Towards quantum optics and entanglement with electron spin ensembles in semiconductors

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(Dated: April 18, 2008)

We discuss a technique and a material system that enable the controlled realization of quantum entanglement between spin-wave modes of electron ensembles in two spatially separated pieces of semiconductor material. The approach uses electron ensembles in GaAs quantum wells that are located inside optical waveguides. Bringing the electron ensembles in a quantum Hall state gives selection rules for optical transitions across the gap that can selectively address the two electron spin states. Long-lived superpositions of these electron spin states can then be controlled with a pair of optical fields that form a resonant Raman system. Entangled states of spin-wave modes are prepared by applying quantum-optical measurement techniques to optical signal pulses that result from Raman transitions in the electron ensembles.

I. INTRODUCTION

Entanglement is the phenomenon that the quantum states of two (or more) degrees of freedom are inseparable, and is arguably the most distinct aspect of quantum theory [1]. It results in non-classical correlations between observable physical properties of the two subsystems. For nonlocal entanglement this concerns two degrees of freedom that are spatially separated over a large distance. The occurrence of such correlations has been thoroughly tested in several experiments, and the results leave little doubt that quantum theory provides the valid predictions. Experimental realizations were, until now, carried out with pairs of elementary particles or photons [2, 3], or with spins in very simple quantum systems as for example trapped ions [4] or alkali atoms [5, 6]. It is nevertheless interesting to continue research on the controlled realization of nonlocal entanglement with other material systems, in particular with degrees of freedom in solid state.

In part this interest is fundamental. Whether entangled states loose their coherence in a different manner than superposition states of individual degrees of freedom is still not fully understood [7, 8]. Recent developments here include an all-optical experiment that showed that entanglement can be lost much more rapidly than the loss of coherence in the two subsystems [9]. Another interesting result from work with entangled photon pairs showed that the relation between the amount of entanglement and the degree of mixedness of a two-particle state can only be represented by a plane of possibilities. Specific points in this plane depend on the nature of the environment that is decohering the initial maximally-entangled pure state [10, 11]. Furthermore, it is still not firmly established that quantum theory does not break down when applied to collective or macroscopic degrees of freedom [12, 13]. This justifies a study of how entangled states can be realized in solid state, and how these states loose their coherence: solid state can provide model systems with complex (collective) degrees of freedom, or systems with elementary degrees of freedom in a complex environment.

Research on the controlled realization of entanglement in solid state systems is also driven by the prospect that it may provide tools for quantum information technologies. Relevant to the discussion here is a proposal for long-distance quantum communication [14] that was until now mainly explored with ensembles of alkali atoms [5, 6], or alkali-atom-like impurities in solids (Refs. [13, 16, 17, 18, 19] and related articles in this issue). However, widespread implementation favors a technique that can be implemented in micron-scale devices that fit inside optical fibers, which are compatible with high-speed opto-electronic operation [20]. Here, the electronic and optical properties of III-V semiconductors outperform the atomic or impurity-based systems. The coherence times of degrees of freedom in these materials, however, tend to be too short for any realization of quantum information technology in the near future, but are long enough for initial experimental studies on entangled states.

We discuss here a technique that enables the controlled realization of nonlocal entanglement between spin-wave modes in ensembles of conduction-band electrons, which are located in two spatially separated pieces of GaAs semiconductor material. We also outline the material properties of a GaAs quantum well system where this technique can be implemented. In Section III we discuss an approach where quantum-optical measurement techniques are used for preparing entangled states of spin degrees of freedom in ensembles of three-level quantum systems. Subsequently, in Section IV we present a GaAs heterostructure material that is suited for realizing such an ensemble of three-level quantum systems.

