Dressed quantum graphs with optical nonlinearities near the fundamental limits

Topics

Nonlinearities of quantum wires with finite δ-potentials (δ-graphs, aka compressed δ-atoms and molecules)

Scaling behavior near the fundamental limits (FL)

Origin of the enhanced nonlinearities

Key results*

Existence of δ-graphs with β_{max} ~ 0.71, γ_{max} ~ 0.6, γ_{min}~ -0.15

Three-levels required for β near its maximum, & for γ near its minimum

Four levels required for γ near its maximum

** all hyperpolarizabilities are x-diagonal and normalized to the FL*

Rick Lytel

August 7, 2013

Reading list

"Analytical solution of the compressed, one-dimensional delta atom via quadratures and exact, absolutely convergent periodic-orbit expansions," R. Blümel, Journal of Physics A: Mathematical and General *39*, 8257 (2006).

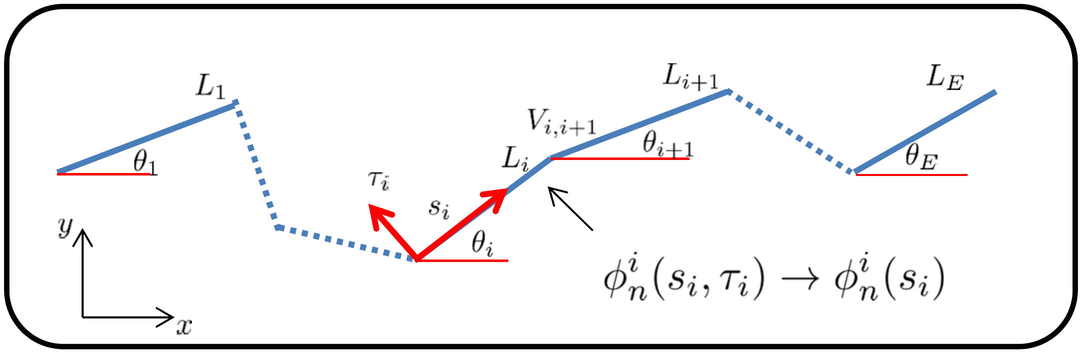
"Scaling and universality in nonlinear optical quantum graphs containing star motifs," R. Lytel, S. Shafei, & M. Kuzyk, *arXiv:1305.4334 (*2013).

" Influence of geometry and topology of quantum graphs on their nonlinear optical properties," Rick Lytel, Shoresh Shafei, Julian H. Smith, and Mark G. Kuzyk, Phys. Rev. A 87, 043824 (2013).

"Sum Rules and Scaling in Nonlinear Optics," M.G. Kuzyk, J. Perez-Moreno, and S. Shafei, Phys. Rep. 529, 297-398 (2013).

"Geometry-controlled nonlinear optical response of quantum graphs," S. Shafei, R. Lytel, and M.G. Kuzyk, J. Opt. Sci. Am. B 29, 3419 (2012).

Quantum mechanics of bare graphs



Extreme confinement along τ

Electron dynamics along s

Self-adjoint Hamiltonian

Zero potential everywhere

Eigenfunctions are unions of edge functions having same eigenvalues

$$H\psi_n(s) = E_n\psi_n(s) \qquad \psi_n(s) = \cup_{i=1}^{E}\phi_n^i(s_i) \qquad H\phi_n^i = E_n\phi_n^i$$

Edge functions: continuity and conservation of probability flux

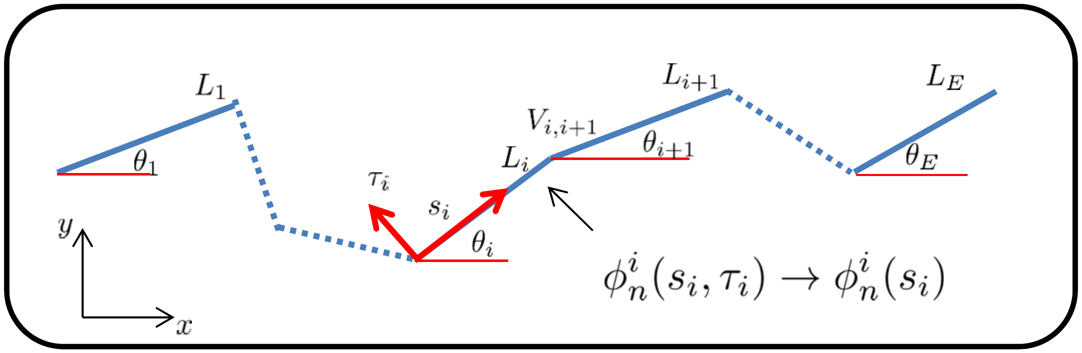
'degree' of vertex

$$\phi_n^i = A_n^{(i)} \sin k_n s_i + B_n^{(i)} \cos k_n s_i \quad \phi_n^i = \phi_n^{i+1}\ at\ V_{i,i+1} \quad \sum_{i=1}^{d} \phi_n'^i = 0\ at\ V_{i,i+1}$$

Homogeneous, coupled linear equations for amplitudes

$$\sum_{i=1}^{2E} a_n^{ij} C_n^{(i)} = 0,\ j = 1, 2, 3...2E,\ C_n^{(1)} = A_n^{(1)},\ C_n^{(2)} = B_n^{(1)}...$$

Quantum mechanics of δ-dressed graphs



Extreme confinement along τ

Electron dynamics along s

Self-adjoint Hamiltonian

V= (g/L)δ(s) at a vertex

Eigenfunctions are unions of edge functions having same eigenvalues

$$H\psi_n(s) = E_n\psi_n(s) \qquad \psi_n(s) = \cup_{i=1}^{E}\phi_n^i(s_i) \qquad H\phi_n^i = E_n\phi_n^i$$

Edge functions: continuity and conservation of probability flux

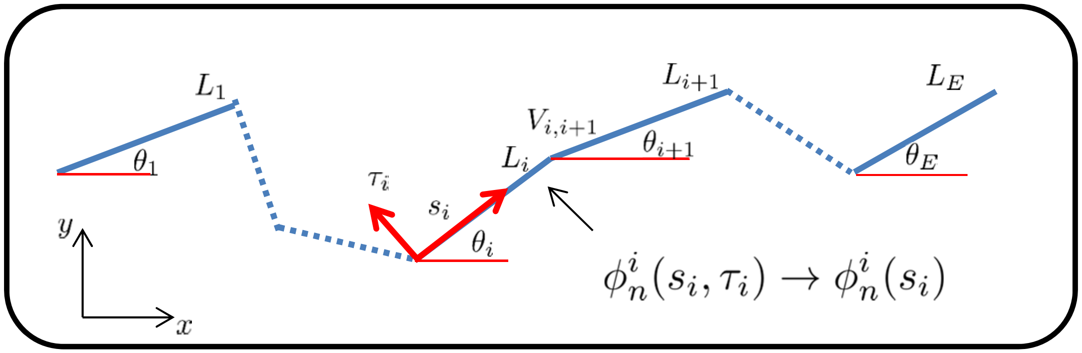
'degree' of vertex

$$\phi_n^i = A_n^{(i)}\sin k_n s_i + B_n^{(i)}\cos k_n s_i \quad \phi_n^i = \phi_n^{i+1}\ at\ V_{i,i+1} \quad \sum_{i=1}^{d}\phi_n'^i = 2(g/L)\phi_n(a)$$

Homogeneous, coupled linear equations for amplitudes

$$\sum_{i=1}^{2E} a_n^{ij}C_n^{(i)} = 0,\ j = 1,2,3...2E,\ C_n^{(1)} = A_n^{(1)},\ C_n^{(2)} = B_n^{(1)}...$$

Quantum mechanics of graphs (cont'd)



Extreme confinement along τ

Electron dynamics along s

Self-adjoint Hamiltonian

Bare or dressed edges

Determinant of coefficients vanishes yielding secular equation for eigenvalues

$$\det a_n^{ij} = 0 \qquad f_{sec}(k_n; L_i, \theta_i) = 0 \;\Rightarrow\; E_n = k_n^2/2$$

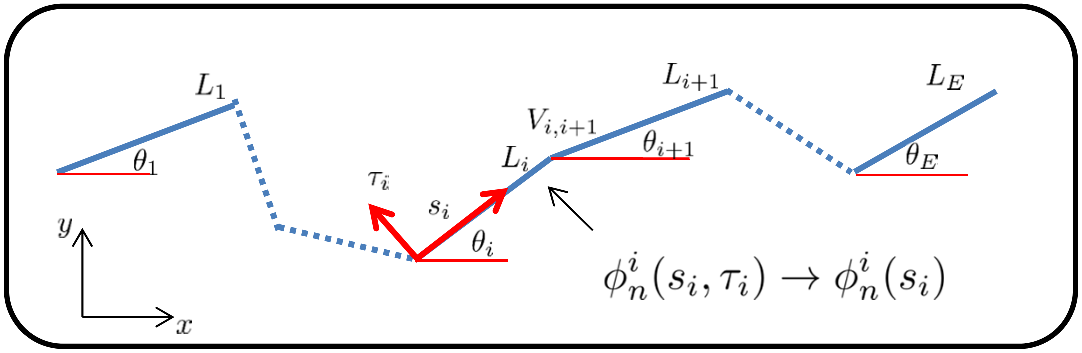
Continuity equations for amplitudes, plus normalization yields amplitudes

$$\phi_n^i = A_n^{(i)} \sin k_n s_i + B_n^{(i)} \cos k_n s_i$$

Transition moments are now calculable

$$r_{nm} = \sum_{i=1}^{E} \int_i ds_i \; r(s_i) \phi_n^{*i}(s_i) \phi_m^i(s_i)$$

Aside: boundary condition equation counting



E edges

V_E external vertices

V_I internal vertices

$V=V_E+V_I$ vertices

Union of edge functions has 2E unknown amplitudes

$$\phi_n^i = A_n^{(i)} \sin k_n s_i + B_n^{(i)} \cos k_n s_i, \;\; i = 1, 2, \ldots E$$

1. **There are exactly V_E amplitude boundary condition eqns on terminated edges**
2. **There are exactly V_I flux boundary condition eqns on internal vertices**
3. **This provides exactly V boundary condition eqns (so far)**

Now, let d^k = degree of each internal vertex, where degree = #edges connected to it. There are exactly d^k-1 amplitude boundary condition eqns at vertex k. Sum over internal vertices. The sum over degrees equals the total # of edges that are internally connected, and is equal to 2E-V_E. The sum of 1 over internal edges is V_I. So we get:

4. There are exactly 2E-V_E-V_I = 2E-V amplitude boundary eqns at internal vertices. QED

First hyperpolarizability tensor

$$\beta_{ijk} = -\frac{e^3}{2}\sum_{n,m}{}' \frac{1}{E_{n0}E_{m0}}\left(r^i_{0n}\bar{r}^j_{nm}r^k_{m0} + r^i_{0n}\bar{r}^k_{nm}r^j_{m0} + r^k_{0n}\bar{r}^i_{nm}r^j_{m0} + h.c\right)$$

Fully symmetric 3rd rank Cartesian tensor

$$\begin{aligned}\beta_{xxx}(\theta) &= \beta_{xxx}\cos^3\theta + 3\beta_{xxy}\cos^2\theta\sin\theta \\ &+ 3\beta_{xyy}\cos\theta\sin^2\theta + \beta_{yyy}\sin^3\theta\end{aligned}$$

Four independent components relating lab and graph frames

$$|\beta| = \sqrt{\beta_{xxx}^2 + 3\beta_{xxy}^2 + 3\beta_{xyy}^2 + \beta_{yyy}^2}$$

Tensor norm (invariant under rotations of graph)

$$\beta_{max} = 3^{1/4}\left(\frac{e\hbar}{\sqrt{m}}\right)^3\left(\frac{N^{3/2}}{E_{10}^{7/2}}\right) \quad \longrightarrow \quad -1 \le \beta_{int} \equiv \frac{\beta}{\beta_{max}} \le 1$$

Normalization for scale-invariant calculations

Second hyperpolarizability tensor

$$\gamma_{ijkl} = (1/6)P_{ijkl}\left(\sum_{n,m,l}{}' \frac{r^i_{0n}\bar{r}^j_{nm}\bar{r}^k_{ml}r^l_{l0}}{E_{n0}E_{m0}E_{l0}} - \sum_{n,m}{}' \frac{r^i_{0n}r^j_{n0}r^k_{0m}r^l_{m0}}{E^2_{n0}E_{m0}}\right)$$

Fully symmetric 4th rank Cartesian tensor

$$\begin{aligned}\gamma_{xxxx}(\theta) &= \gamma_{xxxx}\cos^4\theta + 4\gamma_{xxxy}\cos^3\theta\sin\theta \\ &+ 6\gamma_{xxyy}\cos^2\theta\sin^2\theta + 4\gamma_{xyyy}\cos\theta\sin^3\theta \\ &+ \gamma_{yyyy}\sin^4\theta\end{aligned}$$

