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ADVANCES IN SOLID STATE PHYSICS

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Ultrafast Spectroscopy of Semiconductors

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Summary: Ultrafast laser spectroscopy with time resolution down to 6 femtoseconds has been applied to study optical dephasing, relaxation, and recombination in various semiconductors and semiconductor microstructures. We report some applications of ultrafast laser spectroscopy to bulk and quantum well III-V and II-VI semiconductors in order to illustrate the potential. The characteristic time scales for dephasing, relaxation and recombination are discussed and determined experimentally.

1 Introduction

The magic word *faster* has always been one of the major challenges in the development of semiconductor microelectronics and optoelectronics. For many years this basically has been a task for device and chip design, however, in our days device technology has reached a level, where the characteristic time constants of the underlying physical processes may determine the speed limits. Investigation of these intrinsic time constants thus has become an important field not only in basic research but also from an applied point of view. Recent developments in ultrafast laser technology now allow to study the very initial interaction processes of nonequilibrium carriers in a semiconductor with its environment and thus time resolved laser spectroscopy has become a powerful tool in modern semiconductor physics.

In this article we shall demonstrate the great potential of ultrafast laser spectroscopy for the investigation of the dynamics of nonequilibrium carriers in semiconductors. The present state of the art of laser technology for short optical pulse generation will be summarized briefly and some selected applications for the study of optical dephasing, energy relaxation, and recombination in III-V and II-VI semiconductors and quantum well structures will be discussed.

2 Femtosecond Laser Technology

Femtosecond laser technology is based on mode locking of lasers with sufficiently large gain band width like dye lasers, colour center lasers or semiconductor lasers. Mode locking refers to the coherent superposition of the longitudinal laser cavity modes, which is forced by active or passive gain or loss modulation [1]. Milestones towards ourdays femtosecond laser technology are the first demonstration of mode

locking of a ruby [2] and a Nd:YAG laser [3], the first demonstration of synchronous [4] and passive [5] mode locking of a pulsed dye laser, synchronous mode locking of a cw dye laser by pumping with a mode locked argon laser [6], the development of the colliding pulse mode locked (CPM) ring dye laser [7] and more recently the realization of additive pulse mode locking (APM) [8], which can be considered as a generalized version of the soliton laser [9].

Regardless of the different schemes, the fundamental limit for the direct generation of short optical pulses with mode locked lasers is set by the spectral bandwidth of the longitudinal mode spectrum, $\Delta\nu$, according to the relation

$$\Delta t \cdot \Delta\nu \approx 1 \,. \tag{1}$$

However, in the subpicosecond regime effects like self phase modulation and group velocity dispersion occur and a careful balance of the different pulse shaping mechanisms is required to achieve pulse widths approaching this limit. Pulse widths as short as 27 fs have been generated directly with a CPM laser by carefully optimising the operating conditions and applying intracavity dispersion compensation [10].

Additional pulse shortening can be achieved externally, i.e. outside the laser resonator, by using pulse compression techniques, most commonly an optical fiber for imposing a frequency chirp onto the optical pulses while travelling through the fiber and a dispersive delay line, e.g. a pair of gratings, for subsequent compression of the pulses [11]. In the spectral region corresponding to negative group velocity dispersion of the fibers ($\lambda > 1.3\ \mu m$) compression can be achieved by the fiber itself [12] and pulse shaping then is very much like in soliton lasers. More sophisticated pulse compression schemes applying grating and prism combinations have made it possible to compress pulses of a Rh 6G CPM dye laser down to 6 fs [13].

With respect to spectroscopy applications, the femtosecond laser systems have the drawback that the laser photon energy practically cannot be tuned. To overcome this, different techniques for so-called femtosecond white light continuum generation have been developed [14]. This white light continuum generation involves three steps:

(i) amplification of the femtosecond light pulses by an optical amplifier [15],
(ii) compression of the amplified pulses e.g. by a grating compressor, and
(iii) continuum generation in an optically nonlinear medium, e.g. an ethylene glycol jet, where the new frequency components are generated due to self phase modulation of the incoming light pulses.

White light continuum pulses with typical pulse widths of the order of 100 fs have been generated with center wavelengths at 620 nm [14] as well as 800 nm [16,17].

Altogether, time resolved optical spectroscopy in the visible and near infrared regime with time resolution of 100 fs and – in some cases appreciably higher – is possible, at present. However, one has to keep in mind that the spectral resolution becomes very poor and in general a trade-off between time resolution and spectral resolution has to be found.

3 Hierarchy of Time Constants in Semiconductors

The dynamics of optical excitations in a simple noninteracting homogeneously or inhomogeneously broadened two level system is described by the optical Bloch equations for the time dependence of the polarization and population. In the density matrix formalism the optical Bloch equations for a two level system are [18] ($\hbar = 1$):

$$\delta/\delta t \rho_{12} - i\omega_0\rho_{12} + \rho_{12}/T_2 = -i\mu E(t) N_{inv} \tag{2a}$$

$$\delta/\delta t N_{inv} + [N_{inv} - N_0]/T_1 = -i2\mu E(t)[\rho_{12} - \rho_{21}] \tag{2b}$$

where ρ_{ii} and ρ_{ij} are respectively, the diagonal and off-diagonal elements of the density matrix, μ is the dipole matrix element, i.e. $\mu \cdot E(t) = H_{int}$ is the interaction Hamiltonian. The off-diagonal elements determine the dipole moment p corresponding to the excitation of the two level system with eigenfrequency ω_0 according to:

$$p = \mu\,[\rho_{12} - \rho_{21}] \,. \tag{3}$$

The macroscopic polarization P, which is the driving force for coherent emission from the two level system, is proportional to p, $P = N \cdot p$, where N is the density of two level systems. The population of the levels 1 and 2 is determined by the diagonal elements of the density matrix and the inversion density N_{inv} is given by $N_{inv} = \rho_{22} - \rho_{11}$ and N_0 is its equilibrium value.