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II. PREPARING AND DETECTING ENTANGLED STATES VIA QUANTUM OPTICAL MEASUREMENT

We propose here to use the so-called DLCZ scheme [14] for preparing nonlocal entanglement with solid state devices. The main idea behind this approach is that spontaneous emission of a quantum optical pulse results in quantum correlations (entanglement) between the state of the optical pulse and the state of the system that emits. To illustrate this, consider a two-level system that is initially in its excited state $|1\rangle$. It is emitting a single photon while relaxing to its ground state $|0\rangle$. If we would be able to have control over this process such that it relaxes to a superposition of the states $|1\rangle$ and $|\downarrow\rangle$, the system would emit an optical pulse that is a superposition of the states with 0 and 1 photon, $|0_{\text{puls}}\rangle$ and $|1_{\text{puls}}\rangle$. The quantum state of the system and the optical pulse are then in fact entangled, and the only pure states that can describe the state of the combined system are of the form $|\Psi_{\text{com}}\rangle = c_1 |1_{\text{puls}}\rangle + c_2 |\downarrow_{\text{puls}}\rangle$.

Such control over spontaneous emission can be realized with a three-level Raman system (Fig. 1a). When this system is initially in the state $|\downarrow\rangle$, there will be only spontaneous emission of a Raman photon from the transition $|e\rangle - |\downarrow\rangle$ while a control field is driving the $|\downarrow\rangle - |e\rangle$ transition. Figure 1 illustrates how an extension of this scheme can be used to entangle the states of two three-level systems that are at different locations. Say Alice and Bob both have an identical version of such a three-level system prepared in the state $|\downarrow\rangle$. They both use a classical field to drive the $|\downarrow\rangle - |e\rangle$ transition, in order to get very weak spontaneous emission from the $|e\rangle - |\downarrow\rangle$ transition, such that each system emits an optical pulse that is a superposition of the photon-number states $|0_{\text{puls}}\rangle$ and $|1_{\text{puls}}\rangle$ (note that each of these pulses is then entangled with the system that emitted it). The timing and propagation of these two pulses should be controlled such that they arrive at the same time at a measurement station, that consists of a 50/50 beam splitter with a photon counter at each of its two output channels. If the number of photons in the pulses are now measured after combining the two pulses on the beam splitter, there is some probability that one of the two detectors counts 1 photon and the other 0 photons. In that case, the total number of spin flips in the two three-level systems is 1, but it is impossible to tell which of the two emitted the photon. As a result, the systems of Alice and Bob have been projected onto an entangled state of the form $|\Psi_{AB}\rangle = \frac{1}{\sqrt{2}} (|A\rangle |B\rangle + e^{i\phi} |A\rangle |\bar{B}\rangle)$ (where the phase $\phi$ can be derived from experimental conditions [14, 21]).

Figure 1 depicts in fact emission from ensembles of three level systems. For weak (slightly detuned) driving of the $|\downarrow\rangle - |e\rangle$ transition, the expectation value for the total number of $|e\rangle - |\downarrow\rangle$ photons emitted by an ensemble of identical three-level systems can still be less than 1 photon. Notably, the spin excitation is then not stored on an individual three-level system. Instead, it is stored as a spin-wave mode (collective spin excitation) in this medium, with each three-level system having its spin flipped only by a very small amount. Thus, one can also use this approach for preparing entanglement between spin-wave modes in two different ensembles.

These ensembles should have a long elongated shape that is co-linear with the driving field. An important advantage of using such ensembles is that spontaneous emission becomes highly directional [14], with emission predominantly co-propagating with the driving field. In principle the system will emit very weak in all directions, but an initial spontaneous emission event (extremely weak, far below the single-photon level) is strongly amplified (gain) when it co-propagates with the driving field [22]. For very weak driving, the total energy in all of the spontaneous emission can still be at the single-photon level, and the gain then ensures that emission into the desired direction is exponentially stronger than into other directions. Thus, the collection efficiency for the total number of emitted photons by such ensembles can be near unity. This removes the need for using high-finesse optical cavities as in cavity-QED experiments, which is technically very demanding [23].

