Anomaly in the excitation dependence of the optical gain of semiconductor quantum dots

M. Lorke,[1,*] W. W. Chow,[2] T. R. Nielsen,[1] J. Seebeck,[1] P. Gartner,[1] and F. Jahnke[1]
[1]*Institute for Theoretical Physics, University of Bremen, 28334 Bremen, Germany*
[2]*Semiconductor Materials and Device Science Department, Sandia National Laboratories, Albuquerque, New Mexico 87185-0601, USA*
(Received 2 February 2006; revised manuscript received 21 June 2006; published 26 July 2006)

Optical gain behavior of semiconductor quantum dots is studied within a quantum-kinetic theory, with carrier-carrier and carrier-phonon scattering treated using renormalized quasiparticle states. For inhomogeneously broadened samples, we found the excitation dependence of gain to be basically similar to quantum-well and bulk systems. However, for a high quality sample, our theory predicts the possibility of a decreasing peak gain with increasing carrier density. This anomaly can be attributed to the delicate balance between state filling and dephasing.

DOI: 10.1103/PhysRevB.74.035334 PACS number(s): 78.67.Hc, 71.35.Cc

I. INTRODUCTION

Semiconductor quantum dots (QDs) are currently under intense investigation because of scientific and engineering interests. They are considered as key materials for next generation optoelectronic devices. New applications emerge also in the fields of semiconductor quantum optics with fundamental studies of light-matter interaction[1–3] and for quantum information processing.[4,5] A central issue in various applications is the role of dephasing due to intrinsic interaction processes. Dephasing limits the quantum coherence and determines the homogeneous emission linewidth of QDs.

Crucial to the analysis of fundamental experiments and the realization of engineering advantages is the knowledge of intrinsic QD behavior and the understanding of underlying physics. Extracting the information experimentally is challenging because sample inhomogeneities make the separation of intrinsic from extrinsic properties difficult. Theoretical investigations have been hindered by the lack of a predictive theory. Recently, this hurdle was removed by the development of the elements for a microscopic theory that allows a rigorous treatment of dephasing. These theoretical tools enable the systematic study of fundamental QD behavior. In this paper, we demonstrate the role of intrinsic interaction processes and the resulting excitation induced dephasing for optical absorption and gain spectra. The results are not only of direct relevance for QD lasers but also for other QD applications. On a general level, the role of various interaction processes is quantified. On a more particular level, anomalies in the excitation dependent emission properties are addressed, which are not known both in higher-dimensional semiconductor structures and in atomic systems. Our investigations are performed for typical self-assembled QDs, where the Coulomb interaction of carriers and the carrier-phonon interaction leads to a coupling (i) between the discrete QD states and (ii) to a quasicontinuum of quantum well (QW) delocalized states, e.g., from a wetting layer.

Polarization dephasing directly influences the amplitude and spectral broadening of optical absorption and gain. The primary sources for dephasing are carrier-carrier and carrier-phonon scattering. These processes also contribute to carrier capture and relaxation, which are important for an efficient device operation. The Coulomb interaction leading to carrier-carrier scattering provides efficient scattering channels, especially at elevated carrier density and temperature.[6–8] On the other hand, there was concern over the interaction of carriers with longitudinal optical (LO) phonons, which provides additional scattering channels and thermalization of carrier population. Calculations based on perturbation theory predicted strongly inhibited carrier-LO-phonon scattering, because of mismatch between QD levels and LO-phonon energy.[9] This led to the belief that the QD device performance would be degraded because of a phonon bottleneck problem.[10]

