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Optical instabilities in semiconductor quantum-well systems driven by phase-space filling

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Four-wave mixing (FWM) is one of the best known phenomena in semiconductor optics. Recent experimental results of FWM instabilities and optical switching in atomic systems have renewed the interest in FWM and possible related instabilities in semiconductors. We have recently performed theoretical investigations of FWM instabilities in a variety of semiconductor quantum well systems (single quantum wells, Bragg-spaced multiple quantum wells, and planar semiconductor micocavities) and shown that different systems require different physical processes that potentially can give rise to FWM instabilities. In this contribution, we concentrate on the simple (and largely academic) finding that phase-space filling together with spatial exciton dispersion can lead to FWM instabilities in single quantum wells.

1 Introduction and brief review The issue of optical instabilities driven by four-wave mixing (FWM) processes has been investigated for many decades (see, for example, Refs. [1–5]).

In gaseous atomic or molecular systems and simple Kerr media, FWM processes can lead, among other things, to transverse optical instabilities. The interest in these FWM instabilities has recently been renewed by the demonstration of their effectiveness for all-optical switching at very low light intensities [6, 7]. The question naturally arises whether analogs of the atomic FWM instabilities can be expected in semiconductor systems, notably in quantum well structures. And if so, what are the underlying physical mechanisms that could drive those instabilities in semiconductors.

First, we briefly review our recent work in this area, which includes single semiconductor quantum wells (QWs) [8], Bragg-spaced multiple quantum wells [9], and planar semiconductor microcavities [10–13]. Instead of studying spontaneous off-axis pattern formation induced by these instabilities, we investigated mainly their role in a pump–probe setup as illustrated in Fig. 1. We found that the FWM instabilities can lead to large gain in the probe and (background-free) FWM directions that grows exponentially with the pump pulse duration, limited by the eventual buildup of incoherent exciton/biexciton densities.

In our work on single QWs we showed, on the basis of a microscopic many-particle analysis rooted in a fermionic description of the excited semiconductor, that FWM instabilities can occur via nonlinear excitonic processes. In single QWs, we found that FWM instabilities are most likely to be driven by biexcitonic correlations [8]. However, the question is often asked whether phase-space filling (PSF) effects (which are similar to the nonlinearities found in atomic systems), can – at least in principle – lead to instabilities in single QWs. In this contribution, we address this issue and argue that PSF can indeed lead to instabilities in theory, but the parameter regime in which we find such an effect cannot be considered a realistic option for these instabilities to occur in practice. Nevertheless, the analysis of the purely theoretical PSF instability elucidates an important aspect of FWM instabilities, namely the benefits associated with a pump-induced shift (here: Stark shift) of the resonance towards the pump-light frequency. We will

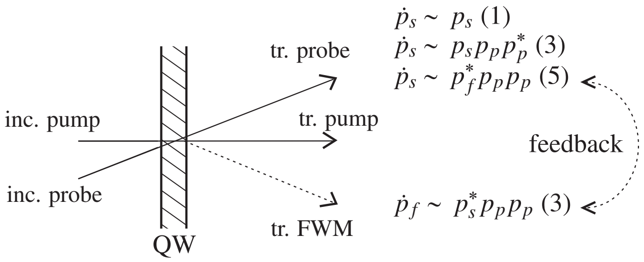

Figure 1 Illustration of the investigated pump–probe setup, including incoming and transmitted light pulses (reflected pulses are omitted for clarity) for probe, pump, and FWM direction (the angle between the different propagation directions is strongly exaggerated). Selected contributions to the probe and FWM polarizations are schematically included in the figure. The numbers in parentheses indicate the lowest order in the external fields in which they appear.

show that the Stark shift combined with spatial exciton dispersion can lead to FWM instabilities. For the biexciton-driven instabilities our earlier investigations have shown that the materials conditions for observing them are more realistic than the PSF instabilities. Admittedly, they are still rather stringent, but it appears that high-quality QW samples [14] might allow for a future experimental verification of our predictions.

We have also investigated FWM instabilities in planar semiconductor microcavities and Bragg-spaced multiple semiconductor quantum wells. Bragg-spaced multiple quantum wells are examples of resonant one-dimensional photonic crystals. Here, the exciton resonance is spectrally inside the photonic bandgap (see, for example, [15–20]). We predicted FWM instabilities in these structures [9], but in contrast to single QWs, we found that the dominant instability here is driven by Hartree–Fock (HF) mean-field Coulomb effects. The HF mean-field effect has been found earlier to dominate stimulated polariton scattering and related instabilities in planar semiconductor microcavities (see, for example, [21–26]). But two-exciton correlations beyond the HF levels have also been found to be important, and in our work [10, 11] we focused on tracing the instabilities and corresponding polariton-polariton interactions to the underlying excitonic correlation functions (T-matrices), which, in turn, are based on a fermionic theory. Finally, we mention that we have shown that the idea of an all-optical switch driven by FWM instabilities can be realized in semiconductor microcavities [12, 13]. In the following, we will restrict ourselves to PSF-driven instabilities in single QWs.