Five independent components relating lab and graph frames

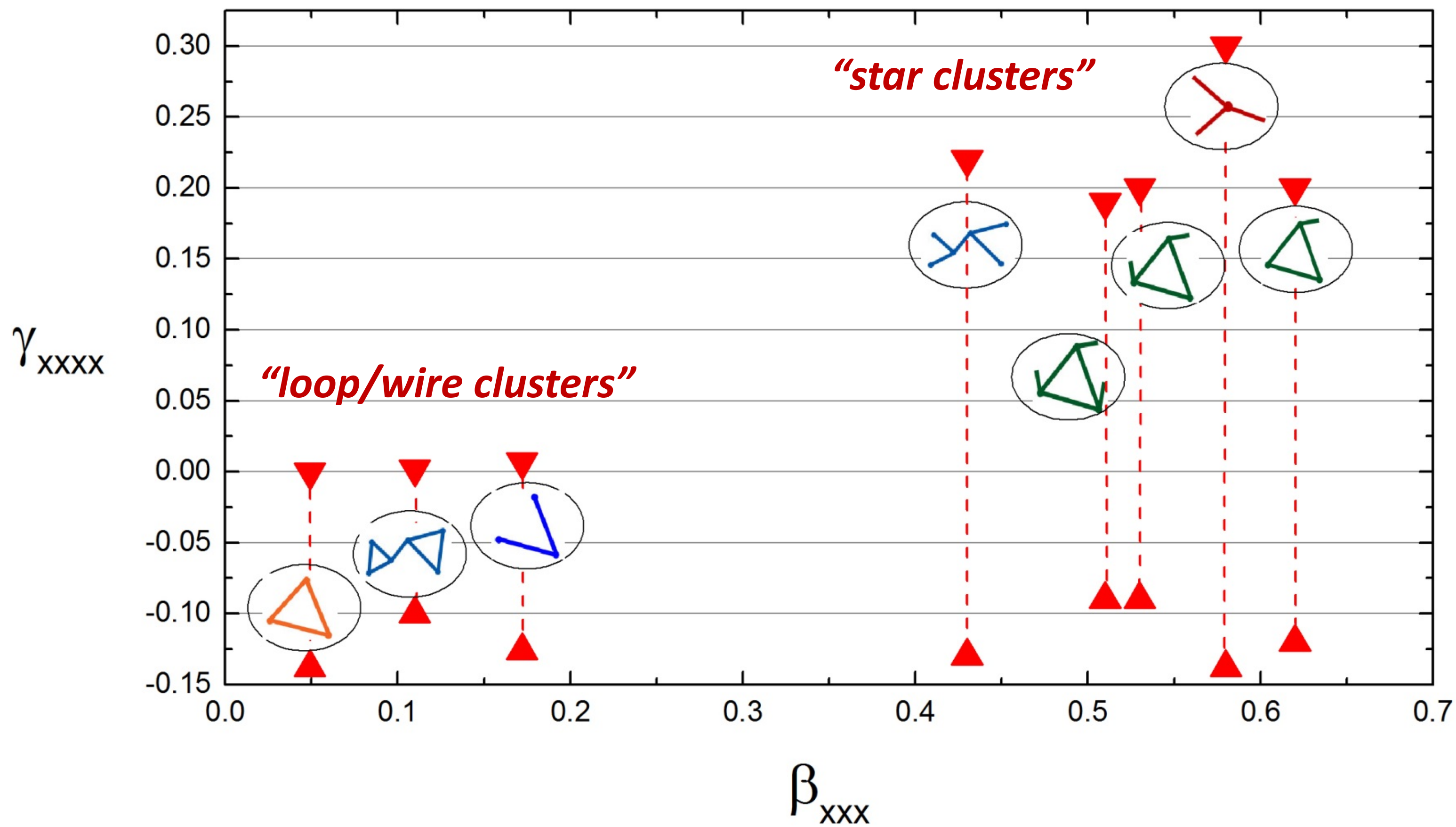
$$|\gamma| = \sqrt{\gamma^2_{xxxx} + 4\gamma^2_{xxxy} + 6\gamma^2_{xxyy} + 4\gamma_{xyyy} + \gamma^2_{yyyy}}$$

Tensor norm (invariant under rotations of graph)

$$\gamma_{max} = 4\left(\frac{e^4\hbar^4}{m^2}\right)\left(\frac{N^2}{E^5_{10}}\right) \longrightarrow -1/4 \leq \gamma_{int} \equiv \frac{\gamma}{\gamma_{max}} \leq 1$$

Normalization for scale-invariant calculations

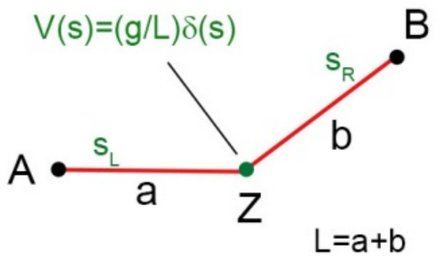
Topological clustering and enhancement of β and γ



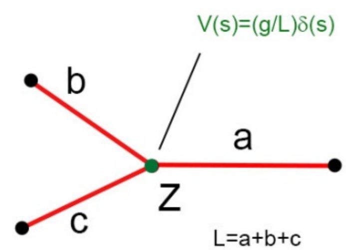
Hyperpolarizability ranges still fall short of (potential optimization) limits

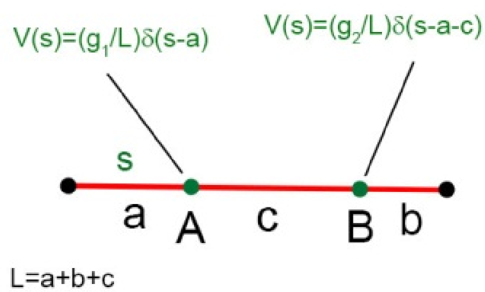
δ-dressed quantum graphs

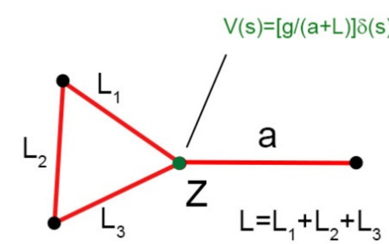
"dressed wires"

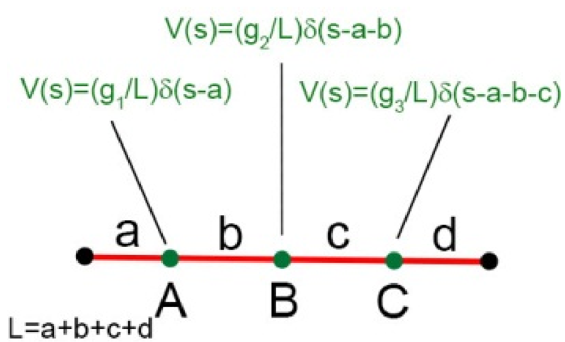


"dressed stars"



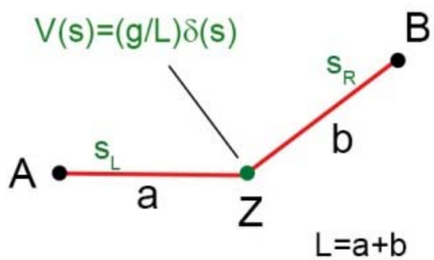






Presence of finite δ potential re-arranges mode shapes
Might we create the optimum states for max β, γ?

Compressed delta atom motif



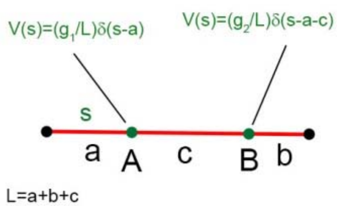
$$\phi_L(s) = \frac{Z \sin k(a - s_L) + A \sin k s_L}{\sin ka}$$
$$\phi_R(s) = \frac{Z \sin k(b - s_R) + B \sin k s_R}{\sin kb}$$

Secular function defined by flow of probability on the graph:

$$Z F_\delta = A \sin kb + B \sin ka \qquad \omega = 2a/L - 1, -1 \leq \omega \leq 1$$

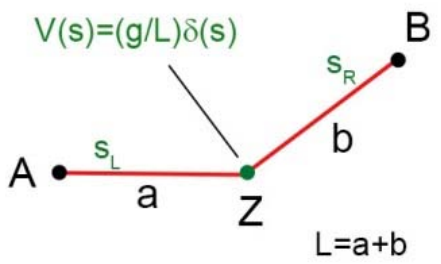
$$F_\delta \equiv F_\delta(g; \omega, kL) = -(1/kL) \left[g(\cos kL - \cos \omega kL) - kL \sin kL \right]$$

Overlapping motifs, with matched endpoints, yields secular functions for larger graphs



$$A F_\delta(g_1; a, c) = B \sin ka$$
$$B F_\delta(g_2; b, c) = A \sin kb$$

Compressed delta atom (A=B=0)



$$\phi_L(s) = \frac{Z \sin k(a - s_L)}{\sin ka}$$

$$\phi_R(s) = \frac{Z \sin k(b - s_R)}{\sin kb}$$

Secular equation determines eigenvalues for graph

$$0 = F_\delta(g; \omega, kL) = -(1/kL)\left[g(\cos kL - \cos \omega kL) - kL \sin kL\right], \ E \geq 0$$

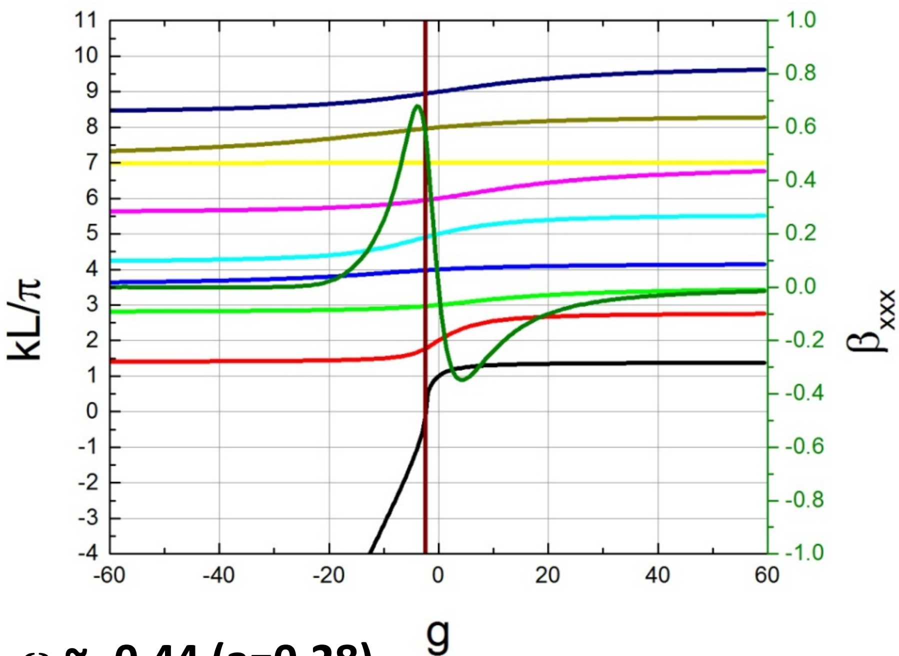
$$0 = F_\delta(g; \omega, \kappa L) = (i/\kappa L)\left[g(\cosh \kappa L - \cosh \omega \kappa L) + \kappa L \sinh \kappa L\right], \ E < 0$$

$$E_n = k_n^2/2, \ \hbar = m = 1, \ k = \imath\kappa, \ E < 0$$

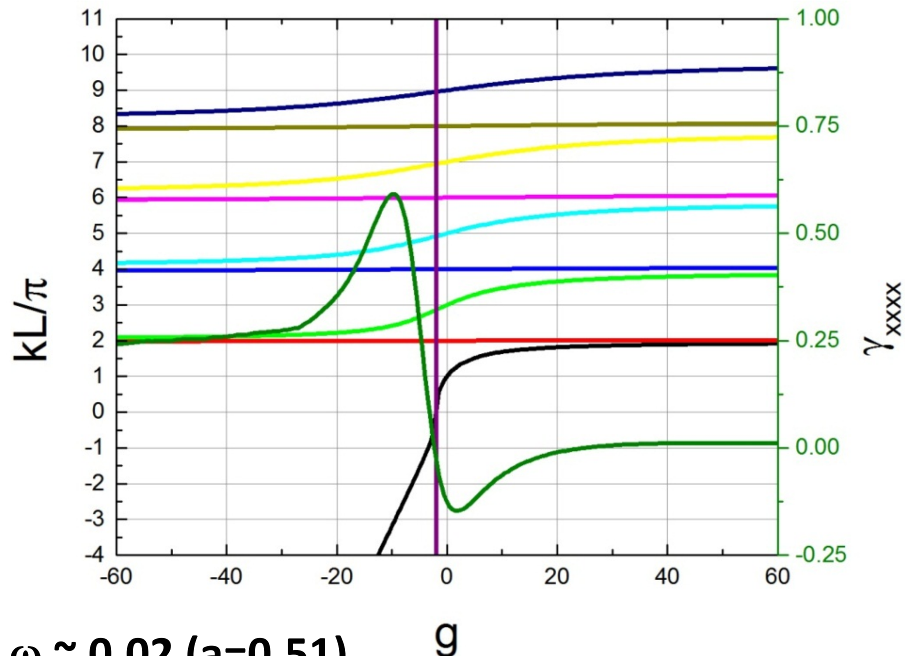
Finite potential allows a single negative energy state for negative $g<g_c$

$$-\imath x F_\delta(g; \omega, \kappa L) = x^2 \left[1 - \frac{g}{g_c}\right] + O(x^4), \ x \equiv \kappa L, \ g_c = 2/(\omega^2 - 1)$$

Spectra and hyperpolarizabilities for finite vertex potentials



ω ~ -0.44 (a=0.28)