Later, nonperturbative treatments within the polaron picture indicated a less serious phonon bottleneck problem,[11] consistent with many experiments, see Ref. 12. A recent quantum-kinetic treatment of carrier-phonon interaction in the polaron picture clearly demonstrates efficient carrier-LO-phonon scattering.[12] This development together with a non-Markovian treatment of polarization dephasing due to carrier-carrier scattering within a QD-QW system[13] allow the completion of a truly predictive theory for QD optical response. The resulting formalism contains a detailed accounting of electronic structure effects and a microscopically consistent treatment of the many-body interaction. The many-body effects include the Hartree-Fock energy renormalizations (band-gap shrinkage and Coulomb enhancement) which contribute to shifts and amplitude modifications of QD absorption and emission resonances, and polarization dephasing due to carrier-carrier and carrier-LO-phonon scattering, which is responsible for the intrinsic (homogeneous) broadening of these resonances and additional energy shifts. Our investigation is based on this QD optical response theory.

We begin by describing the calculation of optical absorption and gain for a sample with a uniform QD size and composition. The computed properties are customarily referred to as being associated with the homogeneously broadened or intrinsic situation. Then, we discuss the incorporation of inhomogeneously broadening effects, such as those arising from QD size and composition variations. This allows one to transform from intrinsic behaviors to what is observed in present experiments. The availability of scattering channels contributing to polarization dephasing depends on the discrete QD energy level spacings, which in turn, depend on the QD confinement potential. By considering $In_{0.3}Ga_{0.7}As$ and

InAs QDs embedded in GaAs QWs, we are able to examine the two very different scenarios of shallow and deep quantum confinement. For these two structures, the focus is on two questions, involving the homogeneously broadened gain spectrum (especially the intrinsic spectral shape and width) and the spectral changes with carrier density. A rigorous treatment of scattering is necessary for addressing these problems. The typical QD gain calculation, which uses the relaxation rate approximation, cannot tell us about the homogeneously broadened spectrum, because it assumes a line shape function and treats the spectral width as a free parameter. A central result of this paper is the discovery of an anomaly in the excitation dependence of QD gain: a decrease in peak gain with increasing carrier density. This result will directly influence QD applications under high excitation conditions. The behavior arises from a delicate balancing of state filling and dephasing, so that a detailed treatment of the carrier density dependence of scattering processes is necessary. Again, this can only be accomplished with the recent quantum-kinetic developments.

II. THEORETICAL DESCRIPTION OF QUANTUM-DOT GAIN SPECTRA

The gain calculation starts with solving for the microscopic polarization p_α, where α represents the discrete levels ν in QD transitions and the in-plane carrier momentum $\mathbf{k}$ in QW transitions. In our confinement situation it is sufficient to consider only the lowest QW subband for gain calculations. Working in the Heisenberg picture and using a many-particle Hamiltonian that includes carrier-carrier interaction, carrier-phonon interaction, and dipole interaction between electron-hole pairs and laser field, the Fourier-transformed polarization equation is[14]

$$(\hbar\omega - \varepsilon_\alpha^e - \varepsilon_\alpha^h)p_\alpha + \hbar\,\Omega_\alpha(1 - n_\alpha^e - n_\alpha^h) = iS_\alpha^{c-c}(\omega) + iS_\alpha^{c-p}(\omega). \tag{1}$$

In (1), the Rabi energy $\hbar\Omega_\alpha$ and transition energy ε_α^β $(\beta = e, h)$ contain the single-particle electronic structure properties and the many-body Hartree-Fock contributions. The complex Coulomb correlation (scattering) term $S_\alpha^{c-c}(\omega)$ is treated in the second Born approximation and non-Markovian limit[13] using self-consistently renormalized energies for the scattering partners.[14] For the carrier-phonon correlation $S_\alpha^{c-p}(\omega)$ we assume the random-phase approximation and use the Fröhlich coupling to monochromatic LO phonons to determine the QD polaron renormalization of the electronic states.[11,12] For linear optical response to a weak laser probe field $E(\omega)$ at frequency ω, the carrier distribution n_α^β is to a good approximation a Fermi-Dirac function. A typical calculation involves simultaneously solving a set of 100–150 equations, each of the form given by (1) and coupled to one another by nondiagonal terms in Ω_α, $S_\alpha^{c-c}(\omega)$and $S_\alpha^{c-p}(\omega)$. The solution is used to give the homogeneously broadened (intrinsic) optical gain of the QD-QW structure:

