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Unusual narrowing of the ESR line width in ordered structures with linear chains of Ge/Si quantum dots

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Electron states in ordered Ge/Si heterostructures with linear chains of quantum dots (QDs) were studied by the electron spin resonance (ESR) method. A new ESR signal with principal g-factor values $g_{zz} = 1.9993 \pm 0.0001$, $g_{xx} = g_{yy} = 1.9990 \pm 0.0001$ was detected. Unlike non-ordered QD structures, where ESR line broadening is usually observed (evidence of Dyakonov–Perel mechanism efficiency), the structures under study demonstrate the narrowing of ESR line when the external magnetic field deviates from the growth direction. The ESR line width is $\Delta H = 1.2$ Oe for perpendicular magnetic field (along the growth direction) and $\Delta H = 0.8$ Oe for in-plane magnetic field. The narrowing of ESR line can be explained by combination of two mechanisms. The first one is suppression of Dyakonov–Perel spin relaxation due to a settled direction of electron motion and finiteness of QD chains. The second one is cancelation of the wave function shrinking effect with decreasing the perpendicular component of the magnetic field.

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Progress in building of the structures with quantum dots [1–3] allows not only to use them as a working element of new electronic devices, but also to use these structures as a model for understanding the fundamental issues of matter. On the basis of artificial atoms one can create objects with different topology (e. g. 2D or 3D crystals, or molecules with different spatial packing), and investigate their electrical, optical and spin properties as functions of the structure of these objects. Recently it has been shown that the spatial configuration of quantum dots (QDs) in two-dimensional (2D) structures can affect the spin dynamics in the system [4]. Self-assembling of isolated groups of closely located QDs leads to a fourfold increase of the longitudinal spin relaxation time T_1 as compared with 2D tunnel-coupled QD arrays with homogeneous in plane distribution of QDs [5]. The effect is related to limitation of electron movement within isolated groups of several QDs.

In the present work, a change of spin dynamics at transition from 2D QD system to a system with finite QD linear chains was demonstrated by electron spin resonance (ESR) measurements. The unusual behavior of ESR line width was obtained: the ESR line narrows when magnetic field deviates from the growth direction of the structure. To explain this effect, we proposed a model of spin relaxation in finite QD linear chains. A principal difference between spin dynamics in infinite QD arrays and that in finite linear QD chains was found.

Samples were grown by molecular-beam epitaxy on the pre-patterned Si(100) substrates with resistivity $\sim 100\,\Omega\cdot$cm. The patterned stripes along the [100] direction with the period of 180 nm were fabricated by nano-imprint lithography and following irradiation by Ge^+ ions through the imprinted resist. The details of the trenches formation are described elsewhere [2]. On such substrates the QD structure with 5 QD layers was grown. Each QD layer was formed by the deposition of 7 Ge monolayers at the temperature of $T = 600\,°$C. Atomic force microscopy (AFM) of the structure with a single uncovered QD layer shows that QDs formed the linear chains (Fig. 1). Each QD has dome-like shape with the base size of about 80 nm and the height of about 16 nm. It is clearly seen that the QD lines are not continuous and consist of finite QD linear chains. Quantum dots are located closely to each other within each linear chain. Si spacers between QD layers were grown at temperature of $T = 500\,°$C. Transmission electron microscopy of covered samples shows that Si overgrowth results in twofold decrease of QD height indicating essential Ge/Si intermixing. On top of the struc-

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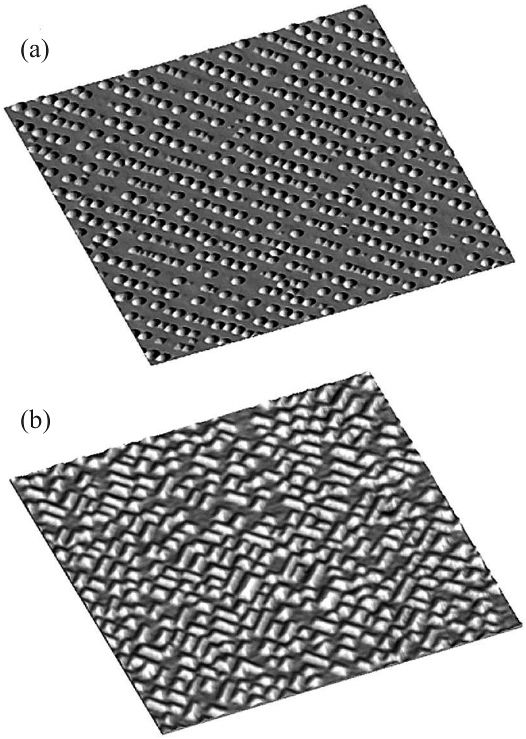