One should also be able to confirm that entangled states have been prepared by reading out the states of...
each ensemble of a pair that has been entangled. Correlations between the spin excitations in the two ensembles (Fig. 1b) can be studied with an optical readout scheme that uses the inverse of the initial Raman transition. For each system separately, the number of flipped spins in its ensemble can be measured using a control field that is now driving the \(|\downarrow\rangle - |e\rangle\) transition. This converts the spin state that is stored in an ensemble into the state of an highly-directional optical pulse (again a superposition of photon-number states \(|0_{\text{puls}}\rangle\) and \(|1_{\text{puls}}\rangle\)) that results from a subsequent \(|e\rangle - |\downarrow\rangle\) transition. This process fully returns the spin excitation into the \(|\downarrow\rangle\) state. The detection should now directly count the number of photons in the emission from the ensemble that is measured (not using a configuration with a beam splitter). Each of the two ensembles should be measured separately in this manner. If the two ensembles were prepared in a state of the form \(|\Psi_{AB}\rangle = \frac{1}{\sqrt{2}} \left( |\uparrow_A\rangle |\downarrow_B\rangle + e^{i\varphi} |\downarrow_A\rangle |\uparrow_B\rangle \right)\), the number of detected photons from the ensemble of Alice can be 0 or 1, each with probability \(\frac{1}{2}\). However, for either measurement outcome, subsequent measurement of the number of photons emitted by Bob’s ensemble must yield that it is perfectly anti-correlated with the result of Alice.

Such measurements can already provide evidence for the quantum nature of these correlations (in particular, the variance of these photon-count correlations should show strong sub-Poissonian statistics \((22)\)). However, it does not yet allow for a formal test of Bell inequalities (testing for non-classical correlations), since this requires the ability to rotate the basis in which the state of each of the two-level systems is measured (with respect to the basis defined by \(|0_{\text{puls}}\rangle\) and \(|1_{\text{puls}}\rangle\)). This cannot be performed directly with a readout technique based on photon-number measurements. To overcome this, the observation of entanglement between two ensembles of alkali atoms \((\mathbf{3})\) used an approach where a local phase shift was applied to one of the two systems, either to the optical signal pulse from readout \((\mathbf{3})\) or to the stored spin excitation. However, the readout then requires once more to combine the signal pulses from readout of the two ensembles on a beam splitter, and to study the interference fringe that results from the local phase shift. A scheme that only relies on local readout of each ensemble can be realized when the states of both Alice and Bob are not stored in a single ensemble but in a pair of ensembles \((\mathbf{1})\). The photon-number readout can then be implemented with a certain setting for a phase difference between the states of these two ensembles. However, both of these approaches require that the path length between the ensembles and detector stations are stabilized with interferometric precision. An alternative more robust approach could be realized with alkali atom ensembles \((\mathbf{3})\) and used the fact that in these systems the states \(|\downarrow\rangle\) and \(|\uparrow\rangle\) consist of multiple (degenerate) Zeeman sublevels. How a spin excitation is distributed over these Zeeman sublevels is then mapped onto two orthogonal polarizations of a signal field, and polarization selective readout then enables to rotate the basis in which signal fields are measured.

Other solutions that are technically even less demanding are currently investigated \((\mathbf{25, 26, 27, 28})\).

Applying this quantum-optical measurement scheme for preparing entangled states in spatially separated electronic devices is an interesting alternative to related research that uses electronic control and measurement techniques. Activities here use for example electron spins in quantum Hall states \((\mathbf{24})\) or quantum dots \((\mathbf{30})\), or superconducting qubits \((\mathbf{31})\). A first advantage of this quantum optical approach is that it naturally allows for having the two devices separated by a large distance, whereas for electronic control coherent interactions are typically limited to short distances. More importantly, it allows one to use photon-number detection. This is a unique quantum measurement tool in the sense that projective measurement can be used for preparing states with very high fidelity. Tools for electronic readout have typically much higher noise levels, which results in a much weaker correlation between a measurement outcome and the state of the quantum system immediately after measurement.

### III. GaAs Quantum Wells as a Medium for Quantum Optics

We now discuss how such an ensemble of three-level systems can be implemented in a GaAs quantum well system. The techniques presented in the previous section have been mainly developed and explored with ensembles of alkali atoms \((\mathbf{5, 6, 32})\), and developing a realization in solid state can thus be viewed as an attempt to mimic an ensemble of alkali atoms. The key properties of the three-level system as in Fig. 1a are then that the splitting \(\Delta E_{\text{spin}}\) is homogeneous for an ensemble, and that superpositions of \(|\downarrow\rangle\) and \(|\uparrow\rangle\) have a long coherence time \(T_{1\downarrow}\). Further, these two states \(|\downarrow\rangle\) and \(|\uparrow\rangle\) must both have a strong optical transition to a common excited state \(|e\rangle\) that can be addressed selectively. For each leg the spontaneous emission life time must be much shorter than \(T_{1\downarrow}\), a requirement that overlaps with conditions for electromagnetically induced transparency \((\mathbf{33})\). Transient signatures of electromagnetically induced transparency were already observed in related work using excitons in undoped GaAs quantum wells (Ref. \((\mathbf{34})\) and references therein).

