PHYSICAL REVIEW B VOLUME 61, NUMBER 8 15 FEBRUARY 2000-II

Interferometric analysis of resonant Rayleigh scattering from two-dimensional excitons

S. Haacke,* S. Schaer, and B. Deveaud
Physics Department, IMO, Swiss Federal Institute of Technology Lausanne, CH-1015 Lausanne, EPFL, Switzerland

V. Savona
Physics Department, IPT, Swiss Federal Institute of Technology Lausanne, CH-1015 Lausanne, EPFL, Switzerland

(Received 17 December 1999)

We have performed spectral interferometry with Rayleigh scattered light coming from a single speckle and emitted after resonant creation of excitons in GaAs quantum wells. The experiment confirms the coherence of the secondary emission at early times, regardless of sample parameters, but underlines the stochastic nature of a single speckle. The scattered electric field is calculated, correctly accounting for the center-of-mass quantization in the disordered potential. We find that the amplitudes of the electric fields emitted by different excitons are uncorrelated, so that the well-known Poisson distribution for speckle intensities applies.

Secondary emission from resonantly excited semiconductor quantum wells is composed of resonant Rayleigh scattering (RRS) conserving temporal coherence and resonant luminescence that implies loss of phase memory.[1–7] Temporal coherence means that the emission interferes with the coherent laser source by which it was created. This effect has been used very recently to discriminate RRS from resonant luminescence and very clear interference features between the emission and femtosecond laser pulses have been demonstrated in the spectral domain.[8,9] Nevertheless, a better theoretical understanding of the effects determining the electric field emitted from an excitonic ensemble needs to be supplied. In particular, the interferometric experiment selects one single speckle and the statistical properties of the speckle emission have to be characterized. Finally, being restricted to a small solid angle of emission, the results of spectral interferometry can differ quite much from those obtained with large angle detection (up-conversion of RRS).

This paper presents a detailed experimental and theoretical analysis of the spectrally resolved interference features. The time dependence of the single speckle emission is compared with the results from speckle-averaging up-conversion experiments. A semiclassical theory fully accounting for the excitonic center-of-mass quantization in the two-dimensional disordered potential is used to calculate the scattered field of a single speckle. We show that the temporal and spectral properties of the single speckle RRS are a result of interference of many excitonic oscillators with uncorrelated random amplitudes, which do not necessarily reflect the underlying disorder situation. As a consequence, only speckle averages convey meaningful information that can be related to statistical properties of the disordered potential.

The interferometric data are obtained in a Mach-Zehnder-type interferometer (see Fig. 1), actively stabilized to a path difference $\leqslant \lambda/10$ by monitoring the interference fringes of a HeNe laser (not shown). 120 fs pulses produced by a Ti:sapphire laser excite the sample and probe the temporal coherence of the secondary emission (SE). The sum of both linearly copolarized electric fields produces interference features on a liquid-nitrogen-cooled charge-coupled device (CCD) after passing through a spectrometer with a focal length of 1 m (resolution 0.7 Å). The finite spectral resolution is improved with respect to previous experiments,[8,9] and allows for a time window of 25 ps in the time-resolved data obtained by Fourier transformation. The delay between the reference beam and the SE can be varied over several picoseconds with a resolution of the path difference better than $\lambda/10$.

We have studied several high-quality GaAs multiple quantum wells (QW's) grown by molecular beam epitaxy with well widths between 18.0 and 9.0 nm, and different inhomogeneous linewidths (0.65–2.0 meV). The samples are held at $T=18$ K in a cold finger cryostat, the excitation density is kept low ($<10^9$ cm^{-2}) and the laser spectrum is tuned to low energies so as to allow for long optical dephasing times.[3] The excitation spot diameter is ≈ 200 μm, resulting in an average solid angle of a single speckle of 1.7 mrad between the sample surface and the SE collection optics. The qualitative trends that we are discussing below are independent of experimental details such as the angle of excitation or of SE collection. We have only taken special care to produce a magnified image of the speckle pattern on the

