Electron spin relaxation as evidence of excitons in a two dimensional electron-hole plasma

S. Oertel, S. Kunz, D. Schuh, W. Wegscheider, J. Hübner, and M. Oestreich

Institute for Solid State Physics, University Hanover, 30167 Hanover, Germany
Institute for Experimental and Applied Physics, University Regensburg, 93040 Regensburg, Germany
Solid State Physics Laboratory, ETH Zürich, 8093 Zürich, Switzerland

(Dated: January 14, 2013)

We exploit the influence of the Coulomb interaction between electrons and holes on the electron spin relaxation in a (110)-GaAs quantum well to unveil excitonic signatures within the many particle electron-hole system. The temperature dependent time- and polarization-resolved photoluminescence measurements span five decades of carrier density, comprise the transition from localized excitons over quasi free excitons to an electron-hole plasma, and reveal strong excitonic signatures even at relatively high densities and temperatures.

PACS numbers: 71.35.-y, 78.47.-p, 78.55.Cr, 78.67.De

Shortly after the big bang, the hot gas of negatively charged electrons and positively charged protons cooled to temperatures below the Rydberg energy and formed a new state of matter, called hydrogen atoms. 13.7 billion years later, researchers shoot short laser pulses on direct semiconductors at liquid helium temperature creating a hot gas of negatively charged electrons in the conduction and positively charged holes in the valence band. The hot carrier gas cools with time by emission of optical and acoustical phonons and forms excitons, i.e., hydrogen like quasi-particles. These quasi-particles have in bulk GaAs a binding energy of 5 meV and strongly influence the photoluminescence (PL) spectrum. Consequently, the PL emission at the exciton transition energy has been used as an indicator for the existence of excitons as the emission and absorption lines of hydrogen are used in astronomy. However, S. Koch and coworkers demonstrated within the framework of many body semiconductor Bloch equations that distinct excitonic like emission lines from an electron-hole plasma do not prove the existence of excitons but can be explained equally by a Coulomb correlated electron-hole plasma.

Manyfold interband pump-probe absorption, reflection, and four wave mixing experiments, high resolution time-resolved PL, and experimentally very demanding quasi-particle THz spectroscopy have been carried out to understand the many-body and quantum-optical character and the diligent interaction of excitons in an electron-hole plasma but besides fifty years of intense research the exciton quest remains a fascinating and active field. The challenge in the interpretation of most interband experiments concerning the incoherent exciton population results from the fact that the interaction of classical light and matter is induced by optical polarization and not directly by incoherent population. The challenge in the interpretation of time-resolved PL experiments results generally from the finite excitation density and that thereby the PL does not result from a two but a many particle interaction which strongly depends on the frequency-dependent strength of light-matter interaction and only to some extend on exciton population. Especially THz experiments support that excitonic like PL lines exist without the population of K=0 excitons. In this letter, we present an entirely different experimental approach to study the existence of excitons in a two-dimensional electron-hole plasma and utilize the electron-hole spin interaction as exciton marker. In general, the spin dynamics in GaAs quantum wells (QWs) is not an appropriate indicator for excitons since the spin of free holes dephases rapidly within the hole momentum scattering time due to the mixing of heavy hole (HH) and light hole (LH) and the electron spin dephases in (001)-GaAs-QWs rapidly due to the Dyakonov-Perel (DP) spin relaxation mechanism, i.e., both electron and hole spin relaxation do not significantly depend on exciton formation. Fortunately, the DP spin relaxation of electrons can be easily suppressed by the symmetry of the QW. Several groups have demonstrated that the DP mechanism vanishes in (110)-GaAs-QWs for electron spins pointing parallel or antiparallel to the growth direction and that the Elliott-Yafet and the intersubband spin relaxation (ISR) mechanism are inefficient in thin QWs. In the case of thin (110)-QWs, the only remaining significant electron spin relaxation mechanisms are the Bir-Aronov-Pikus (BAP) and the exciton exchange spin relaxation. Very recently, Zhou and Wu have calculated by sophisticated semiconductor spin Bloch equations that the BAP mechanism is at moderate carrier densities surprisingly inefficient in two-dimensional samples i.e., efficient electron spin relaxation results in (110)-GaAs-QWs predominantly from excitonic exchange interaction and is thereby a clear measure for the existence of excitons in an electron-hole plasma.