$$g(\omega) = -\frac{K}{\varepsilon w E(\omega)}\,\mathrm{Im}\left[N_{dot}\sum_\nu \mu_\nu p_\nu(\omega) + \frac{1}{A}\sum_{\mathbf{k}} \mu_{\mathbf{k}} p_{\mathbf{k}}(\omega)\right], \tag{2}$$

where ε is the background permittivity, K is the laser field wave vector in the medium, w is the QW width, and N_{dot} is the sheet density of QDs in a QW of area A. The spectral width calculated using (1) and (2) depends solely on dephasing processes described by the real part of the correlation contributions $S_\alpha^{c-c}(\omega)$ and $S_\alpha^{c-p}(\omega)$. Experimental data suggest that present QD gain regions are inhomogeneously broadened by QD size and composition variations. To incorporate these deviations from a perfect situation, we follow gas laser theory,[15] and perform a statistical average of the homogeneous gain spectra assuming an inhomogeneous width, σ_{inh}.

We begin by considering the case of $In_{0.3}Ga_{0.7}As$ QDs embedded in a 4 nm thick GaAs quantum well, which is cladded by $Al_{0.2}Ga_{0.8}As$ layers. To compute the electronic structure, the actual QD shape is approximated by a disk of height and diameter 2 and 18 nm, respectively. Assuming appreciably weaker QD confinement in the QW plane than in perpendicular direction,[16] it is possible to separate the problem into radial and vertical components. With this simplification, the electronic structure for the QD-QW system can already be computed, using as input the bulk material properties, such as the electron effective masses and Luttinger parameters, spin-orbit energy splitting, elastic constants, lattice constants, and deformation potentials.[17] The solutions contain the effects of quantum confinement and mixing between hole states. For the QW states, we use orthogonalized plane waves.[8] The calculation indicates one localized electronic state and one localized hole state located 15 and 25 meV, respectively, below their respective QW band edges. While there are more rigorous approaches to determining the QD single-particle states,[16] the present method adequately describes the essential features necessary for our study of excitation effects in optical response.

III. RESULTS

Figure 1(a) shows the homogeneously broadened gain spectra computed using (1) and (2). The curves are for $T = 300$ K and different carrier densities, which are defined as $N = 2N_{dot}\Sigma_i n_i^e + 2A^{-1}\Sigma_{\mathbf{k}} n_{\mathbf{k}}^e = 2N_{dot}\Sigma_i n_i^h + 2A^{-1}\Sigma_{\mathbf{k}} n_{\mathbf{k}}^h$. For the shallow QD structure, each spectrum has an s-shell resonance and a broad QW contribution. Figure 1(b) depicts the corresponding inhomogeneously broadened spectra. By assuming an inhomogeneous broadening of $\sigma_{inh} = 20$ meV, the spectra in Fig. 1(b) resemble closely those observed in experiment.[18] Comparison of the two sets of spectra reveals significant masking of intrinsic QD properties by QD size and composition variations in present experiments. The homogeneously broadened result clearly indicates a carrier-density dependent energy shift in the QD resonances. There is also noticeable spectral broadening of QD resonances with increasing carrier density, suggesting a strong excitation dependence in the dephasing. Both effects were observed in QD luminescence in single-dot experiments, but not at car-

