Controlling the polarization dynamics by strong THz fields in photoexcited germanium quantum wells

Niko S Köster¹, Andrea C Klettke¹, Benjamin Ewers¹, Ronja Woscholski¹, Stefano Cecchi², Daniel Chrastina², Giovanni Isella², Mackillo Kira¹, Stephan W Koch¹ and Sangam Chatterjee¹,³

¹ Faculty of Physics and Materials Sciences Center, Philipps-Universität Marburg, Renthof 5, D-35037 Marburg, Germany
² L-NESS and Dipartimento di Fisica del Politecnico di Milano, Polo di Como, Via Anzani 42, I-22100 Como, Italy
E-mail: sangam.chatterjee@physik.uni-marburg.de

New Journal of Physics 15 (2013) 075004 (9pp)
Received 19 April 2013
Published 3 July 2013
Online at http://www.njp.org/
doi:10.1088/1367-2630/15/7/075004

Abstract. The interaction of strong single-cycle THz pulses with the optically induced polarization in germanium quantum wells is studied. With increasing THz field strength, it is observed that the excitonic resonances shift toward higher energy and broaden before weak signatures of a splitting of the exciton line occur. In comparison with high-quality GaAs-based quantum wells, where a much clearer Autler–Townes splitting is observed, the germanium system response is significantly more broadened and shows signatures of a quasi-steady-state behavior due to the intrinsic fast dephasing times dominated by intervalley scattering.

³ Author to whom any correspondence should be addressed.
1. Introduction

During the last decade, germanium has experienced a revival as an only weakly indirect silicon-compatible semiconductor material. This renewed interest was sparked, for example, by the observation of the quantum-confined Stark effect \([1, 2]\) and the subsequent demonstration of electro-optical devices such as modulators \([3]\). Followed by the experimental verification of transient and steady-state optical gain \([4, 5]\), the device-related efforts culminated in the realization of an optically and later an electrically pumped laser \([6, 7]\). Concerning the aspects of fundamental physics, several studies compared the optical response of germanium to that of standard direct-gap semiconductor nanostructures pointing out intricate peculiarities due to the germanium-specific band structure subtleties \([8–10]\).

In spite of the recent burst of activities, however, many relevant aspects of the interband response and in particular of the many-body interactions in Ge and its nanostructures remain largely unexplored. Furthermore, besides a few hallmark studies such as \([11]\) or \([12]\), the THz properties of these Ge systems are not yet fully understood despite the realization of THz-related applications such as a p-type germanium laser \([13]\) or quantum-cascade structures \([14]\).

In this paper, we study the interaction of the optical polarization induced by a weak supercontinuum pulse with strong broadband THz pulses. In particular, we are interested in a comparison of the Ge results with those of similar experimental studies in high-quality narrow-linewidth (Ga, In) As multiple quantum wells. There, the excitonic Autler–Townes splitting was observed \([15]\) and multi-photon ionization was shown to occur for THz fluences of few kV cm\(^{-1}\) \([16]\). As a consequence of the reduced exciton binding energy \([17]\), even more pronounced ionization effects can be expected in the germanium system.

2. Experimental detail and sample structure

In our experiments, we irradiate the sample by an optical and a THz beam under normal incidence. The schematics of the setup is shown in figure 1. All pulses are generated from a regenerative Ti:sapphire laser amplifier. The system provides a train of 120 fs laser pulses at 800 nm wavelength with a repetition rate of 1 kHz. The average power of up to 1 W corresponds to 1 mJ pulse energy.

A small fraction of the light is split off and focused into a 1 mm thick (0001)-cut sapphire crystal for white-light supercontinuum generation using an anti-reflection-coated achromatic doublet. Great care is taken in aligning the white-light generation as the best results are achieved for undistorted wave fronts in the sapphire crystal. The effective pulse energies in
Figure 1. Schematic sketch of the experimental geometry and setup including the experimentally determined time trace of the THz pulse (green line).

the spectral vicinity of the germanium quantum well absorption edge are selected with the help of a reflective variable attenuator wheel. The supercontinuum is collimated using a 5 cm focal length near-infrared optimized achromatic doublet and focused onto the sample. The transmitted light is imaged onto a 50 cm grating spectrometer equipped with a 900 line mm\(^{-1}\) grating. The second-order light is detected using a liquid nitrogen-cooled (Ga,In)As photo-diode linear array. Measuring the unperturbed weak transmission and subtracting the appropriate background yield the linear absorption when neglecting nonlinear reflectance effects.