The meaning of the time constants T_1 and T_2 in Eqs. (2a) and (2b) is readily seen when the driving light field is turned off, i.e. $E(t) = 0$. Eqs. (2a) and (2b) then are decoupled and T_2 describes the exponential decay of the macroscopic polarization in the rotating frame

$$P(t) = P(0)\exp(-t/T_2) \tag{4}$$

whereas T_1 accounts for the decay of the inversion

$$N_{inv}(t) - N_0 = [N_{inv}(0) - N_0]\exp(-t/T_1) \,. \tag{5}$$

Since the macroscopic polarization is sensitive to the phase relation of the individual dipoles, T_2 is called the *dephasing time*, whereas T_1 is the *population lifetime*. The dephasing time T_2 is related to the spectral linewidth Γ of the transition at frequency ω_0 ($T_2 = (1/\Gamma)$). The decay of the population of course also results in a decrease of the macroscopic polarization and the relation

$$1/T_2 = 1/2T_1 + 1/T^* \tag{6}$$

holds, where T^* accounts for pure dephasing processes. Eq. (6) defines the hierarchy of time constants for a two level system according to

$$T_2 \le 2T_1 \,. \tag{7}$$

Extension of this formalism to noninteracting inhomogeneously broadened two level systems is straightforward [19]. The macroscopic polarization is now obtained by the spatial average of the dipole moments

$$P(t) = \int d\omega_0\, g(\omega_0)\, p(\omega_0, t) \tag{8}$$

where $g(\omega_0)$ is the distribution function for the eigenfrequencies of the two level systems. The temporal development of the macroscopic polarization now, however, is not only determined by the dephasing time T_2, but also by the inhomogeneous linewidth Δ_{inh}. In particular, the macroscopic polarization decays after δ-pulse excitation with a characteristic time constant of $T \simeq (1/\Delta_{inh})$, even for infinite T_2 (free induction decay). This dephasing, however, is reversible and can be recovered by a subsequent light pulse at $t = \tau$ leading to the emission of a photon echo at $t = 2\tau$. If T_2 is finite, a photon echo can be observed only for delay times of the rephasing pulse in the order of T_2. Photon echoes, in any case, are possible only for inhomogeneously broadened transitions. In turn, time resolved detection of a photon echo provides a reliable determination of T_2 without further assumptions, e.g. on the ratio of homogeneous to inhomogeneous broadening.

The situation is much more complicated in semiconductors and it seems impossible to apply the two level scheme at all to discuss and explain experimental results. Several different approaches to describe coherent interaction of light with intrinsic semiconductors have been developed recently [20]. Nevertheless, we may still apply a generalized two level model at least for the definition of the hierarchy of time constants and qualitative interpretation of experiments. In fact, as single particle excitations, i.e. band to band transitions are concerned, the semiconductor might be considered as an inhomogeneously broadened two level system in momentum space with a broadening determined by the width of the electronic bands. Interaction between these two level systems due to inelastic scattering then can be accounted for phenomenologically by a spectral relaxation time T_3 [20] and thus Eq. (6) has to be extended to

$$1/T_2 = 1/T_3 + 1/T^* + 1/2T_1 \tag{9}$$

in analogy to the case of inhomogeneously broadened two level systems in real space [22].

On the base of Eq. (9) the hierarchy of time constants for the relaxation of optical excitations in semiconductors can be defined as illustrated schematically in Fig. 1. Interaction of a short laser pulse with the electronic states of the semiconductor sets up a coherent macroscopic polarization. This coherent polarization

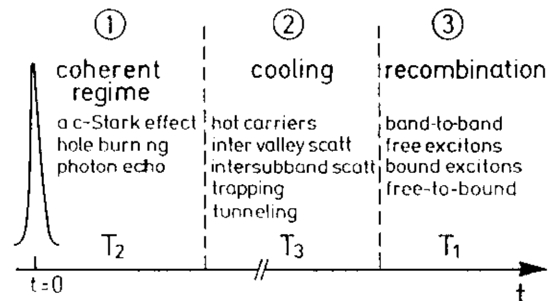

Fig. 1 Schematic representation of the different time regimes in the relaxation of photoexcited carriers in a semiconductor.

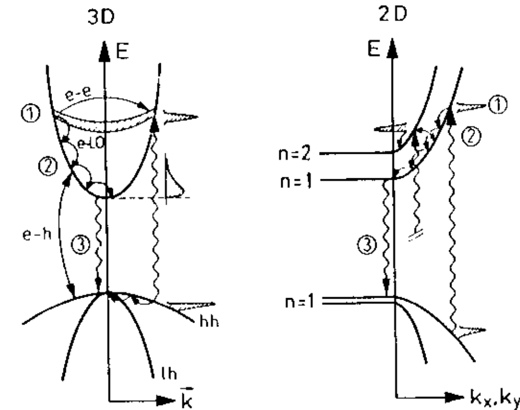

Fig. 2 Schematic illustration of relaxation processes in a bulk (3D) and quantum well (2D) semiconductor. The respective time regimes according to Fig. 1 are indicated by the circled numbers.

decays irreversibly with a time constant T_2 due to elastic and inelastic scattering characterized by T^* and T_3, respectively. Subsequently, after the loss of polarization the population may further relax in energy by subsequent T_3-*processes* and finally the (inversion) population will decay due to recombination with a time constant T_1. The respective processes involved in the different relaxation steps are schematically indicated in a simplified band structure diagram for a direct gap bulk material, e.g. GaAs and a corresponding quantum well structure [23] with quantization along k_z in Fig. 2.

In the following, we shall present and discuss some recent experiments for the different regimes classified in Fig. 1.

4 The Coherent Regime

If two subsequent laser pulses impinge onto a semiconductor within the *coherent regime*, the response or signal reflects the coherent interaction of the fields and induced polarizations of the two excitation pulses, giving rise to e.g. the optical Stark effect [24, 25], coherent emission [26], and photon echoes [27].

A versatile experimental approach to investigate the coherent regime is by time resolved four wave mixing. The general scheme of these experiments is shown in Fig. 3. The sample is excited by three subsequent excitation pulses with wavevector k_1, k_2, and k_3, respectively, at times $t = 0$, $t = \tau$, and $t = T$. A signal then can be emitted into directions 4 and 4′ with corresponding wavevectors $k_4 = k_3 + (k_1 - k_2)$ and $k_4' = k_3 - (k_1 - k_2)$ due to the nonlinear interaction in the sample. This general scheme for time resolved four wave mixing experiments now can be applied

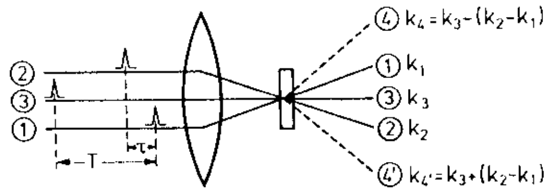

Fig. 3 Scheme of time resolved four wave mixing experiments.