In this letter, we study the existence of excitons in an electron-hole plasma by measuring the density and tem-
FIG. 1: (color online) Initial polarization of the PL of the thick QW in dependence on the excitation energy (black points). The solid blue lines are the PL of the thick QW (left side) and of the thin QWs (middle) with different arbitrary units. The dashed red lines are the calculated HH and LH transitions of the complete structure (the indices correspond to the three lowest energy levels). The light red areas show the calculated broadened transition energies due to monolayer fluctuations in the thin QWs.

The latter proves that the electrons’ spin is conserved while tunnelling from the thin into the thick QW. The carrier tunnelling time is ≤ 3 ps as measured from the thin QW PL decay time at non-resonant excitation. The electron spin polarization is not 100% for resonant excitation of the HH transition of the thin QWs since we also excite the continuum of the wide QW. The degree of the initial PL polarization remains constant at 75 % for temperatures up to 70 K and decreases for higher temperatures exponentially to 22 % at 200 K. The degree of polarization decreases with increasing temperature since the LH contribution rises and, in the case of resonant excitation of the thin QWs, due to the decrease of the excitonic enhancement of the HH transition. We want to point out that such a high efficient optical spin injection structures not only significantly increases the sensitivity of our spin relaxation measurements but is also useful for many other experiments like for optically pumped spin VCSELs or polarization dependent spin relaxation experiments. In the following, all measurements are carried out for resonant excitation of the HH transition of the thin QWs.

In the next step, we distinguish between localized and non-localized carriers by the spectral width and lifetime of the PL and by spin quantum beat spectroscopy. The PL spectrum is at 10 K inhomogeneously broadened due to QW width fluctuations with a full width at half maximum (FWHM) of about 7.5 meV for excitation densities ≤ 1×10^{11} cm^{-2}. The FWHM of the PL is larger than in comparable (001)-GaAs-QWs due to the more complex growth dynamics of (110)-QWs. The PL maximum remains at constant emission energy for densities below 5×10^{9} cm^{-2}, increases sharply by 2 meV with increasing density, and remains constant again for densities between 2×10^{10} cm^{-2} and 2×10^{11} cm^{-2}. This indicates a transition from localized to unlocalized electrons and an electron disorder localization potential of 2 meV. The energy shift of the PL maximum with density vanishes at temperatures of about 20 K to 30 K confirming the potential depth of 2 meV. The PL rise time at 10 K is faster than our time resolution and the decay is purely mono-exponential for low excitation densities showing that the excited carriers are rapidly trapped in the localization potential. In contrast, the PL transient shows at the same temperature but an excitation density of 2×10^{10} cm^{-2} a rise time of about 100 ps which becomes more pronounced and long-lasting with increasing density. The slow rise time vanishes at a lattice temperature of 50 K and is an unambiguous signature for cooling of free carriers, i.e., we observe at 10 K with increasing density a transition from localized to free electrons and for temperatures ≥ 50 K only free electrons. We know that the 2 meV is the electron trapping potential from spin quantum beat experiments whereat the measured electron Landé g-factor g_e depends at low temperatures and low densities on the PL energy and increases linearly from
depicts the low excitation density case, \( n \) excitation density of electrons and weakly bound holes. At room temperature, the electron spin relaxation time is nearly constant.

The spin relaxation of electrons in a plasma of free electrons, free holes, and free excitons is dominated in (110)-QWs at \( T < 200 \) K by spin relaxation due to excitons. The exciton fraction in a 2D electron gas in the Boltzmann limit depends on the carrier density and the temperature and is described by the so-called Saha equation

\[
\frac{(n_{e-h})^2}{n_X} = \frac{k_B T}{2 \pi \hbar^2 \mu_e E_0 / k_B T},
\]

