Excitonic nonlinear optical processes in GaAs quantum-well wires

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In a recent calculation, we obtained exciton and biexciton binding energies and wave functions for a rectangular quantum wire in order to determine its third-order nonlinear optical susceptibility $\chi^{(3)}$. Using a variational calculation involving two parameters—exciton radius and exciton-exciton separation within the biexciton—we computed the binding energies of the exciton and biexciton. In the present paper, we have included a third parameter to allow for the variation of exciton radius within the biexciton. As the result of this added variational freedom, the biexciton binding energies changed by 7–10 % and the magnitudes of all variational parameters and of $\chi^{(3)}$ changed by approximately 20% for wire dimensions between 25×25 and 300×300 Å ^{2}.

In a recent publication, Madarasz *et al.*[1,2] performed a variational calculation of the exciton and biexciton binding energies in a rectangular GaAs quantum-well wire (QWW). That calculation employed two variational parameters: η, the variational parameter for the electron-hole separation along the axis of the wire within the exciton; and ξ, the variational parameter for hole-hole separation along the axis of the wire within the biexciton. However, the exciton radius within the biexciton was taken to be η, i.e., the same as in a free exciton. In the present work, we improve on the above calculation by including a third variational parameter η_{xx}, which allows the electron-hole pair within the biexciton to relax. For clarity, in what follows, we define the set of variational parameters: $\eta_x\equiv\eta$, $\xi_x\equiv\xi(\eta_x)$, and $\xi_{xx}\equiv\xi(\eta_{xx})$.

For complete details of the theory and relevant expressions, the reader is referred to the original work.[1,2] The addition of a third variational parameter does not change the analytic forms of the expressions for the kinetic, potential, and binding energies of the exciton and biexciton except that the newly defined set of parameters must be substituted for the old. On the other hand, while the functional form of $\chi^{(3)}$ does not change, the product of the exciton and biexciton matrix elements explicitly mixes their respective variational parameters; as a result, one must be careful to properly substitute the new parameters inside the expression for $\chi^{(3)}$. With these substitutions, the expression for $\chi^{(3)}$ in the rotating-wave approximation[1–3] for a resonant or near-resonant excitation in a pump-probe experiment is given by[4]

$$\chi^{(3)}=\frac{2}{\pi\sqrt{2\pi}}\left[\frac{n_0}{\eta_x}\right]\left[\frac{\xi_{xx}}{\eta_x}\right]\frac{e^4}{m_0^2\omega_{g0}^4}E_P^2\left[\frac{1}{(\omega_1-\omega_{g0}+i\Gamma)}-\frac{1}{(\omega_1-\omega_{g0}+\omega_{bx}(\eta_{xx},\xi_{xx})+i\Gamma)}\right]$$
$$\times\sum_{r=1}^{2}\left\{\frac{1}{\hbar^3(\omega_r-\omega_2+i\gamma)}\left[\frac{1}{(\omega_{g0}-\omega_2+i\Gamma)}+\frac{1}{(\omega_r-\omega_{g0}+i\Gamma)}\right]\right\}. \quad (1)$$

Here, n_0 is the average areal density of unit cells, m_0 is the rest mass of an electron, e is the charge on an electron, E_P is the Kane matrix element, Γ and γ are the transverse and longitudinal relaxation parameters, respectively, ω_2 and ω_1 are the pump and probe frequencies, respectively, $\hbar\omega_{g0}$ is the exciton ground state energy, and $\hbar\omega_{bx}(\xi_{xx},\eta_{xx})$ is the biexciton binding energy.

In Tables I and II, we compare various quantities obtained in two- and three-parameter variational calculations as functions of L and W, the cross-sectional dimensions of the wire. The difference between the electron-hole variational parameters within the exciton, η_x, and within the biexciton, η_{xx} is 20%±2% over the range of wire dimensions given in Table I. The variation of η within the biexciton results in a value of η_{xx} which is smaller than its value within the exciton η_x. In other words, the mean electron-hole separation within the biexciton is approximately 20% smaller than within an isolated exciton. Consequently, the variation of η within the biexciton leads to a reduction of the hole-hole variational parameter. According to Table I, not only is ξ_{xx} smaller than ξ_x by 20%±2% in the given range of wire dimensions, but also its value is reduced proportionately to the reduction of the corresponding ratio η_{xx}/η_x. In other words, the ratios $\xi_x/\eta_x=\xi_{xx}/\eta_{xx}\approx 3.5$, regardless of exciton relaxation within the biexciton; in either case, then, the hole-hole separation turns out to be 3.5 times the electron-hole separation within the biexciton.