The polarization generated in the sample by the weak optical pulse is modified nonperturbatively using intense, broad-band THz pulses. The single-cycle far-infrared light is generated in a commercially available large-area emitter biased by a rectangular voltage of 50 V with a duty cycle of 50%. The fs-pulses excite the 1 cm\(^2\) active area of the emitter after passing through a diverging lens with a focal length of −5 cm. This way, the emission pattern of the THz radiation completely fills the 2' diameter 2' effective focal length off-axis parabolic mirror used for collecting the far-infrared radiation and avoiding an intermediary focus. The THz light is focused onto the sample collinearly with the optical radiation using a 3' effective focal length off-axis paraboloid. After passing through the sample, the transmitted THz field amplitude is detected by a standard electro-optical sampling system position of the sample. Its spectrum is extracted by Fourier transform from the measured time-resolved electric field; both are shown in figure 1. The THz field strength at the position of the sample calculated from a carefully measured spatial profile using several knife-edge measurements and the THz power measured using a traceable Golay cell and confirmed by a calibrated bolometer.

The sample under investigation is a germanium multiple quantum well sample grown by low-energy plasma-enhanced chemical vapor deposition on a (001)-oriented Si substrate. It contains 50 compressively strained 14 nm wide Ge quantum wells between 20 nm thick tensile strained Si\(_{0.15}\)Ge\(_{0.85}\) barriers. This material sequence yields a type-I band alignment in the quantum wells, the strain lifts the degeneracy of heavy- and light-hole resonances. The quantum
well stack is synthesized on top of a $2 \mu \text{m} \text{Si}_{0.1}\text{Ge}_{0.9}$ virtual substrate grown on a tensile strained Si$_x$Ge$_{1-x}$ graded buffer where the germanium content $x$ is continuously ramped up from 0 to 0.9. A linear absorption spectrum and a band scheme are shown in figure 2; the transitions are confirmed by tight-binding calculations [18].

Figure 3 shows the examples of the measured optical absorption spectra with and without THz excitation. Panel (a) presents the linear absorption spectrum of the 1s heavy-hole exciton resonance, together with the induced modifications for a THz field strength of $13.5 \text{kV cm}^{-1}$ and time delays between the optical and THz pulses of $-250$ and 0 fs, respectively. The corresponding time-resolved data are summarized in panel (b) of figure 3, where the nonlinear absorption spectra are plotted as a function of time delay in false colors. The dashed white line indicates the position of absorption maximum. Comparing this with the time evolution of the THz field amplitude in panel (c) of figure 3 reveals weak signatures of peak splitting in the vicinity of the zero-crossing of the single-cycle THz field amplitude. These observations are very similar to reports of a similar study on a II–VI-semiconductor structure [19] and attributed to the ultrafast dephasing time in germanium of the order of 100 fs [20].

Next, we show the THz field strength dependence for zero time delay between the optical and THz pulses plotted in figure 4. The absorption spectra are plotted in false colors for THz field strengths varying from 0 to $13.5 \text{kV cm}^{-1}$. The white dashed line follows the peak positions. For small THz field strength, we observe a blue shift of the resonance corresponding to a slight THz-induced reduction of the exciton binding energy. Increasing the level of THz excitation, this effect is superimposed to strong broadening and weak splitting signatures of the exciton resonance.