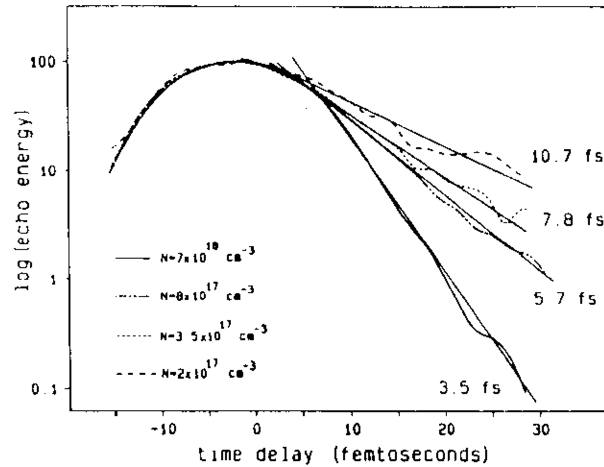

Fig. 4 Self diffraction due to free carrier excitation in bulk GaAs. After Ref. [29].

in different versions. For time integrated, e.g. not time resolved detection of the signals 4 or 4′ and $\tau = 0$, these *transient grating* experiments have been employed to study recombination and diffusion of excitations [28]. If only beam 1 and 2 are used with time integrated detection *self diffraction* ($k_3 = k_2$) generates the signal into direction 4 and 4′. Self diffraction basically is determined by T_2, however T_1 and T_3 as well as the amount of inhomogeneous broadening also enter. Self diffraction with time resolved detection of the signals 4 and 4′ in the case of an inhomogeneously broadened transition corresponds to a *photon echo* experiment, while the most general three beam experiment with time resolved detection of the signals is called a *stimulated photon echo* experiment.

Dephasing of free carrier excitations in bulk GaAs has been recently studied in a self diffraction experiment with 6 fs time resolution by Becker et al. [29]. The experimental result for an excitation photon energy of 2 eV and for different excitation intensities is depicted in Fig. 4, showing the dependence of the diffracted signal intensity into the direction corresponding to $2k_2 - k_1$ as a function of the delay time τ.

The respective decay constants of the self diffraction signal are plotted in the figure, the dephasing times T_2 in the present case of strong inhomogeneous broadening are obtained by multiplying the decay times by four.

For excitation closer to the band gap of bulk GaAs ($E_g = 1.52$ eV) and GaAs/AlGaAs quantum well structures longer dephasing times of the order of 300 fs and 150 fs, respectively, have been determined by spectral hole burning and polarization rotation experiments [30, 31]. Dephasing for band to band excitation in semiconductors at the high excitation intensities generally required because of the low effiency of the nonlinear interaction will be determined by carrier-carrier scattering as confirmed by the results by Becker et al. [29] and recently discussed by Gurevich et al. [32].

The dephasing of resonantly excited free excitons in bulk GaAs and GaAs/AlGaAs quantum wells has been studied by Schultheis and coworkers [33, 34] and in CdSe by Dörnfeld et al. [35]. The dephasing times are typically of the order of a few picoseconds which is appreciably longer than for free carrier excitations. The dephasing at low excitation intensities is determined by exciton-acoustic phonon interaction. Exciton-exciton and exciton-free carrier scattering also contribute at higher excitation intensities with free carrier scattering being much more effective than exciton-exciton scattering. A decrease of the acoustic phonon coupling with decreasing quantum well thickness has also been deduced from self diffraction experiments [36]. In addition to these incoherent scattering processes self diffraction experiments also reveal the coherent exciton-exciton interaction as recently demonstrated by Leo et al. for GaAs/AlGaAs quantum wells [37] and by Wegener et al. for InGaAs/InAlAs quantum wells [38].

The relatively long dephasing times of free excitons of the order of several picoseconds allow the observation of *quantum beats* in the polarization decay as demonstrated very recently for excitons in quantum wells [39] as well as free excitons in a magnetic field in AgBr [26]. The experimental data of a self diffraction experiment in a GaAs/AlGaAs quantum well with GaAs quantum well thickness of $L_z = 7$ nm are depicted in Fig. 5. The exponential decay of the diffracted signal intensity is superimposed by a modulation. This modulation arises from the interference of the macroscopic polarization of two exciton levels separated in energy by an amount corresponding roughly to the beat frequency. In the case of this particular quantum well sample these two levels correspond to excitons excited in different spatial regimes of the quantum well with thickness difference of one monolayer. Recently, quantum beats in quantum wells have also been observed for light and heavy hole excitons [40, 41]. In the AgBr quantum beat experiment the beats are due to the exciton levels splitted in an external magnetic field [26].

The dephasing of weakly localized excitons has been studied first by Hegarty et al. in quantum wells of GaAs/AlGaAs [42] and InGaAs/InP [43] and more recently in detail in CdSSe mixed crystals [44]. Localization of excitons in quantum wells is due to *energy disorder* caused by inavoidable well width fluctuations [45]. In mixed crystals exciton localization arises from *chemical disorder* [46, 47]. It has been shown for CdSSe that dephasing of excitons resonantly excited within the localized state regime, i.e. at the low energy wing of the exciton absorption can be appreciably slower than for free excitons [44]. The most convincing demonstration of these long dephasing times is by a stimulated photon echo experiment [27]. The experimental

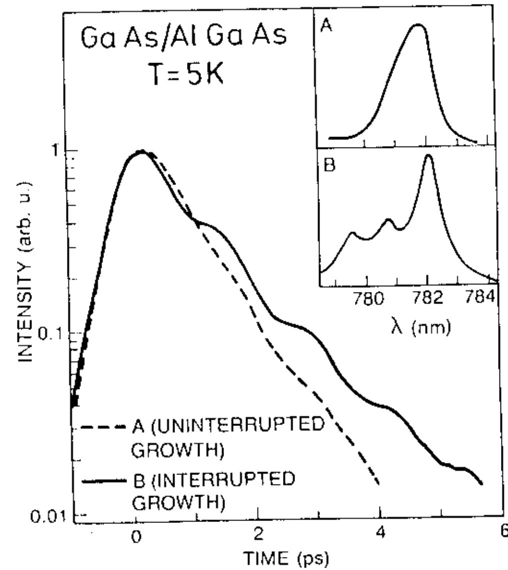

Fig. 5 Self diffraction due to exciton excitation in a GaAs/AlGaAs quantum well with $L_z = 7$ nm. The result shown by the full line exhibits quantum beats arising from excitons with slightly different energy due to one monolayer thickness difference [39].

