Spin relaxation in silicon coupled quantum dots

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We present a detailed investigation for spin relaxation processes in silicon coupled quantum dots. Low-field magnetoconductance measurements have been employed to deduce phase dephasing and spin relaxation rates. On the basis of the dephasing theory containing triplet channel interaction, we have demonstrated that small energy transfer scattering process is the dominant dephasing mechanism, and strong electron-electron interaction results in an interdot spin-exchange relaxation process. Triplet-singlet relaxation is found to be another important spin relaxation process in the inner quantum dots, taking into account the triplet-singlet splitting induced by spin-orbit coupling. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3167817]

The interest on the electron spin in semiconductors has been revived in recent years due to potential applications in spintronics[1] and quantum information processing.[2] Among all the semiconductor structures, quantum dots (QDs) of diverse geometries are considered as promising candidates for implementation of quantum computation devices because the electronic, magnetic, and optical properties can be controlled through the modern growth and nanofabrication techniques.[3] Up to now, extensive investigations have been carried out on electron spins of GaAs, in which the electronic states are well understood.[1,4] Compared with GaAs QDs, less work has been executed on silicon (Si) QDs, though bulk Si has long been predicted a superior semiconductor for spintronics with enhanced lifetime and transport length due to lattice inversion symmetry and low spin-orbit scattering in addition to the market entrenchment in electronics.[5]

So far, several studies have addressed spin related electron transport and/or charge sensing characteristics in Si double QDs, exhibiting the existence of triplet-singlet spin relaxation process.[6–9] Based on these experimental results, the triplet-singlet spin relaxation mechanism has been discussed theoretically in Si QDs.[10] In contrast, less attention has been paid on the experimental spin relaxation rate in Si QDs to illustrate spin relaxation mechanisms in details and further support the development of spin relaxation theory. Therefore, more direct experimental evidence of spin dependent transport through Si multiple QDs are required to investigate spin lifetime, coherence, and entanglement in Si nanostructures. In hydrogenated nanocrystalline Si (nc-Si:H),[11] natural Si QDs embedded in thin (about two to four atomic spacings) amorphous Si tissues with controllable energy band can form the lateral coupled or isolated Si QDs structure via impurity doping.[12] Magnetoconductivity measurements on nc-Si:H with lateral QDs demonstrate the existence of weak antilocalization (WAL) phenomena,[13] from which the spin relaxation rates can be extracted by the aid of WAL theory in low-dimensional semiconductor systems.[14]

A Si coupled QD structure was prepared in a rf (13.56 MHz and power of 75 W) capacitive-coupled plasma enhanced chemical vapor deposition system from silane (SiH_4) and hydrogen (H_2) on weak *p*-type crystalline Si substrate at a temperature of 250 °C with a chamber pressure of 1.0 Torr. The percentage content of silane (SiH_4/SiH_4+H_2) was about 1.0%. Phosphine was used as dopant gas with percentage content (PH_3/SiH_4) of 0.8%. High-resolution transmission electron microscopy revealed a long-range-ordered Si nanocrystals structure.[15] X-ray diffraction and Raman measurements confirmed the good quality of Si QDs system with an average grain size of ~5 nm and a crystalline fraction of 51.7%.[15] Vertical current-voltage[16,17] and magnetic-field dependent Hall effect measurements[18] demonstrated an average lateral confining potential of ~10 meV and minimum vertical confining potential of ~60 meV, making the movement of electrons quasi-two-dimensional (2D). Temperature-dependent magnetoconductivity experiments were carried out to probe the electron spin relaxation characteristics under an Oxford Instruments superconductive magnet.