3. Microscopic model and analysis

To analyze the experimental results, we use our microscopic many-body theory which self-consistently includes the coupling of the Coulomb-interacting semiconductor many-particle...
Figure 3. THz induced splitting of the 1s heavy-hole exciton resonance for a THz field strength of 13.5 kV cm\(^{-1}\). (a) Linear absorption (black curve) as well as nonlinear absorption spectra for time delays of \(-250\) fs (dark red curve) and 0 fs (bright red curve) between optical and THz pulse. (b) False-color plot of the nonlinear absorption as function of time delay and detected photon energy. The delay times for the spectra shown in (a) are indicated by the red horizontal lines. The dashed white line is a guide to the eye at the position of the maximum absorption in each spectrum. (c) Time evolution of the THz amplitude as well as absorption \((1 - T)\) for the same temporal position at the energy of the 1s heavy-hole exciton resonance.

Figure 4. THz field strength dependence of the nonlinear absorption spectra plotted in false colors for zero time delay between optical and THz pulses. The white line is given as guide to the eye highlighting the position of the absorption maximum/maximum.
excitations to the optical and THz fields. The dynamics of the electron–hole polarization \( P_k \equiv P_{c,v,k} \) is determined by the semiconductor Bloch equations \([21,22]\)

\[
i \hbar \frac{\partial}{\partial t} P_k = \left( \epsilon_k - j_k A_{\text{THz}} + \frac{|e|^2}{2\mu} A_{\text{THz}}^2 \right) P_k + \sum_{k'} \Gamma_{k,k'} P_{k'} - \left( 1 - f_k^e - f_k^h \right) \left( d_{c,v,k} E_{\text{opt}} + \sum_{k'} V_{k-k'} P_{k'} \right).
\]

This equation describes the entire polarization dynamics, i.e. its creation through the exciting optical field \( E_{\text{opt}} \), its redistribution by the THz field as well as the eventual decay caused by scattering and dephasing processes modeled via \( \Gamma_{k,k'} \). The dipole matrix element between the conduction and valence band is denoted by \( d_{c,v,k} \) for the momentum \( k \). The occupation probabilities for the electron and hole states are denoted by \( f_k^e \) and \( f_k^h \), respectively. The vector potential of the THz contribution \( A_{\text{THz}} \) drives the transitions between the different quantum states of the optical polarization. It is related to the electric field via \( E_{\text{THz}} = -\partial \phi_{\text{THz}} / \partial t \). The current matrix element \( j_k \equiv -|e|/\hbar \partial \epsilon_k / \partial |k| \cos \phi_k \), where \( \phi_k \) is the angle between \( k \) and the polarization direction of the THz field.

In this study, all dephasing effects are treated via \( \sum_k \Gamma_{k,k'} P_{k'} \) with a dephasing time of 300 fs. This dephasing time was experimentally determined for this sample using coherent oscillation spectroscopy, i.e. analyzing spectrally resolved pump–probe experiments at negative time delays \([20]\). The effective electron (hole) masses used in the numerical analysis are \( m_{e(h)} = 0.041m_0 \) (0.25\( m_0 \)), with \( m_0 \) being the free electron mass.

For a self-consistent solution, we include the back coupling of the polarization on the optical and THz field via Maxwell’s equation. Separating the wave equation for the optical and the THz field, we obtain

\[
\left[ \frac{\partial^2}{\partial z^2} - \frac{n^2(z)}{c_0^2} \frac{\partial^2}{\partial t^2} \right] A_{\text{THz}}(z,t) = -\mu_0 g(z) (J_{\text{THz}} + J_A),
\]

\[
\left[ \frac{\partial^2}{\partial z^2} - \frac{n^2(z)}{c_0^2} \frac{\partial^2}{\partial t^2} \right] E_{\text{opt}}(z,t) = \mu_0 g(z) \frac{\partial^2 P}{\partial t^2}.
\]

Here, the electron–hole confinement function is denoted by \( g(z) \), \( \mu_0 \) is the vacuum permittivity, \( c_0 \) is the speed of light and \( n(z) \) is the refractive index profile of the sample. The propagation of the light through the sample is treated using the standard transfer matrix technique \([23]\). The wave equations describe light propagating perpendicular to the quantum wells and polarized linearly in the \( x \) direction. They have to be solved simultaneously with the semiconductor Bloch equations.

