Effect of nonequilibrium phonons on hot-electron spin relaxation in n-type GaAs quantum wells

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received 6 October 2010; accepted in final form 9 November 2010
published online 10 December 2010

PACS 72.25.Rb – Spin relaxation and scattering
PACS 71.10.-w – Theories and models of many-electron systems
PACS 63.20.kd – Phonon-electron interactions

Abstract – We study the effect of nonequilibrium longitudinal optical phonons on hot-electron spin relaxation in n-type GaAs quantum wells. The longitudinal optical phonons are driven to nonequilibrium states by electrons under an in-plane electric field. The nonequilibrium phonons then in turn influence the electron spin relaxation properties via modifying the electron heating and drifting. When the longitudinal optical phonons are treated as the nonequilibrium rather than the equilibrium ones, the spin relaxation time is increased since the electron heating is enhanced and hence the electron-phonon scattering is strengthened. Meanwhile, the frequency of spin precession, which is roughly proportional to the electron drift velocity, can be either increased or decreased. The former happens in the case with low electric field and/or high lattice temperature, whereas the latter happens in the case with high electric field and/or low lattice temperature. The nonequilibrium phonon effect is more pronounced when the electron density is high and the impurity density is low.

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Introduction. – Understanding spin relaxation is an important issue for the possible application in spintronic devices [1–4]. Among different kinds of spin relaxation mechanisms [5–7], scattering plays an essential role. In general cases, phonons are assumed to form an equilibrium bath. This treatment works well when the carrier system is near the equilibrium. If the carriers are far away from the equilibrium (e.g., driven by an electric field or excited by a laser beam), phonons can be driven away from their equilibrium states significantly by carriers when the carrier energy relaxation mainly goes through the phonon emissions and the phonon relaxation time is comparable with (or longer than) the carrier-phonon scattering time. The nonequilibrium phonons in turn are able to affect the electron dynamics, including the spin relaxation. In fact, the hot-electron transport with nonequilibrium phonons has been investigated [8–16], showing that the calculated electron energy loss rate and mobility fit better with experimental data than those obtained with the equilibrium phonons [10,13,14]. These studies also indicate that it is necessary to treat phonons as nonequilibrium ones in the hot-carrier system because these phonons will affect spin relaxation by modifying the carrier heating and drifting processes.

The hot-electron spin relaxation/dephasing has been studied theoretically in both (001) quantum well structures [17–20] and bulk materials [21], by means of the kinetic spin Bloch equation (KSBE) approach [4]. The spin relaxation/dephasing time is found to increase with electric field when both the temperature and electric field are low, especially in high mobility samples [17–21]. When the electric field is high (for which the multi-subband (in confined nanostructures) [18] and/or multi-valley [19] effect have to be taken into account), the spin relaxation/dephasing time decreases with electric field [17–19,21]. In these studies the phonons are treated as equilibrium ones. This work is to investigate the influence of nonequilibrium phonons on hot-electron spin relaxation in an n-type GaAs quantum well, where the spin-orbit coupling term is of the Dresselhaus type [19,22] and the spin relaxation is limited by the D’yakonov-Perel’ mechanism [5].

Model and KSBEs. – We start our investigation from an n-type [001]||x GaAs quantum well with an in-plane electric field along the [100]||x direction. The well width
\( a = 5 \text{ nm} \). Only the lowest subband is relevant with the proper electron density \( N_c \), lattice temperature \( T_L \) and electric field \( E \) with \( E \leq 1 \text{ kV/cm} \). (At larger electric field, the higher subbands [18] and/or the \( L \) valleys [19] should be taken into consideration.) Due to the electron localization in the \( z \)-direction, the electron-phonon coupling is spatially inhomogeneous, i.e., the emission and absorption of phonons mainly occur in the well where electrons have substantial density. If the phonon relaxation is fast enough or the phonons (particularly, the acoustic (AC) phonons) can easily penetrate through the well interfaces, these phonons can be deemed as in equilibrium with the bulk modes. In our study we assume that the AC phonons keep in equilibrium and the longitudinal-optical (LO) phonons are in nonequilibrium [9–13,15]. In order to investigate the spin relaxation of electrons which are inhomogeneously coupled with the nonequilibrium LO phonons, we combine the rate equation of the LO phonons (eq. (2)), described as “quasi-2D” [10,13,14], with the electron KSBEs (eq. (1)) [4]:

\[
\frac{\partial n_k}{\partial t} \bigg|_{\text{dri}} + \frac{\partial n_k}{\partial t} \bigg|_{\text{coh}} + \frac{\partial n_k}{\partial t} \bigg|_{\text{scat}},
\]

\[
\frac{\partial \rho}{\partial t} = \frac{\partial \rho}{\partial t} \bigg|_{\text{dri}} + \frac{\partial \rho}{\partial t} \bigg|_{\text{coh}} + \frac{\partial \rho}{\partial t} \bigg|_{\text{scat}},
\]

\[
(1)
\]