The last column in Table I lists the exciton ground-state energies as a function of wire dimensions. These energies, which are important for the determination of the energy for the resonant exciton creation, were not given in our earlier work.[1,2] We have specifically limited ourselves to wire dimensions no smaller than 25×25 Å^2, since, from a practical point of view, the ground-state energy cannot exceed the total band offsets, which are about 2.8 eV for a $GaAs/Al_{0.3}Ga_{0.7}$ interface. Since $\eta_x\equiv\eta$, the exciton binding energies do not change from the values reported earlier in Refs. 1 and 2.

In Table II we list the biexciton binding energies as a function of wire dimensions. The biexciton binding energies with η held fixed at η_x, the value within an isolated exciton, are compared to the biexciton binding energies for $\eta=\eta_{xx}$, the related value within the biexciton. For the smallest set of dimensions, $L=W=25$ Å, the change in the biexciton binding energies is 9.4%, whereas for the largest set of dimensions, $L=W=300$ Å, it changes by 7.5%.

In Table II we list the resonant values of $\chi^{(3)}$ determined for the case where $\eta=\eta_{xx}$ is allowed to relax within the biexciton. The relaxation is seen to produce a 20%±2% reduction in $\chi^{(3)}$ for the listed range of wire dimensions. In fact, the percentage reduction is the same as the percentage reduction in the ratio ξ_{xx}/ξ_x of the hole-hole variational parameters. This is easily explained if it is recalled that the original expression for $\chi^{(3)}$ before

TABLE I. Quantities obtained in two- and three-parameter variational calculations as functions of L and W.

Wire dimensions		Electron-hole variational parameter		Hole-hole variational parameter		Exciton ground-state energy (eV)
L (Å)	W (Å)	$\eta_x(a_0)^a$	$\eta_{xx}(a_0)^a$	$\xi_x(a_0)^a$	$\xi_{xx}(a_0)^a$	$E^x_{g0}(\eta_x)$
25	25	216.4	181.1	763.4	634.2	3.552
50	50	276.0	230.8	971.9	806.9	2.014
75	75	322.7	270.2	1135	943.6	1.732
100	100	362.8	302.2	1275	1055	1.635
125	125	398.7	333.0	1400	1161	1.590
150	150	431.6	359.3	1515	1253	1.566
200	200	491.2	408.9	1723	1425	1.543
250	250	544.8	453.8	1910	1581	1.533
300	300	594.1	493.3	2082	1718	1.527
50	25	248.4	208.7	875.3	730.2	2.784
50	100	322.0	269.6	1133	941.7	1.825
50	150	360.4	300.3	1267	1048	1.792
50	200	394.1	329.3	1385	1149	1.782
50	250	424.5	353.6	1491	1234	1.777
50	300	452.2	375.8	1588	1311	1.775
100	25	298.6	250.9	1051	877.1	2.597
100	150	398.2	332.5	1399	1160	1.601
100	200	429.9	357.9	1509	1248	1.590
100	250	458.8	383.0	1611	1336	1.585
100	300	485.6	404.5	1704	1410	1.582
200	25	375.0	312.0	1319	1089	2.554
200	150	461.9	385.5	1621	1344	1.555
200	250	518.3	430.6	1818	1500	1.538
200	300	543.6	452.9	1906	1578	1.535
300	25	436.4	363.1	1534	1267	2.548
300	150	515.8	428.6	1809	1494	1.547
300	250	569.6	473.7	1997	1650	1.530

[a] a_0 is the Bohr radius of a hydrogen atom.

η was allowed to vary within the biexciton containing the ratio ξ_x/η_x, and the new expression for $\chi^{(3)}$ in Eq. (1), in which η is varied within the biexciton, contains the ratio ξ_{xx}/η_x. Therefore, the old and new third-order nonlinear susceptibilities are in the ratio of ξ_{xx}/ξ_x, which accounts for the percent reduction in $\chi^{(3)}$. Moreover, Eq. (1) indicates that $\chi^{(3)}$ also varies inversely with E_{g0}^{x}, the exciton ground-state energy. However, the ground-state energy is a strong function of the fundamental gap energy, especially for wires of dimension larger than 50×50 Å^2, and is independent of any variation within the biexciton. Thus, for resonant excitation, $\chi^{(3)}$ depends on the factor ξ_{xx}/ξ_x.

Finally, we show the graphical representation of the newly calculated $\chi^{(3)}$ for symmetric and asymmetric wires as a function of wire dimensions in Figs. 1 and 2, respectively.

Physically, $\chi^{(3)}$ exists due to the nonlinearity in the interaction between light and the biexciton. The size of the light-induced dipole, in turn, is proportional to the biexciton radius—provided the transition takes place in a direct-gap material and the wavelength of the light emitted in the biexciton formation is large compared to the average radius of the newly formed biexciton.[3,5] Since exciton relaxation leads to a more strongly bound biexciton, the biexciton radius decreases, leading to a reduction of $\chi^{(3)}$. In a forthcoming paper we will address the problem of finite band offsets and the affect of Al interdiffusion from the cladding material into the wire.