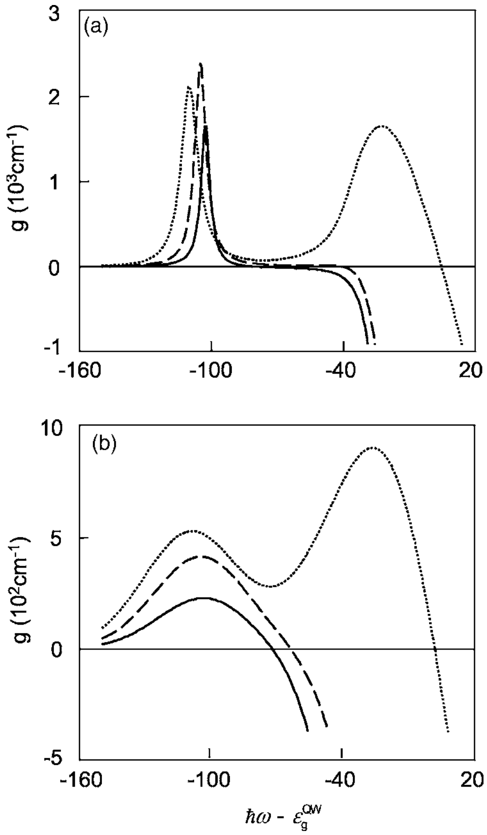

FIG. 1. (a) Homogeneously and (b) inhomogeneously broadened spectra of $In_{0.3}Ga_{0.4}As$ QD embedded in a GaAs QWs at temperature $T=300$ K and carrier densities $N=3$ (solid line), 5 (dashed line), and 10×10^{11} cm^{-2} (dotted line). The energy axis is given relatively to the QW band gap energy.

rier densities sufficiently high to produce laser gain.[19]

Closer examination of the spectra in Fig. 1(a) reveals another effect in the intrinsic QD gain. For $N\geq5\times10^{11}$ cm^{-2}, the s-shell QD peak gain actually decreases with increasing carrier density. To show this anomaly explicitly, we plot the peak gain as a function of carrier density [Fig. 2(a)]. The overall [QD+QW, as defined by (2)] peak gain is chosen because it impacts laser performance more than a transition-specific peak gain. The solid curve, which describes the homogeneously broadened case, clearly contains a carrier density range where the slope dg_{pk}/dN is negative. This counterintuitive behavior is caused by two factors. One is the saturation of the state filling contribution to gain at high carrier densities. This saturation is significantly more pronounced in QD than in QW or bulk systems because of 0 dimensional (0 D) versus 2 D or 3 D density of states differences. The second is the nonsaturating increase in dephasing with increasing carrier density. While increased dephasing always causes gain amplitude reduction, its effect usually lags that of state filling. Consequently, in all active media encountered until now, the peak gain always increases with increasing excitation. The difference with a QD-QW system is that in the moment the excitation produces a non-negligible carrier population in the QW states, there is a sharp increase in scattering channels and a corresponding abrupt jump in the dephasing contribution. As a result, the dephasing overtakes band filling, causing the gain amplitude to decrease throughout the spectral width of the QD resonance. Afterwards, dg_{pk}/dN remains negative until the gain from the QW transitions exceeds that from the QD resonance. The other curves in Fig. 2(a) are for different inhomogeneously broadening width. We note that inhomogeneous broadening always reduces the peak gain, with the decrease being inversely proportional to the width of the gain resonance. The net result is a smoothing of the features of the homogeneously broadened g_{pk} versus N curve, and behavior reverts to what is expected for laser gain structures. Nevertheless, for moderate inhomogeneous broadening [5 meV in Fig. 2(a), 10 meV in Fig. 2(b)] a plateau in the peak gain versus carrier density curve remains clearly visible, so that the anomaly should be observable in high quality QD samples.

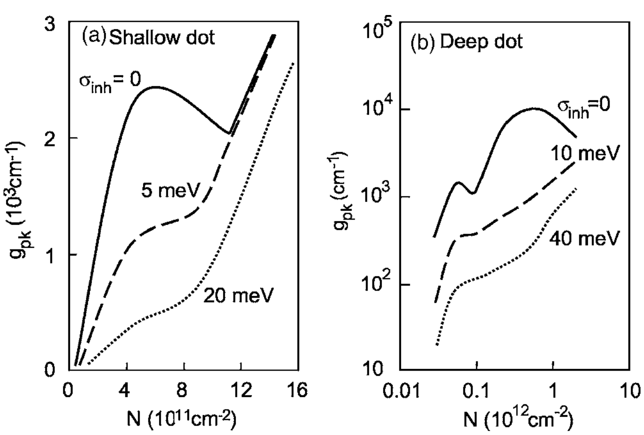

FIG. 2. Peak gain versus carrier density for the shallow QD structure and different inhomogeneous broadening σ_{inh}.