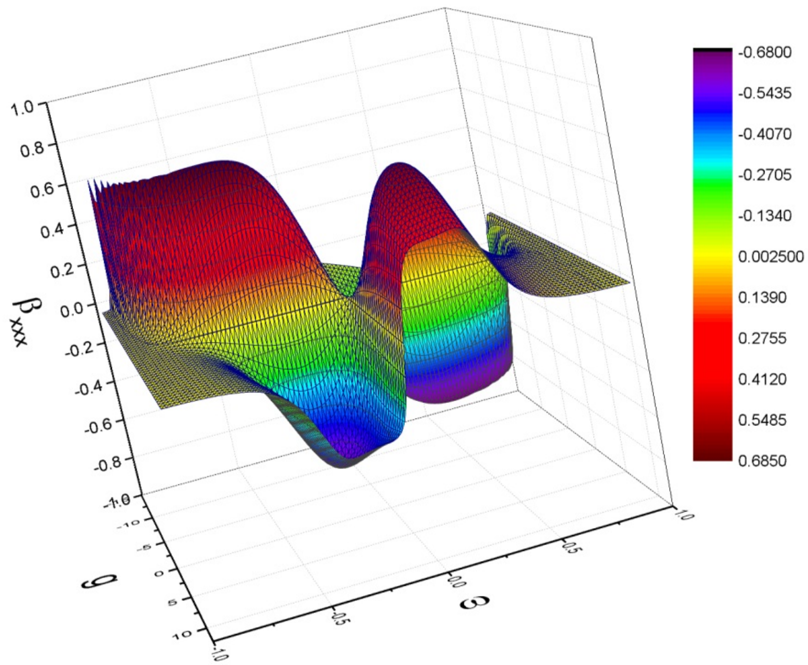
ω ~ 0.02 (a=0.51)

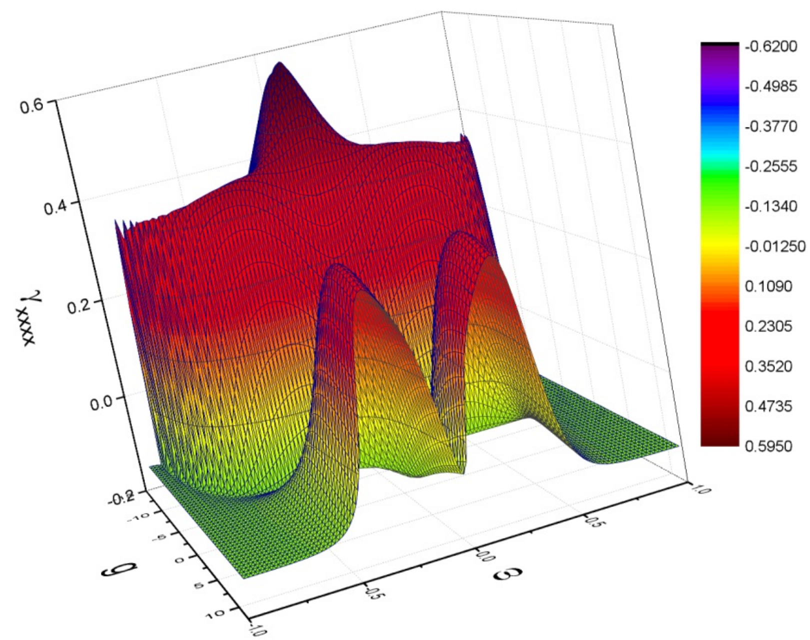
Positive eigenvalues lie between fixed root boundaries at nπ/L

Topologically-induced spectrum shift when negative energy state appears

Shift induces giant enhancement of nonlinearities. WHY?

3D views of ranges of hyperpolarizabilities

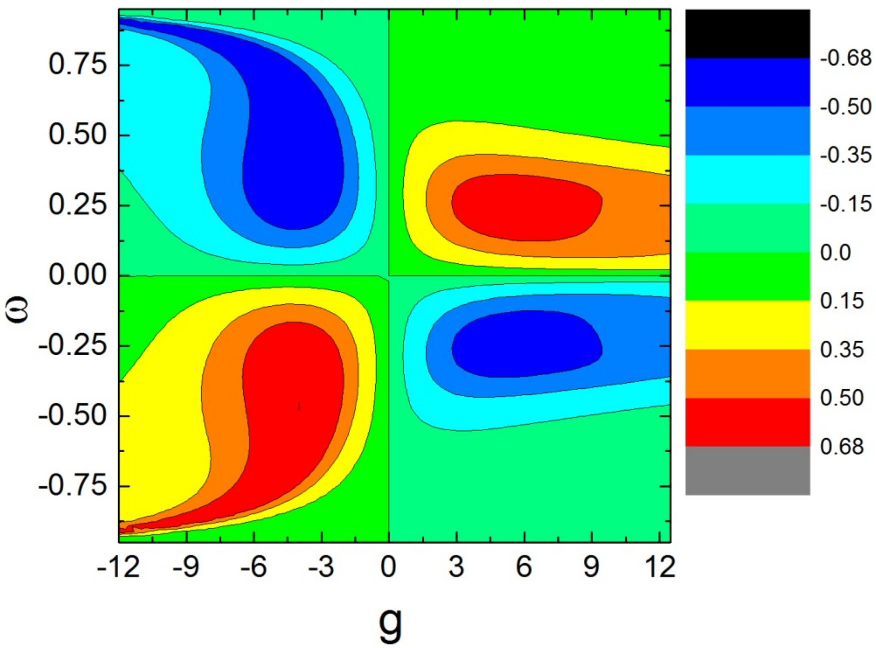


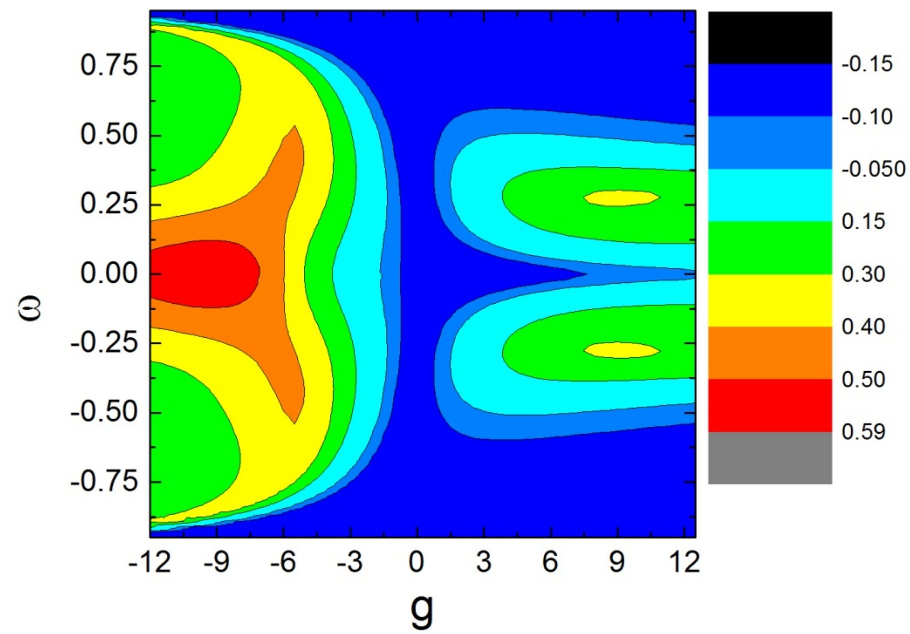


γ saturates at -0.126 when the δ function is moved to either edge (bare wire)

β vanishes when the δ function is moved to either edge or to the center

Contour views of ranges of hyperpolarizabilities





γ saturates at -0.126 when the δ function is moved to either edge (bare wire)

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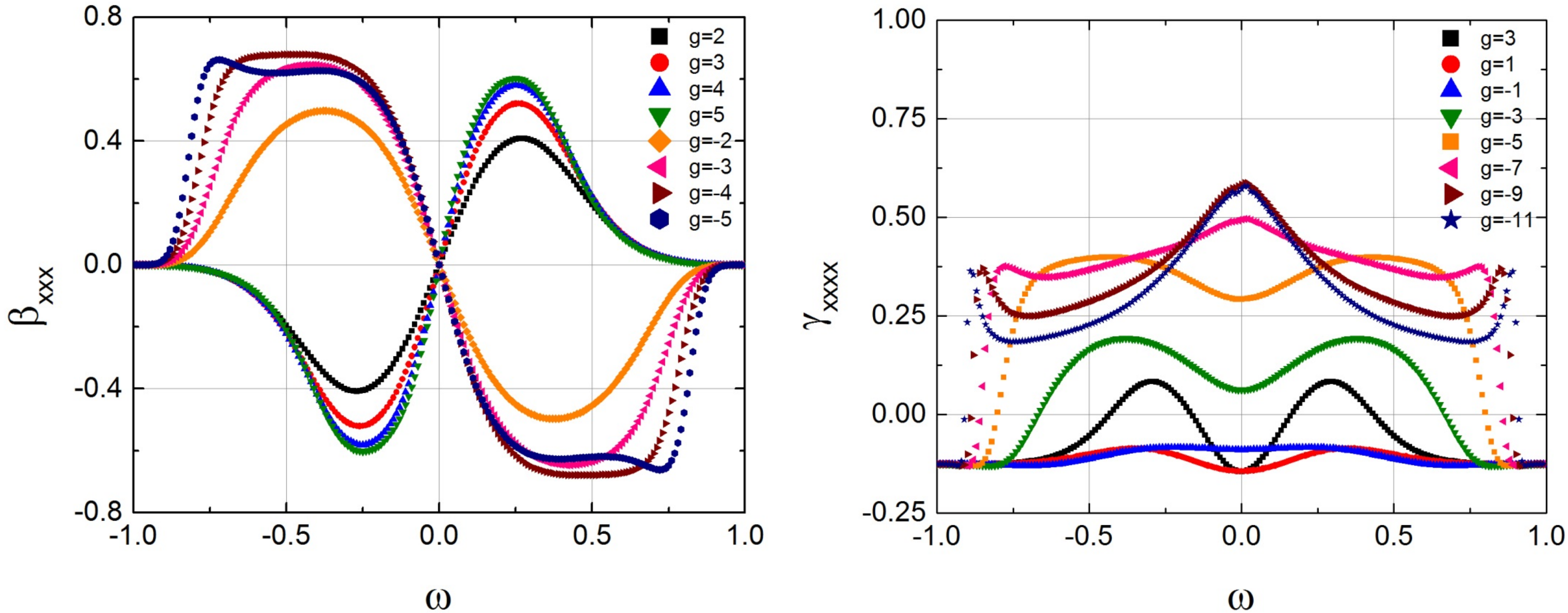
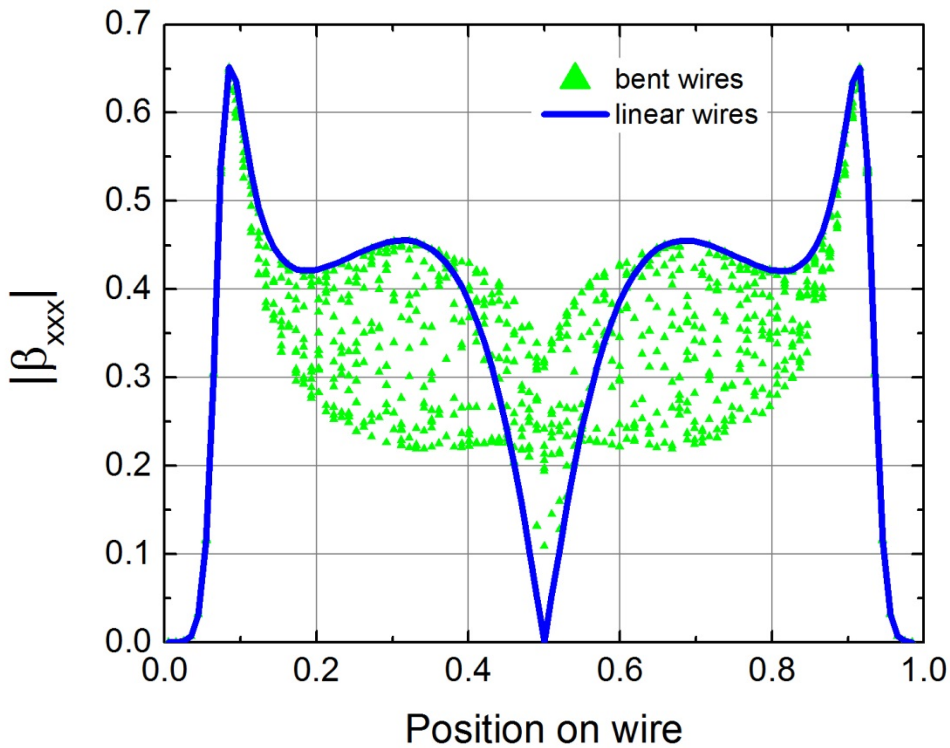
Ranges of hyperpolarizabilities across the atom



γ saturates at -0.126 when the δ function is moved to either edge (bare wire)