In eq. (1), \( \rho_k \) represent the density matrices of electrons with in-plane momentum \( k \), whose diagonal terms \( \rho_k_{\sigma \sigma} \equiv f_{k \sigma} (\sigma = \pm 1/2) \) represent the electron distribution functions and the off-diagonal ones \( \rho_k \pm 1/2 \equiv \rho_k - 1/2 \) describe the inter-spin-band correlations for the spin coherence. \( \frac{\partial \rho_k}{\partial t} \bigg|_{\text{dri}} = eE \cdot \nabla_k \rho_k \) are the driving terms from the external electric field. \( \frac{\partial \rho_k}{\partial t} \bigg|_{\text{coh}} \) are the coherent terms describing the coherent spin precessions due to the effective magnetic fields from the Dresselhaus term [19,22] and the Hartree-Fock Coulomb interaction, as well as the optional magnetic field in the Voigt configuration. \( \frac{\partial \rho_k}{\partial t} \bigg|_{\text{scat}} \) stand for the scattering terms of electrons, including the electron-LO/AC phonon, electron-impurity and electron-electron Coulomb scatterings. \( n_k \) in eq. (2) are the distributions of quasi-2D LO phonons with in-plane momentum \( q \). \( \frac{\partial n_k}{\partial t} \bigg|_{\text{scat}} \) stand for the scattering terms of the LO phonons, including the phonon-phonon and phonon-electron scatterings. Expressions of the coherent and scattering terms of electrons are given in detail in refs. [17] and [20], except that the electron-LO phonon scattering term should be slightly modified here as the LO phonons are described as quasi-2D (this modification makes no difference when the LO phonons are in equilibrium) [10,13,14]. The electron-LO phonon scattering term in eq. (1) reads

\[
\frac{\partial n_k}{\partial t} \bigg|_{\text{scat}} = \frac{\pi}{A} \sum_{k'} M^2_{k \to k'} \{ \delta (\varepsilon_k - \varepsilon_{k'} + \hbar \omega_0) \times (n_{k'} + 1) \rho_k (1 - \rho_k) - n_{k'} \rho_k (1 - \rho_{k'}) \rho_k \} + \delta (\varepsilon_k - \varepsilon_{k'} - \hbar \omega_0) n_{k'} \rho_k \rho_k - (n_{k'} + 1) (1 - \rho_{k'}) \rho_k \} + \ldots \},
\]

\[
(3)
\]

with \{ \ldots \}^{\dagger} \) standing for the Hermitian conjugation of the first term. The scattering term in eq. (2) reads

\[
\frac{\partial n_q}{\partial t} \bigg|_{\text{scat}} - \frac{n_q - n_0^q}{\tau_{\text{pp}}} - 2\pi A M^2 \sum_k \delta (\varepsilon_k - \varepsilon_{k-q} - \hbar \omega_0) \times \{ \text{Tr}[(1 - \rho_k)\rho_{k-q}-\text{Tr}[^1\rho_k(1 - \rho_{k-q})] / (n_q + 1) - \).
\]

(4)