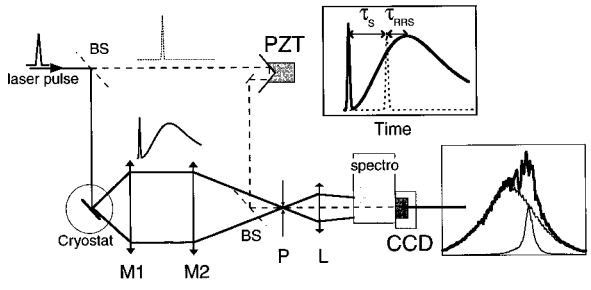

FIG. 1. Schematic of the interferometric experiment. The electric fields of the reference pulse (dashed line) and of some speckles propagate colinearly after the pinhole *P*. The pinhole is used for alignment purposes only. *L*: $f=100$ mm lens; $M1$,$M2$: collection and focusing optics; BS: beamsplitter; PZT: piezoelectric transducer. Upper inset: Timing of reference (dashed) and scattered signals (thick line). Lower inset: Spectrally resolved total intensity $I(\omega,\tau)$ (thick line) and the intensities of I_L and I_{SE} measured individually (thin lines).

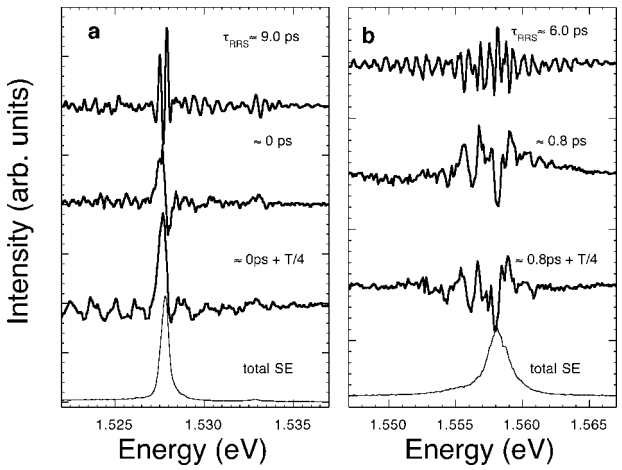

FIG. 2. Spectrally resolved interference $S(\omega)$ recorded for different delays τ_{RRS} and the time-integrated SE (bottom trace). Two GaAs QW structures with different inhomogeneous linewidths Δ are compared [(a) $\Delta = 0.65$ meV, (b) $\Delta = 2.0$ meV].

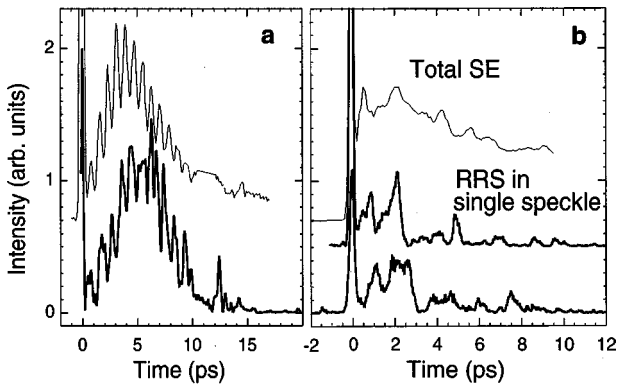

FIG. 3. Time-resolved RRS intensity emitted in a single speckle (thick line) and SE intensity measured in large angle detection, averaging over many speckles (thin line). All spectra are taken at 18 K and low excitation densities. Samples as in Fig. 2. The 18 nm QW's exhibit quantum beats between heavy-hole- and light-hole-excitons (a). The 10 nm QW's show residual fluctuations in the up-conversion data due to incomplete speckle averaging. The results of two different speckles are reported in (b).

CCD and have chosen the focal length and position of lens L accordingly. In this way, we are able to spatially resolve the single speckle emission on the CCD. With the spectrometer set at zero order, one can check that a single speckle corresponds in average to 4×4 pixels on the detector. The beam diameter of the reference laser can be magnified such as to cover several speckles on the CCD, which can all be analyzed by a single exposure. A single speckle is selected through the narrow entrance slit and by selective read-out and binning of 2–3 horizontal lines (spectra) on the CCD. We have thus been able to check that interference features recorded simultaneously from different speckles show different phases and different amplitudes. Finally, one single speckle is selected for the data shown below.