β vanishes when the δ function is moved to either edge or to the center

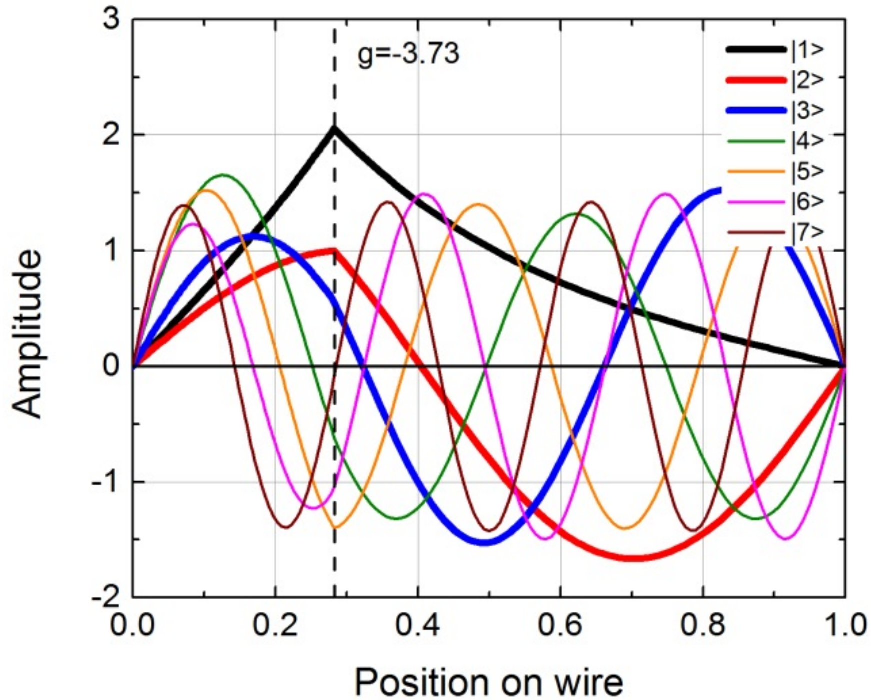
Effect of bending one edge relative to the other

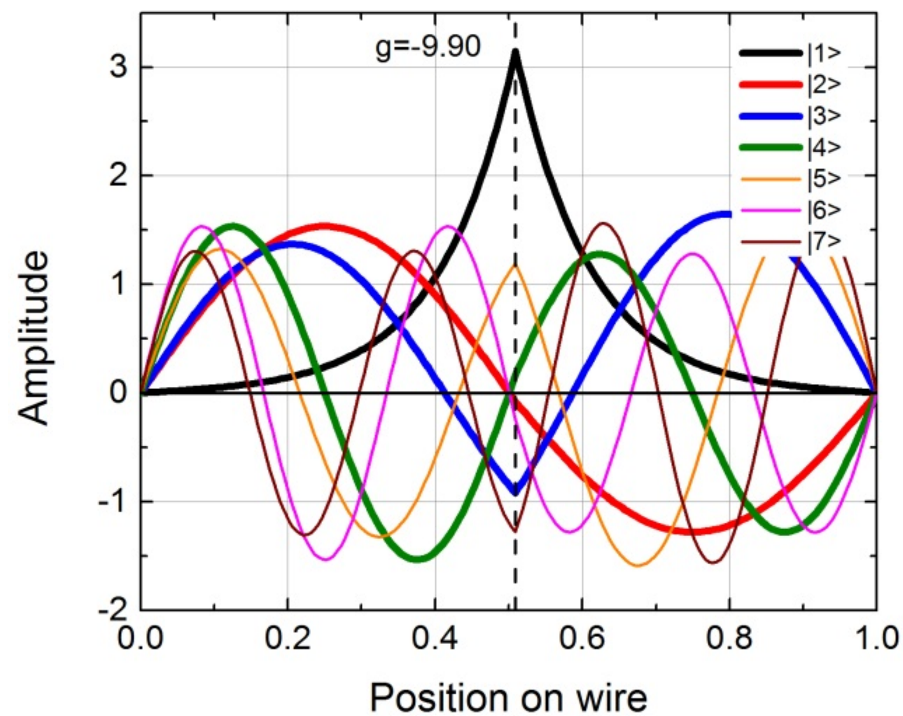


g = -7

Linear graph has the largest β except near the center of the graph

First seven eigenstates for best β and γ

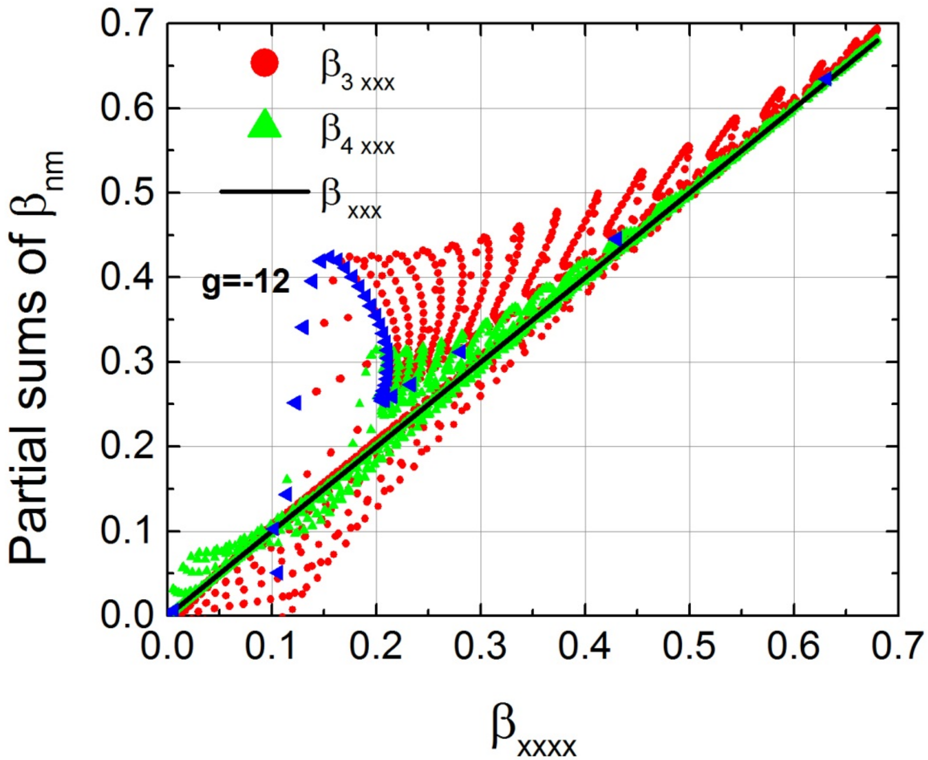


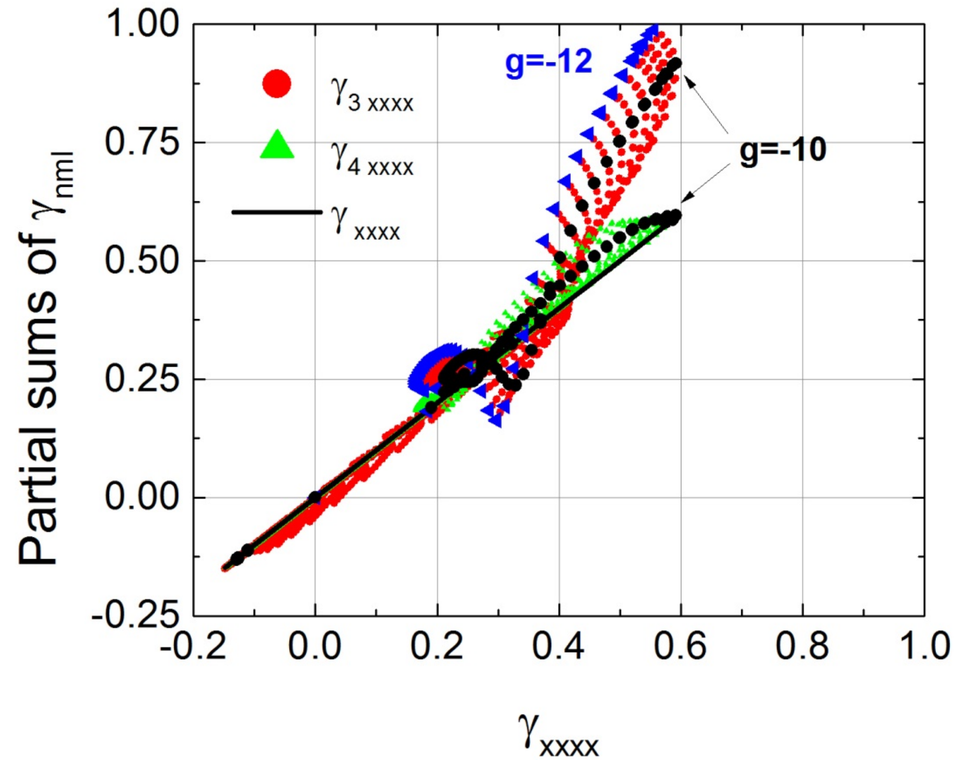


β ~ 0.68 **γ ~ 0.58**

Exquisite tuning of eigenstate overlap for transition moments

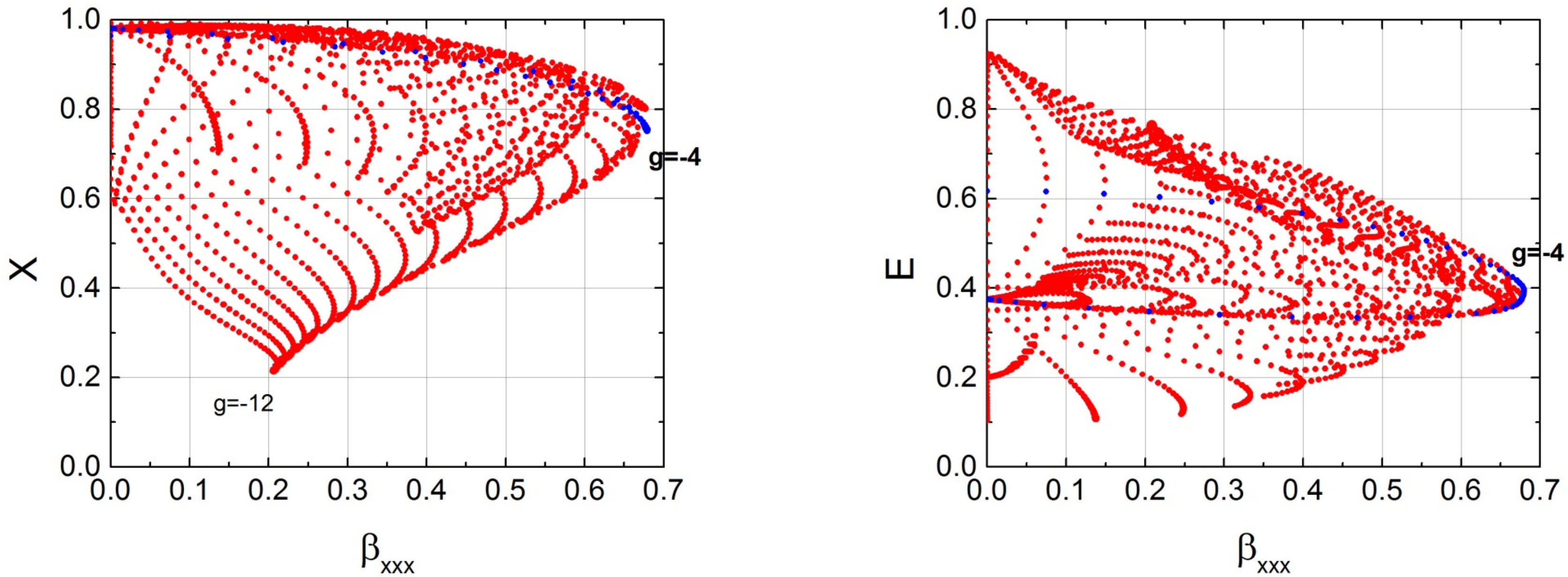
Three- and four-level Ansatz





g=-12 to +12.5 in steps of 0.5, each with 200 positions along the wire

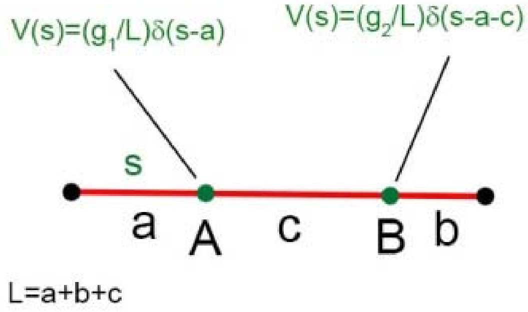
Convergence to universal scaling values for X and E



Normalized dipole moment X asymptotes to 0.79 at maximum β

Normalized energy ratio E asymptotes to 0.39 at maximum β

Compressed 2-delta molecule



$$\phi_a(s_a) = \frac{A \sin k s_a}{\sin ka}$$

$$\phi_c(s_c) = \frac{A \sin k(c - s_c) + B \sin k s_s}{\sin kc}$$

$$\phi_b(s_b) = \frac{B \sin k(s_b - b)}{\sin kb}$$

Overlapping motifs, with matched endpoints, yields secular functions for larger graphs

$$ZF_\delta = A \sin kb + B \sin ka$$

$$AF_\delta(g_1; a, c) = B \sin ka$$

$$BF_\delta(g_2; b, c) = A \sin kb$$

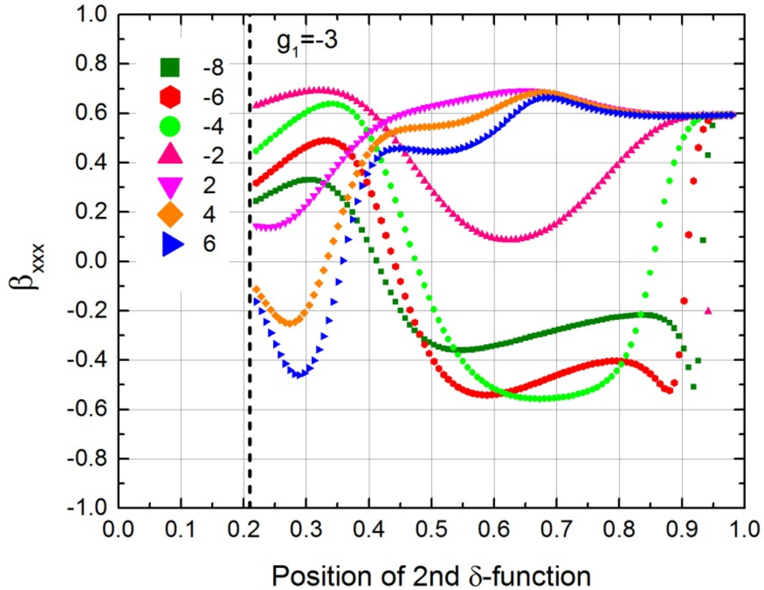
Secular equation determines eigenvalues for graph (two negative energy states possible)

