formed by deposition of 7 ML Ge at the temperature T = 550°C. On top of the structure a 0.3 µm epitaxial n-Si layer (Sb concentration 4·10^{16} cm^{-3}) is grown, the same layer is grown under QDs layers. The STM of structure with single QD layer uncovered by Si shows the bimodal distribution of QDs (huts and domes). The average lateral size of dome-clusters is l = 50 nm and height h = 10 nm. The density of dome-clusters is 10^9 cm^{-2}. The hut-clusters are distributed between dome-clusters with density 10^{11} cm^{-2}.

Measurements were performed with a Bruker X-band EPR spectrometer at the temperatures T = 4.5 – 20 K. To avoid the needless EPR signal from dangling bonds (g = 2.0055) the passivation with atomic hydrogen was done before measurements. To increase the number of registrable spins the sandwiched sample was prepared.

The spin echo measurements were carried out at temperature 4.5 K in resonance magnetic field H = 3470 G (can be slightly varied ±5 G in dependence on resonance conditions) with direction corresponding to the narrowest EPR line width, for the first type structure H || Z, where Z is the [001] growth direction of the structure, for the second type structure the magnetic field deviates from Z on θ = 30°. A two-pulse Hahn echo experiment (π/2 − τ − π − τ− echo) was used to measure T_2 (a detailed explanation can be found in Ref. 3). In order to observe a longitudinal spin relaxation (corresponding time T_1), a different pulse sequence is applied (π − τ − π/2 − T − π − T− echo). In the first and second type of experiments, the durations of π/2 and π pulses were 60 ns and 120 ns, respectively; the interpulse time in the second experiment was kept T = 200 ns.

3. Results and Discussion

Both types of structures demonstrate the EPR signals from electrons localized in QD layers. The characteristic properties of EPR line (g-factor and EPR line width) depends on the type of the structure. The narrowest EPR lines (width ΔH ≈ 0.8 G) was detected for the first type structures. The principal values of g-factor are very close to g-factor values in uniaxially deformed Si (g_{zz} = 1.9995 ± 0.0001 and g_{xx} = g_{yy} = 1.9984 ± 0.0001). These EPR lines have a strong orientation dependence of EPR line width. When external magnetic field deviates from growth direction of the structure the EPR lines become broader and weaker, and for in-plane magnetic field they have the maximal width (ΔH ≈ 3 G). The second type structures show the wider EPR lines, having a weaker orientation dependence of g-factor and EPR line width. In contrast to the single shaped QD samples the maximum of EPR line width for the structures with dome-clusters is observed at θ = 60°. Here the additional peculiarity is observed: at the deviation of external magnetic field on small angles (up to θ = 30°) the EPR line width sharply decreases from 2.3 G to 1.5 G (Fig. 1). The g-factor behavior completely correlates with EPR line width change (Fig. 2). In the magnetic field perpendicular to the plane of QD array the g-factor value is g = 1.9991 ± 0.0001. At the deviation of magnetic field from Z-axis the g-factor value slightly grows up to g = 1.9992 ± 0.0001, then at Θ ≈ 60° the minimal g-factor value g = 1.9987 ± 0.0001 is observed, and in the end Θ = 90° the g-factor value increases and practically returns to its
initial value \( g = 1.9990 \pm 0.0001 \).

**Figure 1.** The angular dependence of EPR-line width for heterostructure with dome-clusters. For \( \Theta = 0 \) the magnetic field is parallel to the growth direction of the structure.

**Figure 2.** The angular dependence of electron g-factor for heterostructure with dome-clusters. For \( \Theta = 0 \) the magnetic field is parallel to the growth direction of the structure.

According to results of two-pulse Hahn echo experiment for both types structures the spin echo behavior can be described by superposition of two exponentially decaying functions (Fig. 3). The decay parameters give two times of spin dephasing: for the first type structure \( T_2^{(1)} \approx 0.9 \mu s \) and \( T_2^{(2)} \approx 20 \mu s \); for the second type structure \( T_2^{(1)} \approx 0.28 \mu s \) and \( T_2^{(2)} \approx 3 \mu s \).