The signal measured by spectral interferometry is a sum of the laser intensity I_L, of the total SE intensity I_{SE}, and of $S(\omega)$. The latter is the term of interference between the reference field E_L and the RRS field E_{RRS}, which depends on the delay τ_{RRS} defined in Fig. 1.[10]

$$I(\omega,\tau_{\mathrm{RRS}}) = I_L(\omega) + I_{\mathrm{SE}}(\omega) + S(\omega,\tau_{\mathrm{RRS}}),$$

$$S(\omega,\tau_{\mathrm{RRS}}) = 2\,\mathrm{Re}\{E_L^*(\omega)E_{\mathrm{RRS}}(\omega)e^{i\omega\tau_{\mathrm{RRS}}}\}. \qquad (1)$$

The first two terms in Eq. (1) are measured separately and subtracted from the total signal $I(\omega)$ so that one remains with $S(\omega)$. For some spectra, we adjust τ_{RRS} to be almost zero, so that the factor $e^{i\omega\tau_{\mathrm{RRS}}}$ is also constant within the small exciton linewidth Δ. The spectral dependence of $S(\omega,\tau)$ is then directly reflecting $E_{\mathrm{RRS}}(\omega)$, since E_L is constant over Δ. This allows us to assess very directly the ''regularity'' of $E_{\mathrm{RRS}}(\omega)$ without any spectral modulation that would be introduced by $e^{i\omega\tau_{\mathrm{RRS}}}$.

$S(\omega,\tau_{\mathrm{RRS}})$ is displayed in Fig. 2 for two samples and different delays τ_{RRS}. The samples have different well widths: 18 nm [Fig. 2(a)] and 10 nm [Fig. 2(b)], and different inhomogeneous linewidths Δ (0.65 meV and 2.0 meV, respectively). For both samples, we observe pronounced interference features in the windows of excitonic emission at 1.5270–1.5285 eV and 1.554–1.561 eV, respectively. The weak periodic signal at other energies is due to interference between laser light scattered at the sample surface (SSL) and the reference beam delayed by τ_S defined in Fig. 1. The interference features observed at the exciton resonance are unambiguously due to interference between the excitonic RRS and the reference laser. Interference between SSL and RRS is excluded as these features would not depend on τ_{RRS} and would be much weaker. Assuming that 100% of SSL interferes with the reference beam, calculations show that 30–50 % of the total SE is temporally coherent RRS.[9] The exact value depends, of course, on the particular speckle that is measured, but in average seems to be sample independent for the set of samples investigated. A sample dependence is only expected for smaller Δ.[11]

Let us now concentrate on the temporal dependence of the electric field $|E_{\mathrm{RRS}}(t)|$, which is obtained as a convolution with the reference pulse by Fourier transformation of $S(\omega)$.[10] The corresponding intensity $I_{\mathrm{RRS}}(t)$ is displayed for both samples in Fig. 3. The zero of the time axis is set by hand to coincide with the maximum of the peak due to SSL. In agreement with the smaller Δ, $I_{\mathrm{RRS}}(t)$ measured for the 18 nm QW's reaches its maximum later than for the 10 nm QW's. This is also expected from the $I_{\mathrm{SE}}(t)$ measured by luminescence up-conversion with large angle detection (averaging over many speckles),[3] which is given for comparison in Fig. 3. The single speckle data confirm qualitatively that RRS dominates in the first picoseconds after excitation. Figure 3(b) shows strong temporal fluctuations similar to those observed in Ref. 12, however over a shorter time domain here. For another speckle direction, reported in Fig. 3(b), we observe a similar duration of the temporal fluctuations, but they occur at different delay times. The 18 nm QW's exhibit a broader ''temporal speckle'' than the 10 nm structure, as expected from the smaller inhomogeneous broadening.[12] If one was able to average the temporal response of many angular speckles one could isolate the RRS contribution in the total transient SE. So far, the comparison between up-conversion and *single* speckle data remains qualitative.

The temporal behavior has its counterpart in the spectrally resolved data $E_{\mathrm{RRS}}(\omega)$, which is also rather different for the

two samples. The spectra $E_{\mathrm{RRS}}(\omega)$ of the 18 nm QW sample are very regular, and an analysis of the data with $\tau_{\mathrm{RRS}} \approx 0$ shows that they represent the real and imaginary part of a single Lorentzian, convoluted by a Gaussian [Fig. 2(a)]. On the other hand, the temporal fluctuations of the electric field observed for the 10 nm structure imply that, according to Eq. (1), the spectrum $S(\omega)$ is a sum of interferences between E_L and many temporal speckles occurring at different delay times τ_{RRS}. These different interferences give rise to various periods for the spectral oscillations, and an irregular structure arises in the spectral domain.

