Monte Carlo Simulations of Ultrafast Resonant Rayleigh Scattering from Quantum Well Excitons: Beyond Ensemble Averaging

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(Received 23 February 1999)

We develop and experimentally verify novel Monte Carlo simulations of ultrafast resonant Rayleigh scattering from quantum well excitons. In contrast to existing theories, these simulations can study the dynamics and spectrum of resonant Rayleigh scattering from a single realization of disorder, and allow direct comparison to experimental data. We find excellent agreement between our experiments and simulations. Our studies demonstrate the high sensitivity of scattering dynamics to a particular realization of disorder, and provide new insights into the nature of spatial correlations of excitons.

PACS numbers: 71.35.Cc, 42.25.Kb, 78.47.+p, 78.66.–w

Recent experimental progress has provided substantial insight into the nature of secondary emission following ultrafast resonant excitation of quantum well (QW) excitons [1–6]. In particular, interferometric techniques have clearly distinguished two components of the secondary emission: incoherent photoluminescence (PL) and coherent resonant Rayleigh scattering (RRS) [3–5]. This progress has underscored the need for a theoretical approach that can analyze and extract information from the newly available experimental data—especially from the RRS component preserving the phase memory of the excitation. Existing theories have established the origin of RRS in the disorder-induced spatially fluctuating resonance frequency and explained basic features of the RRS dynamics using ensemble-averaging techniques [7–10]. However, a direct comparison of the RRS dynamics, obtained using ultrafast spectral interferometry [4], showed large discrepancies and seriously questioned the applicability of these theories. Especially, it is not clear whether theories based on ensemble averaging can be used to describe ultrafast experiments probing only a single realization of disorder. A theory that allows a quantitative comparison with experiment by properly taking the experimental situation into account could provide new information on the role of disorder and spatial correlation of excitons on the RRS dynamics.

In this Letter, we develop and experimentally verify a novel Monte Carlo treatment that enables us to calculate the RRS dynamics and spectrum from a single realization of disorder with prescribed statistical properties. Our Monte Carlo simulations show that the dynamics of RRS can vary strongly with different realizations of disorder. Our experimental results show that there are indeed strong variations in the observed dynamics from different regions of the sample with the same nominal optical properties. This sensitivity of the RRS dynamics to a particular realization of disorder clearly demonstrates the importance of our approach of going beyond ensemble averaging. We show that our simulations are able to model our experiments numerically with remarkable accuracy, and their direct comparison shows an unprecedented quantitative agreement for both dynamics and spectrum of the RRS. We show that the agreement depends on the nature of disorder correlations, and that an exponential correlation function provides better agreement than a Gaussian correlation function. We further show both theoretically and experimentally that the disorder-dependent RRS dynamics is accompanied by shifts in the RRS spectrum. These spectral shifts have not been reported previously, and are related to the Wolf effect [11]. Finally, we extend the previous analytical theory to determine the magnitude of the differences in the RRS dynamics obtained from different realizations, and study the dependence of the decay of the RRS signal on the nature of correlations. These studies thus provide important new insight into the nature of resonant Rayleigh scattering from semiconductors.

We begin our discussion by constructing scattering Monte Carlo simulations, which are intended to theoretically model the ultrafast experiments studying the RRS dynamics and spectrum from a single realization of disorder in QW. The starting point of our analysis is the far-zone expression for the electric field, scattered after a resonant impulsive excitation of QW excitons [8],

$$\mathbf{E_s}(\mathbf{r},t) = \alpha \hat{\mathbf{e}}_\mathbf{s} \frac{\cos\vartheta}{r} \theta(\tau) \int_S d^2 r'_\parallel \, e^{i\mathbf{q}_\parallel \cdot \mathbf{r}'_\parallel - i\Omega(\mathbf{r}'_\parallel)\tau - \Gamma\tau/2}, \tag{1}$$

where the scattering is caused by disorder-induced spatial fluctuations of the local resonance frequency $\Omega(\mathbf{r}_\parallel) = \Omega_R + \tilde{\Omega}(\mathbf{r}_\parallel)$ around its mean value Ω_R. In Eq. (1) t is time; $\mathbf{r} = (x,y,z)$; $\mathbf{r}_\parallel = (x,y)$; $\tau = t - (r/c)$; $\theta(\tau)$ is the Heaviside step function; S is the excitation area; $\hat{\mathbf{e}}_\mathbf{s}$

