Coherent vs Incoherent Emission from Semiconductor Structures after Resonant Femtosecond Excitation

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We show that an interferometric correlation measurement with fs time resolution provides an unambiguous discrimination between coherent and incoherent emission after resonant femtosecond excitation. The experiment directly probes the most important difference between the two emissions, that is, the phase correlation with the excitation pulse. The comparison with cw frequency resolved measurements demonstrates that the relationship between coherent and incoherent emission is similar under femtosecond and steady-state excitation. [S0031-9007(97)02962-1]

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Time-resolved luminescence spectroscopy of semiconductor structures after resonant excitation has received increasing attention in the last few years [1,2]. The photogeneration of excitons at the fundamental transition bypasses the energy relaxation processes and allows a direct investigation of the intrinsic exciton dynamics. Resonant luminescence experiments have indeed provided extremely valuable information on the dephasing processes [2], on spin and momentum relaxation [3,4], and on super-radiant effects related to the 2D exciton polariton in quantum well heterostructures [3–5].

Despite this vast and fruitful activity, the fundamental and debated question of how to discriminate between the coherent [resonant Rayleigh scattering (RRS)] and incoherent [photoluminescence (PL)] emission after ultrashort resonant excitation still needs a conclusive answer, particularly when, as in the case of semiconductor heterostructures, the two signals are expected to decay with comparable time constants [1–5]. The spectral width of the excitation pulse is usually larger than the excitonic linewidth and, under these conditions, the RRS and PL spectral profiles turn out to be very similar in state-of-the-art samples. Therefore in time-resolved experiments, unlike in the cw case [6,7], frequency resolved measurements do not help in the separation of the two signals. Polarization conserving properties have been tentatively used as a possible discriminating tool between RRS and PL; however, the results are not conclusive due to both depolarization effects in RRS and exciton spin relaxation processes [2,3]. Not even resorting to comparative analysis with nonlinear techniques [3,4] and/or additional information such as the power dependence of the emission rise time [4] has succeeded in assessing the nature of the measured emission in a clear-cut way. It has been concluded that "*one must await further theoretical developments and experimental investigations to resolve this question unambiguously*" [8].

We show that an unambiguous discrimination between RRS and PL after resonant excitation in a fully time-resolved experiment can be obtained in a two-pulse interferometric measurement with fs time resolution. The experiment directly probes the most important difference between coherent and incoherent emission, that is, the phase correlation between the emitted light and the excitation pulse. Moreover, a quantitative estimate of the intensity ratio between the RRS and PL signals can be easily obtained. We experimentally demonstrate the validity of the approach and apply it to the investigation of GaAs bulk and quantum well (QW) structures.

The experimental setup is rather simple. The beam of a fs Ti-sapphire laser (130 fs pulse duration, 82 MHz repetition rate) is fed into a Michelson-type interferometer providing at the output two collinear pulses delayed by t_D. The two pulses have the same intensity and polarization and are resonant with the fundamental transition of the system. The emission from the sample is spectrally filtered by a monochromator and detected by a photomultiplier. We have performed a series of experiments on GaAs bulk and QW structures grown by molecular beam epitaxy. The QW structure consists of a 18 nm GaAs layer embedded in thick $Al_{0.3}Ga_{0.7}As$ barriers. The measurements have been performed at low temperature ($T = 13$ K) and low excitation ($n_{\mathrm{exc}} < 10^9$ cm^{-2}). Details on this interferometric correlation technique, as applied to the study of the coherent dynamics in GaAs based heterostructures, can be found in Refs. [9,10].

Let us start with a brief description of the physics involved in the experiments. Among the several theoretical pictures proposed so far for the description of the resonant fluorescence, we refer to the approaches developed in Refs. [11,12] which turn out to be very appropriate for the description of a two-pulse experiment [10,11] even if proposed in a different context. The two-pulse excitation induces two coherent polarization waves that interfere as

long as the delay time is comparable with the coherence relaxation time T_2. The interference of the polarization waves can be probed by detecting the incoherent PL and/or the coherent emission. For example, by solving the optical Bloch equation (OBE) for a two-level system under the assumption of $\delta(t)$-pulse excitation, one finds that the measured signal as a function of the delay time t_D turns out to be an interferogram given by [9]

$$I(t_D) \propto 1 + \cos(\omega_0 t_D) \exp\{-|t_D|/T_2\}, \qquad (1)$$

where ω_0 and T_2 are the frequency and dephasing time of the excitonic transition.