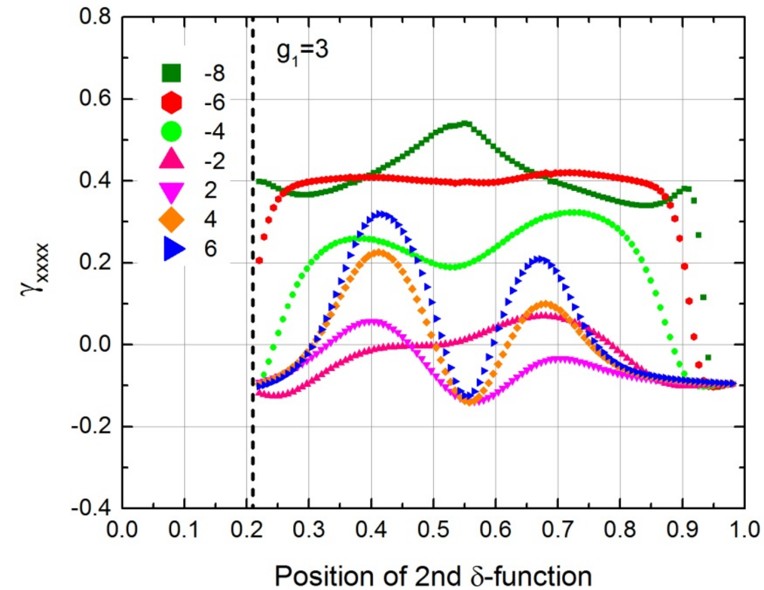
$$F_{2\delta}(g_1, g_2; a, b, c) = F_\delta(g_1; a, c) F_\delta(g_2; b, c) - \sin ka \sin kb = 0$$

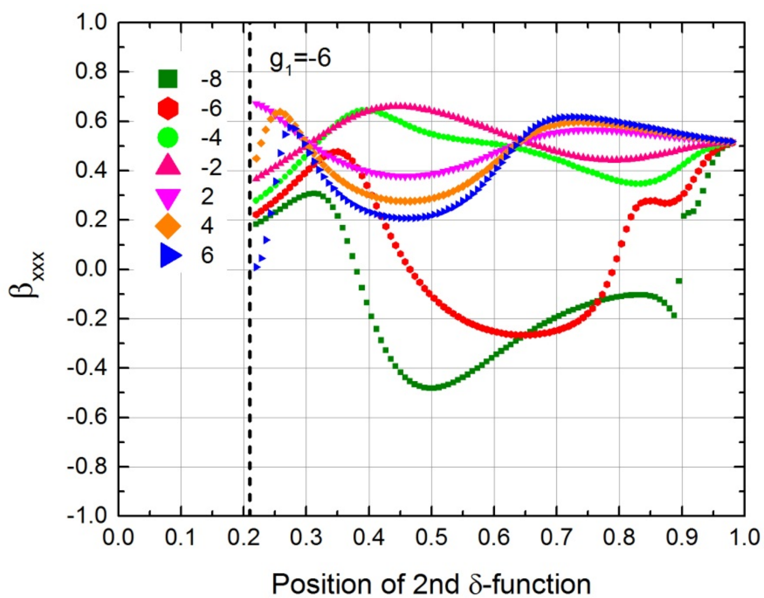
$$F_\delta(g_1; \omega_1, kL) = -(1/kL)\left[g_1(\cos kL_1 - \cos \omega_1 kL_1) - kL \sin kL_1\right]$$

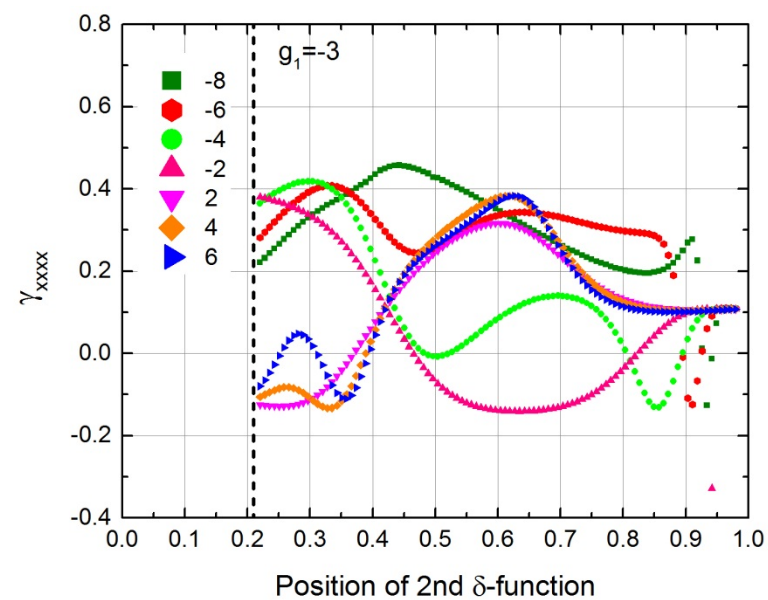
$$F_\delta(g_2; \omega_2, kL) = -(1/kL)\left[g_2(\cos kL_2 - \cos \omega_2 kL_2) - kL \sin kL_2\right]$$

Hyperpolarizabilities across wire

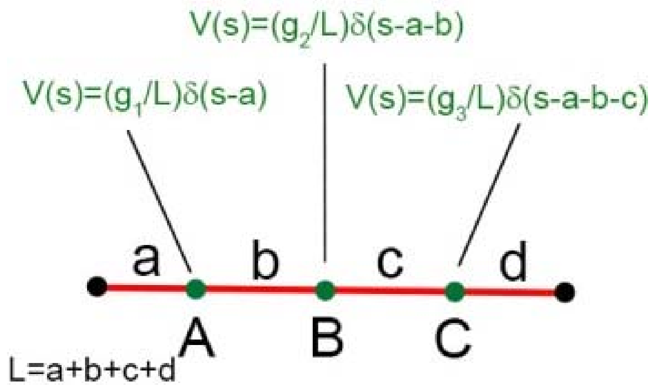








Compressed 3-delta molecule



Overlapping motifs

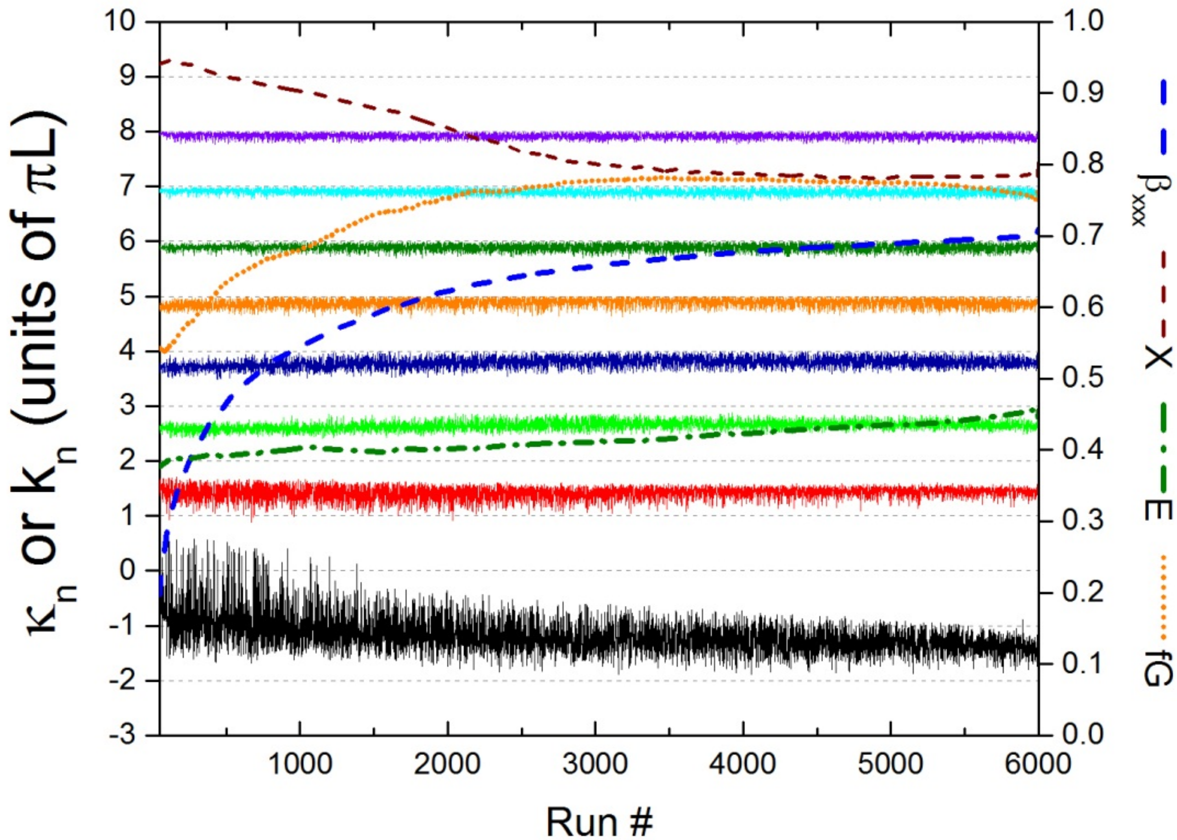
$$ZF_\delta = A\sin kb + B\sin ka$$

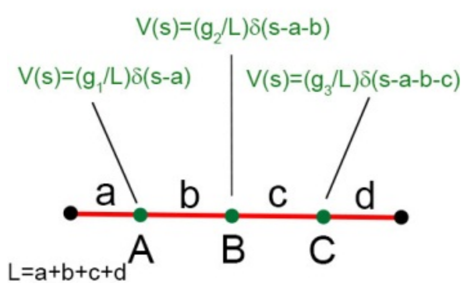
$$\begin{aligned} AF_\delta(g_1;a,b) &= B\sin ka \\ BF_\delta(g_2;b,c) &= A\sin kc + C\sin kb \\ CF_\delta(g_3;c,d) &= B\sin kd \end{aligned}$$

Secular equation determines eigenvalues for graph (three negative energy states possible)

$$\begin{aligned} F_{3\delta}(g_1,g_2,g_3;a,b,c,d) &= F_\delta(g_1;a,b)F_\delta(g_2;b,c)F_\delta(g_3;c,d) \\ &- F_\delta(g_1;a,b)\sin kb\sin kd - F_\delta(g_3;c,d)\sin ka\sin kc \\ &= 0 \end{aligned}$$

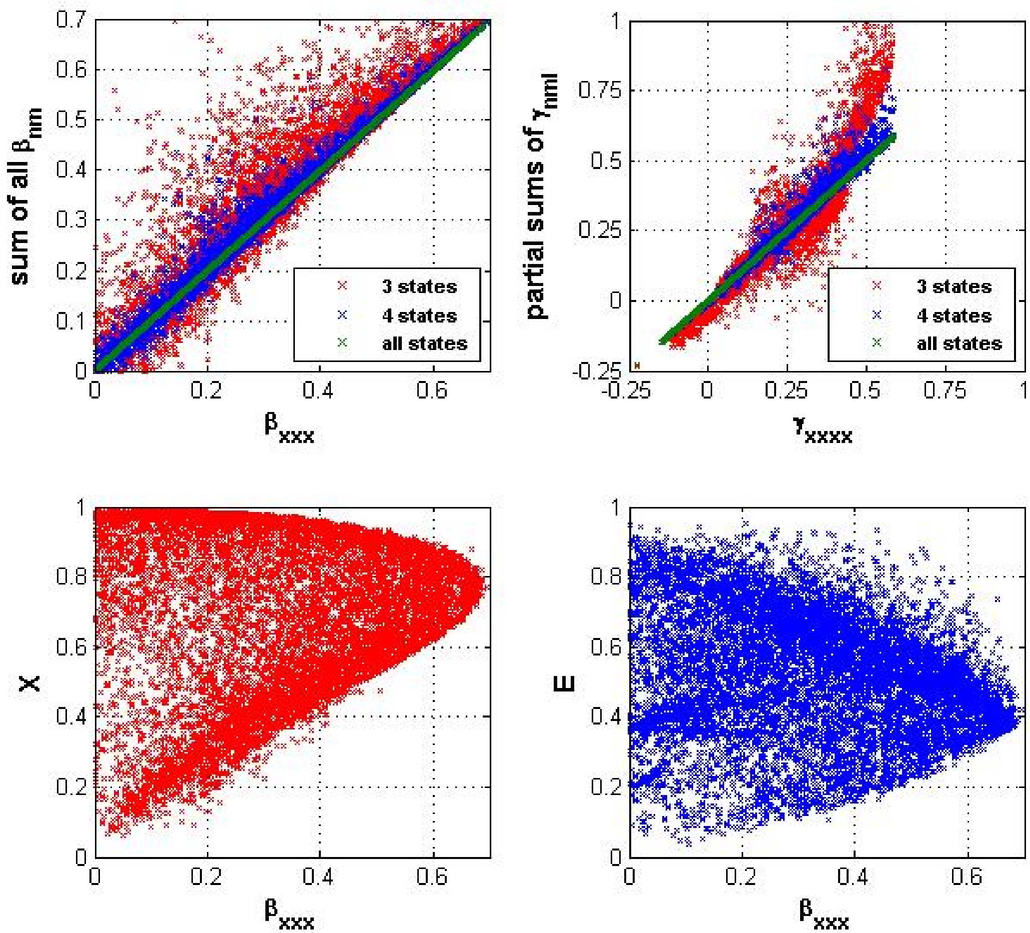
Spectral variation for random δ strengths/locations