is the polarization vector; ϑ is the angle between the unit vectors $\hat{\mathbf{n}}_\mathbf{i}$ and $\hat{\mathbf{n}}_\mathbf{s}$, defining the directions of propagation for the incident and the scattered waves, respectively; $\mathbf{q}_\parallel$ is the projection of the transferred momentum $(\Omega_R/c)(\hat{\mathbf{n}}_\mathbf{s} - \hat{\mathbf{n}}_\mathbf{i})$ on the xy plane; Γ is the homogeneous width of the resonance; and α is a position- and time-independent coefficient, proportional to the amplitude of the incident pulse. We model the fluctuating part $\tilde{\Omega}(\mathbf{r}_\parallel)$ of the resonant frequency by a random, zero-mean, stationary Gaussian process defined by the property

$$\langle \tilde{\Omega}(\mathbf{r}_\parallel)\tilde{\Omega}(\mathbf{r}'_\parallel)\rangle = (\Delta\Omega)^2 W(|\mathbf{r}_\parallel - \mathbf{r}'_\parallel|), \tag{2}$$

where $\langle \cdots \rangle$ denotes the average over the ensemble of realizations of disorder, $\Delta\Omega$ is the rms deviation of $\Omega(\mathbf{r}_\parallel)$ from its mean value Ω_R, and $W(r_\parallel)$ is the correlation function. In calculations we assume that

$$W(r_\parallel) = \exp(-r_\parallel^n/a^n), \tag{3}$$

where n is equal to 1 or 2, and a denotes the exciton correlation length. The Gaussian form ($n = 2$) has been previously employed in the theoretical studies of the RRS [9], while the exponential form ($n = 1$) has not been used in such studies, although this type of correlation is not unknown in QW structures [12].

At each Monte Carlo step we calculate the RRS intensity by generating *a single realization* of $\Omega(\mathbf{r}_\parallel)$ on a fine mesh of points within the excitation area S with standard numerical techniques [13] and then evaluate the integral in Eq. (1) by approximating it with a finite sum. This step models the experiment performed on a given finite excitation area. Note that we have a rare case when the computer can model reality extremely well, since the essential details of a typical excitation area 40 μm $\times$ 40 μm can be easily reproduced with 512×512 mesh used in our calculations. To account for RRS from heavy-hole and light-hole excitons, we refine our model by writing the scattered field (1) as a sum of two correlated integral contributions [9] instead of one. For brevity of the subsequent presentation, the parameters Ω_R, $\Delta\Omega$, a, and Γ will refer to the heavy-hole exciton, since this resonance dominates the RRS dynamics. The parameters characterizing the light-hole exciton resonance and the cross-correlation term will be adjusted to reproduce the observed heavy-hole light-hole beats in the RRS dynamics. To independently check the consistency of our simulations with the analytical models [8,9], we calculated the RRS intensity for 500 realizations of disorder and obtained the ensemble-averaged RRS intensity for several sets of input parameters (Ω_R, $\Delta\Omega$, a, and Γ) and found it to be identical to the analytical result in all cases.

We first use our new theoretical treatment to demonstrate that different realizations of disorder produce different RRS dynamics. Figures 1a–1d clearly show this difference of time-resolved RRS intensities obtained by the simulations for a randomly generated sequence of four disorder realizations described by *identical* input parameters: Gaussian correlation function, $\hbar\Omega_R = 1.5376$ eV, $\hbar\Delta\Omega = 0.4$ meV, $a = 1.0$ μm, $\Gamma^{-1} = 50$ ps, $S = 1600$ μm^2, normal incidence, and $\vartheta = 3°$. The curves in Figs. 1a–1d deviate significantly from each other, and from the ensemble-averaged curve (not shown). Large fluctuations are present in both magnitude of the RRS intensity and the position of its maximum.