In the above equations \( \varepsilon_k = \hbar^2 k^2 / 2m^* \) represents the energy of electron with momentum \( k \) and effective mass \( m^* = 0.067m_0 \). The Sommerfeld factor of LO phonons with in-plane momentum \( \kappa \) is given by [23], \( \delta \) is the area of the well layer. \( M_0^2 = \frac{1}{2} \sum_{i \neq j} q_{pq}^2 |I(iq_\alpha)|^2 \) is the effective electron-LO phonon scattering matrix element with \( g_{pq}^2 = \frac{\epsilon^2 q_w^2}{2m^* \gamma_0 q^2} \) for the form factor under the infinite-depth well approximation. The first term on the right-hand side of eq. (4) represents the contribution from the phonon-phonon scattering in relaxation time approximation. \( n_q^0 = \text{exp} [\hbar \omega_0 / k_B T_L] - 1 \) is the number of quasi-2D LO phonons in equilibrium with the AC phonons. The population relaxation time \( \tau_{\text{pp}} \) is contributed by anharmonic lattice vibrations (especially the third-order anharmonicity) and depends on the phonon momentum as well as lattice temperature. Moreover, distinctly heated nonequilibrium LO phonons (depending on the heating and relaxation of electrons) may have different relaxation times. In spite of these intricate factors involved in the LO phonon relaxation, we assume \( \tau_{\text{pp}} \) to be a constant only depending on the lattice temperature, by adopting the empirical formula fitted by Vallée and Bogani from the time-resolved coherent anti-Stokes Raman scattering experiment [24]. It gives \( \tau_{\text{pp}} (T_L) = \tau_{\text{pp}} (0) \times \left[ \text{exp}(0.2 \omega_0 / k_B T_L) - 1 \right]^{-1} + \text{exp}(0.8 \omega_0 / k_B T_L) - 1 \right]^{-1} \) with \( \tau_{\text{pp}} (0) = 9 \text{ ps} [24] \). This formula in fact depicts the dominant decay route of an LO phonon near the center of the Brillouin zone into a transverse AC phonon and a different LO phonon at the \( L \) critical point of the Brillouin zone. The relaxation time approximation with a constant \( \tau_{\text{pp}} \) related only to the lattice temperature has been widely utilized in the study of hot-electron transport with the presence of nonequilibrium LO phonons [10–14].

Results. – We numerically solve the KSBEs following the scheme mainly laid out in ref. [17], with the rate equation of the LO phonons discretized in the momentum space in a way similar to that for electrons. The impurity density is set as zero and the electric field \( E = -E \hat{x} \) with \( E \geq 0 \). No magnetic field is applied except otherwise specified. The initial conditions at time \( t = 0 \) are chosen as the steady-state solution of eqs. (1) and (2) in the absence of the spin-orbit coupling in the coherent term \( \frac{\partial \rho_k}{\partial t} \bigg|_{\text{coh}} \) [17]. Numerically, they are prepared from a state at
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Fig. 1: (Color online) Typical steady-state distributions of nonequilibrium electrons ((a) and (c)) and LO phonons ((b) and (d)) in momentum space. (a) and (b): $T_L = 200$ K and $E = 0.3$ kV/cm; (c) and (d): $T_L = 50$ K and $E = 1$ kV/cm. Note that in (b) and (d) the LO phonon distributions $n_q$ are rescaled by a factor 2 and $\frac{1}{2}$, respectively.

t = $-t_0$ ($t_0 > 0$) with $f_{k\sigma}(-t_0) = \{\exp[(h^2k^2/2m^* - \mu_\sigma)/k_BT_L] + 1\}^{-1}$, $\rho_{k\sigma\alpha\beta}(-t_0) = 0$ and $n_q(-t_0) = n_0^q$. Here $\mu_\sigma$ are the electron chemical potentials determined by $\frac{1}{A} \sum_k \text{Tr}[\rho_k(-t_0)] = N_e$, and $\frac{1}{A} \sum_k \text{Tr}[\rho_k(-t_0)\sigma_\alpha\beta] = N_eP_0$, where $P_0 = 0.05$ is the spin polarization along the $z$-axis and $N_e = 4 \times 10^{11}$ cm$^{-2}$ is the electron area density. With the driving (by the electric field) and the scattering, the system reaches a steady state at time $t = 0$.

After time $t = 0$, the spin-orbit coupling in the coherent term is switched on and electron spins begin to relax with an initial spin polarization $P(t = 0) = P_0 = 0.05$. The spin relaxation time $\tau$ is obtained from the time evolution of spin polarization $P(t) = \frac{1}{A} \sum_k \text{Tr}[\rho_k(t)\sigma_\alpha\beta]$, the electron drift velocity is the steady value of $v(t) = \frac{1}{A} \sum_k \text{Tr}[\rho_k(t)]h\mathbf{k}/m^* = v_x(t)\hat{x}$ and the hot-electron temperature $T_e$ is obtained by fitting the Boltzmann tail of the drifted Fermi distribution.