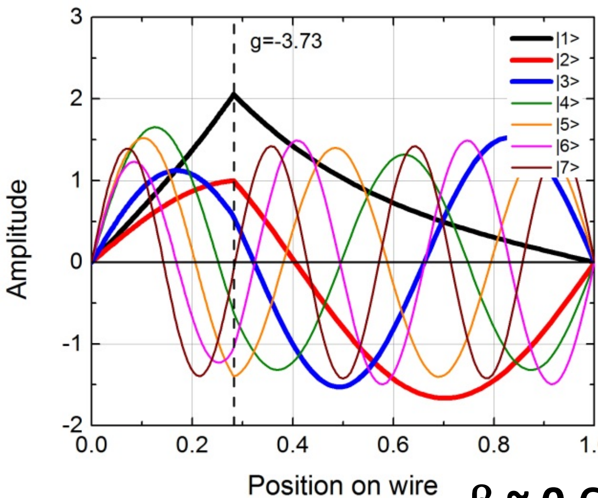


10,000 3-delta, random positions

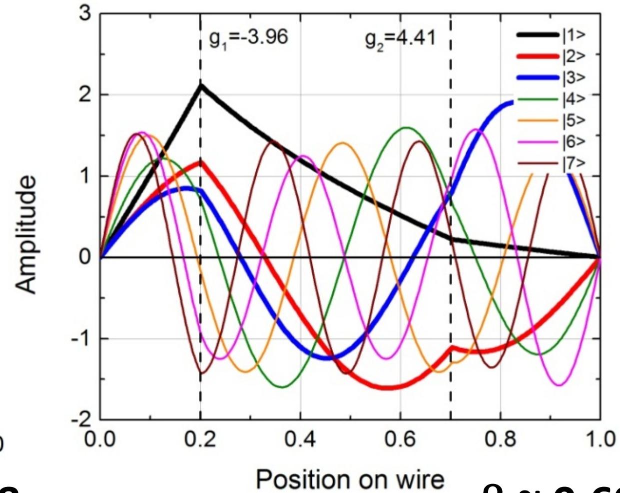
$|g_1| < 8, \ |g_2| < 8, \ |g_3| < 8$



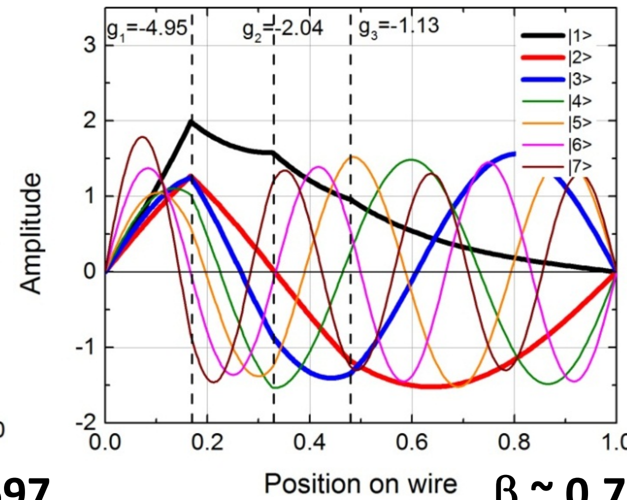
Optimization of eigenstate overlap for max β and γ



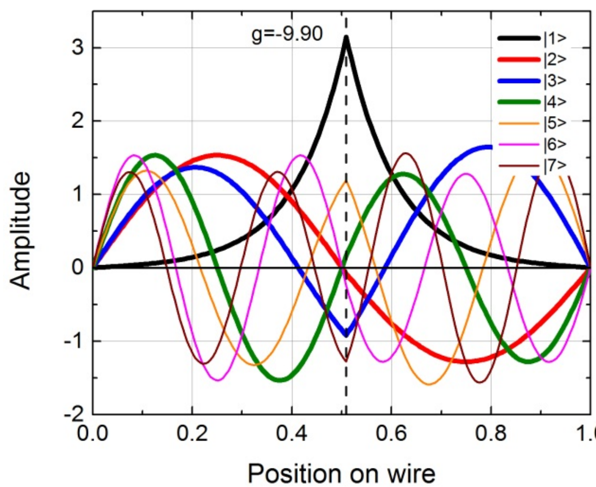
β ~ 0.68



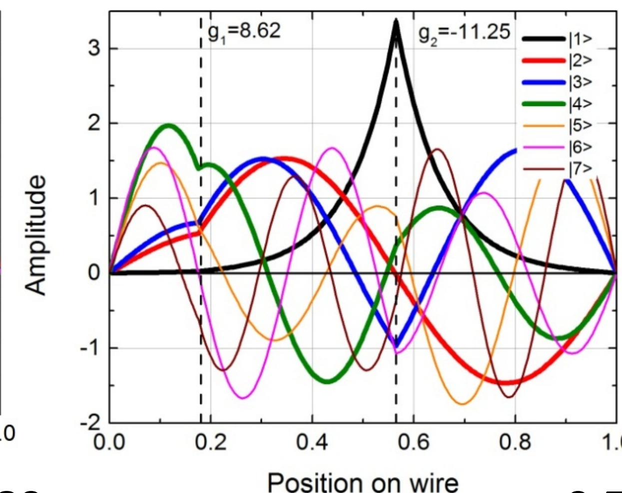
β ~ 0.697



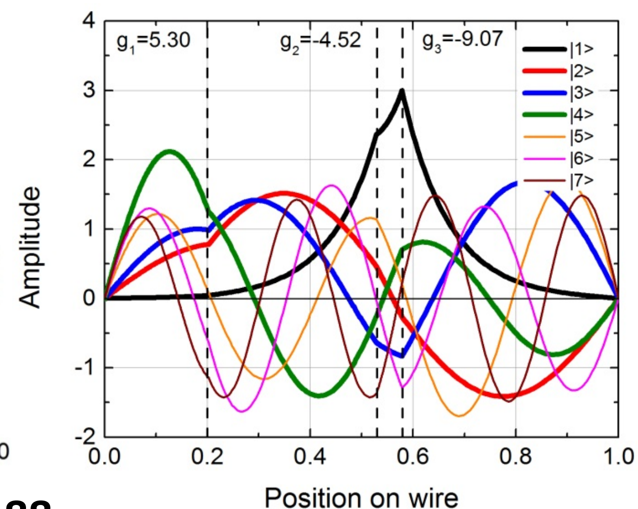
β ~ 0.709



γ ~ 0.580

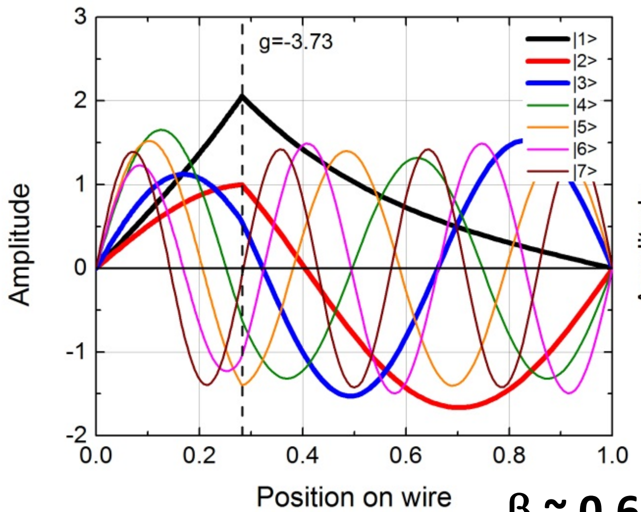


γ ~ 0.588

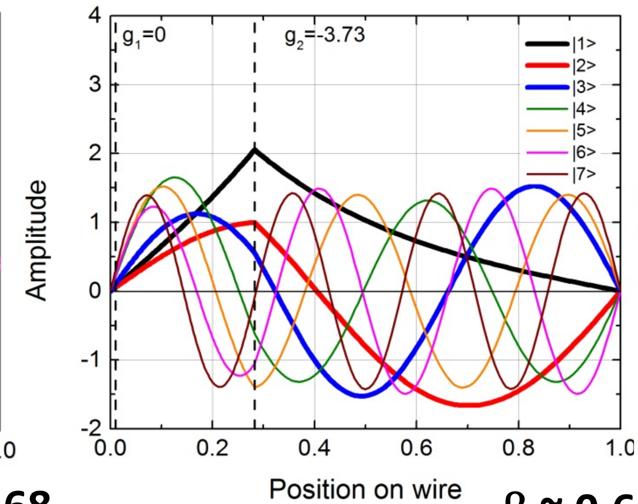


γ ~ 0.590

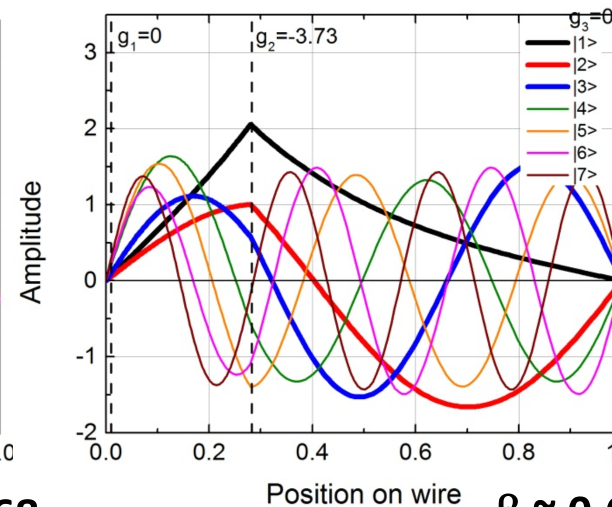
Consistency check for 1,2, & 3 δ atoms



β ~ 0.68

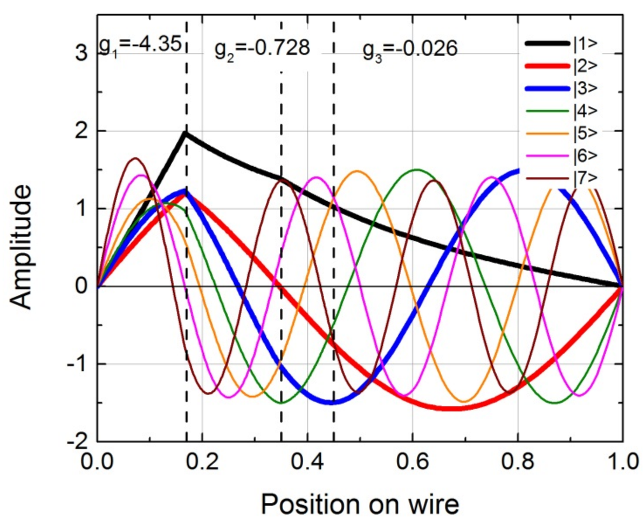


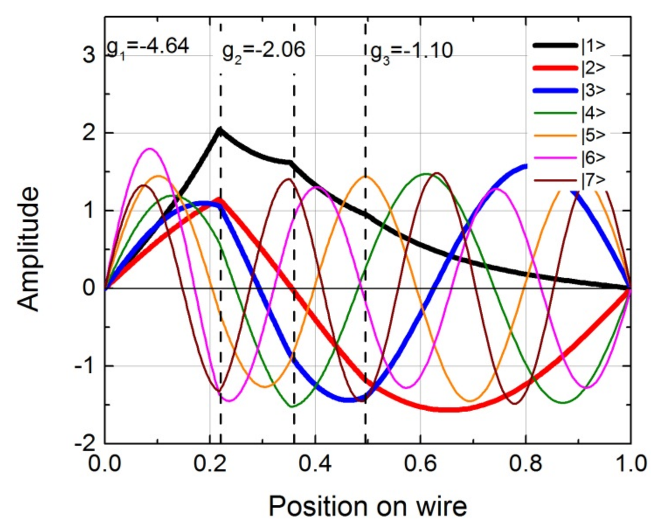
β ~ 0.68



β ~ 0.68

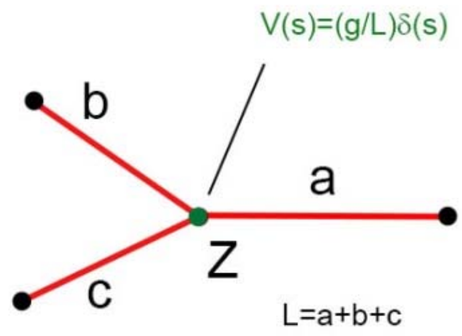
β ~ 0.705





β ~ 0.705

3-star delta graph



$$\phi_a(s) = \frac{Z \sin k(a - s_a)}{\sin ka}$$
$$\phi_b(s) = \frac{Z \sin k(b - s_b)}{\sin kb}$$
$$\phi_c(s) = \frac{Z \sin k(c - s_c)}{\sin kc}$$

