Semiconductor quantum well excitons in strong, narrowband terahertz fields

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Abstract. Optical transitions between exciton states in semiconductors—intraexcitonic transitions—usually fall into the terahertz (THz) range and can be resonantly excited with narrowband, intense THz radiation as provided by a free-electron laser. We investigate this situation for two different quantum well structures by probing the near-infrared excitonic absorption spectrum near the band edge. We observe the dynamical Stark—or Autler–Townes—splitting of the 1s exciton ground state and follow its evolution for various THz photon energies and field strengths. The behavior is considerably more complex as compared to the atomic systems. At the highest field strengths, where the Rabi energy is of the same order of magnitude as the exciton level separation, the system cannot be described within the standard framework of a two-level system in rotating wave approximation. When the ponderomotive energy approaches the exciton binding energy, signatures of exciton field ionization are observed.

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1. Introduction

The coupling of intense electromagnetic waves to electrons in matter is one of the most fundamental problems in physics. In particular, the occurrence of dressed states [1], arising when a quasi-monochromatic electromagnetic wave is coupled resonantly to a two-level system, represents the starting point of any non-perturbative nonlinear optics treatment. Having been studied in atomic and molecular systems for many decades, first investigations of semiconductors date back to the 1980s [2–4]. There are four fundamental energy or frequency scales involved in this problem: (i) the energy level spacing \( \omega_{21} \), (ii) the angular light (terahertz (THz)) frequency \( \omega \), being close to \( \omega_{21} \) in the near-resonant case: \( \omega \approx \omega_{21} \); (iii) the Rabi frequency \( \Omega = \mu_{21} F / \hbar \) describing the light–matter coupling, where \( \mu_{21} \) is the dipole matrix element and \( F \) is the electric field amplitude and (iv) a decay constant \( \gamma \) describing relaxation processes (strictly a population and a polarization decay constant). In the usual model applied in atomic physics, the condition \( \Omega << \omega \) is fulfilled to a very high degree, which allows one to use the rotating wave approximation (RWA) by neglecting rapidly oscillating terms. Then the relevant equations (Schrödinger or density matrix) for the two-level system can be solved analytically, yielding the Autler–Townes [5] splitting in steady state and Rabi oscillations in the time domain before the coherences have decayed.

Although in atomic systems it is easier to choose the Rabi frequency \( \Omega \) larger than the relaxation rates \( \gamma \), in semiconductors, due to the short decay constants of less than picoseconds, relatively large intensities (\( \sim \text{MW cm}^{-2} \)) are necessary to reach this condition. Such intensities cannot be applied to semiconductors in steady state, which is why relevant experiments need to be performed with pulsed (sub)picosecond lasers [6]. For even higher intensities, such that \( \Omega \approx \omega \), the RWA breaks down and the picture becomes more complicated. In fact, by using ultra-short (5 fs) high-power pulses this regime has been explored for interband excitations in bulk GaAs by Mücke et al [7] using intensities up to the TW cm\(^{-2} \) range. Modern laser technology has enabled researchers to yet enter a realm of even higher intensities and electric fields. In this regime of extreme-nonlinear optics or ultra-high electric fields, the ac electric field becomes of the same order of magnitude as the atomic electric field, e.g. the Coulomb field in atoms. Recently, one has been able to access this parameter range in atoms in what is now called attosecond physics [8]. Note that here the electrons follow the electric field quasi-statically if the so-called ponderomotive energy, \( U = e^2 F^2 / 4m\omega^2 \) becomes the largest energy scale in the problem (where \( e \) is the elementary charge and \( m \) is the carrier mass). A quantity often defined to characterize this regime is the so-called Keldysh parameter, being the square root of the ratio of ionization energy and twice the ponderomotive energy. Very closely related to this is a quantity used by Nordstrom et al [9], the ratio between the ponderomotive energy and \( \hbar \omega \).
These high-field limits can be reached at much lower intensities if the fundamental frequencies $\omega$ and $\omega_{21}$ are smaller, as is the case in the THz frequency region. For example, a typical excitation energy of 10 meV corresponds to 2.4 THz. For a long time, nonlinear optics experiments in the THz region have been hampered by the limited availability of high-power THz sources. However, recently there has been significant progress in this area [10, 11], which is one reason for this timely focus issue. In our experiments described below, we will take advantage of a THz free-electron laser (FEL), which, in the overall combination of its parameters, still represents a rather unique THz source.