In fig. 1 the typical steady-state distributions of the nonequilibrium electrons and LO phonons in momentum space are plotted. It is shown by figs. 1(a) and (c) that under the electric field along the $-x$-direction, the electrons gain a drift velocity along the $x$-direction. Figures 1(b) and (d) show the corresponding distributions of nonequilibrium LO phonons. The LO phonons with either very large or small momenta (e.g., in the edge or center of the momentum space shown in figs. 1(b) (the yellow color region) and (d) (the nearly transparent region)) are in equilibrium with the AC phonons due to the weak electron-LO phonon scattering. Therefore, the distribution of LO phonons in these momentum regions varies with the lattice temperature. It is interesting to see that the LO phonons with medium momenta are driven far away from their equilibrium states by the hot electrons. In the case with $T_L = 200$ K and $E = 0.3$ kV/cm (fig. 1(b)), the LO phonons with $q_x > 0$ are emitted (and form a peak in the positive-$q_x$ momentum region) and those with $q_x < 0$ are absorbed (and form a valley in the negative-$q_x$ momentum region). With the decrease of $T_L$ and/or the increase of $E$, the valley in the negative-$q_x$ momentum region is suppressed or even disappears, as shown in fig. 1(d) for the case with $T_L = 50$ K and $E = 1$ kV/cm. In any case, the total phonon density (i.e., the summation of distribution over the momentum space) increases and a net positive momentum is gained by the LO phonons.

In fig. 2(a) the electric-field dependence of electron drift velocity $v_x$ at various lattice temperatures is plotted, with the LO phonons treated as the equilibrium ones in the calculation. The ratio of $v_x$ obtained with the nonequilibrium LO phonons to that with the equilibrium ones is

\[ \frac{v_x}{v_{x0}} = \frac{v_x(T_L = 50 \text{ K}, E = 1 \text{ kV/cm})}{v_x(T_L = 200 \text{ K}, E = 0.3 \text{ kV/cm})} \]

Fig. 2: (Color online) Electric-field dependences of drift velocity $v_x$ (a), hot-electron temperature $T_e$ (c) and spin relaxation time $\tau$ (e), calculated with the equilibrium LO phonons. The ratios of the quantities $v_x$, $T_e$ and $\tau$ with the nonequilibrium LO phonons to those with the equilibrium ones, $\eta_{v_x}$, $\eta_{T_e}$ and $\eta_{\tau}$, are shown in (b), (d) and (f), respectively. In (e), the two curves with open squares and open circles are the electric-field dependences of $\tau$ with a magnetic field ($B = 2$ T) along the $-x$-direction for $T_L = 50$ K and 300 K, respectively (at $T_L = 50$ K ($T_L = 300$ K), when $E > 0.05$ kV/cm ($E > 0.3$ kV/cm) the spin relaxation time coincides with that without magnetic field and thus not shown).
shown in fig. 2(b). From fig. 2(b), one notices that when the nonequilibrium phonon effect is taken into account, $v_\alpha$ can be either increased or decreased. These results are in consistence with the previous studies [9,11,13]. In fact, the influence of nonequilibrium phonons on hot-electron transport consists of two competing effects: the reabsorption of momentum from the nonequilibrium phonons tends to increase the electron mobility, while the enhanced electron heating strengthens the electron-phonon scattering (including both the electron-AC phonon and electron-LO phonon scatterings) and tends to decrease the electron mobility [9,11,13]. Generally the former (latter) effect dominates in the regime with low (high) electric filed and/or high (low) temperature [9,11,13], as indicated in fig. 2(b). As mentioned previously, when the electric field is low and/or the lattice temperature is high, the valley of the LO phonon distribution in the negative-$q_x$ momentum space is pronounced, thus it substantially suppresses the back scattering of electrons in momentum space by absorbing the negative-$q_x$ phonons and hence makes the electron distribution more forward-peaked. Finally, when the lattice temperature is high enough (e.g., $T_L = 300$ K), the effect of nonequilibrium phonons becomes weak and less sensitive to the electric field, mainly due to the shorter LO phonon relaxation time ($\tau_{pp}$) [25]. Here $\mu$ is the electron mobility and $\alpha/T_L$ is the heat transfer coefficient. If both $\mu$ and $\alpha$ are independent of $T_e$, one can infer that $T_e$ increases with $E$ quadratically. However, this is not the case. As both $\mu$ and $\alpha$ depend on the scattering processes and hence $T_e$, the relationship between $T_e$ and $E$ is nontrivial. However, for some special cases, their relationship can be figured out. For example, when the electron-AC phonon scattering serves as the main channel of energy loss, $T_e$ is expected to increase with $E$ quadratically (linearly) when $\mu_0 E \ll v_s$ ($\mu_0 E \gg v_s$) [25]. Here $\mu_0$ is the electron mobility when $T_e = T_L$ and $v_s$ is the sound velocity. The linear dependence of $T_e/T_L$ on $E$ for the case with $T_L = 50$ K, as shown in fig. 2(c), is therefore understood. At this temperature, the energy loss mainly occurs via the electron-AC phonon scattering, and $\mu_0 E$ can easily exceed $v_s$ with the increase of electric field. An estimation with $\mu_0 \approx 5 \times 10^5$ cm$^2$/V$^{-1}$s$^{-1}$ (as indicated by the data shown in fig. 2(a)) and $v_s \approx 5 \times 10^5$ cm/s gives a critical field intensity as small as 5 V/cm when $T_L = 50$ K. As a result $T_e/T_L$ almost shows a linear dependence on $E$ in the whole electric-field region. When $T_L$ increases, the LO phonons come into play and the mobility decreases as well. With these effects, the $T_e/T_L$-$E$ curve deviates from the linearity, as shown by the cases with $T_L \geq 100$ K in fig. 2(c).

