Terahertz-induced depletion of the ground-state population of neutral donors in GaAs measured by resonant elastic light scattering from donor-bound excitons

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Strong resonant elastic light scattering (RELS) from the donor-bound exciton transition in GaAs (1.514eV) occurs at neutral donors in the ground (1S) state, but not at neutral donors in excited hydrogenic states. When 1.6 THz radiation is incident on an ensemble of neutral donors, we observe up to a 30% decrease in the RELS, corresponding to a decrease in the population of neutral donors in their ground states. This optical detection method is similar to quantum nondemolition measurement techniques used for readout of ion trap quantum computers and diamond nitrogen-vacancy centers. In this scheme, Auger recombination of the bound exciton, which changes the state of the donor during measurement, limits the measurement fidelity and maximum NIR excitation intensity.

Neutral shallow donors in semiconductors provide a model system for the study of quantum information in solid state materials for both spin-based and electronic orbital-based qubits. Comprising a positively charged donor and a single bound electron, neutral shallow donors exhibit a hydrogen-like energy spectrum scaled by the electron effective mass ratio \( m^*/m_e \) and the inverse square of the relative dielectric constant \( \varepsilon_r \). In hydrogenic orbital-based neutral donor qubits, the 1S and 2P levels of the bound electron serve as qubit states. For GaAs, the binding energy of electrons to hydrogenic donors, such as S or Si, is \( \sim 5.9\text{meV} \), resulting bound state transition frequencies \( \sim 1\text{THz} \). These are well below the LO phonon energy \( (36\text{meV}, 8.7\text{THz}) \), leading to long excited state lifetimes \( t_1 = 350\text{ns} \).\(^4,5\) Cole, et al. demonstrated Rabi oscillations in ensembles of shallow donors in GaAs, using photoconductivity as the measure of the excited state population.\(^4\) However, photoconductivity is only indirectly sensitive to the state of the donor and completely destroys the qubit. It is desirable to have an alternative means of readout which: (1) is directly sensitive to the state of the bound electron, (2) allows rapid distinguishing between states of the qubit, and (3) achieves a quantum nondemolition (QND) measurement (i.e. leaves the qubit in the state reported by the measurement).

In ion trap quantum computers\(^7\) QND measurement is achieved by using an optical probe. The qubit states are hyperfine states of the trapped ion, and readout is done by resonantly exciting atoms from the lowest hyperfine state to an auxiliary state. Selection rules prevent a spontaneous relaxation from the auxiliary state to other hyperfine states. During readout, atoms in the lowest hyperfine state continually absorb and re-emit, or scatter, the incident light and are visible, whereas atoms in excited hyperfine states remain dark.\(^8\) A photoluminescence-based optical readout technique has been used to observe magnetic resonance\(^9\) and coherent quantum dynamics in single nitrogen-vacancy centers.\(^10\) In this report, we introduce the use of resonant near infrared (NIR) excitation of the neutral donor \((D^0)\) ground (1S) state to donor-bound exciton \((D^0X)\) transition as a means of determining whether neutral donors are in the ground (1S) state. Absorption and re-emission, i.e. scattering, of the NIR light occurs at donors in the 1S state, whereas donors in excited hydrogenic states do not interact with the light. Using this technique we observe changes in the population of donors in the 1S state due to excitation of the donor-bound exciton with THz radiation.

Detection of visible/NIR emission as a means of observing THz/FIR dynamics in semiconductors is a well-used technique. In related work, FIR (THz) modulated photoluminescence (PL) has been used to observe phenomena in a variety of semiconductor structures, including bulk GaAs\(^11\) undoped quantum wells\(^11\) coupled quantum wells\(^11\) and InGaAs quantum dots.\(^11\) McCombe and coworkers have used a variety of optical detection schemes for FIR spectroscopy including both nonresonant (PL) and resonant (specularly reflected) emission.\(^12\) Our technique differs from the above optically detected FIR studies in that we measure scattered, nonspecular emission at the NIR excitation frequency, rather than PL only, or specular reflection.

