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The Relationships between Optical Dephasing Times, Lineshape Functions and Coherent Frenkel Exciton States.

by

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ABSTRACT

A theory for the optical dephasing times and the homogeneous component of the absorption lineshape function to the $k=0$ levels of Frenkel exciton states is presented. It is shown that two different types of phonon-exciton scattering processes can dephase the optical transition to $k=0$. The first is a Raman or Orbach multiple scattering process which results in $k$ scattering in the exciton states and leads to the loss of coherence of the exciton wavepacket. This causes a temperature dependent linebroadening but need not result in temperature dependent frequency shift. The second process is a single phonon interaction where an exciton absorbs and subsequently emits a phonon with no change in phonon or exciton wavevector. This process causes both a temperature dependent linebroadening and a temperature
dependent frequency shift in the optical spectrum. In this instance the optical dephasing time results from energy differences of the phonon branch in the ground and excited states rather than from phonon-exciton scattering and consequently it does not lead to the loss of coherence of the exciton wavepacket. Finally, a simple criterion for distinguishing the two mechanisms is derived that allows a quantitative measure of the phonon-exciton interaction to be determined from experiments.
I. Introduction

Over the years a great deal of effort has been expended\(^1\) trying to relate the linewidths of optical transitions in molecular solids to phonon interactions with Frenkel exciton states\(^2\). It has usually been assumed that phonon-exciton scattering times can be obtained directly from the homogeneous linewidths and that the coherent nature of the exciton state can be established to a first approximation, from the ratio of the linewidth vs. the intermolecular interaction which gives rise to the band dispersion. More recently, electron spin resonance techniques\(^4\)\(^-\)\(^10\) have been used to study the \(k\)-scattering in exciton bands and to determine the coherence times of Frenkel exciton states. It has been shown from these experiments that at low temperatures, exciton migration can be described as coherent because the phonon-exciton scattering times, being \(10^{-6}\) to \(10^{-8}\) seconds, are several orders of magnitude longer than the intermolecular exchange times. In many cases however optical linewidths in emission or absorption would suggest picosecond coherence times. At first sight this might appear to be a contradiction with the EPR results. Linewidths, however, are subjected to inhomogeneous broadening and thus phonon-exciton scattering times determined from linewidth measurements alone are not meaningful unless the linewidth is related to the homogeneous component of the relaxation spectrum. Even more important, as will be shown in this paper, the homogeneous portion of the optical linewidth can result from phonon-exciton interactions which do not cause a loss of the exciton coherence. Still, some theories\(^11\) have assumed a homogeneous lineshape function and have been brought forth to explain away the
apparent discrepancy between the optical lineshape function and the electron spin-resonance lineshape function. More recently, optical coherence experiments\textsuperscript{12-14} have been brought to bear on some of the problems. As yet, however, no clear understanding of the mechanism for optical dephasing times (homogeneous $T_2$ processes), absorption linewidths, and $k$-scattering in the band states has emerged.

In this paper a simple and straightforward theory for the dephasing times of the homogeneous component of the optical absorption linewidth to the $k=0$ levels of the exciton states is presented. It will be shown what explicit features of phonon-exciton scattering are responsible for optical transition linewidths and how these features differ from the scattering that is responsible for the loss of coherence in Frenkel exciton bands as measured from EPR experiments. In addition it will be shown that the optical linewidths associated with the $k=0$ state need not measure features of the exciton coherence at low temperatures (2-50\textdegree{}K) because the optical transition to $k=0$, in many cases, may be in "intermediate exchange" with optic branch phonon transitions. As a consequence, optical dephasing times can result from energy differences of the phonon branch in the excited states relative to the ground state rather than phonon-exciton scattering events which cause a change in $k$.

Finally, it will be shown that even when the transitions can be considered homogeneous from linewidths vs. temperature measurements or from photon echo experiments, the correct phonon scattering times and relaxation times must come from an analysis
of both the homogeneous portion of the linewidth and the frequency shift with temperature.

