Resonant Secondary Emission from Two-Dimensional Excitons: Femtosecond Time Evolution of the Coherence Properties

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Using a novel technique, we simultaneously study the femtosecond dynamics of the amplitude and the coherence of resonant secondary emission from GaAs quantum wells. Two phase-locked pulses resonantly excite excitons in the semiconductor and the subsequently emitted radiation is detected by femtosecond up-conversion. In experiments at various delays and phases between the excitation pulses we identify three different contributions to the secondary emission with different coherence properties in time. [S0031-9007(98)07633-9]

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The time evolution and coherence of light emission after ultrashort resonant excitation of matter is one of the fundamental questions in physics. Light emitted in directions different from the excitation and reflected beams is generically known as secondary emission. Although resonant secondary emission (RSE) has a spontaneous character it is not necessarily incoherent, i.e., a part of RSE may still have a defined phase relation to the excitation pulse. It is well known that a resonantly excited atom emits two components of light [1]. (a) Resonant Rayleigh scattering (RRS): the emitted light has an *electric field* with amplitude and phase because it is driven by the coherently excited polarization of the atom. (b) Resonant photoluminescence (RPL): this *incoherent* component occurs as a result of dephasing of the optical transition. With the decay of the coherent polarization the emitted electric field also decays, and consequently, only the intensity of RPL can be detected.

Resonantly excited excitons in quantum wells emit a considerably large amount of RRS and RPL which was first demonstrated in spectrally resolved experiments with continuous wave excitation [2] and was more recently investigated with a phase-locked pair of excitation pulses [3]. The latter measurements provide information similar to cw experiments as discussed in Ref. [3]. The nature of RRS from an ensemble of excitons may be quite different from that from a single atom. For low excitation intensities RRS is caused by static disorder in quantum wells that is responsible for the inhomogeneous broadening of the excitonic transition. Because of the randomly positioned scatterers one expects, under certain conditions, to observe an irregular speckle pattern in RRS with a granularity depending on the size of the excitation spot and the distance of observation. In addition to dephasing the transient rise and decay of RRS is determined by interplay of constructive and destructive interference between emitted electric fields of the various scatterers. Because of the coherent nature of RRS classical models have been applied to calculate the transient intensity [4] but *not* the electric field of RRS. However, after the decay of RRS there is still RPL driven by the incoherent population of excitons which is absent in any classical description.

The initial dynamics of resonantly excited quantum wells was first investigated by Wang *et al.* [5] and later extended to lower intensities by Haacke *et al.* [6]. Both groups observed after femtosecond excitation a gradual rise of RSE within 5 ps. The time scale and shape of this rise depends strongly on the intensity of the excitation pulse and was interpreted in terms of scattering from static disorder for low intensities (RRS) and in terms of exciton-exciton collisions (RPL) for higher excitation densities. A time-resolved separation of the two components, where the coherence properties of the emitted light are directly measured as a function of time, is still missing.

In this Letter we have taken a novel approach which allows us to separate the different components of RSE as a function of time. The sample is resonantly excited by a pair of phase-locked pulses, and the subsequent emission is detected by femtosecond up-conversion. This allows us to determine the femtosecond dynamics of RSE and its coherence properties *simultaneously* for the first time. In particular, we identify and characterize *three* different components to RSE which have different coherence properties and temporal dynamics.

Our experimental scheme is similar to that in Ref. [7] but we use femtosecond instead of picosecond excitation pulses. Pulses (center wavelength $\lambda = 809$ nm, pulse duration 120 fs) from an 80 MHz Ti:sapphire oscillator travel through an actively stabilized Michelson interferometer in order to produce two phase-locked excitation pulses with a variable separation τ_M (Michelson delay). The relative phase Φ between the excitation pulses is controlled by fine-tuning τ_M within one optical cycle. The pair is focused (diameter: 50 μm) onto a high quality GaAs quantum well sample. It consists of ten periods of 17 nm GaAs wells separated by 15 nm AlGaAs barriers. The sample is cooled in a cryostat to $T = 10$ K. The collected emission is up-converted in $LiIO_3$ using 200 fs gating pulses at 1530 nm from a synchronously pumped OPO. τ_G (gate delay) is the separation between the

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second excitation pulse and the gate pulse. Measurements were made for various Michelson and gate delays, and relative phases Φ between the two excitation pulses. In this Letter we concentrate on the low excitation density data where nonlinear effects are of minor importance. The presented data are recorded for an excitation power of 50 μW which correspond to an estimated excitation density of about 10^9 excitons per cm^2 [8].

