Optically detected measurement of the ground-state population of an ensemble of neutral donors in GaAs

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Strong resonant elastic light scattering (RELS) from the donor-bound exciton transition in GaAs (1.514 eV) occurs at neutral donors in the ground (1*S*) 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 signature allows for optical detection of the ground-state population of donors, and is similar to quantum nondemolition measurement techniques used for readout of ion trap quantum computers. In this scheme, Auger recombination of the bound exciton, which changes the state of the donor during measurement, limits the measurement fidelity and maximum near infrared laser excitation intensity.

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I. INTRODUCTION

Shallow or hydrogenic donors in semiconductors have been the subject of considerable theoretical and experimental work.[1] In particular, the donor bound exciton (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.[2] Measurements of GaAs neutral donor properties have also been made by far infrared (FIR or THz) photoconductivity.[3]

Neutral shallow donors in semiconductors provide a model system for the study of quantum information in solid state materials for both spin-based[4,5] and electronic orbital-based qubits.[6] Comprised of positively charged donor and a single bound electron, neutral shallow donors exhibit a hydrogenlike energy spectrum scaled by the electron effective mass ratio (m^*/m_e) and the inverse square of the relative dielectric constant (ϵ_r). In hydrogenic orbital-based neutral donor qubits, the 1*S* and 2*P* 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 ~5.9 meV, resulting bound state transition frequencies ~1 THz. These are well below the LO phonon energy (36 meV, 8.7 THz), and have long lifetimes (T_1=350 ns).[7,8] Cole *et al.* demonstrated Rabi oscillations in ensembles of shallow donors in GaAs, using photoconductivity as the measure of the excited state population.[6] In their measurements, coherent transfer of the electron population to the excited state was accomplished on the scale of tens of ps. However, photoconductivity measures the conduction band population rather than the state of the electron bound to the donor.[9] Furthermore, photoconductive readout 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 (i.e., $T_{readout} \ll T_1$), 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[10] QND measurement is achieved by using an optical probe for readout. 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 spontaneous relaxation from the auxiliary state to other hyperfine states. During readout, atoms in the lowest hyperfine state continually absorb and reemit the incident light as they cycle between the ground and auxiliary states. The light scattered by this "cycling transition" indicates the atom is in the ground state. Atoms in an excited hyperfine state cannot absorb the incident light due to strict polarization selection rules, as well as being detuned from the laser excitation frequency. Thus atoms in the excited state are dark. Reference 11 describes the experimental details of such a measurement. The term "method of quantum jumps" is synonymous with this light scattering-based readout scheme.[12–14] A similar QND optical readout technique based on photoluminescence has been used to observe magnetic resonance[15] and coherent quantum dynamics in single nitrogen-vacancy centers in diamond.[16]

In this article, we introduce a light scattering technique for readout of neutral donors, which is analogous, in concept, to the method used for readout in ion traps. Rather than hyperfine states of an atom trapped in a vacuum chamber, the qubit states are orbital states of an electron bound to an impurity atom in a semiconductor lattice. The D^0X state serves as the auxiliary state for readout. Under resonant excitation, donors in the ground state continually absorb and reemit, or scatter near infrared (NIR) photons and appear bright [Fig. 1(a)]. Donors in excited states do not interact with the light, and are dark [Fig. 1(b)]. We used this technique to measure changes in the ground-state population of an ensemble of neutral donors (D^0) in GaAs by exciting the electron bound to the neutral donor out of its ground ($D^0 1S$) state with THz light.

Detection of visible or NIR emission as a means of observing THz dynamics in semiconductors is a well-used tech-

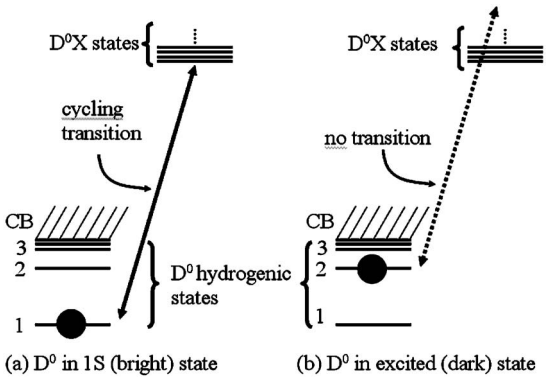

FIG. 1. Schematic energy level diagram (not to scale) illustrating the use of the donor bound exciton state for readout. Initial states are hydrogenic orbitals of the electron bound the the neutral donor (D^0), shown relative to the conduction band (CB), or ionization potential. Several donor bound exciton (D^0X) rotational states are shown. (a) Neutral donors in the $1S$ state (D^01S) absorb near infrared laser light tuned to resonance with the D^0X transition, and return to the ground state with high probability by reemitting light of the same energy (cycling transition). The presence of scattered light indicates the donor is in the ground (bright) state. (b) Donors in excited hydrogenic (dark) states do not scatter light because because optical transitions are weak and excitation is off-resonance.