In the past, we applied this theory to analyze the outcome of similar experiments in \((\text{Ga,In})\text{As}/\text{GaAs}\) quantum-well samples \([16]\). Using the parameters for the Ge system, we now obtain the results summarized in figure 5. The experimental spectra are plotted on the left-hand side and the corresponding theoretical spectra on the right. Exemplarily, we have chosen negative time delays of \(-0.6, -0.3, -0.1\) and 0 ps for both experiment and theory in the top panel. The optical pulse hence arrives before the maximum of the THz electric fields where
Figure 5. Side-by-side comparison of measured (left) and calculated (right) absorption spectra for various time delays between the optical and the THz pulse.

time-zero is defined. Positive time delays are depicted in the bottom two panels, 0, +0.28 and +0.6 ps as well as 0, +0.3 and +0.6 ps for experiment and theory, respectively.

A reasonably good agreement between experiment and theory is found. Most features of the experiments are well reproduced such as the pronounced blue shift of the exciton resonance for negative time delays approaching zero. The shift vanishes for positive time delays, when the optical pulse hits the sample after the maximum of the THz field strength and very strong broadening occurs. To achieve this level of agreement, it was necessary to include excitonic states with large orbital quantum numbers up to 20 indicating the importance of THz-photon-induced exciton ionization processes.

The splitting of the exciton resonance is only barely observable in the experimental data as the sample features a comparatively large, yet predominantly homogeneously broadened exciton line due to the fast, intervalley-scattering-dominated dephasing in germanium [20]. To clearly reveal the origin of this splitting, we perform a switch-off analysis in our microscopic theory. The top panel in figure 6 shows the linear absorption as the gray-shaded area and the modified spectra for the THz field strength of 2 kV cm$^{-1}$ (black solid line). Neglecting the ponderomotive current in the semiconductor Bloch equations significantly shifts the main absorption line toward lower energies (dotted black line). This contradicts the experimental observation where only shifts toward higher energies are observed. No evidence for a peak splitting is found in this case.

In contrast, the splitting is very pronounced when only the ponderomotive $A^2$ term is included as a source (dashed black line) identifying this contribution as the main source of
Figure 6. Exemplary computed transmission spectra \((1 - T)\) resulting from the microscopic many-body theory for a THz field strength of 2 kV cm\(^{-1}\). Top panel: the results of the full calculation are shown as black curve, the linear absorption as gray-shaded area. The dashed curve shows results from the calculation when neglecting the \(A^2\) term in the semiconductor Bloch equations, the dotted curve when only using it as a source. Bottom panel: systematic variation of the ponderomotive contribution in the full calculation for a THz field strength of 2 kV cm\(^{-1}\): 40\% dashed line; 60\% dash–dot line, 80\% dash–dot–dot line, 100\% solid line and 120\% dotted line.

the peak splitting. While the overall line shape does not match the experimental observation in this case, the emergence of two split peaks becomes strongly evident. For the results in the bottom panel of figure 6, we systematically vary the ponderomotive contribution. Increasing its relative strength systematically shifts the peak absorption toward higher energies and increases the visibility of the splitting of the exciton resonance.

4. Conclusions

The interaction of strong, broadband THz pulses with an excitonic polarization in germanium quantum wells is investigated by probing the nonlinear optical transmission. The observed response is very similar to observations in quantum wells composed of direct-gap semiconductors such as GaAs or (GaIn)As, underlining the quasi-direct-gap-like character of the ultrafast optical response in this silicon-technology-compatible material system. The reduced binding energies lead to strong contributions of THz-photon ionization of excitons, strongly broadening the excitonic resonances yet still hinting a splitting of the exciton as
expected for the excitonic Autler–Townes effect; owing to the fast dephasing times in this system, also quasi-steady-state contributions are observed. A microscopic many-body analysis identifies the sources for the observation, underlining the significant role of the ponderomotive contribution on the THz-response.