Fig. 1. (a) – $3\times 3\,\mu$m^2 AFM image of the uncovered sample with linear chains of Ge/Si QDs grown on pre-patterned substrate Si(001). (b) – $0.4\times 0.4\,\mu$m^2 STM image of typical 2D array of self-assembled Ge/Si QDs (growth details are described elsewhere [6])

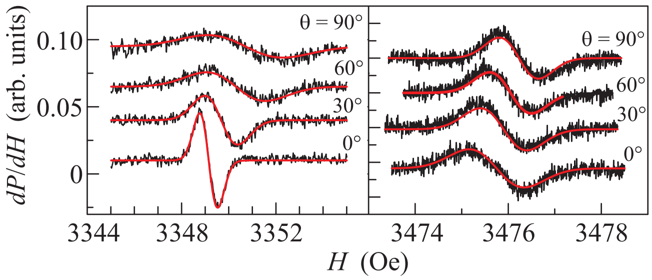

Fig. 2. ESR spectra at different orientations of magnetic field, microwave power $P = 0.063$ mW. Right panel: the results obtained for structures with QD lines (QD arrangement is shown in Fig. 1a). Left panel: the results obtained for typical 2D array of self-assembled Ge/Si QDs (QD arrangement is shown in Fig. 1b). For $\theta = 0^\circ$ the magnetic field is parallel to the growth direction of the structure [001], $\theta = 90^\circ$ corresponds to the magnetic field applied along the crystallographic direction [110]

ture, a 0.2 μm epitaxial n-Si layer with Sb concentration $\geq 5\cdot 10^{16}\,\mathrm{cm}^{-3}$ was grown. At low temperatures electrons should leave the impurities and fill the levels in QDs. The ESR measurements were performed with a Bruker Elexsys 580 X-band ESR spectrometer using a dielectric Bruker ER-4118 X-MD-5 cavity at a temperature of 4.5 K. The sample was glued on a quartz holder, then the entire cavity with the sample was maintained at a low temperature in a helium flow cryostat (Oxford CF935).

New ESR signal with principal g-factor values $g_{zz} = 1.9993 \pm 0.0001$, $g_{xx} = g_{yy} = 1.999 \pm 0.0001$ was detected. Anisotropy of g-factor confirmed that electrons were localized in strained Si regions near QD apexes. The principal values of g-factor and its anisotropy are smaller than that of QD structures grown at lower temperature [6], which is testimony to the larger Ge/Si intermixing and smaller strain in the structures under study. A narrowing of the ESR line in the tilted magnetic fields was observed. The ESR line width is $\Delta H \approx 1.2$ Oe for perpendicular magnetic field (along growth direction Z) and $\Delta H \approx 0.8$ Oe for in-plane magnetic field, $H\|[110]$ (Fig. 2, right panel). This behavior is unusual for tunnel-coupled QD structures. In existing works the opposite effect was observed: the ESR lines were broadened when the magnetic field deviates from Z. For example, in our previous study of a dense array of self-assembled QDs the ESR line width changed four times with sample rotation in the magnetic field [6]. In the magnetic field applied along the growth direction the ESR line width is $\Delta H \approx 0.75$ Oe and, in the perpendicular magnetic field, it increases up to $\Delta H \approx 3$ Oe (see Fig. 2, left panel). Such broadening is the first sign of Dyakonov–Perel spin relaxation mechanism [7] domination and occurs due to the special in-plane arrangement of spin-orbit fields (Bychkov–Rashba fields [8]), leading to the anisotropy of spin relaxation processes in the system. It is easy to show that the transverse spin relaxation time T_2 should decrease in such type 2D systems at deviation of external magnetic field from the growth direction [9]. Since $\Delta H \sim 1/T_2$ for homogeneously broadened ESR lines, then the special orientation dependence of ESR line width is observed. This mechanism can determine spin dynamics in the low-conduction QD system where electron transport occurs in the hopping regime [10], as well as in a 2D electron gas system with high conductivity [11].

The main factor providing the efficiency of Dyakonov–Perel mechanism in the QD system is the randomness of electron tunneling transitions between QDs due to the disorder of the self-assembled QD array. In the structures under study the ESR line width broadening is absent. This effect can be associated with a change of spin dynamics caused by the ordering of QDs in the linear chains. Such type of ordering defines the directions of electron tunneling to be only along