Figure 1(a) presents the temperature dependence of conductance G (squares) of the Si coupled QDs structure without magnetic field ($B=0$ T). Based on previous reports that hopping process is expected to be the dominant transport mechanism in Si coupled QDs below 50 K,[18] we made a least-squares fit to the measured conductance in accordance with the hopping conduction equation[19] $\sigma(T)=\sigma_0 \exp[-(T_0/T)^{-[1/(d+1)]}]$ (here d is the dimensionality, σ_0 is the prefactor, and T_0 is a characteristic temperature). As shown in this figure, the experimental data can be well fitted when

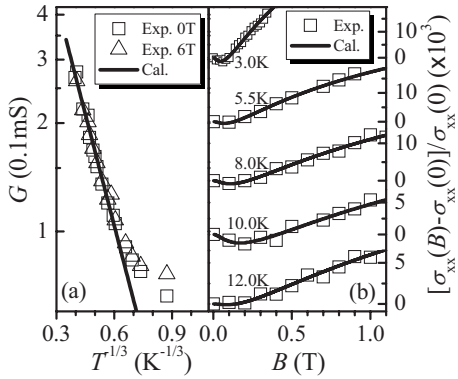

FIG. 1. (a) Conductance $G(T)$ vs $T^{-1/3}$ at magnetic fields of 0 and 6 T of the Si coupled QDs structure. The solid line is the least-squares fitting to the hopping behavior. (b) Experimental and calculated magnetoconductivities.

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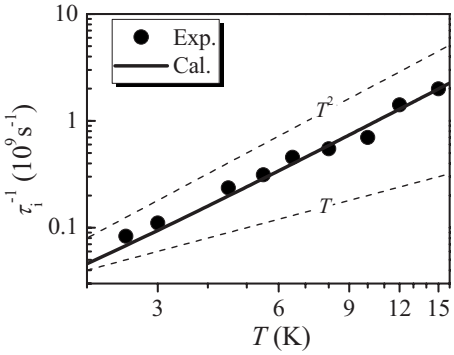

FIG. 2. Experimental and calculated inelastic scattering rate of Si coupled QDs structure. Dashed lines indicate slopes corresponding to T and T^2.

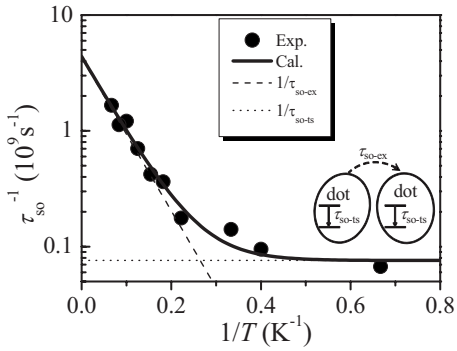

FIG. 3. Experimental and calculated spin relaxation rate of Si coupled QDs structure. Dashed line indicates $\tau_{so\text{-}ex}^{-1}$, and dotted line represents $\tau_{so\text{-}ts}^{-1}$. The inset illustrates the spin relaxation mechanisms in Si coupled QDs.

$d=2$. Such good fitting demonstrates the existence of quasi-2D hopping conducting behavior in Si QDs as we mentioned above, which is induced by strong vertical and weak lateral confining potentials. We have also shown in Fig. 1(a) the temperature-dependent experimental conductance at $B=6$ T (triangles). Since weak localization has been quenched at high magnetic fields, the trivial difference between $B=0$ and $B=6$ T illustrates the existence of strong electron-electron (e-e) interactions, which can be easily understood by the spatially coupled wave function due to the low lateral confining potential. Moreover, e-e interactions will become stronger with decreasing temperature, where Shklovskii–Efros law will play a major role in the electron transport process,[20] leading to the derivation of the hopping conduction fitting results from the experimental data below 3 K in Fig. 1(a).

In materials where hopping is the mechanism responsible for conductivity, positive (negative) magnetoconductivity (magnetoresistance) characteristic has often been observed.[19] The positive magnetoconductivity has been reported in Si QDs structures.[18] Moreover, besides it, the magnetoconductivity at low magnetic fields (<1 T) exhibits much more information related to quantum interference in Si QDs structure. Figure 1(b) displays experimental magnetoconductivities (squares) measured under low magnetic fields (<1 T) at temperatures of 3.0, 5.5, 8.0, 10.0, and 12.0 K, respectively. In addition to the overall positive magnetoconductivity component, there is a very narrow negative magnetoconductivity region under extremely low magnetic fields (<0.15 T), clearly demonstrating WAL effects. The observation confirms the existence of spin-orbit interaction, since the WAL effect appears in the presence of relatively stronger spin-orbit interaction. Here, it should be noted that unlike bulk Si, the inhomogeneity of the confining potential and grain size in Si QDs would result in the structure asymmetry and the incorporation of H atoms in the growth, and its random distribution leads to the inversion asymmetry. Together with in-plane impurity potential, they would yield intrinsic and extrinsic spin-orbit interaction, which eventually suppress the inelastic scattering and cause a change in the sign of the low field magnetoconductance. In order to thoroughly understand the WAL effects and spin related information in Si coupled QDs, theoretical calculation has been performed to interpret quantitatively the experimental results.

