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Electron spin relaxation in GaAs$_{1-x}$Bi$_x$: Effects of spin-orbit tuning by Bi incorporation

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The electron spin relaxation in n-type and intrinsic GaAs$_{1-x}$Bi$_x$ with Bi composition $0 \le x \le 0.1$ is investigated from the microscopic kinetic spin Bloch equation approach. The incorporation of Bi is shown to markedly decrease the spin relaxation time as a consequence of the modification of the spin-orbit interaction. We demonstrate that the density and temperature dependences of spin relaxation time in GaAs$_{1-x}$Bi$_x$ resemble the ones in GaAs. Meanwhile, the Bir-Aronov-Pikus mechanism is found to be negligible compared to the D'yakonov-Perel' mechanism in intrinsic sample. Due to the absence of direct measurement of the electron effective mass in the whole compositional range under investigation, we further explore the effect of a possible variation of electron effective mass on the electron spin relaxation.

Recently, the highly mismatched semiconductor alloys have received considerable interest due to their unusual compositional dependences of electronic and spintronic properties.[1–9] These alloys are typically obtained by isoelectronic doping of very light or very heavy elements into the traditional binary. For example, dilute incorporation of N or Bi into GaAs is found to induce a strong reduction of the band gap.[1–5] This property makes them a focus of current researches in long-wavelength optoelectronics and shows great potential to work as highly efficient solar cell and thermoelectric power generators.[2–5,9–11] The alloys GaAs$_{1-x}$Bi$_x$, which have been less investigated than GaAs$_{1-x}$N$_x$, have great potential for device applications.[9,13,14] One reason is that the incorporation of Bi modulates the band gap primarily through distorting the valence-band edge and leaves the conduction band almost unchanged, while N in GaAs mainly modulates the conduction-band edge and results in a substantial degradation of electron mobility.[7,12–14] Moreover, a strong enhancement of spin-orbit splitting energy is observed in GaAs$_{1-x}$Bi$_x$.[4–6] This indicates a new way of tailoring the spin-orbit coupling (SOC) through Bi incorporation and hence suggests its application in the field of spintronics.[4–6] A systematical calculation of the electron spin lifetime in GaAs$_{1-x}$Bi$_x$ is of fundamental importance for both theoretical and application purposes.

In this work, we employ the microscopic kinetic spin Bloch equation (KSBE) approach,[15–18] which has been demonstrated to be successful in the study of spin dynamics in semiconductors,[18] to calculate the electron spin relaxation time (SRT) in n-type and intrinsic GaAs$_{1-x}$Bi$_x$. We focus on the electron spin relaxation from the D'yakonov-Perel' (DP) mechanism[19] in n-type case and further take into account the Bir-Aronov-Pikus (BAP) mechanism[20] in the intrinsic condition. Both mechanisms are shown to be pronouncedly enhanced in GaAs$_{1-x}$Bi$_x$ and result in enhanced spin relaxation rate when Bi composition increases. Meanwhile, the BAP mechanism is recognized to be still negligible in the intrinsic case as in GaAs.[21] The density and temperature dependences of the SRTs are explicitly studied, where the features are found to resemble those in GaAs.[21] In addition, we discuss the impacts of possible variations of the electron and/or hole effective masses on the spin relaxation in GaAs$_{1-x}$Bi$_x$.

The KSBEs derived from the nonequilibrium Green function method reads[15,16,18]

$$\partial_t \rho_{\mathbf{k}} = \partial_t \rho_{\mathbf{k}}|_{\mathrm{coh}} + \partial_t \rho_{\mathbf{k}}|_{\mathrm{scat}}, \qquad (1)$$

in which $\rho_{\mathbf{k}}$ is the single-particle density matrix of electrons with its diagonal term representing the distribution of each spin band and the off-diagonal term denoting the correlation between the two spin bands. The coherent terms $\partial_t \rho_{\mathbf{k}}|_{\mathrm{coh}}$ describe the spin precession of electrons due to the Dresselhaus SOC[22] as well as the Hatree-Fock Coulomb interaction. $\partial_t \rho_{\mathbf{k}}|_{\mathrm{scat}}$ stands for the scattering terms, including the electron-impurity, electron-phonon, electron-electron and electron-hole Coulomb scatterings, and also the electron-hole exchange scattering. The explicit expressions of both coherent and scattering terms can be found in Ref. 21.