What can we learn from the comparison with the up-conversion data? The initial part of the $I_{\mathrm{SE}}(t)$, dominated by RRS, has decayed to less than 20% of its maximum at $t=10$ ps for the 18 nm structure. It is quite probable that for other speckle directions $|E_{\mathrm{RRS}}(t)|$ also falls within that envelope defined by the up-conversion data. In other words, if, for some speckle, a second temporal maximum appears in $|E_{\mathrm{RRS}}(t)|$ it would be much weaker than the first one. That is why the regular shape of E_{RRS} observed both in time and energy domain for the 18 nm structure is quite likely to be observed also for other speckle directions. For the 10 nm structure, $I_{\mathrm{SE}}(t)$ is only slowly decaying and allows for the observation of many fluctuations of $|E_{\mathrm{RRS}}(t)|$. It has been shown theoretically[13,14] that the speckle-averaged $I_{\mathrm{RRS}}(t)$ is determined by the statistical properties of the disorder potential. We would therefore expect a relation between the regularity of $E_{\mathrm{RRS}}(\omega)$ and the disorder potential, but this requires a statistical analysis of a large number of speckles.

In order to back up the previous discussion of the experimental observations and characterize the excitonic electric field more thoroughly, we have performed calculations for $E_{\mathrm{RRS}}(\omega)$ simulating the emission in a single speckle. We introduce the well-established ''rigid'' exciton approximation,[15,16] namely we assume that disorder affects only the exciton center-of-mass motion. Then, within the simplifying assumptions of an excitation pulse with plane wave geometry, a deltalike time dependence and a far-field detection scheme, the scattered electric field turns out to be proportional to the propagator of the exciton center-of-mass motion along the plane:[17] $E_{\mathrm{RRS}}(\mathbf{k}_{\mathrm{out}},t) \propto D(\mathbf{k}_{\mathrm{in}},\mathbf{k}_{\mathrm{out}},t)$. For the numerical calculations, we have solved the Dyson's equation for this propagator in real space (with respect to the $\mathbf{k}_{\mathrm{out}}$ variable) and frequency domain, for a fixed value of $\mathbf{k}_{\mathrm{in}}$:

$$\left(-\frac{\hbar^2\nabla^2}{2M} + \hbar\omega_0 - \hbar\omega + V(\mathbf{R}) \right) D(\mathbf{k}_{\mathrm{in}},\mathbf{R},\omega) = e^{i\mathbf{k}_{\mathrm{in}}\cdot\mathbf{R}}, \tag{2}$$

where ω_0 is the center $1s$ exciton frequency. The static potential $V(\mathbf{R})$ is the effective disordered potential acting on the exciton center of mass. For the present calculations, the Gauss distributed $V(\mathbf{R})$ is randomly generated obeying the statistical constraint $\langle V(\mathbf{R})V(\mathbf{R}')\rangle = \sigma^2 \exp[-(\mathbf{R}-\mathbf{R}')^2/2\xi^2]$, where σ is the Gauss energy width and ξ is the correlation length in space. The exciton propagator can be equally expressed in terms of the exciton center-of-mass eigenfunctions and eigenvalues derived from the Schrödinger equation $[-\hbar^2\nabla^2/2M + V(\mathbf{R})]\phi_n(\mathbf{R}) = \hbar\omega_n\phi_n(\mathbf{R})$. The electric field measured from a single speckle in direction $\mathbf{k}_{\mathrm{out}}$ is then given by

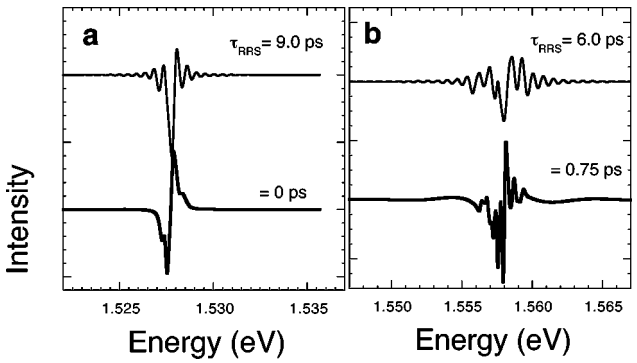

FIG. 4. Calculated spectral interference $S(\omega)$ using adequate parameters to simulate the 18 nm (a) and 10 nm (b) wide QW samples studied. The spectra are convoluted with the experimental spectral resolution. Note the good qualitative agreement with Fig. 2.