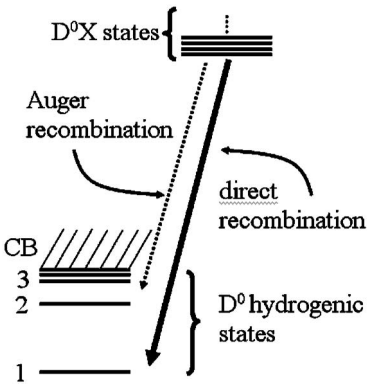

FIG. 2. Nonidealities associated with D^0X decay. The D^0X state decays via one of two pathways: (i) Direct recombination of the bound exciton is fast (<1 ns) and leaves the donor in the ground (D^01S) state. (ii) Exciton recombination with simultaneous excitation of the electron bound to the donor (Auger recombination) changes the state of the donor during measurement. These "two electron satelite" (TES) transitions occur at a much slower rate, and result in leakage of electrons into excited hydrogenic states during measurement (see also Fig. 3).

nique. In related work, THz modulated photoluminescence (PL) has been used to observe phenomena in a variety of semiconductor structures, including bulk GaAs,[17] quantum wells,[18] coupled quantum wells,[19] and InGaAs quantum dots.[20] McCombe and coworkers have used a variety of optical detection schemes for THz spectroscopy including both nonresonant (PL) and resonant (specularly reflected) emission.[21] Our technique differs from the above optically detected THz studies in that we measure scattered, nonspecular emission at the NIR excitation frequency, rather than PL only, or specular reflection.

The D^0X has several properties which make it attractive for use as an auxiliary state for readout of the neutral donor. First, the average time for a bound exciton to recombine is 1 ns (232 ps in a quantum well),[22,23] which is one to two orders of magnitude shorter than the lifetimes of cycling transitions used in ion traps. This allows a donor to scatter many photons during a short readout time. Second, since the binding energy of the exciton to the neutral donor is small (1 meV), excitons do not bind to donors in excited hydrogenic states ($2S$, $2P$, etc.). Therefore, optical transitions between the D^0X and excited hydrogenic states are weak. When an exciton bound to a neutral donor recombines, it will therefore return the donor to the D^01S state with high probability (Fig. 2). In ion traps, transitions from the auxiliary state to excited hyperfine states are so strongly forbidden that they can be entirely ignored. Thus atoms in the ground state are not pumped to excited hyperfine states when the readout laser is turned on. This is critical for QND measurement. However, for excitons bound to neutral donors in GaAs, there is a finite branching ratio. A spontaneous decay from the D^0X state to an excited hydrogenic state changes the state of donor during the measurement. Such Auger transitions, often called two-electron satellite (TES) transitions in semiconductor literature (Figs. 2 and 3), are the limiting factor in our ability to make a QND measurement of the state of the neutral donor. Although our technique 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 electron population in the D^01S state, under low intensity NIR excitation conditions.

Experimentally, the population of donors in the ground state is measured by exciting the D^01S to D^0X transition resonantly and observing the amount of light reemitted from the sample at the laser frequency. In this article, we use the phrase resonant elastic light scattering (RELS) to refer to all the light collected at the frequency of the excitation laser,

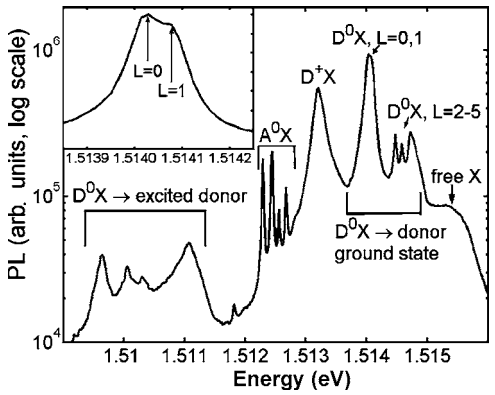

FIG. 3. 5 K PL spectrum (log scale) of GaAs ($N_d-N_a=3\times10^{14}$ cm^{-3}) taken with CCD and grating spectrometer at 35 μeV (0.02 nm) resolution. Laser excitation was at 1.52 eV, 1 mW cm^{-2} intensity. Labels identify transitions associated with the free exciton (*free X*), the neutral donor-bound exciton (D^0X), the ionized donor-bound exciton (D^+X), and the neutral acceptor-bound exciton (A^0X). L denotes the rotational quantum number of the D^0X initial state. (Inset) Linear scale PL of direct exciton recombination from the two lowest D^0X rotational levels.