The incorporation of Bi influences the spin relaxation via both the DP and BAP mechanisms through the modulations of the band gap E_g and the spin-orbit splitting Δ_{SO}. The compositional dependences of E_g and Δ_{SO} are obtained by linear fittings of the experimental results summarized in Fig. 3(c) of Ref. 5: $E_g = (1.435 - 7.0x)$ eV, and $\Delta_{\mathrm{SO}} = (0.329 + 5.27x)$ eV. The electron effective mass m_e^* is taken to be fixed to its value in GaAs m_{GaAs}, which is supported by the direct Shubnikov-de Haas measurement of m_e^{*}[14] and also the mobility experiment.[13] The coefficient of the Dresselhaus SOC takes the form $\gamma_D = (4/3)(m_e^*/m_{\mathrm{cv}})(1/\sqrt{2m_e^{*3}E_g})(\eta/\sqrt{1-\eta/3})$, in which $\eta = \Delta_{\mathrm{SO}}/(E_g + \Delta_{\mathrm{SO}})$, and m_{cv} is a constant

close in magnitude to the free-electron mass m_0.[23] Besides, the longitudinal-transverse splitting, which is a scale of the electron-hole exchange interaction (we neglect the short-range electron-hole exchange interaction due to its marginal effect[21]), is given as $\Delta E_{\mathrm{LT}} = 2e^2\hbar^2 E_P/(3\pi\epsilon_0\kappa m_0 a_{\mathrm{Bohr}}^3 E_g^2)$. Here a_{Bohr} denotes the exciton Bohr radius in bulk, κ is the static dielectric constant, and E_P is the band parameter.[24] In GaAs$_{1-x}$Bi$_x$, κ, E_P and also the hole effective mass (which influences a_{Bohr} together with the electron-hole Coulomb scattering) are still not well investigated and are assumed to be the values in GaAs. In Fig. 1, we plot the Bi-compositional dependences of γ_D and ΔE_{LT}. It is seen that both γ_D and ΔE_{LT} are markedly enhanced with the increase of Bi composition and reach up to roughly four times of their original values when $x = 10$ %. Therefore, the electron spin relaxation from both the DP and BAP mechanisms is expected to be remarkably strengthened thanks to the incorporation of Bi in GaAs. However, considering that the BAP mechanism is demonstrated to be negligible compared with the DP mechanism in the intrinsic GaAs,[21] its effect is still expected to be marginal in the presence of Bi incorporation. In the following, we calculate the SRT in n-type and intrinsic GaAs$_{1-x}$Bi$_x$ from the KSBE approach with all scatterings explicitly included.

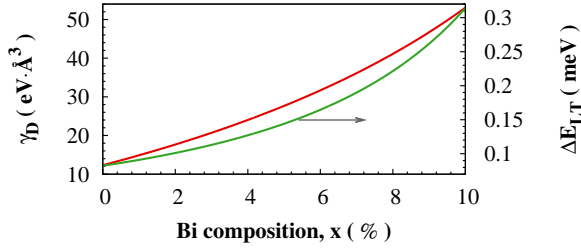FIG. 1: The SOC parameter γ_D and the longitudinal-transverse splitting ΔE_{LT} as function of Bi composition. Note that the scale of ΔE_{LT} is on the right hand side of the frame.