Turning now back to semiconductors, we note that bound electron–hole pairs, excitons, represent a quasi-atomic system, however on an energy scale about 1000 times smaller than in atoms. This is due to the small reduced effective mass ($m^* \sim 0.1m_0$) and large electric permittivity ($\varepsilon \sim 10$) of typical semiconductors. Therefore, excitons represent an interesting model playground for high-field optics in several respects: transitions between exciton states (intraexcitonic transitions) usually fall into the meV or THz range, and the Rabi frequency can reach a similar value already at comparably moderate intensities ($<\text{MW cm}^{-2}$) or fields ($<10 \text{kV cm}^{-1}$). While for decades, excitons were solely studied at their fundamental excitation energy somewhat below the bandgap of semiconductors, only recent experiments have addressed their internal degrees of freedom in the THz range. In fact, the excitonic Mott transition [12, 13], Rabi oscillations [14] and the Autler–Townes splitting [15] have been observed for intraexcitonic transitions.

In the current paper we explore the intraexcitonic nonlinear optics in InGaAs and GaAs quantum wells (QWs) beyond the validity of the two level and rotating wave approximations. By near-resonant pumping of the intraexcitonic 1s–2p transition with the intense, narrowband THz radiation of the FEL, we observe the Autler–Townes (or Rabi or ac Stark) splitting showing significant deviations from the simple two-level RWA model. As the simple two-level model with RWA is still a kind of reference when studying the stronger-field behavior, we give here the well-known analytic expression for the dressed-state energies [1], manifested as resonance peak positions of a probe absorption spectrum (in the present experiment, the probe is the near-infrared (NIR) excitation from the valence band, see section 2):

$$\omega_{\text{NIR}} = \omega_{1s} - \Delta / 2 \pm \frac{1}{2} \sqrt{\Delta^2 + \Omega^2}$$

with the detuning given by $\Delta = \omega - \omega_{21}$. This describes the famous anticrossing behavior of the dressed states. ($\omega_{1s}$ is the main heavy-hole 1s exciton absorption peak.) Note that the other intraexcitonic nonlinear optical effects such as sideband generation have been observed from the perturbational regime [16, 17] up to the extreme nonlinear regime, where field ionization and electron–hole recollisions give rise to high-harmonic sideband generation [18].

2. Experiment

Most data presented here are from a 20-period In$_{0.06}$Ga$_{0.94}$As/GaAs multiquantum well (MQW) sample with 8 nm thick InGaAs QWs separated by $\sim$130 nm thick GaAs barriers. Some data from a GaAs/Al$_{0.34}$Ga$_{0.66}$As MQW sample (QW thickness 8.2 nm, barrier thickness 19.6 nm) with 60 periods will be shown as well for comparison. The InGaAs MQW has the advantage that the GaAs substrate is transparent in the relevant spectral region, whereas the substrate of the GaAs MQW was etched away after transferring the sample to a ZnTe crystal plate. For all measurements, the sample was placed in a LHe flow cryostat equipped with z-cut quartz
Figure 1. Schematic view of the experiment: QW with electron, heavy-hole (hh) and light-hole (lh) subbands (left) and sketch of the exciton levels under intraexcitonic THz pumping and NIR probing (center). The Autler–Townes splitting is indicated (right).

windows and kept around $T = 10$ K. The sample was excited with the THz radiation from the FEL at the Helmholtz–Zentrum Dresden–Rossendorf. This widely tunable laser delivers THz pulses with about 1% spectral bandwidth with pulse energies up to 2 $\mu$J and 13 MHz repetition rate. Under typical conditions 10–100 nJ were employed and focused to a spot size of $\sim 0.5$ mm$^2$ (pulse length 20–30 ps). This yields an intensity of $\sim 1$ MW cm$^{-2}$ or a THz peak field of $12$ kV cm$^{-1}$. Since we are studying excitons, both samples are undoped and excitons are created by band-gap excitation using a NIR Ti:sapphire laser pulse with 12 fs duration. Due to its short temporal width, the pulse spectrum covers all excitonic features and thus we can use this pulse as a probe of the spectral changes induced by the FEL. The NIR pulse is focused on the sample with a smaller spot size, and the transmitted radiation is spectrally resolved with a spectrometer. The Ti:sapphire laser runs at 78 MHz repetition rate, which is reduced to 13 MHz using an acousto-optical pulse picker. FEL and NIR pulses are synchronized electronically to within 1–2 ps and can be delayed with respect to each other. More details of the experimental setup can be found in [15, 19]. The excitation process in the QWs is shown schematically in figure 1.

3. Results and discussion

Figure 2 displays NIR absorption spectra of the InGaAs MQW sample for different THz field strengths as indicated in the figure. The 1s–2p exciton transition energy is approximately 6.8 meV (with $\leq 5\%$ accuracy), thus figure 2(a) (photon energy of 5.6 meV) corresponds to negative detuning, figure 2(b) is near resonance (photon energy 6.8 meV) and figure 2(c) (photon energy 8.2 meV) represents positive detuning. Note that the small peak around 1479 meV represents the 2s exciton absorption, which is energetically very close to the (optically forbidden) 2p exciton.