In conclusion, we have shown that the relaxation of the exciton within the biexciton complex results in 20%±2% reduction in $\chi^{(3)}$ for the range of wire dimensions studied in this work.

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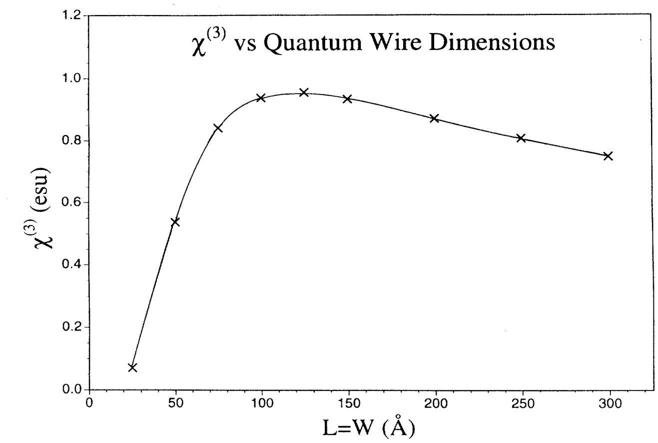

FIG. 1. Nonlinear optical susceptibility for resonant excitation as a function of wire size for a symmetric wire.

TABLE II. Biexciton binding energies as a function of wire dimensions.

Wire dimensions		Biexciton binding energy (meV)		Third-order susceptibility (esu)	
L (Å)	W (Å)	$E_B^{xx}(\eta_x)$	$E_B^{xx}(\eta_{xx})$	$\chi^{(3)}(\eta_x)$	$\chi^{(3)}(\eta_{xx})$
25	25	19.46	21.29	0.08556	0.07109
50	50	13.45	14.64	0.6472	0.5373
75	75	10.65	11.57	1.011	0.8406
100	100	8.957	9.710	1.133	0.9373
125	125	7.798	8.440	1.151	0.9547
150	150	6.944	7.506	1.129	0.9334
200	200	5.755	6.207	1.052	0.8701
250	250	4.954	5.335	0.9739	0.8060
300	300	4.372	4.702	0.9053	0.7468
50	25	15.75	17.19	0.6472	0.5373
50	100	10.67	11.58	0.8219	0.6834
50	150	8.997	9.755	0.7897	0.6535
50	200	7.860	8.509	0.7394	0.6135
50	250	7.025	7.597	0.6932	0.5736
50	300	6.382	6.894	0.6538	0.5396
100	25	11.89	12.93	0.2165	0.1806
100	150	7.805	8.447	1.122	0.9310
100	200	6.964	7.528	1.069	0.8838
100	250	6.316	6.821	1.013	0.8402
100	300	5.800	6.257	0.9627	0.7965
200	25	8.414	9.120	0.1840	0.1520
200	150	6.287	6.788	1.086	0.9003
200	250	5.322	5.735	1.010	0.8336
200	300	4.961	5.342	0.9693	0.8023
300	25	6.688	7.232	0.1596	0.1319
300	150	5.339	5.755	0.9908	0.8180
300	250	4.643	4.997	0.9377	0.7746

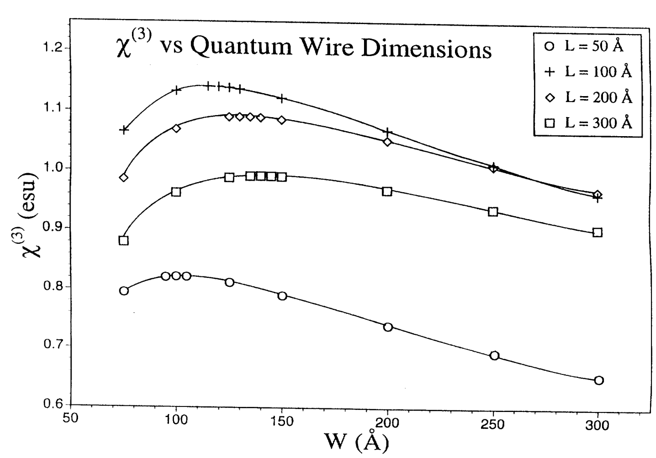

FIG. 2. Same as Fig. 1, but for asymmetric wires.

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[4]In reducing the general terms given in Appendix B of Ref. 2 and regrouping them, some of the indices on the frequencies in the propagator terms of Eq. (35) were misappropriated. Nevertheless, the double Feynmann diagrams and the general terms in Appendix B, as well as the results reported for resonant exciton absorption $\chi^{(3)}$, Eq. (40), are correct. The resulting nonlinear susceptibility, Eq. (38) of Ref. 2, in the rotating-wave approximation is correctly given by Eq. (1) of the present paper.

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