II. General Considerations

In order to equate the lineshape function in optical spectroscopy or EPR experiments to coherent properties of excitons, the correlation function that describes the phonon-exciton scattering must also be responsible for the time dependence of the the electric dipole transition moment responsible for optical absorption to \( k=0 \) or the time dependence of magnetic dipole transition moment responsible for the lineshape function in the EPR experiments. Phonon-exciton scattering is well defined for a Markoffian process and for low temperature is given by the correlation function for the decay of the orbital component of \( k \). The explicit form has been developed by Grover and Silbey, for linear coupling, Munn and Siebrand for quadratic coupling and others from a generalized master equation. It should be self-evident that if the exciton coherence time is to be inferred from a spectroscopic linewidth then the transition lineshape correlation function (optical or EPR) must be associated with changes in the exciton wavevector \( k \) since these are the only events which cause a loss of coherence of the exciton wavepacket.

In the EPR experiments of Harris and co-workers, the lineshape function has been explicitly related to \( k \)-scattering and
was derived by considering the dispersion introduced into the EPR band-to-band transition by the spin-orbit interaction. It has been shown that when the $k$-scattering in the band is slower than the experimental correlation time, a characteristic lineshape is obtained, which for one dimensional excitons, displays a Boltzmann averaged density of states function. The experimental correlation time in the EPR experiment (which measures only one point in the time decay of the correlation function describing the excitons coherence) is essentially the reciprocal of width of the EPR band-to-band transition. The EPR experiments establish whether or not phonon-exciton scattering causes changes in $k$ to occur on a time scale fast or slow relative to the experimental correlation time.

What about homogeneous optical linewidths measured in absorption spectroscopy or optical dephasing times measured in a photon echo or similar experiment? Do they measure the same $k$-scattering or can they result from non-$k$ scattering events as do the EPR experiments? To answer this question it is necessary to look more carefully into the details of the scattering process.

Evidence from a variety of different experiments support the conclusion that the dominant interaction of Frenkel excitons at low temperatures is with the optic phonons. To understand the optical absorption lineshape function and its relation to exciton coherence we have to understand two features of this
phonon-exciton interaction. First, one must know whether or not the interaction of an optic phonon with the exciton state scatters the initial k=0 state (k_o) to another k' state (k'_o) or whether the optic phonon is simply absorbed and re-emitted without a change in q, a process that would not cause a change in k. Second, one must know how the ground zero point energy (|00>) and the exciton k_o state (|k_o>) are mixed with the ground state optic phonon(|0q>) and excited state optic phonon branches (|k_q>) via the scattering events. The effect of specific scattering mechanisms on the transition dipole moment can be qualitatively seen as follows.

Consider for example, the scattering mechanism from k=0 given by equation 1a below and illustrated in figure 1a

\[ |k_o\rangle + |q\rangle \xrightarrow{W^+} |k_q\rangle \xrightarrow{W^-} |k'\rangle + |q'\rangle \] (1a)

\[ |k_o\rangle + |q\rangle \xrightarrow{W} |k'\rangle + |q'\rangle \] (1b)

Equation 1a represents Orbach scattering with a real intermediate state |k_q⟩ while equation 1b represents a Raman scattering process.
In Orbach scattering the oscillating dipole associated with the transition moment \( \langle oo|\mu(t)|k_o0\rangle \) and the corresponding correlation function for describing the lineshape function would decay with the rate at which optic phonons were promoted and subsequently scattered from the state \( |k_0q\rangle \). At low temperatures the scattering rate \( W_+ \) is slower than \( W_- \), consequently, \( W_+ \) is the rate limiting step and is essentially identical to the rate at which the exciton \( k_o \) scatters to the other \( k' \) states. For an Orbach process \( W_+ \) is proportional to the Planck distribution function evaluated at the energy of the optic phonon responsible for scattering; i.e.

\[
W_+ \sim <n(\varepsilon_1)> 
\]  

(2a)

where

\[
<n(\varepsilon_1)> = \exp(\varepsilon_1/kT-1)^{-1} 
\]  

(2b)

which for low temperature is given by

\[
W_+ \sim \exp(-\varepsilon_1/kT) 
\]  

(2c)

For Raman scattering, the rate \( W \) is given by

\[
W \sim <n(\varepsilon_1)> <n(\varepsilon_1)+1> 
\]  

(2d)
The overall effect of Orbach or Raman scattering is to completely dephase the optical transition to \( k=0 \) because the final \( k' \) state has no transition probability for an electric dipole transition from the ground state zero point energy because of the \( \Delta k=0 \) selection rule. In such cases the correlation function for the exciton coherence time at \( k=0 \) and that responsible for the optical lineshape function in absorption to \( k=0 \) would be the same and the optical absorption lineshape function would indeed be a measure of coherence of the exciton state.