A time-resolved measurement of the RSE after resonant excitation with a *single* pulse is shown in Fig. 1(a). The transient intensity is plotted as a function of τ_G. At $\tau_G = 0$ there is an intense peak which is due to nonresonant surface scattering. The subsequent emission shows, first, a gradual rise within 5 ps followed by a fast decay within 9 ps and a slower decay on a 50 ps time scale (not shown). Oscillations with a period of 600 fs are superimposed on the bell shaped part of the curve. This beating between heavy and light hole excitons is present in all transient emission curves and will be ignored in the further discussion.

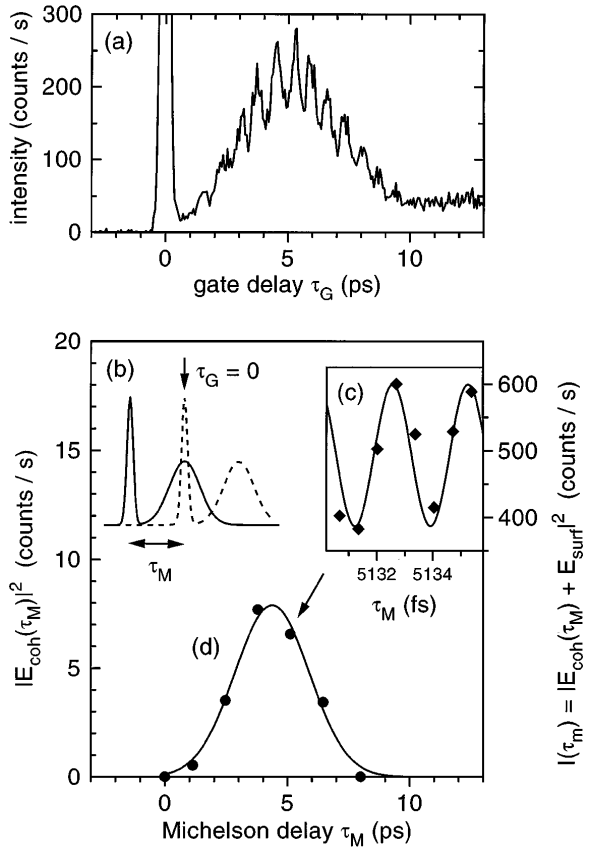

FIG. 1. (a) Transient intensity of the secondary emission after resonant excitation of excitons with a single pulse. The intensity is plotted as a function of the delay τ_G between excitation and gate pulses. The peak at $\tau_G = 0$ is due to nonresonant surface scattering. (b) Schematic of the secondary emission triggered by a pair of phase-locked pulses. Michelson delay τ_M is the separation between the phase-locked excitation pulses. (c) Observed intensity fringes $I(\tau_M)$ (symbols) while fine-tuning τ_M around 5133 fs. (d) Transient intensity $|E_{coh}(t)|^2$ (symbols) of the optically coherent component of RSE obtained in an experiment with a pair of phase-locked excitation pulses. Solid lines in (c) and (d) are cosine and Gaussian fits, respectively.

Now we investigate the coherence properties of RSE. We will show that the total emission shown in Fig. 1(a) consists of *three* different components having different coherent relations to the excitation pulse.