We propose here to realize an ensemble of three-level systems with an \(n\)-doped GaAs quantum well system, building on seminal work by Imamoglu \((\mathbf{35})\). These \(n\)-doped GaAs materials combine relatively long coherence times for electron spin superposition states \((\gtrsim 10 \text{ ns} \ (\mathbf{36, 37, 38})\)), with strong optical transitions across the gap that obey good selection rules \((\mathbf{39})\). Transitions between the highest valence band states and lowest conduction band states follow the selection rules for transitions between a \(p_{3/2}\) and \(s_{1/2}\) manifold \((\mathbf{40})\), as represented for bulk GaAs in Fig. 2a. The continuous density of states for bulk GaAs develops into an atom-like discrete set of levels when a two-dimensional electron system (Fig. 2b)
is brought into the quantum Hall regime by applying a strong magnetic field (Fig. 2), because the electrons then condense into cyclotron orbits. The number of these so-called Landau levels that are filled at a certain magnetic field defines the filling factor $\nu$, where each spin-resolved Landau level is counted individually (it can be shown that $\nu$ equals the ratio of areal electron density and areal density of flux quanta $\Phi_0$). We consider the quantum Hall state at filling factor $\nu = 1$, where the conduction-band Landau levels are fully spin polarized. At $\nu = 1$, the lowest Landau level (further denoted as $|\downarrow\rangle$) is fully occupied, while all higher Landau levels are fully unoccupied. The density of states for the valence band is now also a discrete set of Landau levels that are all fully filled. Note that we assume that the magnetic field is in the $-z$ direction, in order to have Fig. 2 compatible with Fig. 1a (in GaAs the electron g-factor is negative).

The system as sketched in Fig. 2 allows for implementing a three-level system, with the lowest spin-up and spin-down Landau levels in the conduction band serving as the two low-energy spin states $|\downarrow\rangle$ and $|\uparrow\rangle$. These both have, for example, an optical transition to the valence-band $|m_z = -\frac{1}{2}\rangle$ state, as the selection rules only allow for transitions with $\Delta m_z \in \{-1, 0, +1\}$. For GaAs quantum well systems, selective addressing of these two optical transitions must rely on polarization selection rules, since the Zeeman splitting for the conduction-band electrons is not in excess of the narrowest line width that can be obtained for optical transitions. In practice, the narrowest lines ($\approx 0.2$ meV) can be obtained for a quantum well width of $\approx 20$ nm, except for the conduction-band $|m_z = \pm \frac{1}{2}\rangle$ states, which are less intense (see, e.g., Refs. [41, 42, 43]). This gives an electron Zeeman splitting of $\approx 0.25$ meV in a field of 10 Tesla (the electron g-factor is not enhanced for spin-wave modes with a wavelength longer than the magnetic length, as observed in electron spin resonance studies on such ensembles [44, 45]). The other energy splittings (partly shown in Fig. 2) are all larger and allow for using spectral selectivity. These splittings are further discussed below.

For the quantum well width that we choose to consider here ($\approx 20$ nm) there are hole-mixing effects that cannot be neglected: the hole energy levels are then a superposition of two or more different angular momentum states (each characterized by a quantum number $m_z$). These effects, however, can be used for implementing a more convenient control scheme, where the three-level system is formed by $|\downarrow\rangle$, $|\uparrow\rangle$ and the highest Landau level in the valence band. This means that one can work with the first optical transition that becomes available when increasing the photon energy from within the gap, and this transition also has the narrowest line. In practice this an important advantage. Using this transition also has the advantage that it is the most isolated level: lower Landau levels in the valence band for holes with (predominantly) $m_z = \pm \frac{1}{2}$ character may be very close to levels for holes with (predominantly) $m_z = \pm \frac{3}{2}$ character for which the quantum number for confinement in the well or Landau level orbital is increased by one [47] (levels not shown in Fig. 2).