The analysis of an inversion signal recovery, measured in three-pulse echo experiments, shows a non-exponential behavior for both single-shaped and two-shaped QD structures (Fig. 4). The experimental curve can be described by the superposition of two functions:

\[
M(t) = M_{0z} - M_{1}^{(1)} \exp(-\tau/T_1^{(1)}) - M_{2}^{(2)} \exp(-\tau/T_1^{(2)}),
\]

where \( M_{0z} \) is the equilibrium magnetization, \( M_{0z} = M_{0z}^{(1)} + M_{0z}^{(2)} \), \( M_{1}^{(1,2)} = M_{0z}^{(1,2)} - M_{1}^{(1,2)}(0) \), \( M_{2}(0) = M_{2}^{(1)}(0) + M_{2}^{(2)}(0) \) is the magnetization just after applying of an inverting \( \pi \)-pulse. The characteristic times obtained by fitting for the first type structure is about \( T_1^{(1)} \approx 400 \) ns and \( T_1^{(2)} \approx 10 \mu s \). For the second structure \( T_1^{(2)} \approx 2 \mu s \) and \( T_1^{(1)} \approx 20 \mu s \). For interpretation of results the existence of two electron groups with different spin relaxation times is suggested. For the first type structure both groups are formed by electrons in QD layers. These groups located in Si spacers with different width and have the different probability of electron hopping between Ge QDs [4]. In both groups of carriers the special relation between \( T_2 \) and \( T_1 \), \( T_2 \approx 2T_1 \), is observed. The unusual relation \( T_2 > T_1 \) confirms that the spin relaxation is caused by the interaction with the effective magnetic field arising due to the structure-inversion-asymmetry. For the second structure the spin echo signal is also formed by two groups of electrons. There are the free electrons in Sb-doped Si layers with isotropic g-factor 1.9987 and electrons localized in QD layers (\( g_{xx}=1.9995 \) and \( g_{yy}=1.9984 \)). For first group the relation \( T_2 < T_1 \) is observed, while for QD electrons the inverse relation \( T_2 < T_1 \) is obtained.

The orientation dependence of EPR line width for first type structures is well described by model of spin relaxation through precession in the effective magnetic field, arising during tunneling between QDs due to structure-inversion-asymmetry [5]. For the second type structure the orientation dependence of EPR line width is more complicated and can not be explained by simple model. For explanation of this orientation dependence we consider two mechanisms,
which control the EPR line width. The first mechanism is the motional narrowing of EPR line width. It controls the EPR line width at the initial stage of orientation dependence, up to $\theta = 30^\circ$. At $\theta = 0^\circ$ the tunneling transitions between QDs is suppressed due to wave function shrinking. With deviation of magnetic field from Z axis the effect of wave function shrinking becomes weaker, that leads to increasing probability of tunneling transitions between QDs. As result, we observe the narrowing of EPR line down to 1.5 G ($\theta = 30^\circ$). After this point the probability of tunneling transitions between QDs is sufficiently high for coming in force the second mechanism. It is the spin relaxation due to interaction with the spin-orbit fields that works also in the first type structures. Then at larger deviation angles $\theta$ the EPR line width behavior can be described in framework of the same model of spin precession, but with another parameters of model. The main difference is the dependence of characteristic fluctuation time (hopping time) $\tau_c$ on the external magnetic field $\tau_c = \tau_0 \exp(\alpha H^2)$. The theoretical approximation of the orientation dependence of EPR line width for second type structures in the range $\theta = 30^\circ - 90^\circ$ allows to estimate the effective radius of electron localization in second type structures, as 80 nm. While for the first type structures it is about 10-15 nm that is several times smaller than magnetic length at given external magnetic field ($H = 3470$ G) $\lambda \approx 50$ nm. Namely the last leads to independence of $\tau_c$ on the external magnetic field for the first type structures and more simple orientation dependence of EPR line width. The different localization degree leads to different spin dynamics in the structures under study and longer spin relaxation times (up to 20 $\mu$s) in first type structures.

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Figure 3. Results of two-pulse spin echo experiments (points) and approximation by superposition of two exponential functions (line). Time of transversal relaxation.

Figure 4. Results of three-pulse spin echo experiments (points) and theoretical approximation (line). Time of longitudinal relaxation.