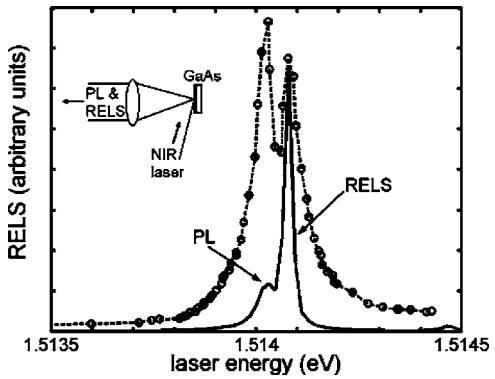

FIG. 4. 5 K resonant emission spectra. The solid line is a typical emission spectrum including both RELS and PL. In this spectrum, the laser is resonant with the D^0X $L=1$ transition (labeled RELS). PL from the D^0X $L=0$ transition is also present. The open circles trace the RELS peak height obtained from a series of spectra at various excitation energies. The dashed line is a guide to the eye. The linewidths of the two transitions are due to inhomogeneous broadening. (Inset) Experimental geometry for collection of NIR emission.

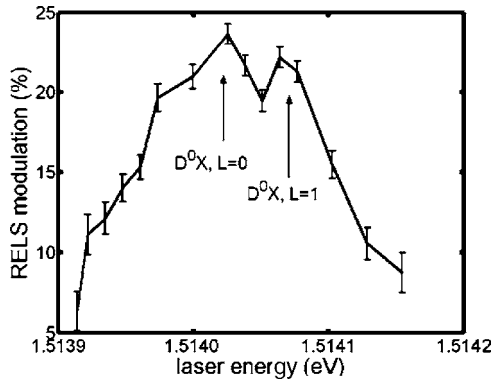

FIG. 5. RELS modulation versus incident NIR laser frequency (intensity ~ 1 mW cm^{-2}). Sample temperature is 5 K and the incident THz power is ~ 20 mW cm^{-2}. The error bars are due to fluctuations in the intensity of the molecular gas laser. Labels denote the spectral features shown in Fig. 3 (inset) and Fig. 4.

which may be a combination of both resonant Rayleigh scattering and resonance fluorescence.[24,25]

II. EXPERIMENTAL METHODS

Samples for all experiments consisted of a 15 μm layer of unintentionally-doped high-purity GaAs grown by molecular beam epitaxy on a 500 μm thick semi-insulating GaAs substrate. The effective donor density ($N_d - N_a$) was inferred to be 3×10^{14} cm^{-3} from Hall effect measurements. THz 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 5 K. The sample was excited in the NIR by a tunable external cavity diode laser (<5 MHz spectral linewidth) with typical intensities ~ 1 mW cm^{-2}, incident at Brewster's angle. Emission, including PL and elastically scattered light, was collected from the surface normal by a lens and focused into a 0.75 m imaging spectrometer and detected by a CCD or PMT (see inset to Fig. 4). A CO_2 laser-pumped difluoromethane gas laser was used for THz excitation. Characteristic incident THz intensities were less than 20 mW cm^{-2}.

III. DATA

Figure 3 shows a typical 5 K GaAs PL spectrum with excitation greater than the free exciton energy. Peak assignments follow those of Ref. 2. Transitions from various D^0X rotational states to the D^01S state (direct recombination) occur in the region 1.5139–1.515 eV. Transitions involving Auger recombination (TES transitions) leave the donor in an excited hydrogenic state. These are visible in the region 1.509–1.5115 eV. The observed recombination of excitons bound to ionized donors (D^+X) near 1.513 eV comes from donors in the ~ 1 μm depletion region near the unpassivated surface of the sample. PL associated with excitons bound to neutral acceptors (A^0X) occurs between 1.512 and 1.513 eV.

When exciting below the band gap, RELS is typically much stronger than PL from nearby D^0X transitions, and is strongest when excitation is resonant with a D^0X transition. The solid line in Fig. 4 shows a typical emission spectrum in the region of the two lowest direct D^0X transitions (labelled $L=0$ and $L=1$ in inset to Fig. 3). The spectrum includes both elastically scattered laser light (RELS) and PL. By fitting a Gaussian curve to the RELS peak in the emission spectrum, the RELS peak position and height can be obtained. Open circles in Fig. 4 trace the RELS peak height from a series of emission spectra taken across a range excitation energies, spanning the $L=0$ and $L=1$ D^0X to D^01S transitions. The transitions are much better resolved by RELS than by PL (compare Fig. 4 with inset to Fig. 3). This improved resolution is not due to the CCD pixel size or spectrometer resolution, but rather, is attributable to the fine-tuning capability of the laser and the sensitivity of a gaussian fit to changes in the center position, even if that change is much smaller than the width of a single CCD pixel. The observed linewidths of ~ 45 μeV in Fig. 4 are primarily due to inhomogeneous broadening.