On the other hand, in the interaction of the exciton with the optic phonons, if a phonon of the same wavevector \( q \) was absorbed and subsequently emitted, the exciton would be returned to the same exciton wavevector state from which it was initially promoted. This is illustrated in figure 1b. The optical transition would be dephased by the process only if the lifetime in the phonon-exciton state \( \langle k_0 q \rangle \) were long compared to the frequency shift associated with a difference between excited and ground state phonon frequencies. From the point of view of exciton coherence it is clear that this mechanism does not lead to the loss of coherence of the exciton state since no change in the wavevector is effected, and thus the correlation functions for the decay of exciton coherence is not related to the linewidth of the \( k=0 \) absorption. In the following section a lineshape theory is developed that allows these processes to be distinguished from one another.
III. Exchange Theory of Optical Absorption Lineshape Functions and Dephasing Times in Frenkel Exciton States.

The question of how phonon scattering causes line broadening can be quantitatively resolved by considering the effects of exchange between the exciton state $|k_0\rangle$ and the phonon promoted state $|k_0q\rangle$. This approach is valid for a simple two state scattering process in all limits that are Markoffian and can be derived from exchange theory.$^{20,21}$ The significant feature of exchange theory that is relevant to the present case is that whenever two modes interact, an excitation of one mode can result in a line broadening and frequency shift of the other mode when the excitation returns to the initial state bringing with it phase memory of the upper state's transition dipole moment.

The time dependent states that can be mixed via exchange which are relevant for optical transitions $k = 0$ are given by all possible combinations of the exciton wavevectors and phonon wavevectors coupled via the electric dipole transition moment operator. They are

$$
<oo|\hat{\mathbf{u}}(t)|k_00>, \\
<oo|\hat{\mathbf{u}}(t)|k_0q>, \\
<oo|\hat{\mathbf{u}}(t)|k'q'>, \\
<oq|\hat{\mathbf{u}}(t)|k_00>, \\
<oq|\hat{\mathbf{u}}(t)|k_0q>, \\
<oq|\hat{\mathbf{u}}(t)|k'q'>,
$$

Of these however, only two

$$
<oo|\mathbf{u}(t)|k_00> \\
and \\
<oq|\mathbf{u}(t)|k_0q>
$$
carry comparable oscillator strength. The others are zero in the weak coupling limit because of $\Delta k=0$ and/or $\Delta q=0$ selection rules for electric dipole transitions.

The application of exchange theory to the optical absorption at $k=0$ or optical dephasing times (in the case of photon echo experiments) is presented in figures 2 and 3. The lowest optic phonon branch in the ground state is taken to be at an energy $\varepsilon_1$, while in the excited state it is $\varepsilon'_1$ and for simplicity only one mode is assumed to be responsible for dephasing of the optical transition in a given temperature range. The transition from the ground state, $<00|$ to the exciton $<k_00|$ state is given by a frequency $\omega_0$ and the phonon scattering or absorption from $<k_00|$ to $<k_0q|$ or from $<00|$ to $<0q|$ is occurring at a rate $W_+$. Finally, $\tau$ is taken to be the lifetime in $<k_0q|$. In such a case the optical transitions $<00| + <k_00|$ and $<0q| + <k_0q|$ will have different frequencies because the optic phonon energy in the excited state is different than that in the ground state. This gives rise to a frequency difference, $\delta \omega$, between the two transitions, as,

$$\delta \omega = \varepsilon'_1 - \varepsilon_1$$

(3)

