```
% Linearity Explanation

kb = 8.617e-5;  %boltzman constant (eV/K)
T = 298;         %Temperature K
Zmax = 60;
Zpos = linspace(0, Zmax, (Zmax+2)/2);

np = 16;
n1 = linspace(1,np,np);

sig = 0.5;  %Potential Profile
Vmax = +0.9;
eta = 0;

% scr function Linear+Expo

a1 = 0.0105;
a2 = 0.0269;
b1 = 2.0603e-06;
b2 = 0.7621;
zeta = a1.*n1+a2+b1.*exp(b2.*n1);

%from oxidation process
Gox = 0.1;       %Oxidation gibb's free energy
lam = 0.01;     %Oxidation Reorganization energy
gam = 1;       %Coupling strength to electrode

kET = zeros(np,size(Zpos,2));    %oxidation state
occ31 = ones(np,size(Zpos,2));    %to count how many molecules are
oxidized
occ22 = zeros(np,size(Zpos,2));    %to count how many molecules are
oxidized

mol22 = zeros(size(n1));
mol31 = zeros(size(n1));

phi = zeros(np,size(Zpos,2));
volt = zeros(np,size(Zpos,2));
series = zeros(np,size(Zpos,2));

for k = 1:np

% Potential Profile START
    for j = 1:size(Zpos,2)

        for i = 1:80
        p1=(Vmax/pi)*(F(i,sig,zeta(k))/
(i*(1+F(i,sig,zeta(k))))*((1+2*(eta-0.5))*sin(pi*i*(Zpos(j)/Zmax))-
((1-2*(eta-0.5))*sin(pi*i*(1-(Zpos(j)/Zmax)))));
        series(k,j)=series(k,j)+p1;
```

```
        end

    phi(k,j) = ((0-(Zpos(j)/Zmax))*Vmax)-series(k,j); % Original
Profile
    end
% Potential Profile END

% oxidation process START
    volt(k,:) = -phi(k,:);        % phi is wrt to vacuum; volt is the
voltage
    for k1 = 1:size(volt,2)
        kET(k,k1) = integral(@(x)(gam/(sqrt(4*pi*lam*kb*T))*exp(-
(lam+Gox-volt(k,k1)+x).^2/(4*lam*kb*T)).*(1-(1./(1+exp(x/
(kb*T))))),-5,+5);
        syms w
        occ22(k,k1) = round(int(dirac(w), w, -Inf, kET(k,k1)-0.6));
        occ31(k,k1) = (1-occ22(k,k1));
    end
% oxidation process END
    mol22(k) = (sum(occ22(k,:),2)/(size(Zpos,2)-1))*100;
    mol31(k) = (sum(occ31(k,:),2)/(size(Zpos,2)-1))*100;       %
Counting the molecules in 22 at kth pulse
end

%}
%%
figure(1) %Fig. S16(a)

R=linspace(0,1,16);
G= linspace(.45,.65,16);
B=linspace(1,0,16);
mypalette=[R',G',B'];
colororder(mypalette)

for k=1:size(phi,1)
    plot(Zpos,phi(k,:),'linewidth',3);
    hold on
end

plot([0 0],[-1 phi(1,1)],'k','linewidth',2);            % Plot left
electrode
plot([Zmax Zmax],[-1 phi(1,end)],'k','linewidth',2);    % Plot Right
electrode
hold off

colormap(mypalette)
cb=colorbar;
clim([1,16*10^3]);
cb.Ruler.Exponent=3;
```

```
xlabel('z (nm)');
ylabel('\Phi_{mol} (n,z) (eV)');
fontsize(gcf,16,"points")
%%
figure(2)

plot(n1,mol22,'Color','#A2142F','linewidth',3)
hold on
plot(n1,mol31,'Color','#77AC30','linewidth',3)
%xlim([0 16])
hold off
xlim("tight")
xlabel('# Pulses (\times10^3)');
ylabel('% Molecules');
title('Raman Data (31 and 22)');
legend('calculated (22)','calculated (31)','Location','northwest')
fontsize(gcf,16,"points")
set(gca,'XTick',(0:2:16))
set(gca,'YLim',[0 120])
%
%%
%Nucleation Theory+NLS Theory+function of z

kN = [0.3, 0.4, 0.5, 0.5, 0.5,   0.5, 0.5, 0.5,  0.5, 0.5, 0.5,  0.5,
0.5, 0.5,  0.5, 0.5, 0.5,  0.5, 0.5, 0.5,      0.5, 0.5, 0.5,
0.5, 0.5, 0.5,      0.5, 0.5, 0.5, 0.5];
chiN = [0.01, 0.05, 0.2, 0.5, 0.7,   1.3, 2.3, 2.7,  3.3, 4.3, 4.7,
5.3, 6.3, 6.7,  7.3, 8.3, 8.7,  9.3, 10.3, 11.3,    11.7, 12.3, 12.7,
13.3,14.3,15.1,    15.8, 16.1, 16.7, 17.1];

n = 1:1:16520;

f22 = zeros(size(n));
f31 = zeros(size(n));
p = zeros(size(n));
z = 1:1:30;

kN=kN*1e+3;
chiN=chiN*1e+3;

Nr = zeros(size(kN,2),size(n,2));

%NLS Eqn with arctan
for i=1:size(n,2)
    for k=1:size(kN,2)
        p(i)=p(i)+((1/pi).*(atan((n(i)-chiN(k))/kN(k)))+0.5);
    end
    f22(i)=(p(i)/30)*100;
    f31(i)=(1-(p(i)/30))*100;
end
```

```
%arctan distribution
for j=1:size(kN,2)
    for m=1:size(n,2)
        Nr(j,m)=((1/pi).*(atan((n(m)-chiN(j))/kN(j)))+0.5);
    end
end

% Applying Spline function
sx = linspace(1,16520,2000);
sr22 = spline(n,f22,sx);
sr31 = spline(n,f31,sx);

%%
figure(13) %Fig. S16(c)

plot(sx,sr22,'Color','#EDB120','linewidth',3)
hold on
plot(sx,sr31,'Color','#0072BD','linewidth',3)
hold off

xlabel('number of pulses (n)');
ylabel('%f_{22}(n)');
legend('22 state','31 state','Location','northwest')
fontsize(gcf,16,"points")
xlim("tight")
set(gca,'XTick',(0:4000:16001))
set(gca,'YTick',(0:40:100))
set(gca,'YLim',[0 100])
box on

%%
figure(16) %Fig. S16(b)

cmap = flip(autumn(30),1);
set(gca(),'ColorOrder',cmap)
hold on
for k=1:size(Nr,1)
    plot(n,Nr(k,:),'linewidth',3);
    hold on
end
hold off

colormap(cmap)
colorbar
clim([1,30]);

xlabel('number of pulses (n)');
ylabel('Nr(z)');
fontsize(gcf,16,"points")
```

```
set(gca,'XTick',(0:4000:16000))
xlim("tight")
box on
%%
function y = F(n,sig,zeta)
%lam = 0.05;
%sig = 0.5;
chi = 0.5*(2*pi*n*sig).^2;
y = 0.5*((sig/zeta).^2)*integral(@(u)(1./u).*exp(chi-u),chi,+inf);
end
```