**FIG. 2:** a) The lowest conduction-band states and highest valence-band states (for electron wavevector $k = 0$) for bulk $n$-GaAs. This system has good selection rules for optical transitions between energy states with well-defined angular momentum in z direction (quantum number $m_z$). b) As panel a), but now for a $p$-doped GaAs quantum well system (in the $x-y$ plane). Quantum well confinement lifts the degeneracy between the heavy-hole ($m_z = \pm \frac{3}{2}$) and light-hole ($m_z = \pm \frac{1}{2}$) states (levels for in-plane electron wavevector $k = 0$). c) As panel b), but now for this system in a magnetic field in the $-z$-direction, bringing the system in the $\nu = 1$ quantum Hall regime.

How such a three-level system can be implemented in a model system where hole-mixing is included is depicted in Fig. 2a). For the highest valence-band Landau level, the heavy-hole $|m_z = -\frac{3}{2}\rangle$ state mixes pre-
dominantly with the light-hole $|m_z = +\frac{1}{2}\rangle$ state \[47\]. In principle, the $|m_z = -\frac{3}{2}\rangle$ state also mixes with the $|m_z = -\frac{1}{2}\rangle$ state, but this mixing is much weaker. Moreover, its contribution to the optical transition is negligible (parity forbidden) if the quantum well is symmetric, since the mixing is in fact with a $|m_z = -\frac{1}{2}\rangle$ state of a different orbital (it has the quantum number for quantum-well confinement increased by one).

Figure 2 depicts the optical transitions that can be used when this highest hole level is used for operating a three-level system. Note that we consider here co-propagating control field $\omega_c$ and signal field $\omega_s$ in the plane of the quantum well (in $x$-direction, see also Fig. 2b), since this allows us to work with long elongated spin ensembles. This also gives convenient polarization selection rules. The control field $\omega_c$ should address a $\Delta m_z = 0$ transition, which couple to fields that are linearly polarized along the $z$-direction. The signal field $\omega_s$ concerns $\Delta m_z = \pm 1$ transitions, which couples for the case of $m$-plane emission to fields with an orthogonal linear polarization (along the $y$-direction, it would be circularly polarized for propagation orthogonal to plane \[39,40\]). This holds both for the path $|\uparrow\rangle - |m_z = -\frac{1}{2}\rangle$ and (as sketched) the path $|\uparrow\rangle - |m_z = +\frac{1}{2}\rangle$. The analogy with Fig. 1 is more evident when describing the process in terms of holes \[35\]. The control field $\omega_c$ is then driving a hole from the fully filled conduction band level $|\downarrow\rangle$ to the highest valence-band level. This hole can then relax to the $|\downarrow\rangle$ level by emission into the $\omega_s$ field (for this picture the arrows in Fig. 2a should point in the opposite direction). Note, however, that the relevant coherence time for the two low-energy states $|\uparrow\rangle$ and $|\downarrow\rangle$ is nevertheless the spin coherence time for electrons in the conduction band.

Figure 3b presents a device structure that can realize this model system. The GaAs quantum well is embedded in an $\text{Al}_x\text{Ga}_{1-x}\text{As}$ single-mode optical waveguide. Narrow (as compared to the optical wavelength) electrical contacts on the side of this waveguide serve for in-situ monitoring of the quantum Hall state of the electron ensemble inside the waveguide. The quantum well inside the piece of waveguide then contains a single electron ensemble, such that the device structure of Fig. 3b represents one of the two spin ensembles in the scheme of Fig. 1b. This device structure naturally implements the situation that the control and signal fields co-propagate and have perfect overlap with a spin ensemble that has a long elongated shape. Furthermore, the engineering of the quantum well and the waveguide with cladding layer naturally fit together, using the fact that for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ material the energy gap increases with increasing Al content, while the index of refraction decreases with increasing Al content \[48\]. Figure 3b presents how this can be realized. The GaAs quantum well is located in the middle of an $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layer that serves as the core of the waveguide. This waveguide core is transparent for the optical fields that are used since the gap of $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ is larger than the gap of GaAs. The $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layer acts as a waveguide core since it is embedded between layers with a lower index of refraction (either formed by $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$ or vacuum).