Considering that the diffusion limit is maintained in Si coupled QDs ($k_B T\tau/\hbar=0.020 \ll 1$ at $T=15$ K, where τ is the mean free time obtained from the mobility), we employ the simple Hikami-Larkin-Nagaoka (HLN) theory[21] to simulate the experimental data, although there are many more precise theories to describe the WAL behavior in 2D semiconductors for both diffusion limit and nondiffusion approximation.[22] According to HLN theory, the correction to the sheet conductance $\Delta G_{\square}(B)$ due to the WAL is described as

$$\Delta G_{\square}(B) = \frac{e^2}{2\pi^2\hbar} \times \left[\Psi\left(\frac{1}{2} + \frac{B_e + B_{so}}{B}\right) - \frac{3}{2}\Psi\left(\frac{1}{2} + \frac{B_i + \frac{4}{3}B_{so}}{B}\right) + \frac{1}{2}\Psi\left(\frac{1}{2} + \frac{B_i}{B}\right) \right], \quad (1)$$

where Ψ is the digamma function, and B_e, B_i, and B_{so} are the elastic, inelastic, and spin-orbit scattering fields, respectively, each corresponding to an associated electron diffusion length $L_x=\sqrt{\hbar/4eH_x}$($x=e$ for elastic, i for inelastic, and so for spin-orbit scatterings). The calculated results shown in Fig. 1(b) as solid curves are in good agreement with the experimental data at various temperatures in the low magnetic field region. There are three parameters in the HLN fitting: B_e, B_i, and B_{so}. Temperature-dependent inelastic scattering rate τ_i^{-1} and spin-orbit scattering rate τ_{so}^{-1} can thus be yielded with the diffusion coefficient D obtained from the mobility, which will be used to analyze the quantum interference process in Si coupled QDs. Figure 2 shows temperature dependence of inelastic scattering rate τ_i^{-1} (circles) extracted from the WAL effects, together with two guiding lines of T and T^2 dependences. We note that the experimental data fall between $\tau_i^{-1} \propto T^2$ and $\tau_i^{-1} \propto T$, which is in good agreement with the previous reported results that $L_i \propto T^{-0.58}$ (here L_i is the dephasing length, and thus $\tau_i \propto T^{-1.2}$).[13] Figure 3 displays the extracted spin-orbit scattering rate τ_{so}^{-1} (circles) versus $1/T$. Within the temperature region of 3–15 K, the spin-orbit scattering rate decreases exponentially with $1/T$, then tends to a steady value with T below 3 K. Compared with the results shown in Fig. 2, the spin-orbit scattering rate is larger than the inelastic scattering rate until temperature rises above 10 K, where the inelastic scattering rate is comparable to the spin-orbit scattering rate, and anomalous (negative) magnetoconductivity originating from WAL effects disappears.

It is well-known that for 2D semiconductor at low temperatures, the dominating dephasing process is quasielastic e-e scattering mechanisms.[23] The standard result for the e-e scattering rate at relative high temperatures $k_B T\tau/\hbar \gg 1$ is proportional to T^2 (large energy transfer processes, or ballis-