To verify that D^0X RELS is directly sensitive to the population of bound electrons in the $1S$ state, we used THz radiation to excite donors from the ground state, and recorded the corresponding change in RELS. If THz 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 should be observed. Figure 5 shows the percent change in the RELS at several NIR excitation energies with light of frequency 1.63 THz (6.73 meV) and 20 mW cm^{-2} intensity incident on the sample. Since the THz excitation energy is greater than the 5.9 meV binding energy of the donors, electrons are excited directly to the conduction band. The percent change in RELS, or RELS modulation, is calculated as $100(A-B)/[(A+B)/2]$, where B

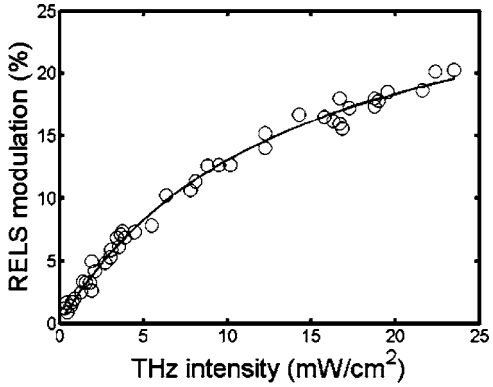

FIG. 6. RELS modulation versus incident THz intensity. Open circles are data points. A fit to a two-level absorption saturation curve of the form $A[1-(1+I/I_0)^{-1}]$ is included as a guide to illustrate the asymptotic behavior (solid line). The fit parameters are $I_0=13.7$ and $A=31$.

and A are the respective RELS signals with and without THz excitation. The curve in Fig. 5 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, THz intensity and sample temperature. The maximum experimentally observed value of the modulation with excitation at 1.6 THz was 30%. Modulation of the RELS due to excitation at both 1.4 THz (5.78 meV, below the electron ionization energy) and 1.04 THz (4.31 meV, slightly detuned from the $1S$-$2P$ bound transition) is similar in magnitude. In future experiments, a freely tunable THz 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 THz radiation is in resonance with the $1S$-$2P$ transition, in order to observe Rabi oscillations, or other coherent effects.

IV. DISCUSSION

Due to the weak binding energy of excitons to shallow donors, RELS efficiency is strongly sensitive to changes in sample temperature (1 meV$=k_BT$ at 11.6 K). RELS efficiency drops (approximately linearly) to 1/7 of the 5 K value by 15 K, and then more slowly to 1/10 of the 5 K value by 20 K. In order to be useful for qubit readout, the measured change in RELS would have to be due to a change in the population of donors in the ground state, and not merely a reduction in RELS intensity due to lattice heating. Figure 6 shows the dependence of the RELS modulation on incident THz intensity. The modulation 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 modulation frequencies $\leqslant$400 Hz, implies the thermal time constant (τ) of the sample is $<1/10$ kHz. The corresponding temperature rise (ΔT) 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 due to changes in the ground-state population. 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^0X to D^01S transitions to the amount of light collected from state-altering Auger transitions. Exciting at 1.5142 eV (above the two lowest direct D^0X transitions), the amount of light collected in the direct transition region (1.5138–1.515 eV) is 133 times larger than light collected in the TES region (1.509–1.5117 eV). Exciting on resonance with the D^0X, $L=0$ transition at 1.5140 eV, 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 provides both superior signal and measurement fidelity than PL and/or off resonant excitation.

Measurement of donor ground-state occupation via D^0X 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. With a D^0X lifetime of 1 ns, 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 100 ns (10 photons per 23 ns in a quantum well). Currently, the collected RELS is limited by the NIR excitation intensity, which must be kept below 1 mW cm^{-2}. Higher NIR intensities result in decreased RELS modulation (−3 dB at 4 ± 1 mW cm^{-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 decreased RELS efficiency due to the presence of free excitons.

We note that D^0X RELS provides an alternative means of detection for studying properties of bound electron quantum states, in addition to absorption saturation spectroscopy and photoconductivity. The technique does not require a magnetic field, external electric bias, and does not require strong THz excitation. Further, it utilizes compact and commercially available semiconductor lasers and can be detected by silicon-based optoelectronics, which typically have their peak responsivity near 1.5 eV. One application of D^0X RELS may be time resolving the recovery of donors to the ground state to determine bound-to-bound state transition lifetimes.

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