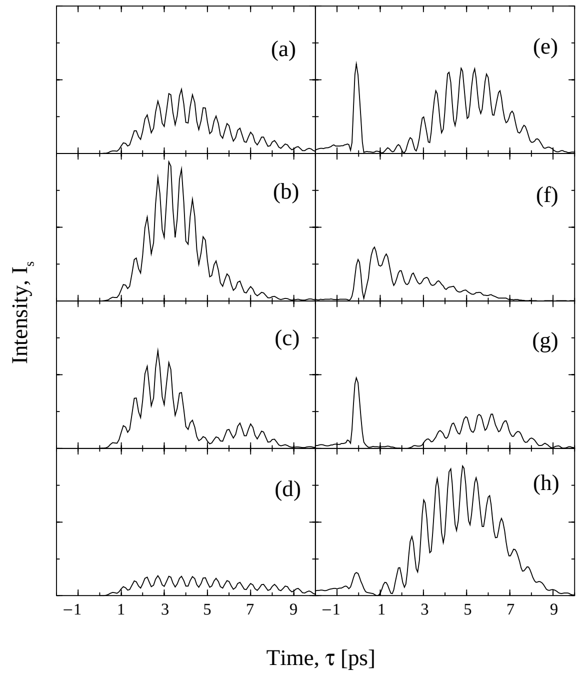

FIG. 1. Time-resolved RRS intensities: [(a)–(d)] simulations based on a random sequence of disorder realizations and [(e)–(h)] experiments on different sample positions of nominally identical optical properties. The scale of intensity is consistent in (a) through (d) and in (e) through (h). The figure illustrates large differences in the RRS dynamics for different realizations of disorder. Note that the theoretical curves *are not* fits to the experiments.

We confirm the existence of such disorder-induced fluctuations with a series of experiments on a multiple quantum well (MQW) structure using ultrafast spectral interferometry. This heterodyne technique allows us to isolate and time resolve the coherent RRS component from the incoherent PL (for further experimental details see Ref. [4]). The exciton density used in the experiments was 2×10^9 cm^{-2}, the temperature was held at 10 K, and the center wavelength of the laser was resonant with the heavy-hole exciton. The time-resolved RRS intensities obtained from four different spots of nominally identical properties on a 13-nm $GaAs/Al_{0.3}Ga_{0.7}As$ MQW structure are shown in Figs. 1e–1h. The peak at $\tau = 0$ appears due to nonresonant surface scattering. This component can be clearly distinguished from the RRS in both time and frequency domains as shown in Ref. [4] and will not be considered further. The comparisons of theoretical and experimental data show significant similarities—delayed rise, beats due to simultaneous excitation of heavy-hole and light-hole excitons [4,5,9], and, most important, the presence of large variations in the RRS dynamics for different disorder realizations. Thus, our results demonstrate that the RRS dynamics is highly sensitive to the particular realization of disorder and that previously employed comparison of a single-realization RRS intensity to the theoretical ensemble-averaged result has to be abandoned.

However, to demonstrate the ability of the Monte Carlo simulations to model the experimental situation and to extract information on the nature of the QW disorder, a quantitative comparison of theory and experiment must be made. Since it is futile to obtain a complete ensemble of experimental RRS intensities (some realizations produce such a weak RRS signal that it cannot be resolved experimentally—see, e.g., Fig. 1d for a theoretical illustration), we must compare the simulations and experiments for single realizations of disorder. We employed the following procedure to take the statistical nature of the problem into account. From a large ensemble of theoretical RRS intensities (500 disorder realizations) we picked those that gave the closest agreement with our incomplete experimental ensemble (six realizations, four of them shown in Figs. 1e–1h). The comparison was carried out for both the RRS dynamics (time domain) and spectrum (frequency domain). Thus, we used the full amplitude and phase information of the scattered field available in our theory and experiment. This procedure was performed for the Gaussian and exponential correlation functions and repeated for a range of model parameters—$\Delta\Omega$ and a with fixed $\Gamma = 0$ (the dephasing term $e^{-\Gamma\tau}$ with a typical value of $\Gamma^{-1} = 50$ ps is insignificant for the RRS dynamics developing on the time scale of 10 ps)—to optimize the agreement with the experimental data.