So far we have assumed the experimental apparatus to have a very large spectral bandpass (i.e., much larger than the RRS linewidth). We now show that filtering the emission through a narrow bandpass filter allows the discrimination between RRS and PL. In fact, as shown in detail in Ref. [10] on the basis of the theory in Ref. [12], the filter induces a distortion of the shape of the interferogram that depends on the coherence properties of the detected emission. In order to clarify the effects of spectral filtering, we report the results obtained in the case of nonresonant excitation of the GaAs bulk structure. The excitation beam has been tuned at 780 nm, that is, more than 70 meV above the fundamental excitonic transition at 818 nm. We have detected the emission at both the excitonic transition (incoherent PL) and the laser frequency [Rayleigh scattering (RS)] [13]; the observed interferograms are reported in Fig. 1. In the case of detection through a broad bandpass filter, the PL and RS interferograms have the same shape [Figs. 1(a) and 1(b); note the different time scales in the two graphs] which reflects the pulse autocorrelation due to excitation in the free carrier continuum. The insertion of a narrow bandpass filter has a dramatic effect on the RS interferogram, Fig. 1(c), which becomes the Fourier transform of the filter shape [10], while no modification is observed in the corresponding PL interferogram [Fig. 1(d)]. In fact, the PL emission occurs after irreversible dephasing and does not keep memory of the phase relationship with respect to the excitation pulse. Then, in a two-pulse experiment, the two PL emissions delayed by t_D have random relative phases so that, on average, they do not give rise to any interference effect and the PL frequency selection does not affect the coherent polarization waves. In fact, the measured interferogram is originated by the interference of the polarization waves, with a consequent modulation of the total absorbed energy according to Eq. (1). In the case of RRS, instead, the absorption and the emission processes are strongly correlated. The RRS emission arises from the coherent polarization, and therefore it has a fixed phase relation with respect to the excitation pulse. The corresponding interferogram reflects the interference between the two coherent phase-locked polarization waves delayed by t_D or, equivalently, between the corresponding emitted fields; it follows that spectral filtering the RRS signal is equivalent to filtering the polarization waves. The consequent removal of some of the Fourier components of the RRS emission does modify the interference pattern. We conclude that the use of spectral filtering allows us to discriminate whether the emitted fields induced by two identical pulses delayed by t_D are mutually phase locked or not.

The interferograms obtained by resonant excitation and detection at the GaAs bulk exciton transition are reported in Fig. 2. The sample emission spectrum and laser spectrum are shown in the inset in Fig. 2(b). In the case of detection through a broad bandpass filter [Fig. 2(a)] we observe a fast component (due to nonresonant RS)

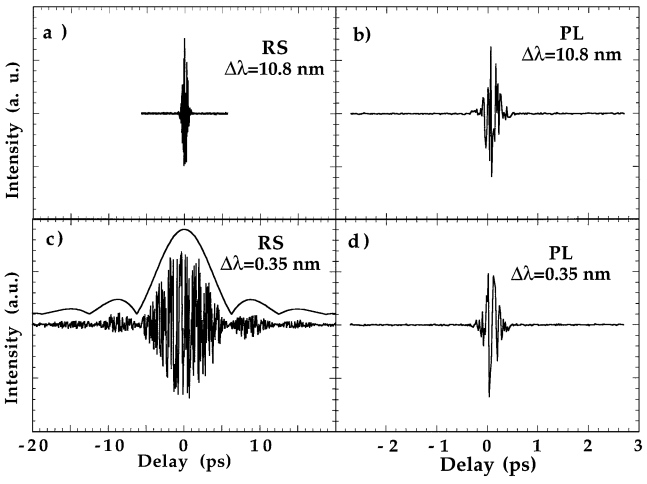

FIG. 1. Effects of spectral filtering on the RS and PL emissions; the value of filter bandpass $\Delta\lambda$ is reported in the figures. The solid line in (c) is the Fourier transform of the bandpass filter.

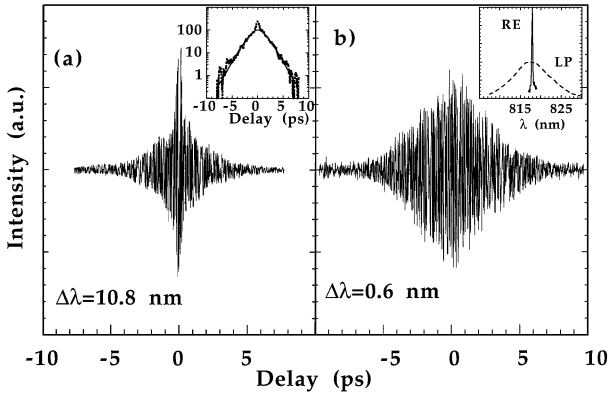

FIG. 2. Resonant excitation of the GaAs bulk structure. (a) Interferogram obtained with a very large bandpass $\Delta\lambda = 10.8$ nm. The inset shows a semilogarithmic plot of the interferogram amplitude (dotted line) together with a fit based on the numerical solution of OBE (solid line). (b) Interferogram obtained with a narrow bandpass $\Delta\lambda = 0.6$ nm detecting the polarization perpendicular to that of the excitation beam. The inset shows the resonant emission (RE, solid line) and the laser pulse (LP, dashed line) spectrum.