where \( \mu = 0.061 m_0 \) is the reduced mass and \( E_0 = 8 \) meV the exciton binding energy. The resulting exciton fraction at 120 K is for example 3.5 % at a density of \( 2 \times 10^9 \) cm\(^{-2} \) and 37 % at \( 6 \times 10^{10} \) cm\(^{-2} \). The spin relaxation time of pure excitons reads \( \tau_s^{exc} = (\Omega^2 \tau_p)^{-1} \) where \( \Omega \) is dominated by the long-range exchange interaction which is according to calculations by Maialle et al. [13] \( \Omega = 69.85 \) GHz for a 9 nm GaAs-QW and \( \tau_p \) is the exciton center of mass momentum scattering time. We calculate \( \tau_p \) by calculating the LO-phonon scattering rate according to Ref. [14] and by introducing phenomenologically a constant scattering rate of 0.77 ps\(^{-1} \) to account for surface roughness scattering and all other residual scattering mechanisms. The resulting electron spin relaxation time in an electron, hole, exciton plasma is the exciton spin relaxation time weighted by the exciton fraction: \( \tau_s = \tau_s^{exc} / f_X \). The solid lines in Fig. 2 depict the calculated \( \tau_s \) for free carriers. For the high density case and \( T < 200 \) K, the calculations are in very good agreement with the experiment while for \( T \geq 200 \) K, the measured \( \tau_s \) is shorter than calculated for two reasons. Firstly, the electrons scatter at high temperatures efficiently into higher subbands and ISR becomes important. Secondly, we observe a significant decrease of the PL lifetime with increasing temperature for \( T > 200 \) K, i.e., electrons jump out of the QW, lose their spin orientation due to the very efficient DP mechanism in the barrier, and a fraction of the depolarized electrons falls back into the QW reducing the average electron spin orientation in the QW. For low densities and \( T \leq 100 \) K, the discrepancy between experiment and theory is tremendous. This is also not surprising since localization dominates the spin relaxation process. More important, for \( T > 100 \) K all carriers become delocalized and the measured \( \tau_s \) approaches theory. For \( T > 120 \) K, reliable measurements become difficult since \( \tau_s \) becomes much longer than the PL lifetime and the laser repetition rate. Please note, that the measured \( \tau_s \) at 120 K is significantly longer in the low density case as predicted by the Saha equation.

We have also measured the in-plane spin relaxation time by applying a transverse magnetic field. The in-plane spin relaxation time is dominated by the DP spin relaxation and directly yields the electron momentum scattering time \( \tau_p^{s} \). This extracted \( \tau_p^{s} \) is at temperatures below...
200 K much shorter than the calculated LO-phonon scattering time and thus related to electron-electron scattering. Nevertheless, the LO-phonon scattering time dominates the excitonic \( \tau_s \) confirming the theoretical prediction that electron-electron scattering does not yield motional narrowing in the case of excitonic electron spin relaxation.

Next, we study the density dependence of \( \tau_s \) for temperatures between 50 K and 300 K (see Fig. 3). At \( T \geq 50 \text{ K} \), the electron spin decays in good approximation monoeponential for all carrier densities which is consistent with the picture of delocalized electrons. The slight deviation from a monoeponential decay results from the change of carrier density in the measurement window due to radiative recombination. Most interestingly, the measured electron spin relaxation time is for densities \( \ll 10^{12} \text{ cm}^{-2} \) not consistent with the calculated spin relaxation by the BAP mechanism which is at least one order of magnitude less efficient for densities \( \leq 10^{11} \text{ cm}^{-2} \) (see Ref. [11]). The times are also not consistent with electron spin relaxation times where holes are absent since previous spin noise spectroscopy measurements in n-doped (110)-QWs with an electron density of \( 1.1 \times 10^{11} \text{ cm}^{-2} \) yield two orders of magnitude longer electron spin relaxation times.[15] In fact, the measured spin relaxation times can only be explained by the existence of excitons and efficient electron spin relaxation by exciton exchange interaction, i.e., the density dependent measurements confirm that \( \tau_s \) is really a measure of the excitonic influence in an electron hole plasma. For densities between \( 10^{11} \text{ cm}^{-2} \) and \( 10^{12} \text{ cm}^{-2} \), \( \tau_s \) converges with increasing carrier density to approximately the same \( \tau_s \) for all temperatures between 50 K and 300 K. This measured \( \tau_s \) of \( \approx 450 \text{ ps} \) at a carrier density of \( 10^{12} \text{ cm}^{-2} \) is in good agreement with sophisticated calculations by semiconductor Bloch equations for (001)-QWs. Thereby, the calculations by Zhou and Wu prove that the electron spin relaxation is dominated at very high densities not by excitonic spin relaxation but by the BAP mechanism. Our experiments also confirm the predictions by Zhou and Wu that the BAP mechanism is surprisingly temperature insensitive.