Secular function for star graph

$$F_{star}(a,b,c) = \frac{1}{4}\left[\cos k_n L_1 + \cos k_n L_2 + \cos k_n L_3 - 3\cos k_n L\right]$$
$$L = a + b + c,\ L_1 = |a + b - c|,\ L_2 = |a - b + c|,\ L_3 = |a - b - c|$$

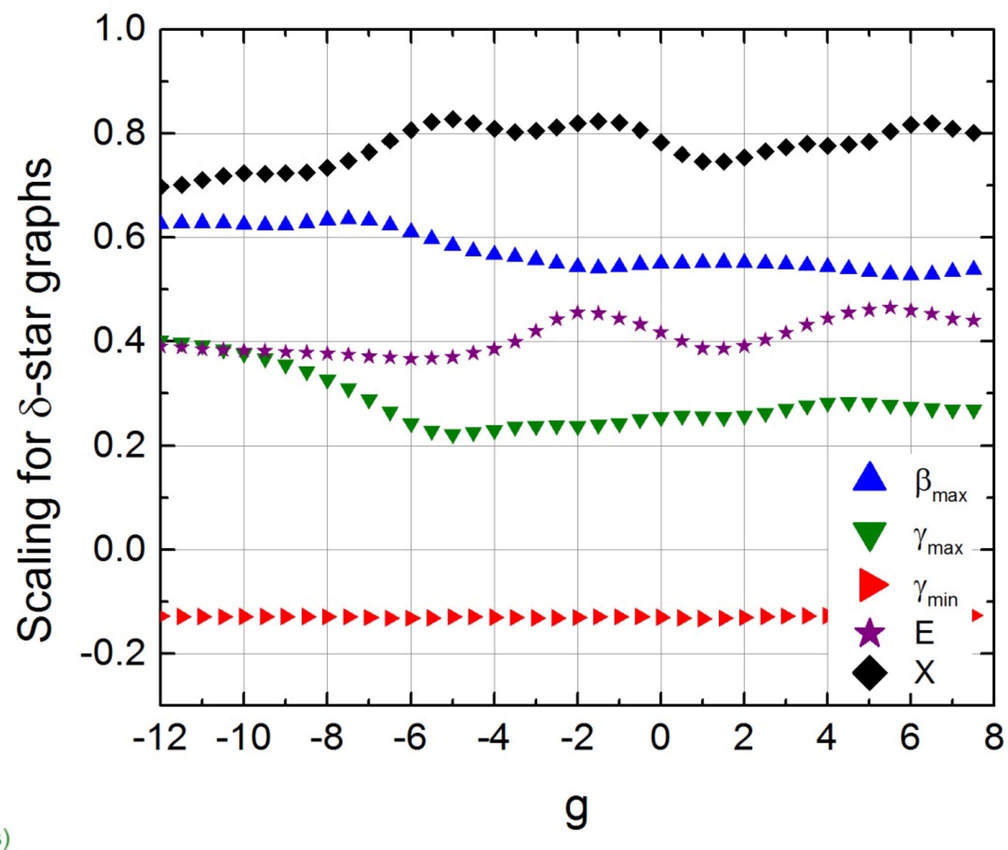
Secular function for star delta graph

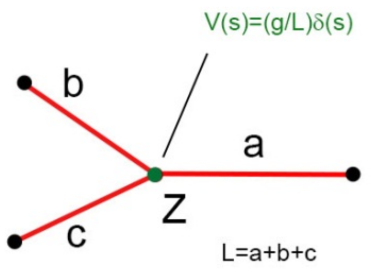
$$F_{star}(a,b,c) + (2g/kL)\sin ka \sin kb \sin kc = 0$$

$$g_c = -\frac{(a+b+c)(ab+ac+bc)}{2abc}$$

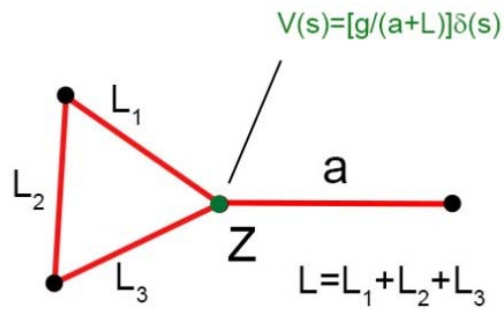
$g<g_c$ yields one negative energy state

Scaling of β, γ, X, and E for delta star graph





Delta lollipop graph



$$\phi_a(s) = \frac{Z \sin k(a - s_a)}{\sin ka}$$

$$\phi_L(s) = \frac{Z \sin k(b - s_L) + Z \sin k s_L}{\sin kL}$$

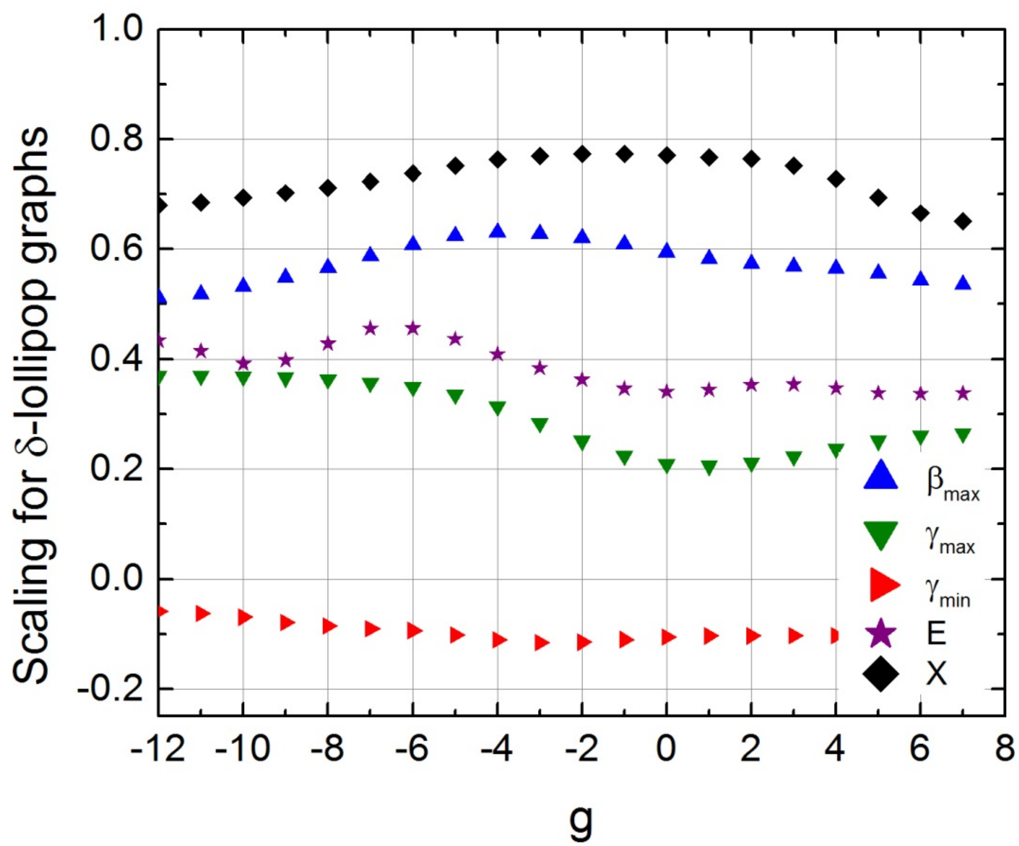
Secular function for lollipop graph

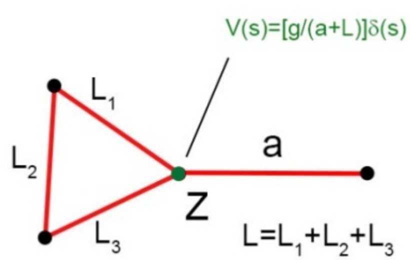
$$F_{pop}(a, L) = \frac{1}{2}\left[3 \cos k(a + L/2) - \cos k(a - L/2)\right]$$

Secular equation for lollipop delta graph

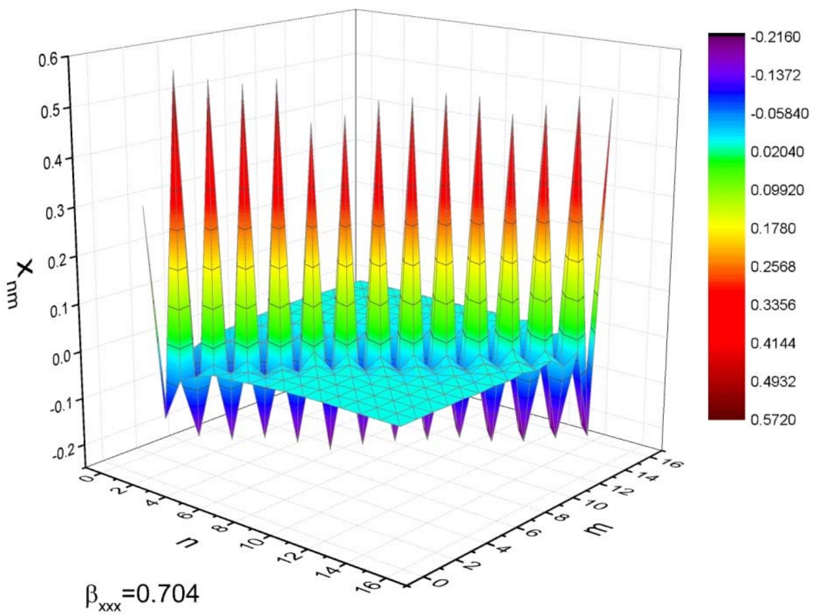
$$F_{pop}(a, L) + \frac{2g}{k(a + L)} \sin ka \cos kL/2 = 0$$

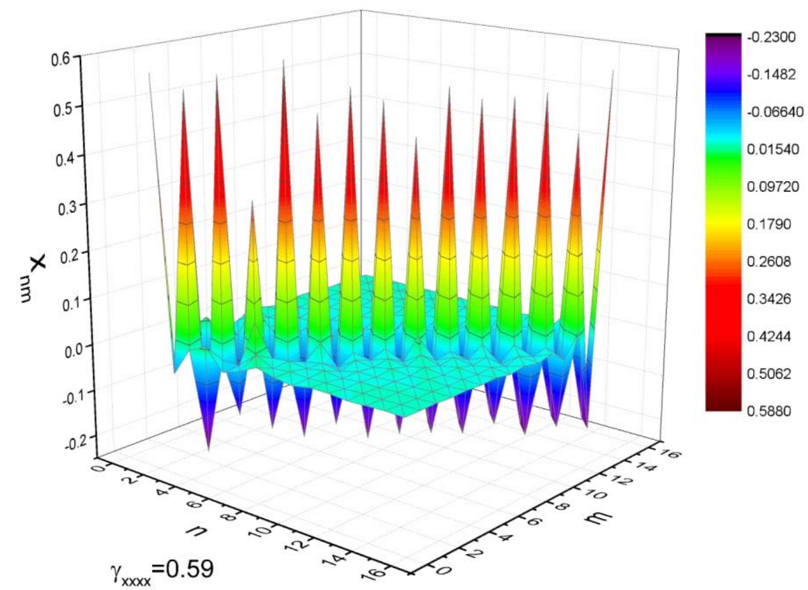
Scaling of β, γ, X, and E for delta lollipop graph





Transition moments for 3δ graphs at max β, γ



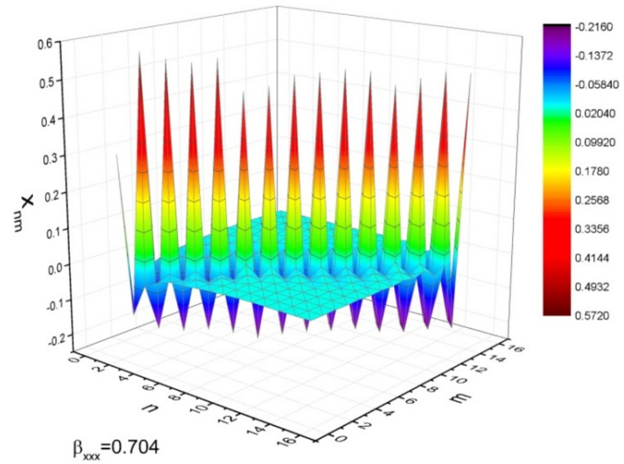


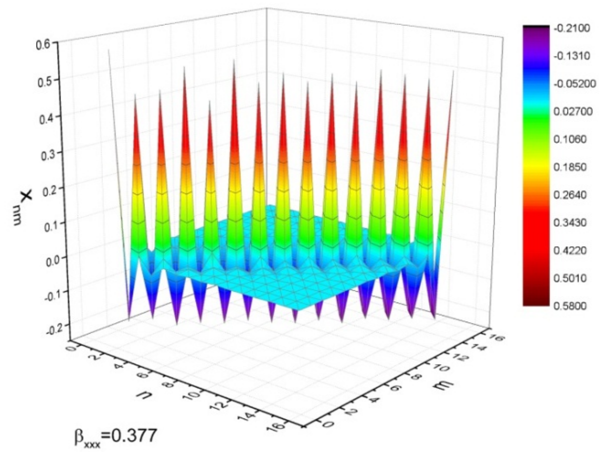
$$\beta_{xxx} = \sum_{n,m}{}' \beta_{xxx}(n,m) = 3^{3/4} E_{10}^{7/2} \sum_{n,m}{}' \frac{x_{0n} \bar{x}_{nm} x_{m0}}{E_{n0} E_{m0}}$$