QD lines. Indeed the recent results of conductance measurements of similar structures with QD lines demonstrate a strong conductance anisotropy [12]. The conductance along QD lines is two orders larger than in perpendicular direction. During tunneling along a linear QD chain the electron spin rotates around the effective magnetic field direction determined by vector product $\mathbf{H}_{\text{eff}} \sim [\mathbf{n} \times \mathbf{e}_z]$, where $\mathbf{n}$ is the tunneling direction, $\mathbf{e}_z$ is the growth direction of the structure [6]. For example the electron tunneling from the beginning to the end of the QD chain is accompanied by the clockwise spin precession, and the tunneling in the opposite direction – by the anticlockwise one. In the ideal case of the infinite chain of identical tunnel-coupled QDs this does not lead to a loss of spin orientation in zero magnetic field. The nonzero external magnetic field applied along $\mathbf{e}_z$ provides an additional Larmor precession and the spin relaxation process depends on the relation between the frequency of Larmor precession ω_{L} and the mean time of hopping between dots τ_h. Recently we have demonstrated that for a ring-like group of closely spaced QDs at $\omega_l \tau_h \ll 1$ the stabilization of S_z-polarization occurs [4]. Now we simulate the spin relaxation process for the linear chain of QDs.

The model is based on the existing results of the experimental study of hopping transport in Ge/Si QD structures [12, 13] and the theoretical study of spin relaxation during hopping through QD arrays [14]. The model includes a strong tunnel coupling between QDs in the line. Hopping between any neighboring QDs is permitted with an equal probability for the back and forth motion. Each tunneling transition is accompanied by a spin rotation by the small fixed angle $\alpha = 0.01$. This value is taken based on results of tight binding calculation performed for holes localized in Ge/Si QDs [14] and scaled for electrons taking into account the weakness of spin-orbit interaction in Si. The external magnetic field provides the Larmor precession between tunneling events. The time intervals between tunneling events are distributed exponentially with a mean value τ_h. In our recent study of similar structures [6] the value of τ_h was estimated to be of order 10^{-11} s. Such short times are a consequence of small width and height of the potential barrier separating electron states in neighboring closely located QDs (width ~ 10 nm, height ~ 10 meV according to simulations of strain-induced potential). The spin dynamics at the hopping between dots was simulated by the Monte-Carlo method for different numbers of QDs in the line and different $\omega_{\text{L}} \tau_h$. For comparison, the simulation for square lattice was performed. Despite of the ordered positioning of QDs in the last case the electron transport through this structure must follow a random walk in 2D plane, since the direction of each hop between QDs is chosen using a random number generator.

The simulation results show that for infinite line and square lattice the behavior of T_2 is conventional, T_2 decreases when the external magnetic field deviates from the growth direction (Fig. 3). This corresponds to experimental broadening of the ESR line in 2D systems with fluctuating in-plane spin-orbit fields [6, 11]. The only difference between a square lattice and an infinite line consists in a twofold increase of spin relaxation times. This can be easily explained by the exclusion of one degree of freedom, transition from a two-dimensional to one-dimensional case. However, for the finite line with a QD number $N \sim 10$ dots the unusual behavior of T_2 is observed, T_2 increases for tilted magnetic fields (Fig. 3). To understand this, let us use the simple picture (Fig. 4) for the 3 QD line. Hopping between dots is accompanied by the spin turning at the small angle α around the effective magnetic field direction. Let $\mathbf{h}$ be a unit vector along the effective magnetic field. Let us introduce the reference frame that rotates by the same angle as spin at each hopping event and thereby eliminates the spin rotation caused by hopping. In this system only Larmor precession can change the spin orientation. But the axis of Larmor precession deviates at the small angle from the original direction at each hopping event, and this provides spin relaxation. The angular velocity of Larmor precession $\mathbf{\Omega}'$ in the new reference frame is

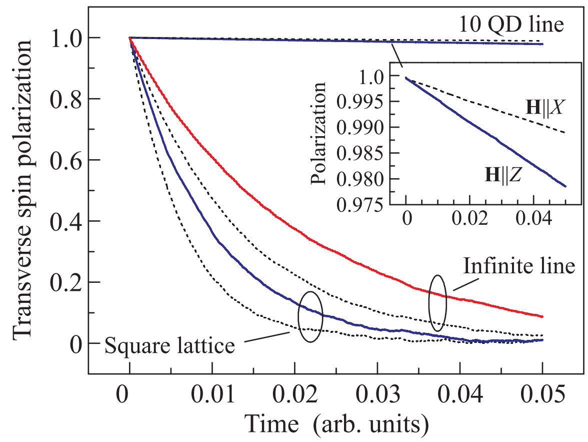

Fig. 3. Results of transverse spin relaxation simulation for a square lattice, infinite QD line and a 10 QD line for two orientations of magnetic field, $\mathbf{H}\|Z$ (solid lines) and $\mathbf{H}\|X$ (dashed lines). The direction of QD line is taken like in the experiments at the angle of 45° to the X direction, [100]. The relation between the hopping time and Larmor frequency is taken as $\omega_{\text{L}} \tau_h = 0.01$. For the 10 QD line the unusual anisotropy of transverse spin relaxation time is observed