From fig. 2(d), where the ratio of $T_e$ obtained with the nonequilibrium LO phonons to that with the equilibrium ones is plotted, one finds that when the phonons are treated as nonequilibrium ones, electrons are further heated as expected [11–13]. Moreover, when $T_L$ is low, $\eta_e$ shows a nonmonotonic behavior. That is caused by the decay of the heating efficiency with the increase of electric field in the presence of nonequilibrium phonons: With the increase of electric field, the number of the LO phonons increases and the rate of electron energy relaxation through the electron-LO phonon scattering increases as well. This effect is more pronounced when the lattice temperature is low, where the nonequilibrium LO phonons can be considerably accumulated with the increase of electric field, due to the long phonon relaxation time ($\tau_{pp} = 7.3$ ps, when $T_L = 50$ K) [24].

The electric-field dependence of the spin relaxation time $\tau$ under various temperatures is plotted in fig. 2(e). From the figure, one notices that $\tau$ generally increases with $E$ in the regime under investigation. Two reasons lead to this phenomenon: I) Under the electric field along the $-\hat{x}$-direction, a net effective magnetic field along the $-\hat{x}$-direction is induced via the Dresselhaus spin-orbit coupling [17,19]. With this effective magnetic field, spins begin to precess around it and thus the in-plane spin relaxation is mixed with the out-of-plane one [26,27]. As the in-plane spin relaxation rate of the two-dimensional electron system in (001) GaAs quantum well is smaller than the out-of-plane one in the framework of the D’yakonov-Perel’ relaxation mechanism, when the electric field is applied, $\tau$ is increased due to the effective magnetic field [26,27]. The effective magnetic field decreases with the increase of $T_L$, because it is proportional to $v_x$ [17,19] and $v_x$ decreases with increasing $T_L$ (as shown in fig. 2(a)). Therefore the mixing of the in-plane and out-of-plane spin relaxations is obvious in the low-temperature regime. In fact, when $E = 0.05$ kV/cm, the effective magnetic field is $\sim 1$ T when $T_L \sim 50–100$ K and $\sim 0.1$ T when $T_L = 300$ K. As a result, the effective magnetic field causes an abrupt increase of $\tau$ [26] when $E$ sweeps from 0 to 0.05 kV/cm for the cases with $T_L \sim 50–100$ K, but a slow increase of $\tau$ when $E$ goes from 0 to 0.3 kV/cm for the case with $T_L = 300$ K. II) The heating of electrons by the electric field enhances the electron-phonon scattering and thus increases the spin relaxation time $\tau$ in the strong-scattering limit [4,17–21]. This effect, only important in the low-temperature regime where the heating effect is strong (as shown in fig. 2(e)), is responsible for the continuing increase of $\tau$ with $E$ when $E > 0.05$ kV/cm and $T_L \sim 50–100$ K. To make the underlying physics depicted above more pronounced, a
magnetic field $B = 2\, \text{T}$ is applied along the $-\hat{x}$-direction for the cases with $T_L = 50 \, \text{K}$ and $300 \, \text{K}$. The corresponding electric-field dependences of $\tau$ are plotted as curves with open squares ($T_L = 50 \, \text{K}$) and open circles ($T_L = 300 \, \text{K}$) in fig. 2(c). With this large external magnetic field, the in-plane and out-of-plane spin relaxations are efficiently mixed even when $E = 0$. One finds that for the case with $T_L = 300 \, \text{K}$, $\tau$ almost keeps unchanged with the increase of electric field, while for the case with $T_L = 50 \, \text{K}\tau$ keeps on increasing with $E$ due to the strong-heating effect. Finally, it is noted that when $T_L = 300 \, \text{K}$, there is a marginally decreasing tendency of $\tau$ with $E$ when $E$ is near $1 \, \text{kV/cm}$. This is caused by the enhanced inhomogeneous broadening of the effective magnetic field from the Dresselhaus spin-orbit coupling due to the drifting and heating of the electric field [4,17–21]. This effect takes place more easily when the lattice temperature is high [17,19].