The lineshape function for $k=0$ absorption is given by
the Fourier transform of the electric dipole autocorrelation function including exchange, i.e.,

\[ I(\omega) = \int_{-\infty}^{\infty} \exp(i\omega t)\langle \mu(0)\mu(t) \rangle dt. \] (4)

When the interaction which is responsible for the frequency shift \( \delta \omega \) does not appreciably mix the optic branch with the \( k = 0 \) exciton state, then the off-diagonal elements are small and the effect of the scattering at rates \( W_+ \) and \( W_- \) (Cf. figure 3) can be written in terms of a time dependent frequency \( \omega_o(t) \).

In this case

\[ \mu(t) = \mu(0)\exp(i \int_0^t \omega_o(t')dt') \] (5)

and the correlation function is given by

\[ \langle \mu(0)\mu(t) \rangle \approx \langle \exp(i \int_0^t \omega_o(t')dt') \rangle \]

\[ \equiv \phi(t) \] (6)

If \( W_+ \) and \( W_- \) are time independent and \( \omega_o(t) \) is a Markoffian function the autocorrelation function \( \phi(t) \) is given by

\[ \phi(t) \approx W \cdot \exp[t(\omega_o + \pi)] \cdot 1 \] (7)

The matrix \( \omega_o \) contains all possible values of \( \omega_o(t) \) and is diagonal. The transition matrix \( \pi \) contains the scattering rates between all possible \( \omega_o(t) \). \( W \) is a row vector of the steady state probability distribution while \( 1 \) is a column vector whose elements are all unity. Under these conditions, the lineshape is given by
$I(\omega) \sim \int_{-\infty}^{+\infty} \exp(-i\omega t) \left\{ \tilde{W} \exp[\tau(i\omega + \pi)] \tau^{-1} \right\} dt$

$= \text{Re} \tilde{W} \cdot A^{-1} \cdot \tilde{1}$

where $A = i(\omega - \omega_0) + \pi$. The row vector $\tilde{W}$ for the processes illustrated in figure 3 is

$\tilde{W} = \left[ \frac{W_+ \tau}{(1 + W_+ \tau)} \quad (1 + W_+ \tau)^{-1} \right]$  \quad (9)

and the matrix $A$ is given by

$A = \begin{bmatrix} i(\delta \omega / 2 - \omega^{-1}) \tau^{-1} & \tau^{-1} \\ W_+ & i(-\delta \omega / 2 - \omega^{-1}) - W_+ \end{bmatrix}$  \quad (10)

where $\omega' = \omega - \omega_0 - \delta \omega / 2$. At low temperatures, $W_+ \tau < 1$ and one expects a large absorption near $\omega_0$ and a small one near $\omega_0 + \delta \omega$. Near $\omega = \omega_0$, $I(\omega)$ is given by a Lorentz lineshape function by setting $\omega' = -\delta \omega / 2 = -\delta \omega$. This form is

$I(\omega) = \frac{W_+ \tau^2 (\delta \omega)^2}{[D(1 + W_+ \tau)]}

\sqrt{(\omega' + \delta \omega / 2 - W_+ \delta \omega / D)^2 + W_+^2 \delta \omega^2 \tau^2 / D^2}$

where $D = 1 + (\delta \omega)^2 \tau^2$. The characteristic feature of this lineshape function is that it contains both a line broadening term and a frequency shift term. The lineshape function can be re-expressed in terms of an effective relaxation time and frequency. The effective relaxation time is given by:

$(T_{\text{eff}})^{-1} = \frac{W_+(\delta \omega)^2 \tau^2}{(1 + (\delta \omega)^2 \tau^2)}$  \quad (11)
while the effective frequency is given by:

$$(\omega)_e = \omega_o + (\delta \omega \dot{W}_+, \tau) / (1 + (\delta \omega)^2 \tau^2)$$  \hspace{1cm} (13)

The lineshape function in terms of the effective relaxation time and frequency is given as:

$$I(\omega) = \frac{1}{1 + (\omega_e - \omega)^2 (T_{\text{eff}})^2}$$  \hspace{1cm} (14)

$T_{\text{eff}}$ is given in sec$^{-1}$ and $\omega$ is in radians/sec.