Shallow or hydrogenic donors have been the subject of considerable theoretical and experimental work.\(^13\) In particular, the \(D^0X\) state has received a great deal of attention. Karasyuk et al. utilized \(D^0X\) transitions to identify donors in GaAs as well as determine donor binding energies and central cell corrections.\(^14\) (Measurements of GaAs neutral donor properties have also been made by FIR absorption spectroscopy and photoconductivity.\(^15\))

The donor-bound exciton state \((D^0X)\) is a short-lived \(H_2\)-like complex (binding energy \( \sim 1\text{meV} \)) with several rotational bound states. The bound exciton recombines optically with an average lifetime of 1ns \((232\text{ps in a quantum well})\).\(^16,17\) Donor-bound excitons may be created by above band gap excitation of free excitons followed by capture via phonon emission (Fig. 1). In that case, the light emitted from bound exciton recombination is
PL (see Fig. 2). Alternatively, the sample may be excited below the band gap, at the D\textsuperscript{0}X resonance (Fig. 1b). Light emitted from the sample at the laser frequency may be due to either resonant Rayleigh scattering or resonance fluorescence (light absorbed and re-emitted by D\textsuperscript{0}X recombination). We use the term resonant elastic light scattering (RELS) to refer to all the light collected at the laser frequency \( \lambda \). Since the probability of decay from a D\textsuperscript{0}X state to the neutral donor 1S state (D\textsuperscript{0}X) is high, donors measured to be in the 1S state continually cycle between the 1S state and the D\textsuperscript{0}X state, elastically scattering many NIR photons. Donors in excited hydrogenic states do not absorb the incident light, and are dark.

When the bound exciton decays, there is a nontrivial probability of transferring energy to the electron bound to the donor, leaving it in an excited state. If that occurs, the state of the donor is changed during the measurement. This Auger recombination process is the limiting factor in the ability of the D\textsuperscript{0}X RELS technique to make a QND measurement of a neutral donor. Although D\textsuperscript{0}X RELS does not yet provide a high fidelity QND measurement, the probability of return to the ground state is high enough to enable sensitive detection of population electrons in the D01S state, under low intensity NIR excitation conditions.

Samples for all experiments consisted of a 15\textmu m layer of unintentionally-doped high-purity GaAs grown by molecular beam epitaxy on a 500\textmu m thick semi-insulating GaAs substrate. The effective donor density \( N_d - N_a \) was inferred to be \( 3 \times 10^{14} \text{cm}^{-3} \) from Hall effect measurements. FIR photoconductivity indicates the dominant donor impurities are S and Si, and the PL spectrum confirms acceptors are a relatively minor impurity. For optical measurements, a GaAs sample was mounted in a strain-free manner on the cold finger of a liquid helium flow cryostat with a minimum sample temperature near 5K. The sample was excited in the NIR by a tunable external cavity diode laser with typical intensities \( \sim 1 \text{mWcm}^{-2} \), incident at Brewster’s angle. Emission, including both PL and elastically scattered light, was collected from the surface normal by a lens and focused into a 0.75m imaging spectrometer and detected by a CCD or PMT (see inset to Fig. 3). A CO\textsubscript{2} laser-pumped difluoromethane gas laser was used for FIR (THz) excitation. Characteristic incident intensities were less than 20mWcm\(^{-2}\).

Fig. 2 shows a typical 5K GaAs PL spectrum. Peak assignments follow those of Ref. 17. Transitions from various D\textsuperscript{0}X rotational states to the D\textsuperscript{0}1S state (direct recombination) occur in the region 1.5139-1.515eV. Transitions involving Auger recombination, often called two electron satellite (TES) transitions in semiconductor literature, leave the donor in an excited state. These are visible in the region 1.509-1.51515eV. The observed recombination of excitons bound to ionized donors (D\textsuperscript{+}X) near 1.513eV occurs primarily in the \( \sim 1 \mu m \) depletion region near the unpassivated surface. The PL associated with excitons bound to neutral acceptors (A\textsuperscript{0}X) occurs between 1.512 and 1.513eV.

The solid line in Fig. 3 is a typical emission spectrum with excitation in the region of the two lowest direct D\textsuperscript{0}X transitions (labelled L=0 and L=1 in Fig. 2 inset). RELS is much stronger than PL, and strongest when excitation...
is resonant with a $D^0X$ transition. Open circles in Fig. 3 trace the RELS peak height from a series of emission spectra taken across a range excitation energies. The two inhomogeneously broadened transitions are much better resolved by RELS than by PL (compare Fig. 3 with inset to Fig. 2).