(i) In a first series of experiments, we investigate whether there is an optically coherent component to RSE, i.e., light with a macroscopic electric field which can interfere with a replica of the excitation pulse. For this, we scan the nonresonantly surface-scattered light of the second excitation pulse ($\tau_G = 0$) across the RSE triggered by the first pulse by *fine-tuning* the Michelson delay τ_M [Fig. 1(b)]. This leads to pronounced constructive or destructive interference with the electric field of the surface-scattered light of the second pulse [Fig. 1(c)]. Since surface-scattered light is a replica of the laser pulse, this observation shows that there is an *optically* coherent part of the RSE. We want to emphasize that such a component of RSE has not been reported or discussed in previous studies [3,5,6]. The measured intensity [Fig. 1(c)] results from the superposition of the two electric fields: $I(\tau_M) = |E_{coh}(\tau_M) + E_{surf}|^2$. The transient intensity of the optically coherent part of RSE $|E_{coh}(\tau_M)|^2$ calculated from this equation is shown in Fig. 1(d). Since we do not know the exact spatial pattern of the surface-scattered light it is not possible to give an exact value for the amplitude of this contribution. Nevertheless, our experiment gives reliable relative values at different times after excitation. A similar experiment and analysis as in Ref. [3] on our sample have shown that this optically coherent component must be a large portion if not the major part of the total RSE. In classical formulation, the intensity of any light is connected to an electric field with defined amplitude and phase. Using the model of spatially correlated disorder with correlation length ξ [4] we have numerically calculated the electric field of RRS in this classical picture. For a spatial disorder on a length scale comparable to the wavelength we calculate transients similar to those observed in our experiment shown in Fig. 1(d). The amplitude and the decay of the signal are essentially determined by the spatial correlation length ξ. For long dephasing times we find that the rise and decay of this classical RRS is mainly governed by constructive and destructive interference between the various emitters of the inhomogeneously broadened ensemble.

(ii) Figure 1(d) shows that the optically coherent RSE in response to the first pulse vanishes >8 ps after the pulse, i.e., it has *no longer* an electric field with a defined phase relation to the excitation pulse. Nevertheless, the RSE intensity at $\tau_G = 15$ ps exhibits well-defined fringes as a function of the fine-tuning of τ_M [Fig. 2(b)]. We define a gate-delay dependent fringe contrast as $FC(\tau_G) = (I_{max} - I_{min})/(I_{max} + I_{min})$. For a given τ_M, the FC is nearly independent of τ_G for $\tau_G > 15$ ps.

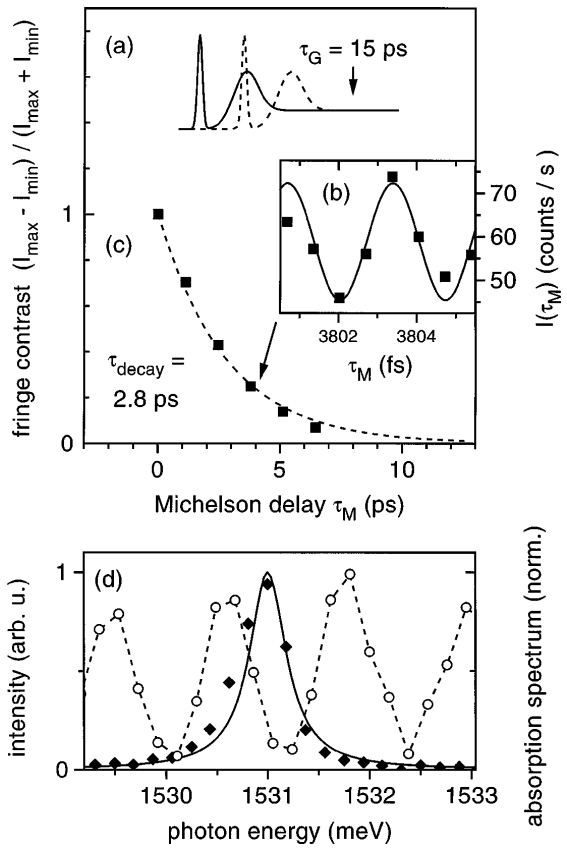

FIG. 2. (a) Scheme of two pulse excitation and subsequent gated detection at $\tau_G = 15$ ps. (b) Intensity fringes $I(\tau_M)$ (symbols) while fine-tuning of τ_M around 3803 fs (solid line: cosine fit). (c) Symbols: fringe contrast of RSE defined by $(I_{max} - I_{min})/(I_{max} + I_{min})$ as a function of τ_M. The exponential fit (dashed line) gives a decay time of 2.8 ps. (d) Diamonds: time-integrated RSE spectrum as a function of the detection frequency. Circles: intensity spectrum of two phase-locked pulses ($\tau_M = 3.8$ ps). Solid line: excitonic absorption spectrum (Lorentzian) calculated from the exponentially decaying fringe contrast.