Figure 2(b) shows that the $dg_{pk}/dN<0$ behavior is more pronounced in a deeply confined QD structure of InAs QDs embedded in a GaAs QW. This example is interesting because its emission wavelength at around 1.5 μm is useful for optical fiber communication. The calculated gain spectrum consists of two QD resonances, from s-shell and p-shell states, and a broad QW contribution. The homogeneously broadened g_{pk} versus N curve indicates two regions with $dg_{pk}/dN<0$, which correspond to the s- and p-shell emission. Dephasing is significantly weaker in the deep QD structure because of a scarcity of allowable collisions. As a result, the homogeneously broadened QD peak gain is considerably higher and the spectral width is corresponding narrower than is the case for the shallower $In_x\,Ga_{1-x}As$ QDs. This leads to a greater sensitivity to inhomogeneous broadening, as evident in Fig. 2(b).

One gathers from the above results that intrinsic QD gain properties are strongly governed by dephasing. Figure 3 illustrates the complicated nature of dephasing as predicted by quantum-kinetic theory. Plotted is the full width at half height (FWHH) of the homogeneously broadened QD resonance as a function of carrier density. This quantity provides a measure of the net effect of the real and imaginary, diagonal and nondiagonal contributions of the scattering terms, S_α^{c-c} and S_α^{c-p} in (1). The solid curve in the inset shows a typical QD gain resonance; in this case, s-shell, shallow dot

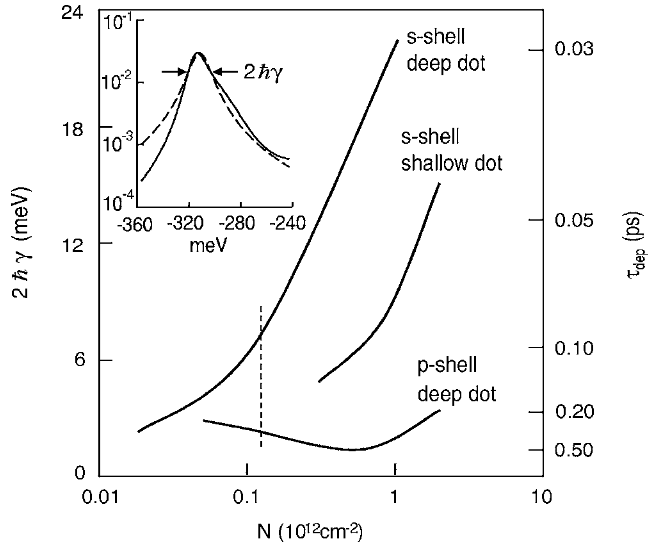

FIG. 3. Full width at half height (FWHH) of QD resonances versus carrier density. The inset shows a typical QD resonance and best fit with a Lorentzian function.

at $N=5\times10^{11}$ cm^{-2}. Comparison with a Lorentzian function with the same amplitude maximum and FWHH (dashed curve) indicates a basically good fit except for the asymmetry in the actual resonance and deviations at the spectral tails. The differences may be traced to the nondiagonal correlation contributions and QW influence. The main curves indicate significant variation in dephasing with carrier density and QD structure. For the shallow QD, the *s*-shell dephasing increases appreciably with carrier density. For the deep QD, there is a further complication of the *s*- and *p*-shell resonances experiencing different dephasing effects. When g_{pk} moves from the *s*-shell to the *p*-shell resonance (intersection of solid curve and dashed line), there is a sharp decrease in dephasing. The *p*-shell dephasing dependence on carrier density first decreases before increasing, with the increase caused by the onset of QW contributions. The dephasing widths or times (right axis) is consistent with experiment.[20] The complex dependences shown in the figure are describable only with a rigorous treatment of scattering effects.