2 Theoretical basis

In our theoretical approach to FWM instabilities, we investigate a pump–probe setup as illustrated in Fig. 1 with the light propagating in quasi-normal incidence and with all optical pulses spectrally centered near the 1s heavy-hole (hh) exciton resonance. Assuming all other resonances to be sufficiently far away, the coherent response of the system in the lowest-order nonlinear regime ($\chi^{(3)}$-regime) has been well studied. We focus our analysis on small pump intensities where the many-particle effects listed above are dominant for the coherent optical QW response (i.e., we neglect higher than four-fermion or two-exciton correlations). The influence of higher-order correlations and incoherent exciton contributions is discussed later in our analysis. We start from the nonlinear equation for the optically induced interband polarization $p^{\pm}$ (+, − label the circular polarization states) and perform a spatial Fourier decomposition of $p^{\pm}$ and the exciting field $E^{\pm}$ with respect to the in-plane wave vector $\boldsymbol{k}$. We label the Fourier components with the subscripts s, f and p for probe (also called signal, with $k = k_s$, assumed to be small but nonzero), background-free FWM ($k = -k_s$), and pump ($k = 0$) direction, respectively. The resulting equations are linearized in the weak probe field $E^{\pm}_s$ but solved self-consistently in the pump field $E^{\pm}_p$ [27, 28]. The equations for $p^{\pm}_{s,f}$ read:

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} p^{\pm}_{s,f} &= (\varepsilon_x - i\gamma) p^{\pm}_{s,f} \\ &\quad - [\varphi^*_{1s}(0) - 2A^{PSF}|p^{\pm}_p|^2]\, d_{cv} E^{\pm}_{s,f} \\ &\quad + 2 d_{cv} A^{PSF}[p^{\pm}_{s,f} p^{\pm *}_p E^{\pm}_p + p^{\pm *}_{f,s} p^{\pm}_p E^{\pm}_p] \\ &\quad + \text{Coulomb terms}\,. \end{aligned} \quad (1)$$

The "Coulomb terms" are given in Ref. [8]; they are omitted here because in the following we discuss only the PSF terms. (We note that PSF contributions and the Coulomb terms, derived here from a microscopic theory [29] can be related to simple few-level models as outlined in Ref. [30].) The nonlinear pump equation for $p^{\pm}_p$ (not shown) involves the same nonlinear processes. We note that the solution to Eq. (1) goes beyond the $\chi^{(3)}$-limit and includes the pump polarization up to arbitrary order. Here, ε_x is the 1s–hh exciton energy, γ a phenomenological excitonic dephasing constant (not including the radiative decay), d_{cv} the interband dipole matrix element, $\varphi_{1s}(\boldsymbol{r})$ the two-dimensional exciton wavefunction (in the following assumed to be real and positive), and A^{PSF} the excitonic PSF constant. The propagation of the optical field $E^{\pm}$ across the QW is described with a transfer-matrix method accounting for radiative corrections/decay and assuming the QW to be infinitely thin (e.g., Ref. [31]).

Within the framework of a linear stability analysis, we re-write the homogenous part of Eq. (1) in the form of a linear matrix equation. For steady-state pump excitation ($p_p(t) = \tilde{p}_p\, e^{-i\omega_p t}$ with $\dot{\tilde{p}}_p = 0$), we use the ansatz $p_{s,f}(t) = \tilde{p}_{s,f}(t)\, e^{-i\omega_p t}$. We then arrive at the general form

$$\frac{d}{dt} \tilde{\boldsymbol{p}}(t) = M\, \tilde{\boldsymbol{p}}(t)\,, \quad (2)$$

with the vector of polarization components[1]

$$\tilde{\boldsymbol{p}}(t) = [\tilde{p}^{+}_s(t),\ \tilde{p}^{+*}_f(t),\ \tilde{p}^{-}_s(t),\ \tilde{p}^{-*}_f(t)]^T\,, \quad (3)$$

[1] In Ref. [8], this vector also contains elements describing the contribution of bound biexcitons, which is important for the correct analysis of biexciton-driven FWM instabilities.

where the matrix M follows from Eq. (1). If at least one of the eigenvalues λ_i of M fulfills $\mathrm{Re}\{\lambda_i\} > 0$, the system is unstable. An arbitrarily small seed of $p^{\pm}_{\mathrm{s,f}}$ would grow exponentially, until the matrix M ceases to describe the system correctly.