Acknowledgments

SC is particularly thankful to Martin Koch for granting him access to the regenerative amplifier system. The Politecnico group is partially financed by the EC GREEN Silicon project. MK thanks the Deutsche Forschungsgemeinschaft for financial support.

References

[1] Kuo Y H, Lee Y K, Ge Y, Ren S, Roth J E, Kamins T I, Miller D A B and Harris J S 2005 Nature 437 1334
[2] Chaisakul P, Marris-Morini D, Isella G, Christina D, Roux X L, Gatti E, Edmond S, Osmond J, Cassan E and Vivien L 2010 Opt. Lett. 35 2913
[3] Chaisakul P, Marris-Morini D, Rouifed M S, Isella G, Christina D, Frigerio J, Roux X L, Edmond S, Coudecyele J-R and Vivien L 2012 Opt. Express 20 3219
[4] Lange C, Köster N S, Chatterjee S, Sigg H, Christina D, Isella G, von Knell H, Schäfer M, Kira M and Koch S W 2009 Phys. Rev. B 79 201306
[5] Liu J, Sun X, Kimerling L C and Michel J 2009 Opt. Lett. 34 1738
[6] Liu J, Sun X, Camacho-Aguilera R E, Kimerling L C and Michel J 2010 Opt. Lett. 35 679
[7] Camacho-Aguilera R E, Cai Y, Patel N, Bessette J T, Romagnoli M, Kimerling L C and Michel J 2012 Opt. Express 20 11316
[8] Lange C, Köster N S, Chatterjee S, Sigg H, Christina D, Isella G, von Knell H, Kuhnert B and Stolz W 2010 Phys. Rev. B 81 045320
[9] Gatti E, Grilli E, Guzzi M, Christina D, Isella G and von Känel H 2011 Appl. Phys. Lett. 98 031106
[10] Gatti E, Grilli E, Guzzi M, Christina D, Isella G, Chernikov A, Bornwasser V, Köster N, Woensch R and Chatterjee S 2011 Phys. Rev. B 84 245319
[11] Grischkowski D, Keiding S, van Exter M and Fattinger C 2009 J. Opt. Soc. Am. B 7 2006
[12] Hebling J, Hoffmann M C, Hwang H Y, Yeh K-L and Nelson K A 2010 Phys. Rev. B 81 035201
[13] Bergner A, Heugen U, Bründermann E, Schwaab G, Havenith M, Chamberlin D R and Haller E E 2005 Rev. Sci. Instrum. 76 063110
[14] Paul D J 2010 Laser Photon. Rev. 4 610
[15] Wagner M, Schneider H, Stehr D, Winnerl S, Andrews A M, Scharner S, Strasser G and Helm M 2010 Phys. Rev. Lett. 105 167401
[16] Ewers B, Köster N S, Woensch R, Koch M, Chatterjee S, Khitrova G, Gibbs H M, Kira M and Koch S W 2013 Phys. Rev. B 85 075307
[17] Madelung O, Rössler U and Schulz M (ed) 2002 Group IV Elements, IV–IV and III–V Compounds vol 41A1b Part b—Electronic, Transport, Optical and Other Properties, Landolt Börnstein—Group III Condensed Matter Numerical Data and Functional Relationships in Science and Technology (Berlin: Springer) pp 2820–1
[18] Virgilio M and Grosso G 2006 J. Phys.: Condens. Matter 18 1021–31
[19] Hirori H, Nagai M and Tanaka K 2010 Phys. Rev. B 81 081305
[20] Kolata K et al 2010 Phys. Rev. B 86 201303
[21] Haug H and Koch S W 2009 Quantum Theory of the Optical and Electronic Properties of Semiconductors 5th edn (Singapore: World Scientific)
[22] Kira M and Koch S W 2006 Prog. Quantum Electron. 30 155
[23] Khitrova G, Gibbs H M, Jahnke F, Kira M and Koch S W 1999 Rev. Mod. Phys. 71 1591

New Journal of Physics 15 (2013) 075004 (http://www.njp.org/)