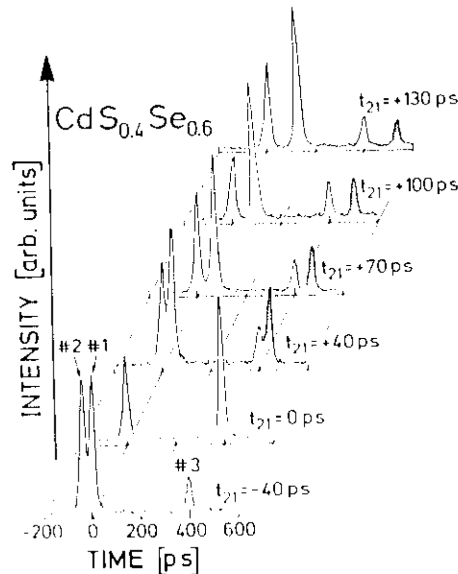

Fig. 6 Stimulated photon echo due to localized exciton excitation in CdSSe. The pulses labeled 1–3 correspond to the excitation pulses. The shaded signal is the stimulated photon echo [27].

result is depicted in Fig. 6. Time resolved detection of the photon echo emitted into the phase matching direction k_4 is performed by a streak camera with 20 ps time resolution. The signals labelled 1, 2, and 3 correspond to the excitation pulses, the time delay between pulse 1 and 2 ($\tau = t_{21}$) is given in the figure. Pulse number 3 is delayed by a fixed amount of $T = 400$ ps. The signal marked by shaded area corresponds to the stimulated photon echo which is emitted at a time $t = T + \tau$. Stimulated photon echoes are observed up to delay times of the order of 100 ps which demonstrates that T_2 can be of the order of 400 ps for excitons excited at very low energies. The dephasing times become much shorter at higher photon energies, where localization is weaker.

Dephasing of excitons in InGaAs/InAlAs quantum wells has been investigated recently by Wegener et al. [48]. Results of a self diffraction experiment are shown in Fig. 7 for a quantum well with $L_z = 20$ nm at three different temperatures. The InGaAs/InAlAs quantum well system is of particular interest because of the combined effect of energy disorder and chemical disorder. However, in pretended contrast to the results on CdSSe mixed crystals, the dephasing times as listed in Fig. 7 are much shorter than for free excitons in GaAs and CdSe. Thus it seems obvious, that the problem of dephasing of localized excitons is quite complicated and general conclusions are difficult at present. This in fact, is strongly supported by recent theoretical studies of disorder induced dephasing in semiconductors [49]. It is demonstrated that a quasi continuous transition from a two level system in momentum space without disorder to a non-interacting two level system in real space for strong disorder exists. In the intermediate regime, dephasing by disorder alone without any quasi particle interaction occurs and thus dephasing of excitons in disordered semiconductors may strongly depend on the detailed nature and strength of the disorder.

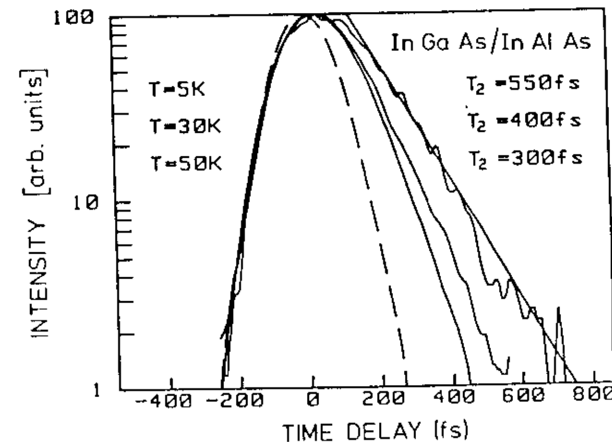

Fig. 7 Self diffraction due to exciton excitation in an InGaAs/InAlAs quantum well with $L_z = 20$ nm at three different temperatures of 5 K, 30 K, and 50 K [48].

5 The Cooling Regime

5.1 Carrier cooling

The subject of carrier cooling in bulk semiconductors and quantum well structures has been of considerable interest over the last years, because these studies provide a very direct access to the interaction of nonequilibrium carriers with the lattice. Time resolved luminescence has been mainly employed. Several excellent reviews covering this topic have been published recently [50–53]. We will here discuss only a few examples to illustrate the concept and potential of these experiments. In a very crude picture, the cooling regime can be described as follows: After the initial scattering of nonresonantly excited carriers out of the optically coupled states, which causes the decay of the macroscopic polarization, the carriers lose their excess energy by the emission of phonons and thermalize amongst each other to a thermal but hot distribution due to carrier carrier scattering. Depending on carrier density, phonon scattering or carrier-carrier scattering dominates initially. The change of the carrier distribution function with time can be directly measured very conveniently by time resolved photoluminescence or excite and probe experiments with white light continuum probe pulses. If a thermalized distribution function is established, the cooling can be characterized by the variation of the effective carrier temperature with time. An example for these *cooling curves* is shown in Fig. 8 for bulk GaAs [54]. The effective carrier temperature is plotted versus delay time, the zero on this scale corresponds to 20 ps after excitation. The effective temperature decreases with increasing time reflecting the cooling of the carriers. Carrier cooling obviously is reduced at higher excitation intensities which can be attributed to hot phonon effects [55]. Screening of the Fröhlich interaction, which is the dominant cooling mechanism in the polar III–V and II–VI semiconductors, would also result in a reduction of the cooling rates, however, seems to be less important even up to carrier densities of the order of 10^{18} cm^{-3} [56].

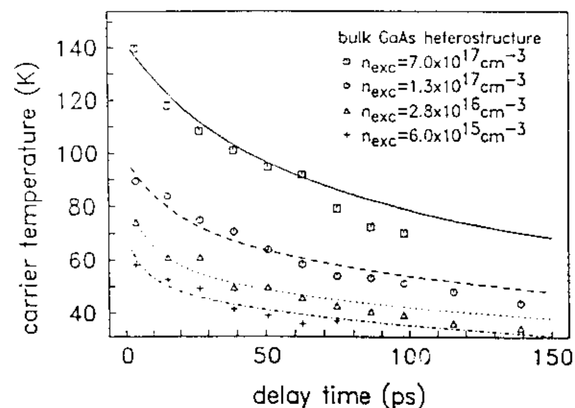

Fig. 8 Cooling curve for free carrier excitations in bulk GaAs. The effective carrier temperature is plotted versus delay time. After Ref. [54].