tic term in Ref. 24), while at low temperature, where small energy transfer scattering processes dominate ($k_BT\tau/\hbar \ll 1$), it is proportional to T.[23] However, in all these calculations, only the contribution of singlet channel interaction is considered, and this kind of approximation is not accurate to describe the *e-e* scattering behavior in Si coupled QDs system with strong *e-e* and spin-orbit interactions. Narozhny *et al.*,[24] have considered the dephasing scattering rate including triplet channel interaction in both diffusive ($k_BT\tau/\hbar \ll 1$) and ballistic ($k_BT\tau/\hbar \gg 1$) regimes. They have predicted that the temperature dependence of τ_i^{-1} in the small energy transfer (SET) scattering processes ($k_BT\tau/\hbar \ll 1$) is given by

$$\begin{aligned}\tau_i^{-1}(T) = &\{1 + 2(F_0^{\sigma})^2/[(1 + F_0^{\sigma})(2 + F_0^{\sigma})]\} \\ &\times [k_BT/(G_0\hbar)]\ln[G_0(1 + F_0^{\sigma})] \\ &+ (\pi/4)[1 + 3(F_0^{\sigma})^2/(1 + F_0^{\sigma})^2] \\ &\times [(k_BT)^2/(\hbar E_F)]\ln(E_F\tau/\hbar), \end{aligned} \quad (2)$$

where F_0^{σ} is the interaction constant in the triplet channel, $G_0 \equiv 2\pi\hbar G/e^2$, and E_F is the Fermi energy. The Fermi energy in the Si coupled QDs structure is 68.8 meV calculated from $E_F = (\hbar\sqrt{2\pi n_e})^2/2m^*$ with $n_e = 9.2 \times 10^{12}$ cm^{-2} and $m^* = 0.32m_0$.[25] The solid line in Fig. 2 is the theoretical value of τ_i^{-1} calculated from Eq. (2) with the above experimentally determined E_F, G_0, τ, and a restricted fitting parameter $F_0^{\sigma} = -0.2823$, showing the good agreement with the experimental results. Such a good agreement confirms that the dephasing process in Si coupled QDs is dominated by the SET mechanism, in which triplet channel interaction plays an important role.

When electron hops from one Si dot to another, it experiences not only phase relaxation (dephasing) mentioned above, but also spin relaxation (scattering) process. The negative value of F_0^{σ} shows an existence of spin-exchange interaction, which makes a contribution to spin relaxation process. It is well-known that strong spin-exchange interaction of $F_0^{\sigma} = -1$ will make spin aligned.[26] In Si coupled QDs structure, the spin-exchange interaction is not so strong to align the spin ($F_0^{\sigma} = -0.2823 > -1$) but already enough to cause spin relaxation/diffusion, resulting in the contribution of a temperature-independent relaxation rate $\tau_{\text{so-ex}}^{-1}$.[27] On the other hand, the spin-orbit coupling can mix orbit and spin states, and make the triplet-singlet spin splitting.[28] The triplet-singlet spin relaxation process has been described in Si single and/or double QDs,[10] and can be expressed as an exponential function of the reverse of temperature $\tau_{\text{so-ts}}^{-1} = A\exp(-\varepsilon/k_BT)$ with ε representing triplet-singlet splitting.[29] This law is in line with the observed exponential decay behavior of the spin relaxation rate from 3 to 15 K in Fig. 3. Due to the existence of spin-orbit coupling and the fact that our Si QDs structure has an abundant isotope with zero nuclear spin, we have ignored the spin relaxation mechanism by nuclear spin, which is thought to be an important spin relaxation mechanism in many QDs structures.

Considering cocontribution of the spin-exchange relaxation and triplet-singlet spin relaxation in Si coupled QDs structure, we can employ the equation of $\tau_{\text{so}}^{-1} = \tau_{\text{so-ex}}^{-1} + \tau_{\text{so-ts}}^{-1}$ to simulate the experimental spin-orbit relaxation rate. The solid curve in Fig. 3 is calculated results with the energy gap ε between the singlet and triplet states of about 1.3 meV. The experimental fact that both the temperature-independent spin-exchange interaction and temperature-dependent triplet-singlet spin relaxation mechanisms coexist in the Si coupled QDs system demonstrates that spin relaxation in Si coupled QDs structure happens not only in the hopping process from one dot to another but also in the inner of Si QDs, as shown in the inset of Fig. 3.

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