$$E_{\mathrm{RRS}}(\mathbf{k}_{\mathrm{out}},t) \propto \sum_n M_n(\mathbf{k}_{\mathrm{in}}) M_n^*(\mathbf{k}_{\mathrm{out}}) e^{-i\omega_n t - \Gamma_h t}, \tag{3}$$

where $M_n(\mathbf{k}) = \int d\mathbf{R}\phi_n(\mathbf{R})\exp(i\mathbf{k}\cdot\mathbf{R})$ is the overlap between exciton center-of-mass eigenfunctions ϕ_n and the electric field in the direction $\mathbf{k}$. $\hbar\Gamma_h$ is the homogeneous broadening. Equation (3) has the form of a sum of electric fields emitted by different oscillators with the individual amplitudes A defined by the product $\mathrm{M}_n(\mathbf{k}_{\mathrm{in}})\mathrm{M}_n^*(\mathbf{k}_{\mathrm{out}})$. It is important to note that the sum runs over all excitons created in the excitation spot and in the different wells of the multiple QW structure. It does not average to a finite value if $n \to \infty$ (not self-averaging[18]) so that the signal fluctuates strongly in time and frequency as we observe it here. Theories that make use of ensemble averages[13,14,17] apply only for large solid angle detection and are inadequate for the signals measured from a single speckle. As the single speckle is only one particular stochastic realization of the sum (3), the temporal and spectral properties cannot be used to verify general models, for instance, concerning the type of spatial correlation of the disordered potential.

The numerical solution of Eq. (2) is computed on a discretized two-dimensional square region of 2 μm of side using a mesh of 256×256 points. The result is then Fourier transformed back into the wave vector domain. All the computed results that follow are obtained for an incident normal beam and an out-scattering direction of 60° in air. Moreover, they always correspond to a single discrete $\mathbf{k}_{\mathrm{out}}$ value, and no configuration average is performed. Figure 4 displays the spectral and temporal results for $\xi = 40$ nm, a typical value for the samples investigated,[9] $\hbar\Gamma_h = 70$ μeV, and for two different σ. As the precise value of the phases critically depend on the speckle, we do not expect to obtain a quantitative agreement between calculations and experiment, but we find good qualitative agreement with the measured spectra: a regular spectrum for the sample with low σ and more pronounced phase fluctuations for larger σ. It should be noted that, in contrast to the conclusions of Ref. 8, the 18 nm MQW shows a very regular $E_{\mathrm{RRS}}(\omega)$ even though ξ is much smaller than the optical wavelength λ.

Fourier transforming Eq. (3) one gets the expression for the spectrally resolved electric field

$$E_{\mathrm{RRS}}(\mathbf{k}_{\mathrm{out}},\omega) \propto \sum_n \frac{M_n(\mathbf{k}_{\mathrm{in}})M_n^*(\mathbf{k}_{\mathrm{out}})}{\omega-\omega_n+i\Gamma_h}. \quad (4)$$

In analogy to the classical formalism of speckle formation,[19] $E_{\mathrm{RRS}}(\mathbf{k}_{\mathrm{out}},\omega)$ is a sum of Lorentzians with complex amplitudes $M_n(\mathbf{k}_{\mathrm{in}})M_n^*(\mathbf{k}_{\mathrm{out}})$. One may ask whether, due to the spatial correlation in the disordered potential, the amplitudes of different excitonic emitters might be correlated. We have computed the center-of-mass wave functions ϕ_n in a one-dimensional (1D) disordered potential of 1 μm length, with the same σ and ξ as used above. These calculations show that the amplitudes of excitons emitting in the same narrow spectral window $\hbar\omega \ldots \hbar\omega + d(\hbar\omega)$ are uncorrelated, since the wave functions are localized at different regions of the disordered potential, which are in average more than ξ apart from each other. Eigenstates that fall in the same localization site have a larger difference in their respective energies. These general considerations are also valid for a 2D disordered potential. Equation (4) is thus a sum over random complex numbers, such that the intensity I_{RRS} in a narrow energy interval $d(\hbar\omega)$ ($\approx$ spectral resolution) is stochastic. It obeys therefore the well-known Poisson distribution[19] $p(I_{\mathrm{RRS}}) = 1/\langle I_{\mathrm{RRS}}\rangle \exp(-I_{\mathrm{RRS}}/\langle I_{\mathrm{RRS}}\rangle)$, where $\langle I_{\mathrm{RRS}}\rangle = N_{\mathrm{RRS}}\langle \epsilon^2\rangle$ with $\langle \epsilon^2\rangle$ being the mean intensity of the individual oscillators emitting in $d(\hbar\omega)$.