It can be shown$^{25}$ that the relaxation time including temperature independent $T_1$ and other $T_2$ process can be incorporated into equation (12) as:

$$(T_{\text{eff}})^{-1} = (W_+) (\delta \omega^2 \tau^2 / (1 + (\delta \omega \tau)^2)) + T_1^{-1} + T_2^{-1}$$  \hspace{1cm} (15)

The temperature independent terms will be left out of the following equations for clarity, however, one should realize that as $W_+ \to 0$, then the other terms limit the lineshape function.

(a) **Optical lineshape function for Orbach or Raman phonon-exciton scattering processes**

The important point to note about phonon-exciton scattering that results in a change in the exciton and phonon wavevector, is that scattering can only effect the linewidth and to first order can not affect the frequency of the transition at low temperatures because the lifetime in the decay channels are long. This means that the transition $<\Omega|\mu(t)|k_o>\Omega$ which can be exchange coupled to $<\Omega|\mu(t)|k_o>\Omega$ can not contribute
to the autocorrelation function for \( \mu(t) \) because the recurrence time, from \( \langle k_0 q \rangle \) to \( \langle k_0' q \rangle \) is long. The effect on the lineshape function can be seen by letting \( \tau \) get long such that \( \delta \omega \tau > \delta \text{lin} \) equation 12 and 13. In this limit

\[
(T_{\text{eff}})^{-1} = W'' \tag{16}
\]

and

\[
(\omega)_e = \omega_0 \tag{17}
\]

The scattering rate constant \( W'' \) is \( W_+ \) or \( W \) for Orbach and Raman process respectively (cf. equation 2) and is given by the appropriate Planck distribution function for optic phonons evaluated at an energy equal to the separation between the \( k=0 \) state of the exciton and the energy of the optic phonon branch responsible for scattering. This is given in figure 3 as \( \epsilon'_{\text{1}} \).

One concludes, therefore, that if scattering of the type illustrated in figure 1a is responsible for the lineshape function (or optical dephasing times from photon echo or other similar experiments), the temperature dependent linewidth will reflect the Planck distribution for the number of phonons at \( \epsilon'_{\text{1}} \) and that at low temperatures over the temperature range where linebroadening occurs, little if any frequency shift will occur. Such is not the case for absorption and emission of phonons of the same wavevector.
b. **Optical lineshape function for phonon absorption and emission processes**

The lineshape function (or dephasing time) for phonon absorption and emission process as illustrated in figure 1b and given by

\[
<k_{00}| + <q_{0}| \quad \begin{array}{c} \text{W}_{+} \\ \text{W}_{-} \end{array} \quad <k_{0q}| \quad (18)
\]

can be simply derived. For optic phonons, the relationship between \( W_{+} \) the absorption rate constant, \( \tau \), the lifetime in \( <k_{0q}| \), and the Planck distribution function \( <n(\epsilon_{i})> \) is:

\[
W_{+} \tau = \frac{<n(\epsilon_{i})>}{<n(\epsilon_{i})> + 1} \quad (19)
\]

This satisfies microscopic reversibility and for low temperatures becomes:

\[
W_{+} \tau = (\exp (\epsilon_{i}/kT) - 1)^{-1} \quad (20)
\]

\( \epsilon_{i} \) is the energy of the optic phonon branch that is absorbed by the \( k=0 \) state of the exciton. Because the Planck distribution function falls to small values for temperature where \( \epsilon_{i} \geq kT \), the most effective optic branch would be the one of the lowest energy. Inserting 20 for low temperature cases into (12) and (13) and rearranging, expressions for the temperature dependent
portion of the effective relaxation time and the frequency shift are obtained. These are given by:

\[
(T_{\text{eff}})^{-1} = \delta \omega^2 \tau \left[ \frac{1}{(1 + (\delta \omega t)^2)} \right] \left[ \frac{1}{e^{\frac{i \epsilon_i}{kT} - 1}} \right],
\]

and

\[
\Delta_{\text{eff}} = \delta \omega \left[ \frac{1}{(1 + (\delta \omega t)^2)} \right] \left[ \frac{1}{e^{\frac{i \epsilon_i}{kT} - 1}} \right],
\]

respectively.