We first investigate the Bi-compositional dependence of the SRTs in GaAs$_{1-x}$Bi$_x$. In Fig. 2, the SRTs as function of the Bi composition are plotted for both n-type and intrinsic conditions at $T = 300$ K. We consider three electron densities from n-doping or photoexcitation, i.e., 10^{16}, 10^{17} and 10^{18} cm^{-3}, corresponding to the nondegenerate, intermediate, and degenerate regimes, respectively. One observes that the SRTs significantly decrease with the increase of Bi incorporation. In all conditions, the SRTs decrease with the increase of Bi composition and reach values around one order of magnitude smaller than the corresponding Bi-free ones at $x = 10$ %. Another feature shown in Fig. 2 is that, for both n-type (solid curves) and intrinsic (dashed curves) cases, the SRTs first increase and then decrease with increasing electron densities. This nonmonotonic density dependence of the SRT resembles the one in GaAs and is at-
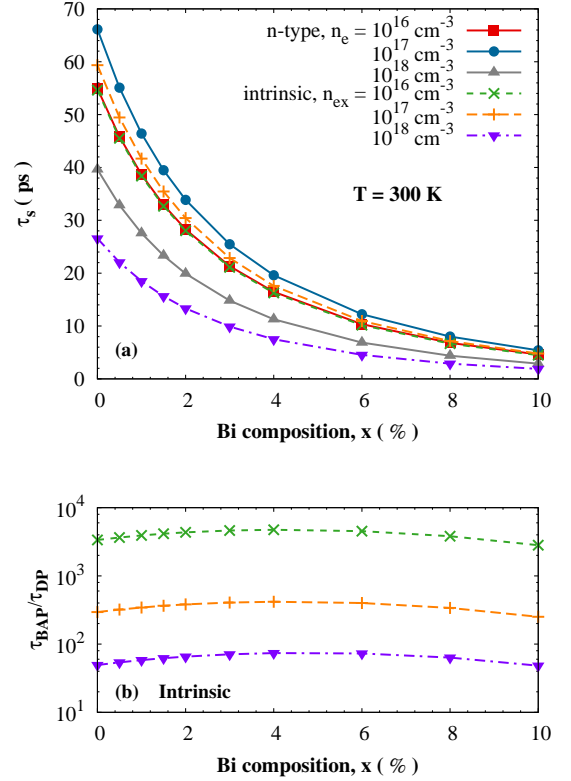

FIG. 2: (a) SRT τ_s as function of Bi composition at temperature $T = 300$ K. For n-type condition with electron densities $n_e = 10^{16}$, 10^{17} and 10^{18} cm^{-3} (solid curves) and for intrinsic condition with photoexcitation densities $n_{\mathrm{ex}} = 10^{16}$, 10^{17} and 10^{18} cm^{-3} (dashed curves). (b) The ratio of the SRT due to the BAP mechanism τ_{BAP} to that due to the DP mechanism τ_{DP} corresponding to the intrinsic condition in (a). Note that the same color and type of line and point are used for the corresponding photoexcitation density.

tributed to the crossover from the nondegenerate to degenerate limit.[21,25] It is also noticed that the curves corresponding to the n-type and intrinsic cases at electron density 10^{16} cm^{-3} coincide with each other. The underlying physics is that at high temperature and low electron density, the electron-phonon scattering is dominant while all other scatterings are irrelevant. With the increase of the electron density and hence also the corresponding impurity or hole density, the electron-impurity and electron-hole scatterings are strengthened, which leads to a considerable difference between the n-type and intrinsic cases. Accordingly, the differences of the corresponding curves with the same electron density are observed. We further address the relative importance of the DP and BAP mechanisms in intrinsic GaAs$_{1-x}$Bi$_x$. In Fig. 2(b), we plot the ratio of the SRT due to the BAP mechanism, τ_{BAP}, to that due to the DP mechanism, τ_{DP}, in the conditions corresponding to Fig. 2(a). It is seen that $\tau_{\mathrm{BAP}}/\tau_{\mathrm{DP}}$ is insensitive to the composi-