The ratio of coherently versus incoherently emitting excitons $N_{\mathrm{RRS}}/N_{\mathrm{PL}}$ is an interesting parameter, but the previous discussion shows that it cannot be inferred from the visibility $I_{\mathrm{RRS}}/I_{\mathrm{tot}}$ of the interference features in a single speckle. The visibility measured for the different speckles does not exceed 50% which gives, however, an upper limit of the order of one for $N_{\mathrm{RRS}}/N_{\mathrm{PL}}$. Again, only an analysis of many speckles would allow to trace the distribution $p(I_{\mathrm{RRS}})$ and thus to determine $\langle I_{\mathrm{RRS}}\rangle$.

In conclusion, we have reported an experimental and theoretical investigation of the spectrally resolved electric field emitted by an ensemble of two-dimensional excitons after resonant femtosecond excitation. The results confirm the temporal coherence of the early secondary emission. We have discussed in detail the origin of the speckle formation due to interference between numerous exciton states, and explained qualitative aspects (degree of fluctuations) of the spectral and temporal dependence of the electric field emitted in a single speckle. The comparison with speckle-averaged SE transients obtained under very similar experimental parameters demonstrate that a statistical analysis of many speckles is required in order to use $E_{\mathrm{RRS}}(\omega)$ as a source for more general information (e.g., temporal dependence of the RRS field, mean exciton localization length).

The authors thank R. Zimmermann, G.R. Hayes, and P. Selbmann for helpful discussions. Contacts with M. Joffre have also been very useful and are acknowledged. This work was financially supported by the Swiss National Priority Program for Optics.

*Present address: IPMC, University of Lausanne, CH-1015 Lausanne, Switzerland.
Electronic address:stefan.haacke@ipmc.unil.ch.

[1] J. Hegarty *et al.*, Phys. Rev. Lett. **49**, 930 (1982).
[2] H. Stolz *et al.*, Phys. Rev. B **47**, 9669 (1993).
[3] S. Haacke *et al.*, Phys. Rev. Lett. **78**, 2228 (1997).
[4] M. Gurioli *et al.*, Phys. Rev. Lett. **78**, 3205 (1997).
[5] X. Marie *et al.*, Phys. Rev. Lett. **79**, 3222 (1997).
[6] M. Woerner and J. Shah, Phys. Rev. Lett. **81**, 4208 (1998).
[7] G. R. Hayes *et al.*, Phys. Rev. B **58**, R10 175 (1998).
[8] D. Birkedal and J. Shah, Phys. Rev. Lett. **81**, 2372 (1998).
[9] S. Haacke *et al.*, in *Proceedings of the 24th International Conference on the Physics of Semiconductors*, edited by D. Gershoni (World Scientific, Singapore, 1999), p. 81.
[10] L. Lepetit *et al.*, J. Opt. Soc. Am. B **12**, 2467 (1995).
[11] M. Colocci (private communication).
[12] W. Langbein *et al.*, Phys. Rev. Lett. **82**, 1040 (1999).
[13] R. Zimmermann, Nuovo Cimento D **17**, 1801 (1995).
[14] V. Savona and R. Zimmermann, Phys. Rev. B **60**, 4928 (1999).
[15] R. Zimmermann, Phys. Status Solidi B **173**, 129 (1992).
[16] S. Glutsch and F. Bechstedt, Phys. Rev. B **50**, 7733 (1994); S. Glutsch, D. S. Chemla, and F. Bechstedt, *ibid.* **54**, 11 592 (1996).
[17] D. S. Citrin, Phys. Rev. B **54**, 14 572 (1996).
[18] R. Loudon, *The Quantum Theory of Light* (Oxford Science Publications, Oxford, 1983).
[19] J. W. Goodman, in *Laser Speckles*, edited by J. D. Dainty, Topics in Applied Physics Vol. 9 (Springer, Heidelberg, 1975).