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1 function [logprob , varargout]=BXkinetics(params,data,sigma,t,powers)
2 % Function to evaluate kinetic model for biexciton and exciton states with
3 % poisson statistics:
4 %
5 % Full fluence series, no hot carrier thermalization, BX and X states, -
6 % k1,k2,XC
7 % BX-->k1-->X-->k2
8 % XC=cross section parameter (N/uW)
9 % Evaluates component dynamics
10 %
11 % Function evaluated the log likelihood function. The component
12 % spectra and fitted TA data are returned as optional outputs so that the
13 % same function can be used to evaluate the fit and produce visualizations
14 % after running the MCMC itself.
15 %
16 % Inputs:
17 % params = kinetic model parameters - rate constants and cross section
18 %         parameters
19 % data = Fluence dependent TA data stored as a 3D array, wavelength x time x
20 %         fluence
21 % sigma = standard deviations of the individual points in data, typically
22 %         calcualted as the sample standard deviation from the set of scans
23 % t = Time delays in ps, stored as a row vector
24 % Powers = Vector of pump powers from each measurement, in uW
25 %
26 % Outputs:
27 % logprob = log likelihood calculated from the sum of squared error
28 % spec = Component spectra (optional)
29 % datafit = Fitted TA spectra (optional)
30 %
31 % -Matthew Ashner, 2018
32
33 params(1:2)=10.^params(1:2); %Convert log rate constants to linear space
34 [specpts,tpts,numtrace]=size(data); %Determine data size and preallocate arrays
35 dyn=NaN(2,tpts,numtrace);
36 datafit=NaN(specpts,tpts,numtrace);
37
38 %Solve kintetic model by eigenvalue decomposition for each fluence
39
40 %Define kinetic model as a matrix, edit this line to change kinetic model
41 K=[-params(1),0;params(1),-params(2)];
42 [U,D]=eig(K); %Perform eigenvalue decomposition
43
44 numt=length(t);
45 numk=length(diag(D));
46
47 krep=repmat(diag(D),1,numt);
48 trep=repmat(t,numk,1);
49 eigdyn=exp(krep.*trep); %Dynamics in eigenbasis
50

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51 %Evaluate kinetics with fluence-dependent initial conditions, edit this
52 %section to change model for component initialization
53 for i=1:numtrace
54     %BX and X source terms from poisson distribution
55     BXyield=1-poisscdf(1,params(3)*powers(i));
56     Xyield=poisspdf(1,params(3)*powers(i));
57
58     %Generate source term in component basis, generate matrix transform
59     %that transforms source term to eigenbasis, evaluates dynamics, and
60     %transforms back to component basis
61     source=[BXyield;Xyield];
62     A2=U\diag(U\source);
63     dyn(:,:,:,i)=A2*eigdyn;    %Dynamics evaluated at input time points
64
65 end
66
67 %Concatenate fluence along time dimension to convert from 3D to 2D for
68 %linear least squares
69 data2=reshape(data,specpts,tpts*numtrace);
70 dyn2=reshape(dyn,2,tpts*numtrace);
71
72 spec=data2/dyn2; %Perform linear least squares
73
74 %Evaluate fitted TA data, back in 3D form
75 for i=1:numtrace
76     datafit(:,:,:,i)=spec*dyn(:,:,:,i);
77 end
78
79 %Evaluate log probability
80 logprob=-1*sum((datafit-data).^2./sigma.^2,'all');
81
82 %Output component spectra and fitted TA data if requested
83 varargout{1}=spec;
84 varargout{2}=datafit;
85 end

```