Carrier cooling in quantum well structures has also been investigated by many groups recently and the conclusions drawn in detail are still controversial. Leo et al. [57] have performed detailed studies of cooling in bulk GaAs and modulation doped as well as undoped GaAs/AlGaAs quantum wells. They conclude that carrier cooling in bulk samples and quantum well structures is much the same for the same volume densities in accordance with earlier theoretical conclusions [58]. In particular, they demonstrate that carrier scattering with longitudinal optical phonons due to the Fröhlich interaction is independent on well width. Instead a slight increase of the acoustic deformation potential scattering with decreasing well width is observed. Ryan and coworkers reported a reduction of the carrier cooling for undoped GaAs quantum wells with wide wells [59] due to the slow light to heavy hole scattering [60]. Nevertheless, the well width dependence of carrier cooling in quantum wells is small and the overall reduction of the cooling rates at high excitation intensities can be attributed to hot phonon effects in both, bulk and quantum well samples. Whether different cooling behaviour might be observed in very thin quantum wells e.g. due to the modifications of the phonon spectrum (interface phonons [61]) remains an open question at present [62, 63].

Hot carrier cooling has also been studied in materials other than GaAs. For example, the cooling in InGaAs has been investigated by Kash et al. [64] and recently by Rieck et al. [65]. Cooling in bulk InGaAs and InGaAs/InAlAs quantum well structures was compared by Lobentanzer et al. [66]. Carrier cooling has also been studied by time resolved luminescence in CdS at high excitation intensities [67]. It turns out that hot phonon effects are less pronounced in this II–VI semiconductor due to the shorter phonon lifetime [68]. At low excitation intensities, hot excitons are formed in the wide gap II–VI materials and thermalization and cooling proceeds via hot exciton relaxation, as recently demonstrated for CdZnTe/ZnTe multiple quantum wells by means of time resolved luminescence and self diffraction experiments [69]. In the nonpolar semiconductor Ge cooling has been investigated by means of subpicosecond excite and probe experiments by Roskos et al. [70].

A complementary and very elegant approach to study carrier-phonon interaction by time resolved techniques is the inverse of the above described cooling experiments, i.e. the observation of the heating of resonantly excited cold carriers by a *warm* lattice.

This technique has been introduced by Rühle et al. [71] using time resolved luminescence to investigate electron-phonon interaction in GaAs. The particular advantage of this approach is that hot phonon effects are eliminated. A similar concept has been applied by Knox et al. [72] to study the dynamics of exciton dissoziation at room temperature in a GaAs/AlGaAs quantum well by means of femtosecond excite and probe experiments. An exciton ionization time due to optical phonon scattering of about 300 fs is obtained. Finally it should be mentioned that transient free carrier absorption applying picosecond infrared laser pulses ($\lambda \simeq 7$ μm) also has a great potential for the study of carrier cooling in semiconductors [73].

Energy relaxation of carriers within localized continuum states of semiconductors can be appreciably slower than in delocalized extended states because energy relaxation then requires spatial relaxation. Energy relaxation can be described by multiple trapping or hopping in the two extremes of high and low temperatures, re-

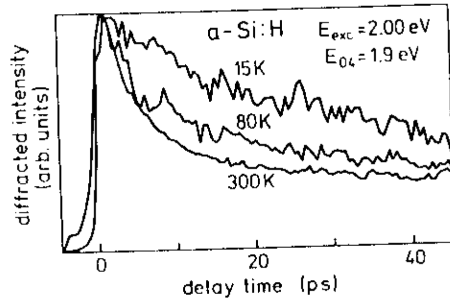

Fig. 9 Transient grating signal in hydrogenated amorphous Silicon at three different temperatures of 10 K, 70 K, and 300 K [79].

spectively, as investigated in great detail for hydrogenated amorphous silicon. Time resolved spectroscopy again has been proven to be a powerful tool to explore energy relaxation in disordered crystalline [74,75] as well as amorphous semiconductors [76–78]. The transition of photoexcited carriers from the extended state regime at higher energies into the localized states across the so-called mobility edge has been studied in amorphous silicon and related alloys by Noll et al. [79] applying the transient grating technique. Results for hydrogenated amorphous silicon at three different temperatures are shown in Fig. 9. The first order diffracted light intensity is plotted versus delay time. The transient grating signal exhibits an initial fast component with a time constant of a few picoseconds at room temperature and is almost constant on a picosecond time scale afterwards. The initial fast transient can be attributed to refractive index changes caused by the relaxation of carriers across the mobility edge [79]. The increase of these trapping times with decreasing temperature reflects the saturation of the shallow localized states due to slower relaxation within these localized states at lower temperatures.

5.2 Intervalley scattering

The conduction band structure of III–V semiconductors exhibits several minima at different points within the Brillouin zone with energy separation of the order of several 100 meV. In the direct gap materials like GaAs and InP the minimum at the center of the Brillouin zone (Γ-point) is lowest and the minima at or close to the edges of the Brillouin zone (L and X, respectively) are at higher energies. Free electrons with sufficiently high energy then can scatter inbetween these different minima, which is referred to as intervalley scattering. Due to the different effective masses corresponding to the different conduction band minima the transport properties depend strongly on the position of the carriers in k-space as seen most clearly in the Hilsum-Gunn effect.

Intervalley scattering in bulk semiconductors requires the participation of large k-vector phonons. Determination of the intervalley scattering time constants thus again provides direct information on the strength of the respective electron-phonon interaction.

Different ultrafast spectroscopy methods have been applied recently to study intervalley scattering in GaAs bulk and quantum well samples [80–84], AlGaAs [80,81,85] and InP [84], even though in some cases stationary hot luminescence experiments also may be applied [86–89]. Excite and probe as well as transmission correlation experiments in GaAs have provided intervalley scattering times of the order of 10 to 100 fs for Γ–X as well as Γ–L scattering. Longer intervalley scattering times have been obtained in some of the cw-experiments [88]. However, since the experimental conditions (excess energy, excitation intensity etc.) may not be always the same one should be careful in the comparison of different data. The femtosecond luminescence experiments by Shah et al. [83] have shown that the reverse scattering process, e.g. from the L-conduction band to the central minimum is much slower (of the order of a few ps) due to the different masses and thus density of states of the different bands.

Whereas in elementary or binary bulk semiconductors intervalley scattering requires participation of large k-vector phonons, intervalley scattering in mixed alloy semiconductors can take place without phonon participation as demonstrated recently by Kalt et al. [85] in AlGaAs by means of time resolved luminescence.