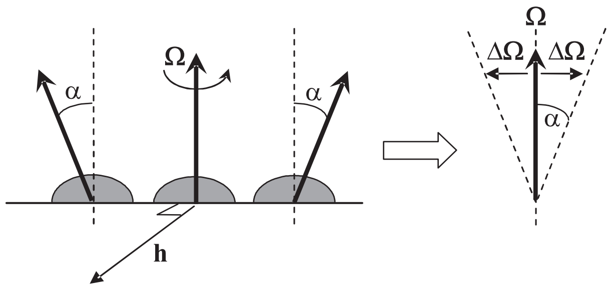

Fig. 4. Elimination of spin rotation caused by hopping by means of transition to the reference frame rotating together with the spin around the effective magnetic field at each hopping event. In this reference frame the axis of Larmor precession deviates at the small angle from the original direction at each hopping event, and this provides the spin relaxation. The angular velocity of Larmor precession $\mathbf{\Omega}'$ in the new reference frame is $\mathbf{\Omega}' = \mathbf{\Omega} + \Delta\mathbf{\Omega}$, where $\mathbf{\Omega}$ is the angular velocity in the fixed reference frame, $\Delta\mathbf{\Omega} = 0$ at the central QD and $\Delta\mathbf{\Omega} \approx \pm\alpha[\mathbf{\Omega} \times \mathbf{h}]$ at the ends of the QD chain

$\mathbf{\Omega}' = \mathbf{\Omega} + \Delta\mathbf{\Omega}$, where $\mathbf{\Omega}$ is the angular velocity in the fixed reference frame, $\Delta\mathbf{\Omega} = 0$ at the central QD and $\Delta\mathbf{\Omega} \approx \pm\alpha[\mathbf{\Omega} \times \mathbf{h}]$ at the ends of the QD chain. The rate of spin relaxation is proportional to the mean square of $\Delta\Omega$. This provides the following orientation dependence of spin relaxation times T_1 and T_2:

$$T_{1,2}^{-1} \sim \sin^2 \phi,$$

where ϕ is the angle between the external magnetic field and the effective magnetic field, i.e. between $\mathbf{\Omega}$ and $\mathbf{h}$. So, deviation of magnetic field from the growth direction (decreasing of ϕ) results in slower spin relaxation. In our experimental conditions the external magnetic field changes its direction from [001] to [110], that corresponds to varying of angle ϕ from 90° to 45°. So, for external magnetic field with in-plane orientation the spin relaxation time should increase as compared to the case of magnetic field along the growth direction. This corresponds to a narrowing of the ESR line observed in experiments.

There is one more possible mechanism of ESR line narrowing in the structures under study. This is a change of electron localization radius through the wave function shrinking effect [15]. The localization radius in the investigated structures is comparable with the magnetic length λ. Magnetic length $\lambda = \sqrt{c\hbar/eH}$ in our experimental setup ($H = 3470$ Oe) is about 45 nm, that is very close to the size of QD base edges (≈ 50 nm) and, correspondingly, to the size of electron wave function. In these conditions, the magnetic field, applied along the growth direction [001], can effectively shrink the tails of the electron wave functions, resulting in the enhancement of the electron localization. In the tilted magnetic field the shrinking effect vanishes and the localization of electrons becomes weaker. This leads to a more effective overlapping of wave functions of electrons localized in neighboring QDs. An increase of the overlapping can promote two possible processes leading to the narrowing of the ESR line. The first process is the electron hopping between QDs. The hopping results in the ESR line narrowing, provided that electrons move along the same trajectories determined by QD linear chains. Thus, the effective averaging of local magnetic fields produced by nuclear spins ^{29}Si and averaging of the QD parameters take place. The second process is the averaging through the exchange interaction between electrons in neighboring QDs. This interaction also depends on the overlapping of wave functions and becomes more intensive at the deviation of magnetic field from the Z direction. As a result the narrowest ESR lines are observed for in-plane magnetic field.

However, if one take into consideration only the second mechanism of narrowing, then the orientation dependence of ESR line width will be unmonotonic. Firstly, one will observe the ESR line narrowing then at larger angles θ the ESR line is broadened, like in the recent work [4], where spin relaxation in inhomogeneous QD arrays was studied. Then the presence of finite QD linear chains in the samples is responsible for the observed behavior of ESR line width.

To summarize, we observed the unusual narrowing of ESR line in ordered structures with linear QD chains. The mechanism of this narrowing was proposed. The organization of QDs in finite lines leads to suppression of Dyakonov–Perel mechanism that rules the spin behavior in dense 2D QD arrays.

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