tion of Bi, which results from the comparable increasing rates of γ_D and ΔE_{LT} with Bi-doping as shown in Fig. 1. Meanwhile, $\tau_{\mathrm{BAP}}/\tau_{\mathrm{DP}}$ increases with increasing electron density but remains larger than 40. Thus the BAP mechanism is negligible in intrinsic GaAs$_{1-x}$Bi$_x$ as expected.[21] Furthermore, taking into consideration the experimental evidence of an increase of the hole effective mass in GaAs$_{1-x}$Bi$_x$,[5,26,27] we perform the calculation of the SRT in intrinsic GaAs$_{1-x}$Bi$_x$ by doubling the heavy- and light-hole effective masses. The obtained SRTs for all three photoexcitation densities remain almost unchanged, thanks to the marginal effects of the electron-hole Coulomb and exchange scatterings at room temperature.
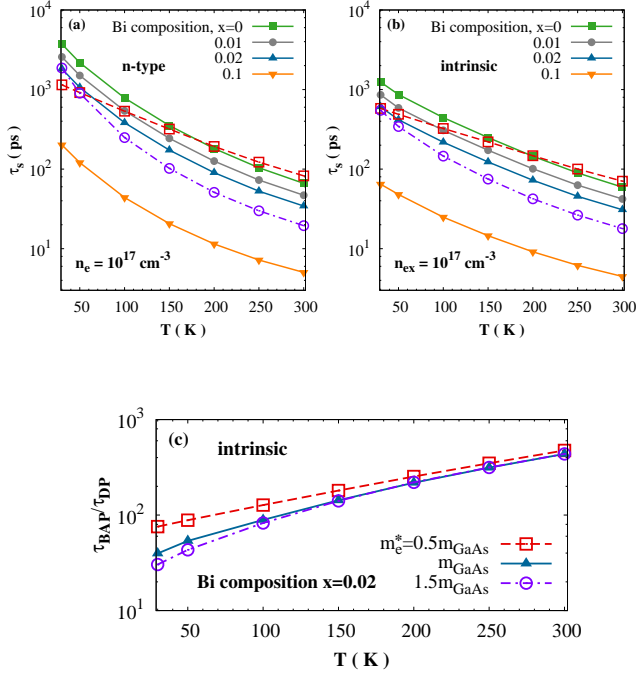
experimentally[28,29] and theoretically[21,25] in GaAs and the underlying physics lies in the drastic increase of the inhomogeneous broadening[15,16] due to the ascending temperature. Moreover, the giant tailoring of SRTs resulting from the incorporation of Bi at all temperatures is seen in the figures.

TABLE I: Coefficient of the Dresselhaus SOC γ_D and longitudinal-transverse exchange splitting ΔE_{LT} in GaAs$_{0.98}$Bi$_{0.02}$ for different electron effective masses.

m_e^* (m_{GaAs})	0.5	1.0	1.5
γ_D (eV$\cdot$ Å^3)	24.98	17.67	14.43
ΔE_{LT} (meV)	0.02	0.10	0.22

It is noted that the electron effective mass in GaAs$_{1-x}$Bi$_x$ from a direct measurement is only available for Bi composition up to $x = 0.01$.[14] Although it is argued to stay untouched under Bi incorporation,[9,13,14] there are also works suggesting an increasing[30,31] or a nonmonotonic[32] behavior with the increase of Bi composition. Therefore, it is meaningful to perform an investigation to include the possible modification of the electron effective mass in GaAs$_{1-x}$Bi$_x$. For simplicity, we calculate the temperature dependence of the SRTs in both n-type and intrinsic GaAs$_{0.98}$Bi$_{0.02}$ by artificially setting the electron effective mass $m_e^* = 0.5m_{\mathrm{GaAs}}$ and $1.5m_{\mathrm{GaAs}}$. The possible variation of the electron effective mass is reasonably covered within this range.[31,32] We indicate the obtained SRTs with dashed curves in Figs. 3(a) and 3(b), accordingly. It is seen that, the variation of m_e^* markedly changes the specific values, together with the temperature dependence of the SRTs. In order to elucidate this behavior, we calculate the variation of γ_D and ΔE_{LT} from the change of electron effective mass. From Table. I, one notices that γ_D visibly decreases with the increase of m_e^*, while the reverse is true for ΔE_{LT}. One may hence expect the BAP mechanism to play a role in the spin relaxation in intrinsic GaAs$_{1-x}$Bi$_x$ if m_e^* does increase. However, a detailed calculation of $\tau_{\mathrm{BAP}}/\tau_{\mathrm{DP}}$ shown in Fig. 3(c) indicates that the BAP mechanism is still negligible in the intrinsic case. This is analyzed as follows. The overall coefficient of the long-range electron-hole exchange scattering is actually proportional to $a_{\mathrm{Bohr}}^3\Delta E_{\mathrm{LT}}$ rather than ΔE_{LT} alone,[21] and hence is independent of m_e^*. Therefore, only slight change of $\tau_{\mathrm{BAP}}/\tau_{\mathrm{DP}}$ with the increasing m_e^* is observed in Fig. 3(c). We then focus on the DP mechanism. The variation of the electron effective mass influences the DP spin relaxation in two ways: (i) it modulates the coefficient of the Dresselhaus spin-orbit coupling γ_D; (ii) it changes the energy spectrum together with the electron distribution and hence modifies both the inhomogeneous broadening and the scattering. With the increase of m_e^*, on one hand, γ_D decreases and hence tends to suppress the spin relaxation; on the other hand, the electrons are more easily thermalized to high k states in the case of a flatter energy spectrum. This results in a larger inho-