3 Discussion Equation (1) and the corresponding matrix Eq. (2) are the basic ingredients of our recent instability studies of single quantum wells [8], Bragg-spaced quantum wells [9] and microcavities [10–12]. In the following, we illustrate the linear stability analysis of a single QW for the simple case of PSF nonlinearites, in other words we neglect the Coulomb terms in Eq. (1). The second term in the third line is the one describing the feedback between the signal and FWM polarizations. It is this feedback that is necessary for an instability to occur. In the literature, this term in the equation is often labelled 'phase-conjugate oscillation' (PCO) term. Since the PSF-PCO terms do not couple the two circular polarization components, we can restrict ourselves to, say, the "+" polarized fields, $\tilde{\boldsymbol{p}}(t) = [\tilde{p}^+_{\mathrm{s}}(t),\ \tilde{p}^{+*}_{\mathrm{f}}(t)]^T$. Neglecting also radiative decay, the matrix M reduces to

$$\hbar M = \begin{pmatrix} a & b \\ b^* & a^* \end{pmatrix}, \qquad (4)$$

with $a = -\gamma + i(\Delta - \varepsilon^{\mathrm{PSF}})$ with $\Delta = \hbar\omega_{\mathrm{p}} - \varepsilon_x$, $\varepsilon^{\mathrm{PSF}} = 2A^{\mathrm{PSF}} d_{\mathrm{cv}}\,\tilde{p}^{+*}_{\mathrm{p}}\tilde{E}^+_{\mathrm{p}}$ and $b = -i2A^{\mathrm{PSF}} d_{\mathrm{cv}}\,\tilde{p}^+_{\mathrm{p}}\tilde{E}^+_{\mathrm{p}}$. We denote the nonlinear terms in the diagonal matrix elements "self-wave mixing" (SWM) terms, as they do not couple the signal and FWM polarizations. The off-diagonal elements we call PCO terms. First, it is easy to see that, if we had only the PCO terms, we would predict instabilities for all pump parameters, as one of the two eigenvalues $\lambda_{\pm} = \pm|b|^2$ is positive real. However, the situation is quite different if we include the diagonal matrix elements in the analysis. Now, the eigenvalues are $\lambda_{\pm} = \mathrm{Re}\,(a) \pm \sqrt{|b|^2 - \mathrm{Im}^2(a)}$. Clearly, instability requires the square root to have a real part that can overcompensate the dephasing (γ) and power broadening ($\mathrm{Im}\,(\varepsilon^{\mathrm{PSF}})$) included in $\mathrm{Re}\,(a)$.

Let us first discuss a "best-case scenario" in which there is no dephasing and power broadening, i.e. $\mathrm{Re}\,(a) = 0$. We can get an analytical prediction for the instability if we treat the pump polarization stationary and in first order in the pump amplitude, $\tilde{p}^+_{\mathrm{p}} = -\varphi_{1\mathrm{s}}(0) \times d_{\mathrm{cv}}\tilde{E}^+_{\mathrm{p}}/(\Delta + i\gamma)$. Defining the exciton density $n_x \equiv |\tilde{p}_{\mathrm{p}}|^2$, we find $|b|^2 - \mathrm{Im}^2(a) = -\Delta^2(1 + 4A^{\mathrm{PSF}} n_x/\varphi_{1\mathrm{s}}(0)) < 0$. Hence, the square root is purely imaginary and there is no instability.

We can rationalize this result by noting that the Stark shift, which is part of the diagonal matrix elements a and given by $-\mathrm{Re}\,(\varepsilon^{\mathrm{PSF}})$, shifts the exciton resonance away from the light field frequency, and therefore increases the effective detuning. To confirm the analytical finding of the absence of PSF instabilities, which was based on a first-order evaluation of $\tilde{p}_{\mathrm{p}}$, we show in Fig. 2 a numerical