followed by a slower tail characterized by a nearly exponential decay, with time constant $T_2 = 1.45$ ps, as more clearly shown in the inset in Fig. 2(a) where a semilogarithmic plot of the interferogram amplitude is reported together with a fit based on the numerical solution of OBE that includes inhomogeneous broadening [10]. This kind of free induction decay of the laser induced macroscopic polarization has been recently observed also in pulse distortion experiments both in transmission [14] and reflection [15]. However, since these techniques probe the coherent dynamics, they do not give any information on the incoherent emission. Moreover, the experiment presented here has the advantage, with respect to the transmission experiments [14], to be sensitive even in opaque samples such as in the GaAs bulk case. Also note that, unlike other measurements of the polarization decay, the measured signal decays with T_2 and not $T_2/2$ in the two-pulse correlation technique, as shown in Eq. (1). The insertion of a narrow bandpass filter drastically modifies the interference pattern, showing that the detected signal is dominated by the coherent RRS emission. Similar distortions of the interference pattern, and therefore a prevalence of RRS upon PL, are obtained when detecting different wavelengths within the emission band or even when the polarization component perpendicular to the one of the excitation pulse is detected, as shown in Fig. 2(b). In fact, depolarization of the RRS emission in GaAs bulk samples has been reported recently in cw experiments as well [16].

Experiments of the same kind have been performed on the GaAs QW structure. In Fig. 3 we report the measured interferograms, for the emission perpendicular [Fig. 3(a)] and parallel [Fig. 3(b)] to the linear polarization of the excitation pulse after filtering through a narrow bandpass filter (0.08 nm). Also reported in the insets are the semilogarithmic plots of the interferogram envelope (together with a fit based on the numerical solution of OBE) and the spectra of the laser pulse and of the detected emission. Note also, in the inset in Fig. 3(b), the two peaks corresponding to the emission at the heavy-hole (HH) and light-hole (LH) exciton transitions. The interferograms obtained by filtering through a broad bandpass filter (not shown here) are, for both polarization channels, very similar to the ones shown in Fig. 3(a) and clearly show the beating of the two excitonic transitions with a period of 0.79 ps, in agreement with the HH-LH energy separation of 5.3 meV, superimposed onto a slow exponential decay with time constant $T_2 = 2$ ps.

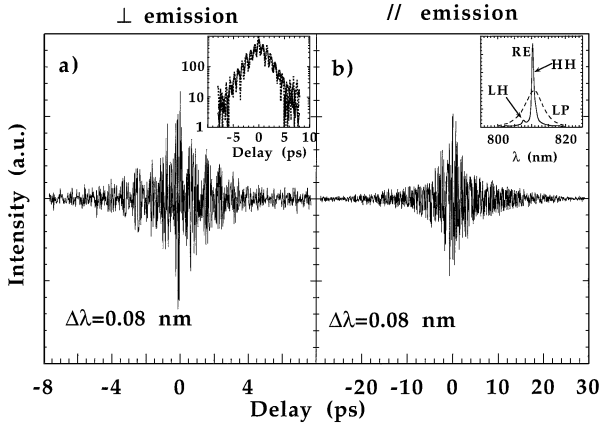

FIG. 3. Resonant excitation of the 18 nm GaAs QW and detection at the same wavelength ($\lambda = 810.4$ nm) through a narrow bandpass filter $\Delta\lambda = 0.08$ nm. (a) Interferogram obtained detecting the polarization perpendicular to that of the excitation beam. The inset shows a semilogarithmic plot of the interferogram amplitude (dotted line) together with a fit based on the numerical solution of OBE (solid line). (b) Interferogram obtained detecting the polarization parallel to that of the excitation beam; the inset shows the resonant emission (RE, solid line) and the laser pulse (LP, dashed line) spectrum.