Let us attempt to grasp the key features of this sequence of spectra in the vicinity of the heavy-hole 1s peak. For negative detuning (figure 2(a)) the main peak continuously shifts to lower energy upon increasing field strength, and a small satellite peak develops at higher photon energy (Rabi sideband), qualitatively resembling the two dressed-state solutions (equation (1)). At even higher fields the main peak again moves to higher energies. This behavior of the main peak for $\omega < \omega_{21}$, a red shift followed by a blue shift, is nicely consistent with the previous
Figure 2. Excitonic absorption spectra of the InGaAs MQW under THz pumping for different photon energies from negative detuning (a), resonant pumping (b), to positive detuning (c) for different electric fields as indicated (increasing from top to bottom). The photon energies and corresponding FEL wavelengths are indicated as well as the hh (1s) and (2s) peaks (only in the left panel). Note that the spectra in the right panel (c) were recorded on a different sample spot.

observation of Nordstrom et al [9], who explained it as effect of ac Stark shift and dynamical Franz–Keldysh effect, respectively [20].

Near resonance (figure 2(b)) the 1s peak splits up into the two dressed states; while initially the high-energy peak is slightly larger, this asymmetry is reversed for higher fields and the low-energy peak becomes clearly stronger. Above resonance, i.e. for positive detuning (figure 2(c)) the main peak shifts to the right and a shoulder develops on the low-energy side. For small electric fields, this corresponds qualitatively to a mirror image of figure 2(a). However, for higher fields, the asymmetry is totally reversed and the low-energy peak starts to dominate and finally the overall spectrum shifts to higher energies, which is again reminiscent of the dynamical Franz–Keldysh effect [9, 20]. Such an asymmetry reversal has been observed in the previous work by some of the present authors [15], but cannot be explained in the framework of the RWA and two-level approximations. The same is true for the peak positions at higher field strength.

In order to directly show the instantaneous response of the excitons to the THz field, we present some of the data in a 2D color map of NIR photon energy versus delay time between pump and probe, with the color code indicating the absorption strength. This is shown in figure 3(a) for an FEL pulse of 151 μm wavelength (8.2 meV photon energy, i.e. as shown in figure 2(c)) with a peak electric field of 12.5 kV cm⁻¹. This corresponded to an average FEL power of 107 mW. The lower left panel (figure 3(b)) depicts time-sliced spectra, representing the same information as in figure 2(c), however recorded not by adjusting the FEL power, but measuring the NIR transmission at different time delays during the FEL pulse. Naturally, spectra at many different field strengths can be extracted this way. In fact, we can use this procedure to reconstruct the FEL pulse shape, which is shown in figure 3(c). The points correspond to the time-sliced spectra in figure 3(b), the red line is a guide to the eye. The asymmetric shape with exponential rise and Gaussian tail is typical for spectrally narrow FEL pulses, which require relatively large cavity detuning [21].
Figure 3. (a) 2D color map of InGaAs MQW absorption (color scale: red represents strong absorption, blue is weak absorption) as a function of NIR photon energy (horizontal) and delay time (vertical) for a THz photon energy of 8.2 meV and a peak electric field of 12.5 kV cm\(^{-1}\). (b) Horizontal cuts, i.e. time slices taken from (a). Since the intensity and the field vary during the pulse, each time slice corresponds to different THz field strengths. (c) FEL pulse shape, reconstructed from (b) using the measurement from figure 2(c) as a reference. The nominal average power on the vertical axis also refers to the values from the reference measurement (i.e. at 107 mW average power the peak electric field was 12.5 kV cm\(^{-1}\)). The points correspond to time slices, the red line is a guide to the eye.