Our calculations showed that for both correlation functions $\Delta\Omega$ primarily defines the initial RRS dynamics with maximum intensity fluctuating around the value $\tau \sim (\Delta\Omega)^{-1}$ for different disorder realizations. The correlation length a determines the magnitude of these fluctuations and the overall RRS intensity, but has only minor influence on the time dependence of the RRS intensity. Therefore the value of $\Delta\Omega$ can be determined with high accuracy, whereas a cannot be determined very accurately. The values found by our optimization procedure were $\hbar\Delta\Omega = 0.34$ meV for the exponential ensemble and $\hbar\Delta\Omega = 0.32$ meV for the Gaussian ensemble, and a was found to be in the range of 0.4 to 3.0 μm for both ensembles. Our simulations showed that the form of the correlation function and not the numerical value of a plays a dominant role in determining the RRS dynamics. We observed that the exponential ensemble outperformed the Gaussian one in producing consistently better agreement with the experimental curves—for a given experimental realization frequently the 3rd or 4th best match from the exponential ensemble was still found to be better than the overall best match from the Gaussian ensemble. Our results for the RRS dynamics are well illustrated by Fig. 2, which shows theoretical curves generated using the optimized model parameters for one of the experimental realizations. The agreement between the experimental (full) and theoretical (dotted) curves is remarkable and unprecedented in the literature on the RRS. We have thus confirmed the validity of the model [Eqs. (1) and (2)] and have shown that the use of exponential correlations with optimized $\Delta\Omega$ and a yielded a full numerical reproduction of the experimental RRS dynamics and spectrum (see also Fig. 3).

It is important to realize that the fluctuations appearing in the RRS dynamics also manifest themselves in the spectral domain. This leads to a remarkable and yet previously unnoticed effect illustrated in Figs. 3a–3d, which show the RRS emission spectra in the vicinity of heavy-hole

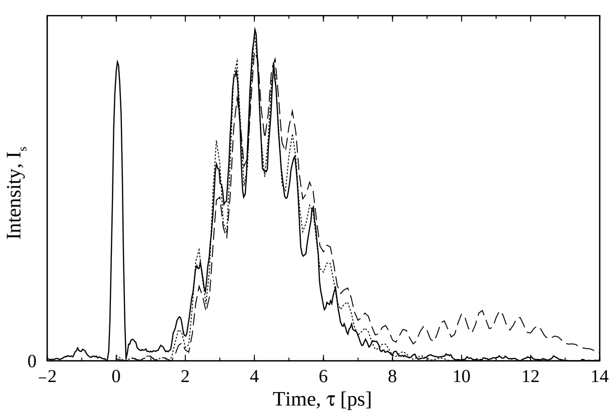

FIG. 2. Time-resolved RRS intensities for a single realization of disorder. Solid curve: experiment; dashed and dotted curves: closest matches found by simulations with Gaussian and exponential correlations, respectively.

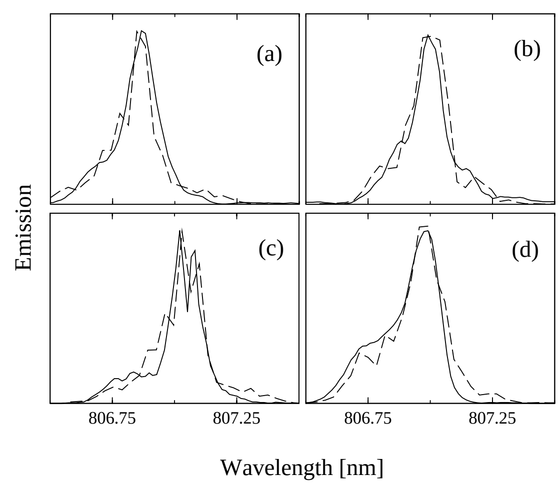

FIG. 3. Spectra of coherent RRS. Solid curves in (a)–(d) represent experimental spectra for realizations used in Figs. 1e–1h; dashed curves are numerically generated for matching theoretical realizations with exponential correlation functions.

resonance for disorder realizations used in Figs. 1e–1h (in the same order), and the spectra for the four matches, found for these realizations in our theoretical exponentially correlated ensemble of disorder realizations. We observe that the theory and experiment are consistent, as expected, and that emission spectra, corresponding to different realizations, differ from each other. These different spectral lines are formed in a complicated process of constructive and destructive interference of scattered waves, whose spectral components are correlated due to spatial correlations of excitons in QW. The spectral changes, induced by correlated disorder in the secondary source (QW, in our case), can be viewed as a form of Wolf spectral shifts predicted for the scattering of polychromatic light from systems with volume and surface correlated disorder [11].