Last, we study the density dependence of \( \tau_s \) at \( T = 10 \text{ K} \) where localization and Pauli blockade plays an important role. At this lattice temperature, the spin relaxation is monoeponential at low densities with \( \tau_s \approx 500 \text{ ps} \) and becomes biexponential at densities \( \geq 2 \times 10^9 \text{ cm}^{-2} \) whereat Fig. 3 depicts the initial fast decay. A clear transition from mono- to biexponential spin relaxation appears at the same density whereat the PL shows a transition from localized to free electrons, i.e., this initial fast \( \tau_s \) results from free electrons. The electron spin relaxation time decreases with decreasing localization (increasing density) since the spin relaxation time of localized holes is extremely long in comparison to free holes, as e.g., in semiconductor quantum dots[16]. An increase of \( \tau_s \) in the low density regime due to an admixture with unpolarized electrons originating from an unintended background doping can be excluded since the initial degree of polarization is constant within the error bars for densities from \( 1.7 \times 10^7 \) to \( 1.7 \times 10^{11} \text{ cm}^{-2} \), i.e., for all densities where bleaching can be neglected. The density dependence of \( \tau_s \) at 10 K not only traces the transition from localized to free carriers but also does not converge to the same \( \tau_s \) at \( 10^{12} \text{ cm}^{-2} \) as at all other temperatures. This difference can not result from localization but reveals the different strength of the BAP spin relaxation mechanism for Boltzmann and Fermi statistics.

In conclusion, we have performed time- and polarization-resolved PL measurements with a high efficient spin injection structure that enables us to measure the spin relaxation in a (110)-GaAs-QW over a wide range of densities. The special growth direction provides access to exciton induced electron spin dynamics in an electron, hole, exciton plasma that is for other growth direction masked by the highly efficient DP spin relaxation mechanism. The spin dynamics reveals unambiguously the existence of excitons and proves the decrease of the exciton fraction in an electron-hole plasma at thermal equilibrium with decreasing densities and increasing temperatures. Although the electron, hole, exciton plasma is well described by the Saha equation, the currently inherent disorder in (110)-QWs and the complex spin physics adds two degrees of complexity. We demonstrated qualitative measurements of the exciton population in thermal equilibrium whereat future calculations by semiconductor spin Bloch equations, which include the commonly neglected exciton population, will enable the extraction of the quantitative exciton population and the influence of localization. We have additionally verified that the BAP spin relaxation mechanism dominates at high den-

---

**FIG. 3:** (color online) Spin relaxation time versus excitation densities for different temperatures ranging from 10 to 300 K (the lines are guides to the eye).
ities and more importantly that the dependence of the BAP mechanism on temperature is extremely weak.

We thank W. W. Rühlle for helpful discussions. This work has been supported by the German Science Foundation (DFG–Priority Program 1285 “Semiconductor Spintronics”) and the excellence cluster QUEST at the university Hannover.

[1] Feldmann et al., Phys. Rev. Lett. 59, 2337 (1987).
[2] Koch et al., Nat. Materials 5, 523 (2006).
[3] Knox et al., Phys. Rev. Lett. 54, 1306 (1985).
[4] Malinowski et al., Phys. Rev. B 62, 13034 (2000).
[5] Noll et al., Phys. Rev. Lett. 64, 792 (1990).
[6] Szczytko et al., Phys. Rev. Lett. 93, 137401 (2004).
[7] Kaindl et al., Nature 423, 734 (2003).
[8] Kaindl et al., Phys. Rev. B 79, 045320 (2009).
[9] Lombez et al., Phys. Stat. Sol. (c) 4, 475 (2007).
[10] Wu et al., Physics Reports 493, 61 (2010).
[11] J. Zhou and M. W. Wu, Phys. Rev. B 77, 075318 (2008).
[12] The degree of PL polarization is less than 50 % for non-resonant excitation besides 50 % electron spin polarization since hot $k \neq 0$ HHs have an admixture of LHs.
[13] Maialle et al., Rev. B 47, 15776 (1993).
[14] B. K. Ridley, Quantum Processes in Semiconductors (Oxford University Press, New York, 1988).
[15] Müller et al., Phys. Rev. Lett. 101, 206601 (2008).
[16] Eble et al., Phys. Rev. Lett. 102, 146601 (2009).