The similarity of the interferograms obtained with and without the narrow bandpass filter points out the incoherent character of the perpendicular emission in the QW that we investigated. More complicated is the situation for the polarization conserving emission; the interferogram obtained by detecting through a narrow bandpass filter shows a two component decay of the interference envelope [Fig. 3(b)]. The fast component follows the 2 ps decay of the excitonic transition while the slow one reflects the Fourier transform of the filter line shape [10]. This result clearly shows that the emission contains both the RRS and PL components. From the fit of the interferogram one gets an estimate of a ratio of 4 to 1 of the PL with respect to the RRS intensity, when detecting at the emission peak. The ratio strongly depends on the detected energy: We find that it suddenly increases at the tails of the emission band where the PL dominates. The RRS to PL ratio of 4 to 1 has been obtained from the intensity of the time-integrated signal: A larger RRS to PL ratio is therefore expected in an experiment with real-time resolution. Note also that the slight asymmetry in the interferogram line shape shown in Fig. 3(b) is only an experimental artifact due to a small misalignment of the interferometer; in fact, a correlation experiment with identical pulses must produce symmetric signal.

Let us finally compare the findings concerning the RRS to PL intensity ratio obtained from the fs interferograms with the results under steady-state excitation. We have performed a series of cw frequency-resolved measurements on the 18 nm QW, tuning the laser excitation at steps of 0.1 meV. Details on the experimental apparatus can be found in Ref. [7]. A typical emission spectrum after near resonant cw excitation is shown in the inset in Fig. 4. The sharp peak in the spectrum has a line shape reflecting the instrumental spectral resolution and is clearly due to the Rayleigh coherent signal. The incoherent PL emission has a broadening not smaller than the inverse of the measured T_2 [11,12], which is larger than our instrumental resolution, and corresponds to the underlying broader emission band. The separation of the RRS and PL emissions is therefore extremely simple owing to their very different line shapes;

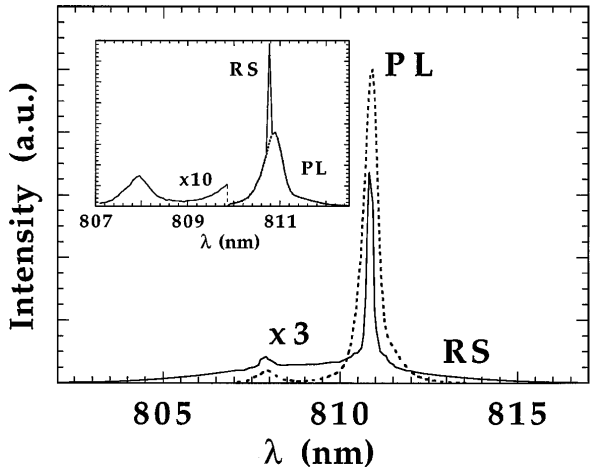

FIG. 4. Inset: emission spectrum of the 18 nm GaAs QW after resonant cw excitation at 810.8 nm. Main figure: simulation of the RS and PL emission profiles after fs excitation through the cw measurements, as described in the text; the RS profile has been multiplied by 3 for better visibility.

moreover, the nonresonant contribution to the RS signal can be easily estimated from a nonresonant experiment. In order to simulate the corresponding PL and RRS spectra after fs excitation we have added up the RRS and PL contributions at the different excitation wavelengths weighted by the pulse spectrum envelope. The obtained profiles are reported in Fig. 4; the broad pedestal below the RRS signal is due to nonresonant RS and reflects the laser pulse shape. We find that the RRS profile is sharper than the PL one and that the PL to RRS ratio at the emission peak is of the order of five. It follows that, despite the very different fs and cw excitations, the cw results agree very nicely with the findings from the fs interferometric measurements. A correct interpretation of the resonant time-resolved experiments can therefore be obtained even with a careful cw characterization, but the fs interferograms certainly provide a much more direct and unambiguous method.

In conclusion, we have shown that a two-pulse experiment allows us to point out the phase relation between the emitted fields and the ultrashort laser pulse used for excitation, thereby solving the long-standing debate on the discrimination between RRS and resonant PL. It is also worth noting that autocorrelation measurements of the detected emission in a single pulse experiment cannot discriminate between RRS and PL because of their similar first order coherence; only a cross correlation of the emission in a two-pulses experiment can point out the difference between the phase-locked RRS and the phase-random PL emissions. Examples have been given in GaAs bulk and QW structures. In addition, a quantitative estimate of the relative magnitude of the PL to RRS signals is obtained. The comparison with cw frequency resolved measurements demonstrates that the relationship between RRS and PL is similar under femtosecond and steady-state excitation, at least in the low excitation limit.

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Note added.—In a recent experiment with real-time resolution, the RRS and PL signals have been discriminated by looking at the dependence of the rise time on the exciton density [17].

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