When FIR photons with enough energy to promote bound electrons to a higher hydrogenic state, or the conduction band, are incident on the sample, fewer donors are in their ground state, and hence less RELS from $D^0X$ to $D^01S$ transitions is observed. Fig. 4 shows the percent change in the RELS at several NIR excitation energies with FIR light of frequency 1.63THz (6.73meV) and 20mWcm$^{-2}$ intensity incident on the sample. Since the FIR excitation energy is greater than the 5.9meV binding energy of the donors, electrons are excited directly to the conduction band. The percent change in RELS, or RELS modulation, is calculated as $100\left(\frac{A-B}{A+B}\right)$, where $B$ and $A$ are the respective RELS signals with and without FIR excitation. The curve in Fig. 4 indicates the modulation is greatest when the NIR is resonant with a $D^0X$ transition. The experimentally observed modulation depends on the relative sizes and overlap of the NIR and FIR laser spots, the NIR laser frequency and intensity, FIR laser intensity and sample temperature. The maximum experimentally observed value of the modulation with FIR excitation at 1.6THz was 30%. Modulation of the RELS due to FIR excitation at 1.4THz (5.78meV, below the electron ionization energy) or 1.04THz (4.31meV, slightly detuned from the 1S-2P bound transition) is similar in magnitude. In future experiments, a freely tunable FIR source such as a free electron laser or energy level tuning via magnetic field could be used to investigate RELS modulation under conditions where the FIR is in resonance with the 1S-2P transition, in order to observe Rabi oscillations, or other resonant or coherent effects.

RELS efficiency, like photoconductivity, is sensitive to changes in sample temperature. RELS efficiency drops (approximately linearly) to 1/7 of the 5K value by 15K, and then more slowly to 1/10 of the 5K value by 20K. (This behavior is expected, because the binding energy of excitons to the neutral donors is only 1meV, and $k_BT = 1$meV at 11.6K.) In order to be useful for qubit readout, the measured change in RELS would have to be due to a change in the donor ground state population, and not merely a reduction in RE intensity due to lattice heating. Fig. 5 shows the dependence of the RELS modulation on incident FIR intensity. The mod-
ulation initially saturates at 30%, a value much lower than is achievable by thermal effects ($B = 1/7$ or 90%). The fact that the change in RELS is constant to within 1% for FIR modulation frequencies $\leq 400$Hz, implies the thermal time constant ($\tau$) of the sample is $< 1/10$kHz. The corresponding temperature rise ($\Delta T$) in terms of the absorbed power ($P_{abs}$), mass ($m$) and specific heat ($c_p$) is approximately given by $\Delta T = P_{abs} \tau / (mc_p) = 0.2 K$.

Also, the linear response at low THz intensities, where thermal effects are smallest, is consistent with absorption modulation. Therefore, we are confident the modulation is dominated by changes in the population of donors in their ground states, not heating.

For qubit readout, an important figure of merit related to measurement fidelity is the ratio of the amount of light emitted from direct D$^0$X to D$^1$S transitions to the amount of light collected from state-altering Auger transitions. Exciting at 1.5142eV (above the two lowest direct D$^0$X transitions), the amount of light collected in the direct transition region (1.5138-1.515eV) is 133 times larger than light collected in the TES region (1.509-1.5117eV). Exciting on resonance with the D$^0$X, L=0 transition at 1.514eV, the ratio increases to a maximum of 1250, while the integrated emission in the direct transition region increases by a factor of 60. This indicates resonant excitation is superior to PL and/or resonant excitation from both a signal detection and measurement fidelity standpoint.

Measurement of donor ground state occupation via D$^0$X RELS may provide a solution for single qubit readout, provided adjacent donors can be optically resolved, and Auger transitions can be reduced to a manageable level via, for instance, choice of magnetic field, crystal orientation and NIR polarization. The D$^0$X lifetime is 1ns, so a single donor may scatter up to $10^9$ photons per second, if excited near saturation, and no Auger transitions to excited states occur. For a collection efficiency of 0.1, 10 photons could be collected in 100ns (10 photons per 23ns in a quantum well). Currently, the collected RELS is limited by the NIR excitation intensity, which must be kept below 1mWcm$^{-2}$. Higher NIR intensities result in decreased RELS modulation (−3dB at 4 ± 1mWcm$^{-2}$). The reason for the decrease in RELS modulation with increasing NIR intensity remains to be investigated, but could be due to donors becoming shelved in long-lived excited hydrogenic states via Auger transitions, or increased scattering from free excitons.

We note that D$^0$X RELS provides an alternative means of detection for studying properties of bound electron quantum states, in addition to absorption saturation spectroscopy and photoconductivity. One such application may be time resolving the recovery of donors to the ground state to determine bound-to-bound state lifetimes.

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