Figures 1–3 unambiguously prove that our numerical simulations give an excellent qualitative and quantitative description of the RRS. However, our new results for disorder-dependent fluctuations and the importance of the correlation function $W(r_{\|})$ allow a simple analytical explanation. To understand the origin of the fluctuations, we have to go beyond the traditional analysis [8,9] aimed at the calculation of the average of the RRS intensity,

$$\langle I_s \rangle \propto \theta(\tau) e^{-\Gamma\tau} \int_S d^2 r'_{\|}\, e^{i\mathbf{q}_{\|}\cdot\mathbf{r}'_{\|} - (\Delta\Omega)^2 \tau^2 [1 - W(r'_{\|})]}, \qquad (4)$$

and calculate its variance. The result, somewhat cumbersome in the general case, has an especially simple form in the early-time limit ($\Delta\Omega\tau \ll 1$),

$$\langle \delta I_s(\mathbf{r},t) \rangle = [\langle I_s^2(\mathbf{r},t) \rangle - \langle I_s(\mathbf{r},t) \rangle^2]^{1/2} = \langle I_s(\mathbf{r},t) \rangle . \qquad (5)$$

Equation (5) means that the deviations of the single-realization time-resolved intensity from the ensemble average are expected to be as large as the ensemble average itself. This is consistent with the fluctuations observed in the Monte Carlo and experimental RRS intensities in Fig. 1.

We next carefully analyze Eq. (4) to understand the influence of exciton correlations on the RRS. This analysis brings a new and important result: *the nature of exciton correlations does not determine the dynamics of RRS at early times but does so at later times*. Indeed, $\langle I_s \rangle$ rises quadratically in time for $\Delta\Omega\tau \ll 1$ regardless of the form of $W(r_{\|})$—in fact, the quadratic dependence holds even for nonaveraged I_s for any given realization of disorder. However, the form of $W(r_{\|})$ plays a crucial role in the long-time limit $\Delta\Omega\tau \gg 1$, e.g., for $W(r_{\|})$ defined in Eq. (3) and $\Gamma = 0$ we obtain

$$\langle I_s \rangle \propto 1/\tau^{4/n}. \qquad (6)$$

Although the average time-resolved intensity is never reproduced in a single-realization experiment, it determines the generic behavior of dynamics for the entire ensemble. We find that the large τ behavior of the intensity in Figs. 1e–1h is a much better fit by a $1/\tau^4$ decay ($n = 1$) than a $1/\tau^2$ decay ($n = 2$). This is why our Monte Carlo simulations with exponential correlations essentially reproduced the experimental data, while the ensemble with Gaussian correlations did not yield such a good agreement.

In conclusion, we have demonstrated pronounced disorder-induced fluctuations in the RRS dynamics and spectrum in experiment as well as in theory. These fluctuations are inherent to the ultrafast resonant scattering experiment and make impossible any direct comparison of the experimental RRS dynamics with theoretical predictions obtained by ensemble averaging. We developed Monte Carlo simulations that allow direct comparison to experimental data, since by construction, these simulations model the experiment by studying a single realization of disorder at a time. An unprecedented quantitative agreement of our simulations and experiments for both RRS dynamics and spectrum shows that our theoretical approach will be valuable in understanding other RRS experimental data [3,5,6]. We also believe that our new findings on the role of exciton correlations and on disorder-induced spectral shifts in RRS show a high potential of our theoretical treatment in guiding future investigations.

We are grateful to R.S. Knox, E. Wolf, A.A. Maradudin, P.S. Carney, and V. Chernyak for useful discussions, to P. Tran for providing us the computer code for enerating 2D Gaussian random processes, and to L.N. Pfeiffer for providing us the high quality MQW sample. D.B. acknowledges financial support from the Danish Natural Sciences Research Council.

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