A very special intervalley scattering process can take place in type-II GaAs/AlAs quantum well and superlattice structures. In type-II quantum wells and superlattices electrons and holes are spatially separated and recombination is *indirect in real space* [90], similar to nipi structures. In the GaAs/AlAs system a transition from the usual type-I structure to a type-II system can be achieved by a decrease of the GaAs well thickness. The band alignment of the GaAs/AlAs system is depicted in Fig. 10 to illustrate this behaviour. The full lines correspond to the alignment of the Γ-conduction and valence band edges, the dashed line shows the position of the X-conduction band edge. The top of the valence band is at the center of the Brillouin zone for both, GaAs and AlAs. However, since AlAs is an indirect gap semiconductor, the X-conduction band minimum is lowest in the AlAs, whereas Γ is lowest in the GaAs. In addition, the electron mass at the Γ-minimum in GaAs

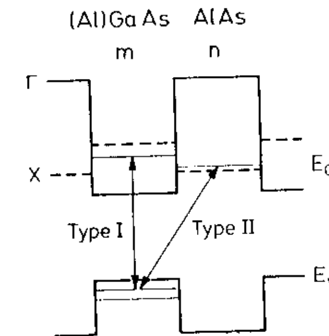

Fig. 10 Band alignment of a type-II (Al)GaAs/AlAs quantum well or superlattice structure.

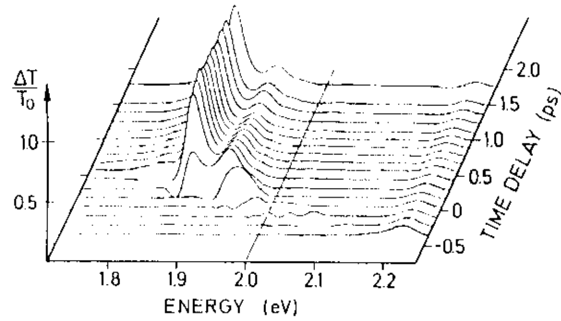

Fig. 11 Differential transmission spectra of a type-II GaAs/AlAs quantum well at various delay times ($T = 5$ K) [91].

is much smaller ($m_e = 0.067\ m_0$) than the highly anisotropic effective mass of the X-minimum in AlAs ($m_\ell = 1.1\ m_0$, $m_t = 0.2\ m_0$). The heavy longitudinal mass m_ℓ is the quantization mass for the X_z minima aligned along the [001] growth direction, whereas the light transverse mass m_t is the quantization mass for the X_x and X_y minima aligned along the [100] and [010] directions, respectively. In any case, the confinement energy increases much faster with decreasing well and barrier thicknesses for the Γ-states confined in the GaAs layers as compared to the X-states confined in the AlAs layers because of the much smaller electron mass of the Γ GaAs states. Eventually, at some critical thickness of the GaAs slabs, the Γ-conduction band states of the GaAs will be higher in energy than the X AlAs states, as indicated in Fig. 10. This critical layer thickness in the GaAs/AlAs system is about 12 monolayers of GaAs ($a_0 = 0.283$ nm). Electrons created optically in the Γ-conduction band of the GaAs then will relax into the lower X states of the AlAs. Opposite to intervalley scattering in bulk semiconductors, this requires an appreciable spatial transfer across the interface of the GaAs/AlAs heterobarrier. This unique intervalley scattering process in type-II GaAs/AlAs and AlGaAs/AlAs quantum wells and superlattices has been studied recently in great detail by Feldmann et al. [91,92] applying various femtosecond spectroscopy techniques. A typical result of a femtosecond excite and white light continuum probe experiment is depicted in Fig. 11 for a type-II GaAs/AlAs sample with 11 and 24 monolayers of GaAs and AlAs, respectively. The sample is excited by 100 fs pulses of a Rh 6G CPM dye laser. This excitation pulse creates holes and electrons in, respectively, the Γ valence band and conduction band states of the GaAs. The transmission changes induced by this pump pulse are monitored with the white light continuum probe pulses.

The differential transmission spectra ($\Delta T/T$) are plotted for different delay times between the pump and the probe pulses. The spectra exhibit two strong positive peaks at about 1.9 eV corresponding to bleaching of the light and heavy hole exciton transition of the GaAs slabs and a weaker positive peak at about 2.2 eV corresponding to the split-off exciton. The exciton bleaching recovers partly on a subpicosecond

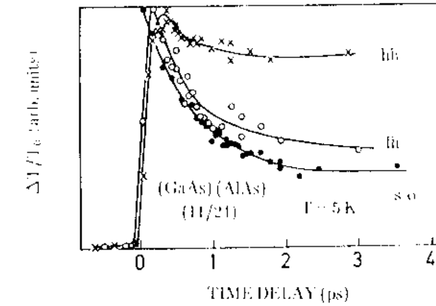

Fig. 12 Peak differential transmission (from Fig. 11) versus delay time for the heavy hole (hh), light hole (lh) and split-off (s-o) exciton transition [91].

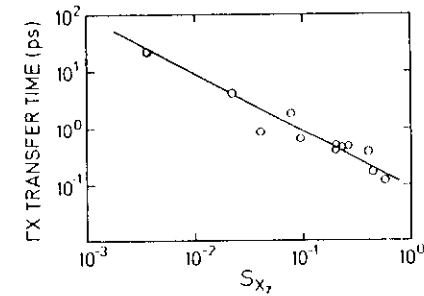

Fig. 13 Intervalley (Γ–X) transfer times ($T = 5$ K) versus the squared overlap integral for different type-II GaAs/AlAs structures [92].

time scale in case of the type-II structures, opposite to the behavior of the usual type-I quantum wells and superlattices [93]. This partial recovery of the bleaching signal is more clearly seen in Fig. 12, where the maximum signal is plotted for each transition versus delay time. The fast partial recovery of the exciton bleaching in type-II structures can be attributed to intervalley scattering of electrons out of the Γ-conduction band states of the GaAs into the X-conduction band states of the AlAs. Due to this scattering the state filling of the Γ-electron states of the GaAs disappears, which explains the partial recovery of the exciton bleaching. A Γ–X scattering time of about 650 fs is deduced for this particular sample. Different samples with different GaAs (AlGaAs) and AlAs layer thicknesses show different Γ–X scattering times. The time constants are basically determined by the spatial overlap of the Γ and X wavefunctions, which have maximum amplitudes in the GaAs and AlAs, respectively. This is demonstrated in Fig. 13, where data for the Γ–X scattering time constant of different samples are plotted versus S_{X_z}, which corresponds to the sum over the squared overlap integrals of the Γ and X_z Kronig-Penney envelope

wavefunctions including all the possible X_z final states. Scattering into these X_z states is possible without phonons and due to the potential step at the interface, which mixes Γ and X_z states. Mixing of Γ and X_x and X_y states is also possible due to interface roughness, but seems to be less important as compared to $\Gamma - X_z$ mixing [92]. For AlGaAs/AlAs type-II quantum wells with much thicker layer thicknesses, Γ-X scattering also takes place by optical phonon scattering [92].