FIG. 3: SRT τ_s as function of temperature for four Bi compositions: $x = 0$, 0.01, 0.02 and 0.1. (a) n-type condition with electron density $n_e = 10^{17}$ cm^{-3} and (b) intrinsic condition with photoexcitation density $n_{\mathrm{ex}} = 10^{17}$ cm^{-3}. The STRs in GaAs$_{0.98}$Bi$_{0.02}$ calculated by artificially using electron effective masses $0.5m_{\mathrm{GaAs}}$ (dashed curves with □) and $1.5m_{\mathrm{GaAs}}$ (dashed curves with ⊙) are also plotted. (c) The ratio of the SRT due to the BAP mechanism τ_{BAP} to that due to the DP mechanism τ_{DP} in intrinsic GaAs$_{0.98}$Bi$_{0.02}$ for three electron effective masses. Note that the same color and type of line and point are used for the corresponding m_e^* in (b).

We then investigate the temperature dependences of the SRTs in both n-type and intrinsic GaAs$_{1-x}$Bi$_x$ with the electron density 10^{17} cm^{-3} for different Bi compositions $x = 0$, 0.01, 0.02 and 0.1. One observes from Figs. 3(a) and 3(b) that in all cases, the SRTs decrease monotonically with the increase of temperature. This behavior has formerly been studied both

mogeneous broadening and hence tends to enhance the spin relaxation (which is obvious when the change of the inhomogeneous broadening overcomes that of the scatterings). At high temperature, the latter factor dominates, so larger m_e^* implies smaller τ_s. At low temperature, the situation is reversed. Accordingly, the crossovers are seen in Figs. 3(a) and 3(b) [although less obvious in Fig. 3(b)].

In summary, we have investigated the electron spin relaxation in n-type and intrinsic $GaAs_{1-x}Bi_x$ with Bi composition $0 \leq x \leq 0.1$ by applying the fully microscopic KSBE approach. The variations of the band gap and the spin-orbit splitting energy are explicitly considered. We show that the electron spin relaxation time decreases significantly by the incorporation of Bi, which suggests potential applications of $GaAs_{1-x}Bi_x$ in the engineering of spintronic devices. The density and temperature dependences of the SRT are studied for different Bi

compositions, with their behaviors found to resemble the ones in GaAs. Meanwhile, we show that the BAP mechanism is negligible compared to the DP mechanism in the intrinsic condition. Moreover, due to the absence of unambiguous experimental data of m_e^* in the whole compositional range under investigation and also the existing contradictions in the literature, we perform calculations to include the possible variation of m_e^* with the change of Bi incorporation. Profound impacts on the explicit value together with the temperature dependence of the SRT are found. This makes the precise measurement of m_e^* in $GaAs_{1-x}Bi_x$ highly desirable from both theoretical and engineering points of view.

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