Figure 2(c) shows that the fringe contrast at $\tau_G = 15$ ps (solid symbols) decays almost exponentially as a function of τ_M. An exponential fit (dashed lines) gives a decay constant of 2.8 ps. The constant fringe contrast for $\tau_G > 15$ ps shows that the observed interference has nothing to do with the emission process, i.e., it has to be attributed solely to interference in the absorption process. This is incoherent luminescence (RPL), i.e., a sequential process where first a photon is absorbed and then at later times another photon is emitted. Between these independent processes any phase memory about the excitation process is gone and, consequently, the interference *exclusively* takes place in the absorption process. Here, the well known picture of coherent control of exciton generation applies [9]. The intensity spectrum of two phase-locked pulses is simply the spectrum of a single pulse times a phase-dependent fringe function $F(\tau_M) = 1 + \cos(\omega_0 \tau_M)$, where ω_0 is the carrier frequency of the excitation pulses. A measured spectrum of such pulses is shown as a dashed line in Fig. 2(d). The experimentally observed fringe pattern is simply the product of $F(\tau_M)$ times the excitonic absorption spectrum $\alpha(\omega)$. Plotting the fringe contrast as a function of τ_M as in Fig. 2(c) provides direct information about the line shape of the excitonic absorption process [3,9]. The exponential fit corresponds to a Lorentzian absorption line with a width of 0.5 meV shown as a solid line in Fig. 2(d). An emission spectrum (diamonds) obtained from a time-integrated measurement gives a comparable linewidth. It is important to note that the fringes at late times do not contain information about whether the absorption line is homogeneously or inhomogeneously broadened. At higher excitation intensities we observe a broadening of this absorption line (not shown) which is probably due to excitation induced dephasing [10]. In contrast the width of the measured time-integrated luminescence spectrum [diamonds in Fig. 2(d)] does not broaden pointing again to the independence of absorption and emission processes.

(iii) Measuring the fringe contrast for intermediate gate delays reveals the surprising existence of a third regime of RSE with coherence properties between cases (i) and (ii). Typical experimental results are shown in Figs. 3(a)–3(c). The intensity of the RSE is plotted as a function of the gate delay τ_G for opposite phase relations ($\Phi = 0$ and $\Phi = \pi$) and various Michelson delays τ_M between the two excitation pulses. For $\tau_M = 0$ (not shown) the fringe contrast is 100% at all τ_G, as expected. For all nonzero τ_M [Figs. 3(a)–3(c)], the signal shows two peaks at the positions of the excitation pulses due to nonresonant surface scattering, and the RSE after the second pulse shows a more complex structure in the transient intensity as well as the phase of the fringes. For instance, RSE shown in Fig. 3(b) exhibits a phase change by π for gate delays around $\tau_G = 4.5$ ps. The most striking feature is the fact that in the time interval $0 < \tau_G < 12$ ps the fringe contrast is much larger than at later times [cf. Fig. 2(d)]. The components discussed in (i) and (ii) cannot explain this behavior for the following reasons. We know from Fig. 1(d) that the coherent field emitted by the sample in response to the first pulses vanishes $\approx$8 ps after the pulse. Therefore, no interference between RSE of the two pulses is expected from picture (i) more than 8 ps after the first pulse, a time position indicated by the vertical lines in Figs. 3(a)–3(c), and yet strong interference is observed (Fig. 3). Picture (ii) corresponds to the interference in the excitation process; i.e., interference between the polarization created by the first pulse and the electric field of the second pulse creates a certain incoherent population that is *exclusively* determined by τ_M. This population, and hence the fringe contrast, is independent of the gate delay τ_G. We conclude, therefore, that the varying fringe contrast and phase changes as experimentally observed in Figs. 3(a)–3(c) are possible only if some additional interference takes place *in the sample*.