The Γ-X scattering discussed above is an example for a process within the *cooling regime* only possible in the artificial low dimensional structures like quantum wells or superlattices. Other examples for processes which are unique to these low dimensional semiconductors are *intersubband scattering* [94–96], *carrier trapping* from barrier layers into the quantum well [97] and *resonant* or *non-resonant tunneling* between different wells. This latter process will be discussed briefly as a final example for the cooling regime, even though tunneling might be considered as a coherent process and thus could be treated as well in Section 4. Nevertheless, it should be realized that the coherence in the tunneling process refers to an electron or hole state, whereas in Section 4 we have been concerned with optical coherence.

5.3 Tunneling

One of the most fundamental processes in quantum mechanics is tunneling of particles through potential barriers. Initiated by the work by Esaki and Tsu [98], tunneling in semiconductor quantum wells and superlattice structures has gained great interest over the last 15 years also because of the great potential for devices [99]. The traditional approach to study resonant and nonresonant tunneling phenomena is by means of electrical transport measurements [99–101]. More recently, however, picosecond time resolved luminescence has been applied very successfully to study tunneling in quantum well structures [102–110]. In particular, the difference between resonant and nonresonant tunneling can be addressed in asymmetric double quantum well structures by applying external electric fields [104, 108–110]. Results of time resolved luminescence of a GaAs/AlGaAs asymmetric double quantum well tunneling structure with nominal thicknesses of the two GaAs quantum wells of 7 nm and 5 nm as reported by Oberli et al. [104] are shown in Fig. 14. The upper part in Fig. 14 illustrates the alignment of the $n = 1$ subband of the narrow well and the $n = 1$ and $n = 2$ subbands of the wide well at zero electric field (a), at the resonance field (b), where the $n = 1$ and $n = 2$ subband of the narrow and wide well, respectively, are in resonance, and above resonance (c). At resonance the two wells are strongly coupled and the energy levels are splitted according to the strength of the coupling. An electron created in either one of the wells will oscillate coherently back and forth if no damping occurs. The measured tunneling times as depicted in the lower part of Fig. 14 clearly reveal the resonance behavior showing the shortest tunneling time at resonance. However, the tunneling times are appreciably longer than expected for coherent oscillation of the electron wavepacket, indicating that incoherent processes have to be considered in addition [104, 108].

A quite different experimental approach to study tunneling in asymmetric quantum wells has been applied recently by Leo et al. [111] using the two beam self diffraction technique as described in Section 4. The self diffracted beam intensity is measured

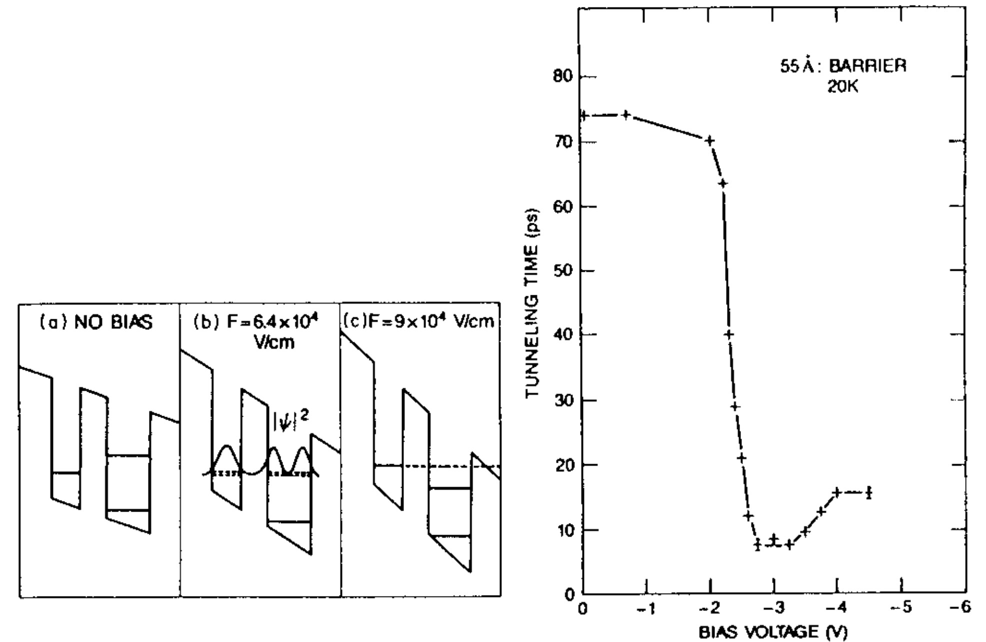

Fig. 14 Tunneling time versus bias voltage for a GaAs/AlGaAs asymmetric double quantum well structure. The upper part shows the conduction band alignment and electronic sublevels at zero bias (a), resonance (b), and above resonance (c). After Ref. [94].

in these experiments for a GaAs/AlGaAs asymmetric double quantum well with 4.2 and 6.7 nm thickness of the narrow and wide well, respectively, and a AlGaAs ($x = .37$) barrier thickness of 4.8 nm for different external electric fields and for resonant excitation of either the lowest exciton transition in the wide well (WW) or the narrow well (NW). Results are shown in Fig. 15. The dephasing times T_2 as obtained from the decay of the self diffracted signal intensity (c.f. Section 4) are independent on electric field in case of resonant excitation of the wide well. Opposite, the dephasing times show a pronounced minimum at the resonant field corresponding to resonance of the $n = 1$ level of the wide and narrow well for resonant excitation of the narrow well. The experimental results are summarized in Fig. 16, where the decay times of the self diffracted signal intensity are plotted versus external electric field strength for the case of resonant excitation of the narrow well. The dephasing times T_2 are obtained by multiplying the decay times by four, because of the dominant inhomogeneous broadening. The important difference between resonant excitation of the wide and narrow well is illustrated in the inset of Fig. 15: Resonant excitation at the lowest possible energy in case of the narrow well causes also nonresonant free carrier excitation in the wide well, whereas resonant excitation of the wide well cannot produce any intrinsic excitation in the narrow well, because