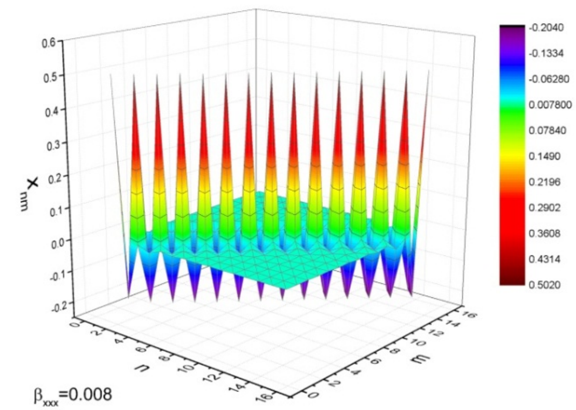
$$\beta_{xxx}^{3L} = 3^{3/4} E_{10}^{7/2} \left(\frac{|x_{01}|^2 \bar{x}_{11}}{E_{10}^2} + \frac{|x_{02}|^2 \bar{x}_{22}}{E_{20}^2} + \left[\frac{x_{01} x_{20} x_{12}}{E_{10} E_{20}} + c.c. \right] \right)$$

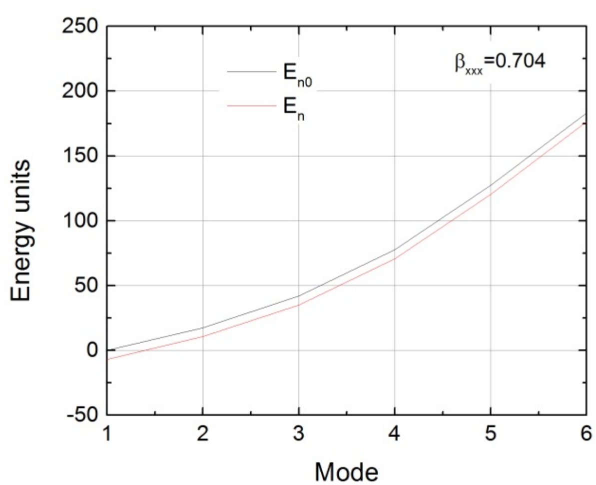
$$\beta_{xxx}^{4L} = \beta_{xxx}^{3L} + 3^{3/4} E_{10}^{7/2} \left(\frac{|x_{03}|^2 \bar{x}_{33}}{E_{30}^2} + \left[\frac{x_{03} x_{10} x_{31}}{E_{10} E_{30}} + c.c. \right] + \left[\frac{x_{03} x_{20} x_{32}}{E_{20} E_{30}} + c.c. \right] \right)$$

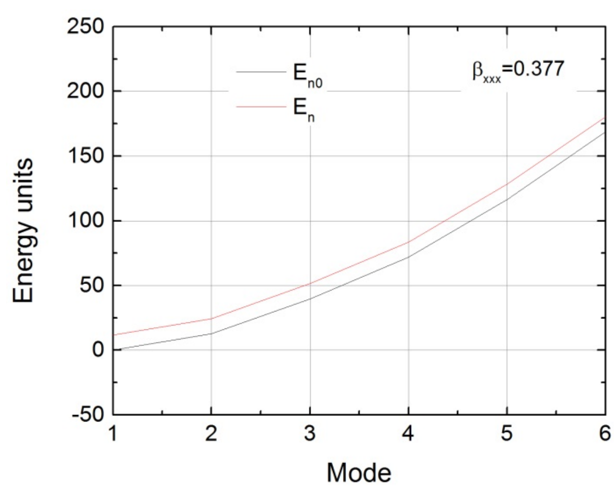
These are described by the 3L model

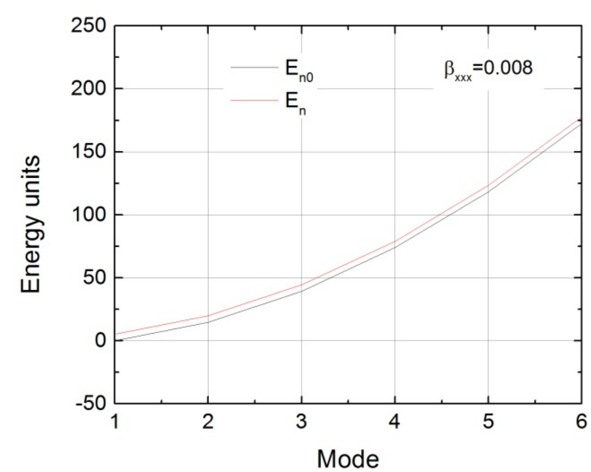




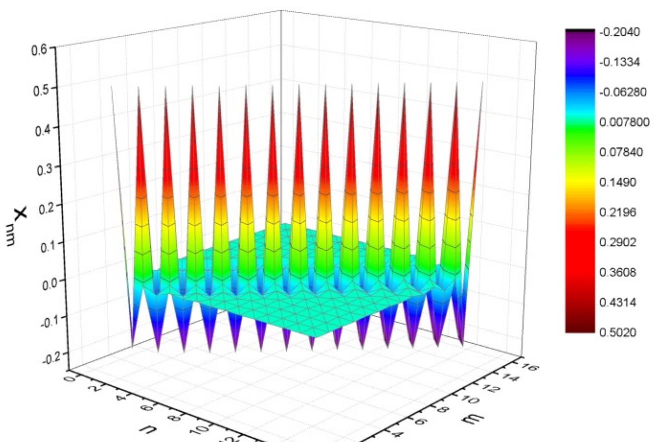




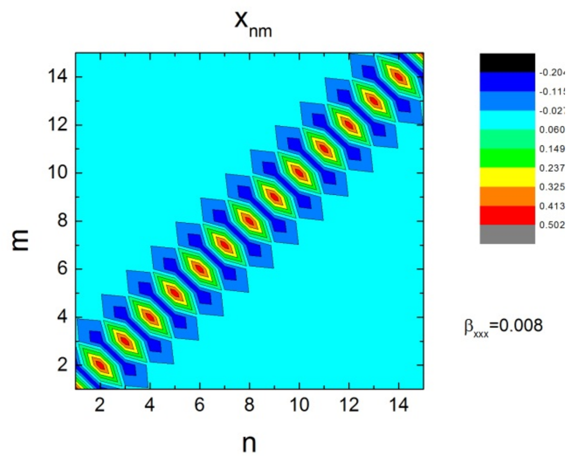


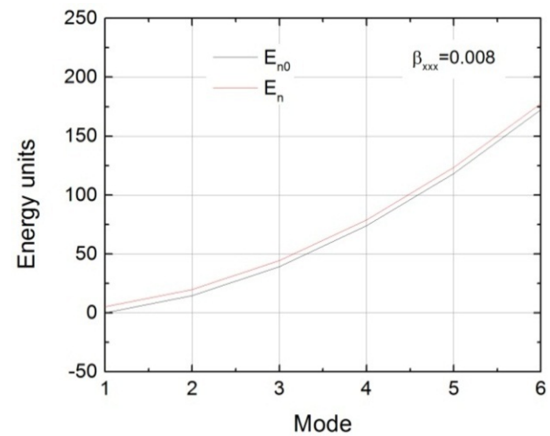


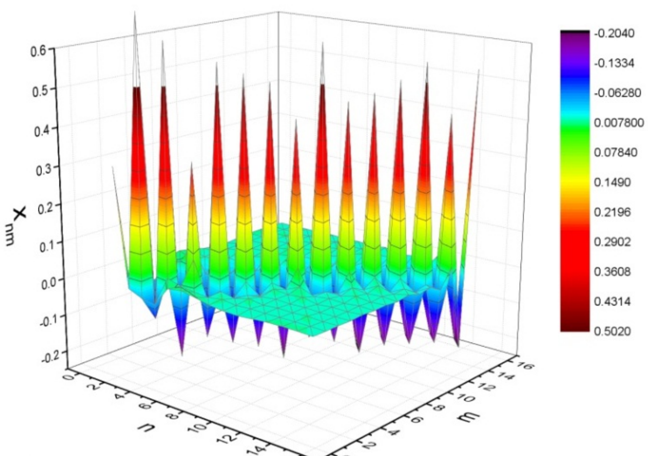
3L model for δ graphs with different g but β ~ 0



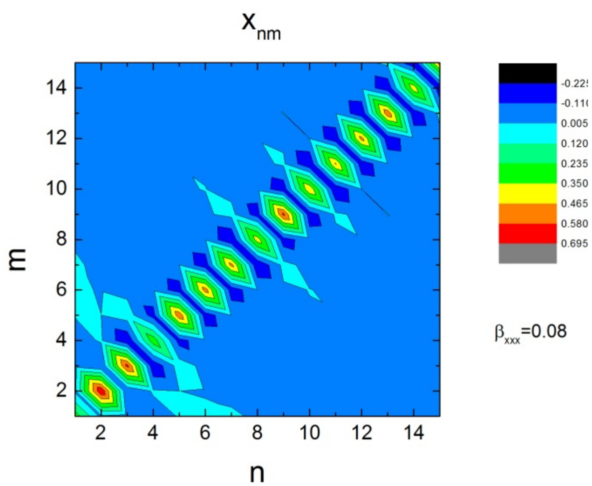
β_{xxx}=0.008

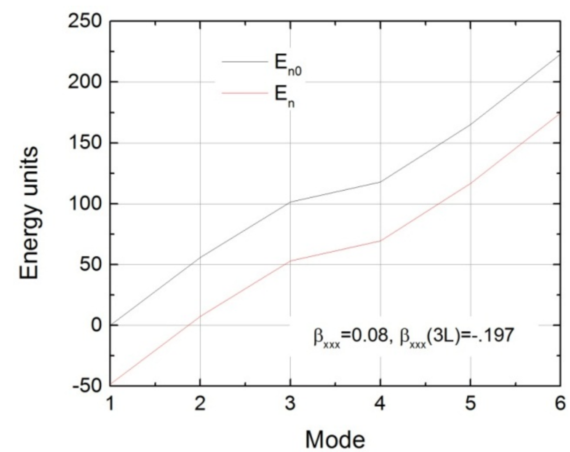






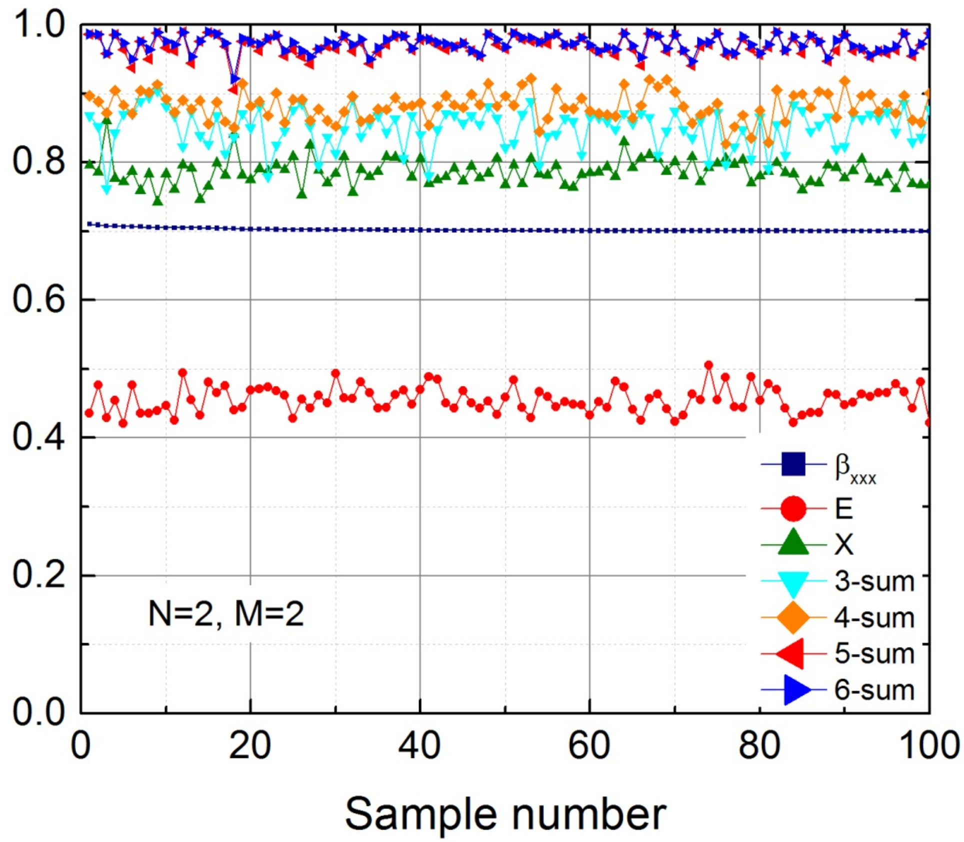
β_{xxx}=0.08, β_{xxx}(3L)=-.197





β_{xxx}(4L)=0.058

Partial sums when β_{xxx} is near its maximum value



Rick Lytel

August 7, 2013

Summary: For δ-dressed quantum graphs

Negative energy ground state significantly alters low-end spectrum,

enabling larger contributions from low-lying states for small β, γ

Scaling is nevertheless maintained near optimum values

Fine tuning of states and spectra possible in these graphs

Existence of δ-graphs with β_{max} ~ 0.71, γ_{max} ~ 0.6, γ_{min} ~ -0.15

Saturation of values with number of δ potentials ~ 3.

Rick Lytel

August 7, 2013