In fig. 2(f), the ratio of the spin relaxation time obtained with the nonequilibrium LO phonons to that with the equilibrium ones is shown. With the nonequilibrium LO phonons taken into account, $\tau$ is generally increased due to the strengthened electron-phonon scattering. Therefore the increase of $\tau$ corresponds to the increase of $T_e$, as shown in fig. 2(d). However, when the lattice temperature is high enough (e.g., $T_L = 300 \, \text{K}$), the modification on $\tau$ induced by the nonequilibrium phonons cannot be seen. Under the electric field along the $\hat{x}$-axis, an effective magnetic field along the $\hat{x}$-axis with the magnitude proportional to the drift velocity $v_x$ is induced via the Dresselhaus spin-orbit coupling term [17,19,28]. This magnetic field makes spins precess around it. Therefore, when $v_x$ is affected strongly by the nonequilibrium LO phonons, the spin precession frequency has a modification with the magnitude roughly proportional to the change of $v_x$. In fig. 3(a) we show the typical spin precession signals with the equilibrium and nonequilibrium LO phonons, respectively. In fig. 3(b), the typical ratio of spin precession frequency obtained with the nonequilibrium LO phonons to that with the equilibrium ones is plotted in the region of the electric field where $v_x$ is large enough that spin precession signal with clear periods can be distinguished.

Finally, further calculations show that the effect of nonequilibrium phonons on electron mobility, electron heating and electron spin relaxation decays with the decrease of electron density and the increase of impurity density. This can be easily understood as the LO phonons are driven to the nonequilibrium states by electrons and the increase of the electron-impurity scattering suppresses the effect caused by the electron-LO phonon scattering.

**Conclusion.** – In this work, we have studied the effect of nonequilibrium LO phonons on hot-electron spin relaxation in $n$-type (001) GaAs quantum wells. Under an in-plane electric field, the LO phonons can be driven away from their equilibrium states by electrons and then in turn affect the electron transport, electron heating as well as electron spin relaxation.

In the presence of the nonequilibrium LO phonons, the electron drift velocity under electric field can be either increased (at low electric field and/or high lattice temperature) or decreased (at high electric field and/or low lattice temperature). This phenomenon is caused by two competing effects: the momentum reabsorption from phonons and the strengthened electron-phonon scattering [9,11,13]. The former tends to increase the electron mobility whereas the latter tends to suppress it. The nonequilibrium LO phonons also impede the energy relaxation of electrons and thus the electrons are further heated, especially when the lattice temperature is low. The nonequilibrium LO phonons effectively affect the hot-electron spin relaxation through the strengthening of the electron-phonon scattering, which tends to increase the spin relaxation time. This effect also dominates in the low-temperature regime. Moreover, as the spin precession frequency under the electric field is proportional to the electron drift velocity, it can be either increased or decreased when the nonequilibrium LO phonons are taken into account. Finally, it should be noticed that the effect of nonequilibrium phonons is more pronounced in systems with high electron density and low impurity density.

This work was supported by the Natural Science Foundation of China under Grant No. 10725417.

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