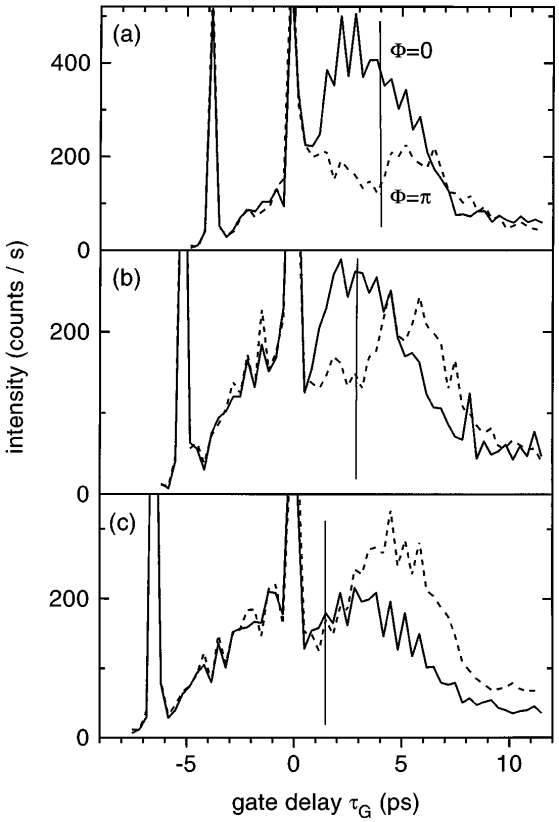

FIG. 3. Transient intensity of RSE after excitation with phased-locked pulses for various separations τ_M between the excitation pulses of (a) 3.8 ps, (b) 5.13 ps, and (c) 6.47 ps. Solid and dashed lines represent relative phases between the excitations pulses of $\Phi = 0$ and $\Phi = \pi$, respectively. The vertical lines indicate the respective time positions at 8 ps after the first pulse where the optically coherent component of RSE triggered by the first pulse has totally vanished [Fig. 1(d)].

A qualitative explanation for this third component of the resonant secondary emission from quantum well excitons (Fig. 3) is as follows. The occurrence of RRS is directly connected with the fact that the excitonic line is inhomogeneously broadened. Thus, in addition to the coherent control of the total incoherent population [component (ii)] the two phase-locked pulses introduce also quantum coherence between excitons of different transition frequencies within the ensemble [11–13]. The time scale of such excitonic interstate coherence might be much longer than the dephasing time of the individual optical transitions. This can result in quite complex interference phenomena for intermediate gate delays; e.g., the impulsively excited interstate coherence leads also to quantum beats in the optically incoherent luminescence as proposed in Ref. [14]. This qualitative picture is fully confirmed by model calculations which will be published in a separate paper.

In conclusion, we have investigated the initial dynamics of resonantly excited excitons in quantum wells by a novel technique in which the sample is resonantly excited using a pair of phase-locked pulses and the resulting emission at a variable delay from the excitation pulses is detected using femtosecond up-conversion. We have identified and characterized for the first time three different contributions to RSE with different coherence properties and temporal dynamics. We have directly time-resolved measured an optically coherent component to RSE which has a macroscopic electric field. After the decay of optical coherence in RSE, phase memory of the excitation persists in the sample and results and complex phase sensitive behavior in RSE.

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[1] R. Loudon, *The Quantum Theory of Light* (Clarendon Press, Oxford, 1983), 2nd ed.
[2] J. Hegarty *et al.*, Phys. Rev. Lett. **49**, 930 (1982).
[3] M. Gurioli *et al.*, Phys. Rev. Lett. **78**, 3205 (1997).
[4] R. Zimmermann, Nuovo Cimento Soc. Ital. Fis. **17D**, 1801 (1995).
[5] H. Wang *et al.*, Phys. Rev. Lett. **74**, 3065 (1995).
[6] S. Haacke *et al.*, Phys. Rev. Lett. **78**, 2228 (1997).
[7] X. Marie *et al.*, Phys. Rev. Lett. **79**, 3222 (1997).
[8] Y. Masumoto *et al.*, Phys. Rev. B **32**, 4275 (1985).
[9] A. P. Heberle *et al.*, Phys. Rev. Lett. **75**, 2598 (1995).
[10] H. Wang *et al.*, Phys. Rev. Lett. **71**, 1261 (1993).
[11] E. O. Göbel *et al.*, Phys. Rev. Lett. **64**, 1801 (1990).
[12] M. Koch *et al.*, Phys. Rev. B **48**, 11 480 (1993).
[13] S. T. Cundiff, Phys. Rev. A **49**, 3114 (1994); S. T. Cundiff *et al.*, Phys. Rev. Lett. **77**, 1107 (1996).
[14] H. Stolz, *Time-Resolved Light Scattering from Excitons*, Springer Tracts in Modern Physics Vol. 130 (Springer, Berlin, 1994); H. Stolz *et al.*, Phys. Rev. B **47**, 9669 (1993).