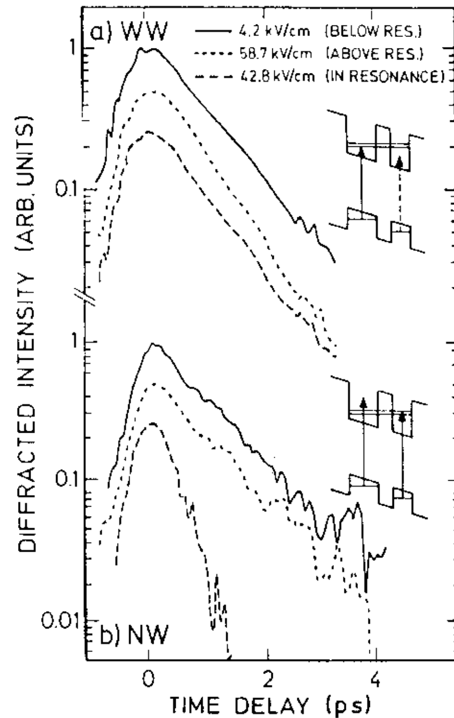

Fig. 15 Self diffraction due to exciton excitation in a GaAs/AlGaAs asymmetric double quantum well structure ($T = 5$ K) at different bias voltages for resonant excitation of the wide well $n = 1$ heavy hole exciton (a), and $n = 1$ narrow well heavy hole exciton (b) [111].

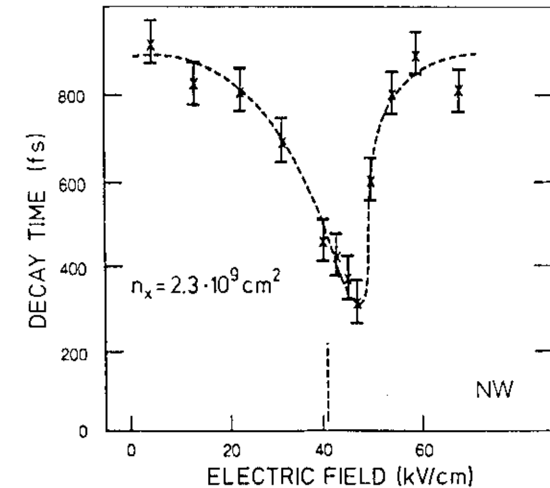

Fig. 16 Decay time constants of the self diffracted signal intensity of the same sample as shown in Fig. 15 versus electric field for resonant excitation of the narrow well [111].

the confinement energy is larger in the narrow well: At the resonant field for the $n = 1$ electron subbands, electrons created resonantly in either well can tunnel into the neighbour well. In case of NW-excitation scattering with the nonresonantly created electrons of the wide well will immediately destroy the phase of the tunneling electrons, which results in the loss of optical coherence of the excitonic excitation. In the case of WW-excitation the electrons can tunnel back and forth several times and the dephasing of the exciton is determined by the processes already discussed in Section 4.

6 Recombination

Recombination in high quality intrinsic direct gap semiconductors takes place on a nanosecond, in some cases even much longer time scale. Thus ourdays recombination is not considered to be *ultrafast* and will not be discussed in detail in this article. However, a few comments will be made for the sake of completeness according to the scheme of Fig. 1.

Recombination in intrinsic, direct gap bulk crystals as well as quantum well and superlattice structures is strongly determined by excitonic effects. Recombination in direct gap bulk crystals therefore has to be analyzed in the polariton picture. Exciton recombination in ultrathin heterostructures, in particular quantum wells, instead may be discussed more straight forward in terms of exciton transition strengths. The free exciton recombination lifetime of bulk GaAs at low temperatures has been determined to be about 3 ns [112]. Excitonic effects are enhanced in quantum wells due to the confinement resulting in shorter exciton lifetimes [113, 114]. The effect of exciton confinement and exciton coherence on the exciton recombination lifetime has been studied in detail by Feldmann et al. [115–117]. Results for the low temperature exciton recombination time constants for GaAs/AlGaAs multiple quantum well structures with different quantum well thickness L_z are shown in Fig. 17. The decay times of the exciton recombination decrease with decreasing well thickness from about 1.7 ns for $L_z = 15$ nm to about 0.32 ns for $L_z = 2.5$ nm. This decrease is consistent with the increase of the exciton binding energy and the increase of the oscillator strength, yet, it has to be considered that apart from the exciton oscillator strength the dephasing time T_2 also determines the exciton recombination lifetime [115–117]. The exciton decay times increase again with further decrease of the quantum well thickness due to the penetration of the wavefunctions into the barrier material [118]. The radiative exciton decay times also increase again in

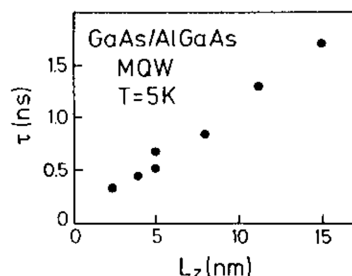

Fig. 17 Exciton luminescence decay times in GaAs/AlGaAs multiple quantum wells as a function of quantum well thickness L_z [115].

(i) superlattices where miniband formation occurs [119–120] revealing the transition from a two dimensional quantum well to an anisotropic three dimensional superlattice and

(ii) modulation doped quantum wells due to the reduction of the exciton oscillator strength by phase space filling [120–122].

Recombination in the type-II quantum well and superlattices is much slower due to the spatial separation of electrons and holes with characteristic recombination times of the order of μs or even ms [90] comparable to indirect gap semiconductors.

7 Conclusion

As we have shown in this article, the *life* of photoexcited electron hole pairs may cover a time scale of the order of 10 decades if the time constants of the very initial scattering processes resulting in the loss of coherence are taken as the unit of time. To visualize this number, one might keep in mind that 10 decades with a year as the unit of time corresponds roughly to the age of our universe since *big bang*. To stay within this picture, femtosecond laser technology today has made it possible to study experimentally the history of a photoexcited electron hole pair from *big bang* up to its *annihilation*. So far, however, we are still at the beginning, in particular as the very initial processes in light — semiconductor interaction are considered. The potential of ultrafast laser spectroscopy has been demonstrated, but many questions still remain open. It thus seems very likely that ultrafast laser spectroscopy of semiconductors and semiconductor microstructures will be a very active and exciting field also in the near future.

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Extract of the Foreword