IV. SUMMARY

The intrinsic properties of QD gain is investigated within the context of a microscopic theory, with carrier correlations treated at the level of quantum kinetics. It was found that a QD active medium exhibits unique optical gain properties that are usually masked by inhomogeneous broadening. The most interesting is the decrease in peak gain with increasing excitation for certain carrier density ranges. This anomalous behavior depends on the delicate balancing of band-filling and dephasing contributions in a Coulomb coupled QD-QW system.

ACKNOWLEDGMENTS

The work was supported by the Deutsche Forschungsgemeinschaft, NIC of Forschungszentrum Jülich, U. S. Department of Energy under Contract No. DE-AC04-94AL85000, and Humboldt Foundation.

*Electronic address: www.itp.uni-bremen.de

[1] M. Bayer, F. Weidner, A. Larionov, A. McDonald, A. Forchel, and T. L. Reinecke, Phys. Rev. Lett. **86**, 3168 (2001).
[2] J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel, Nature (London) **432**, 197 (2004).
[3] T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, and D. G. Deppe, Nature (London) **432**, 200 (2004).
[4] P. Michler, A. Imamoglu, M. D. Mason, P. J. Carson, G. F. Strouse, and S. K. Buratto, Nature (London) **406**, 968 (2000).
[5] X. Q. Li, Y. W. Wu, D. Steel, D. Gammonand, T. H. Stievaterand, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Science **301**, 809 (2003).
[6] U. Bockelmann and T. Egeler, Phys. Rev. B **46**, 15574 (1992).
[7] I. Magnusdottir, S. Bischoff, A. V. Uskov, and J. Mørk, Phys. Rev. B **67**, 205326 (2003).
[8] T. R. Nielsen, P. Gartner, and F. Jahnke, Phys. Rev. B **69**, 235314 (2004).
[9] H. Benisty, C. M. Sotomayor-Torres, and C. Weisbuch, Phys. Rev. B **44**, 10945 (1991).
[10] M. Sugawara, K. Mukai, and H. Shoji, Appl. Phys. Lett. **71**, 2791 (1997).
[11] T. Inoshita and H. Sakaki, Phys. Rev. B **56**, R4355 (1997).
[12] J. Seebeck, T. R. Nielsen, P. Gartner, and F. Jahnke, Phys. Rev. B **71**, 125327 (2005).
[13] H. C. Schneider, W. W. Chow, and S. W. Koch, Phys. Rev. B **70**, 235308 (2004).
[14] M. Lorke, T. R. Nielsen, J. Seebeck, P. Gartner, and F. Jahnke, Phys. Rev. B **73**, 085324 (2006).
[15] J. W. E. Lamb, Phys. Rev. **134**, A1429 (1964).
[16] A. D. Andreev and E. P. O'Reilly, Phys. Rev. B **62**, 15851 (2000).
[17] *Numerical Data and Functional Relationships in Science and Technology*, Landolt-Börnstein (Springer-Verlag, Berlin, 1982), Vol. 17.
[18] S. Osborne, P. Blood, P. Smowton, J. Lutti, Y. C. Xin, A. Stintz, D. Huffaker, and L. Lester, IEEE J. Quantum Electron. **40**, 1639 (2004).
[19] K. Matsuda, K. Ikeda, T. Saiki, H. Saito, and K. Nishi, Appl. Phys. Lett. **83**, 2250 (2003).
[20] P. Borri, W. Langbein, J. M. Hvam, F. Heinrichsdorff, M. H. Mao, and D. Bimberg, IEEE J. Sel. Top. Quantum Electron. **6**, 544 (2000).