We end this section with further quantifying the material parameters. We already argued that a symmetric GaAs quantum well system with a width of 20 nm is the optimal choice. To bring it in the quantum Hall state $\nu = 1$ with a magnetic field of about 10 T, the quantum well should contain a high-mobility electron gas with a
density of $n_s \approx 2.4 \cdot 10^{15} \, \text{m}^{-2}$. In a field of 10 T, the Landau levels are as depicted in Fig. 2d, and we will discuss here the other energy splittings of this system. The literature is not very conclusive about the effective g-factor for the light- and heavy-hole levels. This results from the fact that these depend on the quantum well width, Al content of the Al$_x$Ga$_{1-x}$As barriers, strength and direction of the magnetic field, and hole mixing effects. Nevertheless, most results indicate that the Zeeman splittings for holes at 10 T in a 20 nm system are substantially larger than $\approx 0.2 \, \text{meV}$ [17, 49]. Typical values are close to the energy spacings to the next hole Landau levels (with identical value for $m_z$, but with the Landau-orbital quantum number one higher). These splittings are under these conditions all $\gtrsim 3 \, \text{meV}$, while the splittings between conduction-band Landau levels are much larger thanks to the low effective mass of electrons [47]. Finally, the energy spacings between the subbands due to confinement are for this system $\approx 25 \, \text{meV}$ for electrons, $\approx 5 \, \text{meV}$ for heavy holes and $\approx 20 \, \text{meV}$ for light holes [39, 42]. This also sets the scale for the splitting between the highest heavy- and light-hole levels (Fig. 2b), which is about $\approx 5 \, \text{meV}$ [39].

IV. CONCLUSIONS

The reasonably long coherence times for electron spin ensembles in $n$-doped GaAs materials allows for studies of how such ensembles can act as a medium for quantum optics. We showed that this idea is feasible, and that this allows for preparing entanglement between states of spin wave modes in two different ensembles. For initial studies, an $n$-doped GaAs quantum well system in the quantum Hall $\nu = 1$ state provides the most promising model system. The electron ensembles are addressed by placing the quantum wells inside optical waveguides, with in-plane propagation of optical control and signal fields. Realizing such systems is compatible with standard epitaxial growth techniques for GaAs/Al$_x$Ga$_{1-x}$As heterostructures. We analyzed that an optimal system is formed by a symmetric GaAs quantum well of about 20 nm width. In this system one can address electron spin degrees of freedom inside ensembles of three-level quantum systems with optical transitions across the gap. The most suitable three level system uses transitions between the conduction band spin states and the highest Landau level of the valence band. Selective control over these two transitions is possible with polarization selection rules and using hole-mixing effects that naturally occur in this system.

Progress towards the realization of entanglement with such a system first requires spectroscopy with fields that propagate in plane to confirm the optical selection rules (in particular with respect to the hole mixing). Also Pauli blocking when driving a completely filled Landau level needs to be demonstrated. A crucial next step is then to demonstrate electromagnetically induced transparency (EIT), as this provides evidence that a medium is suited for the quantum optical techniques that we discussed here. If these steps are successful, this clean material system is a very promising candidate for studies of entanglement with ensembles of conduction band electrons in solid state. In particular, the observed long spin coherence times for electron spin ensembles imply that the Zeeman splittings are very homogeneous in these ensembles. This allows to generate Raman scattered fields from two different ensembles that are centered at identical optical frequencies, while their spectral width is tuned by the EIT bandwidth [33]. Consequently, the two signal pulses then have very good spectral overlap, and preparing entanglement by interfering these two pulses on a beam splitter should indeed be possible.

Acknowledgments

We thank X. Liu, D. Reuter, D. Gerace, H. E. Tureci and A. Imamoglu for help and stimulating discussions. This research is supported by the Netherlands Organization for Scientific Research (NWO).

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