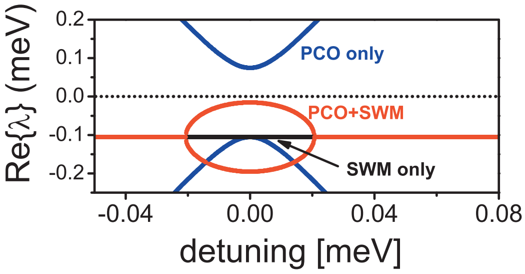

Figure 2 (online colour at: www.pss-b.com) Linear stability analysis for a circularly polarized pump for steady-state total coherent exciton density $n_x^{\mathrm{total}} = 1.7 \times 10^{11}\ \mathrm{cm}^{-2}$ and without in-plane exciton dispersion. Shown are the real parts of the eigenvalues λ_i of the matrix M vs. pump detuning. The dotted line separates the stable ($\mathrm{Re}\,\{\lambda\} < 0$) from the unstable ($\mathrm{Re}\,\{\lambda\} > 0$) regime.

solution[2] of the detuning dependence of the eigenvalues obtained with the stationary solution of the nonlinear pump equation. For all detunings, we adjust the pump intensity such that the exciton density n_x is $1.7 \times 10^{11}\ \mathrm{cm}^{-2}$. We chose this rather unrealistically high density to illustrate the point that PSF without in-plane excition dispersion does not yield instabilities (which can be seen from Fig. 2, because the real parts of all the eigenvalues are negative), whereas inclusion of spatial dispersion does yield instabilities in principle. However, as will be discussed in the following, even with spatial dispersion we had to choose the high (unrealistic) density in order to obtain positive eigenvalues.

One feature of spatial dispersion (i.e. ε_x entering the pump equation is different from that entering the signal and FWM equations) that facilitates instabilities is the fact that the Stark effect can actually shift the exciton into resonance with the light field. This is illustrated in Fig. 3. At a finite incidence angle, which is equivalent to a finite in-plane wave vector, the signal has generally a different

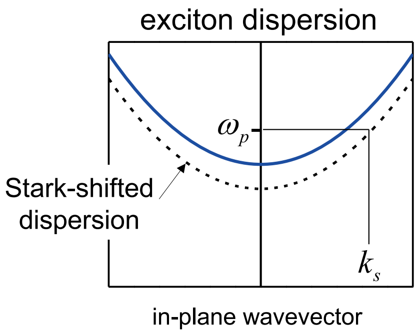

Figure 3 (online colour at: www.pss-b.com) Sketch of the in-plane exciton dispersion (solid line: unshifted, dashed line: Stark shifted). For the indicated pump frequency and in-plane signal wave vector, the Stark-shifted dispersion is in resonance with the light frequency.

[2] Parameters: $\varepsilon_x = 1.4965$ eV, $d_{cv} = 4$ eÅ, $\gamma = 0.01$ meV, $A^{\mathrm{PSF}} = 4a_0^x\sqrt{2\pi/7}$, with $a_0^x \approx 170$ Å, $E_b^x \approx 13$ meV.

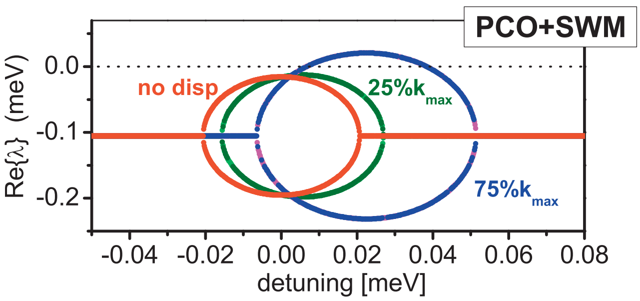

Figure 4 (online colour at: www.pss-b.com) Same as Fig. 2, but including spatial in-plane dispersion of the exciton resonance.

detuning from the exciton resonance than the normal-incidence pump. However, the Stark shifted exciton can be in resonance with the signal beam, as shown in the figure. For this case, we have performed a linear stability analysis, and the results are shown in Fig. 4. We show the case of no-dispersion as well as two different in-plane wavevectors of the signal. Here, k_{max} is the maximum in-plane vector for the given frequency. Clearly, we find a region of positive eigenvalues (i.e. instabilities) for the case of $k_s = 0.75 k_{max}$, which indicates that spatial dispersion is indeed beneficial for the PSF-driven instability. However, we stress again that this calculation is only meant to elucidate the interplay of light-induced energy shifts and FWM instabilities; it is not meant to predict PSF-driven instabilities in single QWs in actual experiments.

4 Conclusion We have briefly discussed FWM instabilties in semiconductor quantum well systems. We have restricted the detailed discussion to instabilities driven by phase-space filling effects in single QWs. We have found PSF-driven instabilities to be unrealistic and in principle possible only if spatial in-plane dispersion of excitons is taken into account. While not necessarily realistic, the PSF-driven instabilities in single QWs illustrate the benefits from optically-induced exciton shifts toward the frequency of the pump pulse.

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