Often the Autler–Townes or Rabi splitting is displayed using an anti-crossing plot, where the (NIR) peak positions are plotted versus the (THz) driving-field photon energy (or the detuning) at constant electric field. This is shown in figure 4 for electric fields of 1.3 (panel a), 2.2 kV cm\(^{-1}\) (panel b) and 3 kV cm\(^{-1}\) (panel c). At relatively small electric fields (figure 4(a)), the data can still be fitted with a two-level model using the RWA, as shown by the solid lines. The fitting also yields the proper excitonic 1s–2p energy separation of 6.8 meV. Note that such an analysis considers only the peak positions, but not the peak strengths. For a somewhat higher electric field of 2.2 kV cm\(^{-1}\) (figure 4(b)), the fit still works, however it yields a slightly larger 1s–2p separation of 7.0 meV. At a larger, but still moderate electric field of 3 kV cm\(^{-1}\) (figure 4(c)) it is evident that the data points cannot be fitted well with the simple model anymore. The best, but still unsatisfactory fit now shows that the experimental
Figure 4. Anticrossing plot, i.e. NIR peak photon energy versus THz photon energy at peak electric fields of 1.3 kV cm$^{-1}$ (a), 2.2 kV cm$^{-1}$ (b) and 3 kV cm$^{-1}$ (c). The symbols correspond to experimental points and the lines represent a fit using a two-level model and the RWA. The zero on the horizontal scale corresponds to the unperturbed 1s–2p energy of 6.8 meV and the zero on the vertical scale corresponds to the unperturbed NIR peak position of 1472.3 meV. The vertical line represents the detuning value where the peak splitting is minimized (dynamically shifted 1s–2p energy).

The high-energy peak is significantly blue shifted, in particular for negative detuning. In addition, it yields a further increased 1s–2p separation of 7.6 meV. This dynamic level shift in the strong THz field (here of $\sim$0.2 and 0.8 meV, respectively, see vertical lines in figures 4(b) and (c)) may be related to the Bloch–Siegert shift, which is the lowest-order (in $\Omega^2/\omega^2$) non-RWA correction [22]. Both observations clearly indicate the breakdown of the two-level model with RWA. It has been shown [23], however, that if one uses a full microscopic model on the basis of the semiconductor Bloch equation without applying the RWA [24, 25], the experimental points and even the observed line shapes can be reproduced very well. A more detailed comparison between experimental spectra and the full theory can be found in [23] and shows that taking into account higher excitonic levels is of key importance.

For fields larger than 3 kV cm$^{-1}$ the shape of the absorption curves is distorted so radically (cf figure 2) that an anticrossing plot is no longer useful. This field strength corresponds to a Rabi energy of about 1.6 meV, already a significant fraction of $\hbar\omega_{21}$. At the highest field investigated here, 12.5 kV cm$^{-1}$, the Rabi frequency $\Omega$ is already approximately equal to $\omega_{21}$. On the other hand, the ponderomotive energy $U = e^2F^2/4m\omega^2$ is already 11 meV under these conditions, i.e. it becomes the largest parameter in the system. This indicates that field ionization [26] becomes relevant. The loss of carriers to higher levels and/or the continuum is also reflected in the overall bleaching of the NIR absorption at high THz fields seen in figure 2.

So far we have discussed one specific InGaAs QW sample with a 1s–2p exciton energy of 6.8 meV. One may ask how the above described effects vary for different sample parameters. Thus, we inspect data recorded on a 8.2 nm wide GaAs/AlGaAs QW (see [15, 19]) at positive detuning, namely at a wavelength of 88 $\mu$m (14.1 meV photon energy), shown in figure 5. The qualitative similarity of this set of spectra to figure 3(b) is obvious. We note that the peak at 1583 meV for small THz fields represents here the light-hole exciton, which was not visible.
Figure 5. Excitonic absorption spectra of the GaAs MQW under THz pumping ($\hbar \omega = 14$ meV or $\lambda = 88 \mu m$) for positive detuning and different electric fields as indicated (increasing from top to bottom). The three exciton peaks (hh(1s), hh(2s), lh(1s)) are indicated as well.

in InGaAs due to the strain-induced shift, whereas the shoulder at 1576 meV is due to the 2s exciton. Let us compare the relevant parameters for the two samples: the GaAs QW has a 1s–2p exciton energy of 9 meV (compared to 6.8 meV for InGaAs), mainly due to the somewhat larger (reduced) effective mass. On the other hand, the exciton linewidth is 3 meV for the GaAs sample (versus 1.5 meV for InGaAs). Both relations support the tendency that higher THz fields are needed in GaAs to obtain similar spectra as in InGaAs, and similar values of $\Omega/\omega_{21}$. The fact that the larger $\omega_{21}$ of GaAs implies a smaller dipole matrix element additionally supports this trend.

4. Conclusions

Using excitons in semiconductor QWs as a generic quasi-atomic system with binding energy in the THz range, we have explored the high-field nonlinear behavior when driving the 1s–2p intraexciton transition with an intense THz laser. Probing the induced spectral changes of the 1s exciton NIR absorption, Rabi splitting has been observed, but going well beyond the behavior of a two-level system under RWA. Both peak positions and strengths vary in a more complicated manner, accompanied by a significant line broadening and overall bleaching, that can be taken as an indication of field ionization. Nevertheless, a qualitatively similar behavior is observed on two different QW systems, GaAs and In$_{0.06}$Ga$_{0.94}$As, corroborating the generality and scalability of the observed effects.

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