The absorption and emission of a phonon by the k=0 exciton state is allowed for all phonon wavevectors q and it is the lowest order phonon process that can effect both the linewidth of the optical absorption to k=0 and simultaneously frequency shift the transition with temperature. The frequency shift arises from the retention of phase memory in the scattering processes because the scattering process connects two transitions of comparable oscillator strength as illustrated by the diagram in figure 3. Two requirements, however, are necessary for frequency shifts. First, the optic phonon energy in the excited state must be different than in the ground state. The second requirement is that the lifetime in the excited phonon state \(\tau\) be long enough to partially dephase the correlation function for the transition dipole to k=0 but short enough to bring back phase memory of the upper state frequency \(\omega_0 + \delta \omega\). The frequency shift can be to either higher or lower frequency and is given by the sign of \(\delta \omega\).
If the optic phonon's energy is lower in the excited state than in the ground state the frequency shift is to lower frequency with increasing temperature.

From the ratio of the temperature dependent portion of linewidth, \( (T_{\text{eff}}^{-1}) \) and the frequency shift, \( (\Delta_{\text{eff}}) \) one can determine whether the optic lineshape function is exchange broadened or narrowed by the phonon absorption and emission mechanism. When the ratio, given by,

\[
\frac{(T_{\text{eff}}^{-1})/\Delta_{\text{eff}}}{\delta \omega t} = 1
\]

is near unity, the frequency shift with temperature would be evidence for phonon-absorption and emission causing dephasing of the optical transition to \( k = 0 \). Indeed, in the absence of inhomogeneous broadening, one would observe a narrower optical absorption when \( \delta \omega t < 1 \) than one might expect on the basis of the transition rate \( W_+ \) alone since \( (T_{\text{eff}}^{-1}) \) would be less than \( W_+ \) in this instance. If \( \delta \omega t >> 1 \), one can not say with certainty what mechanism is responsible for dephasing. This fact points out the caution that must be exercised in interpreting optical dephasing times from linewidths or even from photon echo experiments. The correct scattering mechanisms and relaxation times must come from an analysis of both the linewidth and the frequency shift with temperature.
IV. Summary

It has been shown that two different phonon exciton scattering processes can dephase the optical transitions to the \( k = 0 \) state of the exciton states. The first is a multiple scattering processes \( (k_0 + q + (k_0 q) + k' + q') \) which results in \( k \) scattering in the exciton states and thus leads to the loss of coherence of the exciton wavepacket. The effect of this scattering mechanism is to line broaden the transition from the ground state to \( k = 0 \) of the exciton with a temperature dependence related to the Planck distribution function evaluated at the energy \( \epsilon_i \) of the optic phonon which is initially coupled to the exciton state. No significant frequency shifts of the transition are expected at low temperatures.

The second mechanism that can account for optical dephasing times (\( T_2 \) processes) and absorption linewidths is the absorption and emission of a phonon of the same wavevector, i.e. \( k_0 + q \neq k_0 q \). This process does not lead to the loss of exciton coherence and is expected to be more probable than the multiple scattering mechanism at low temperatures because it is a single phonon process. The effect on the optical transition can be to broaden and frequency shift the transition with temperature because of the retention of phase memory in the phonon absorption and emission process. The frequency shift will be observed when the lifetime \( <k_0 q|\tau, k_0 q| \) and the difference in the phonon frequencies between the ground and excited state, \( \delta \omega \) satisfy the condition \( \delta \omega \tau \neq 1 \).
In another paper\textsuperscript{27} an analysis of the experimental linewidths and frequency shifts associated with the electronic origin in the one dimensional exciton states of 1,4 dibromo-napthalene\textsuperscript{28} are presented which show that the prevalent mechanism for the dephasing of the transition in this system is the absorption and emission of optic phonons of the same wavevector.

V. Acknowledgement

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Figure 1: Schematic representation of phonon-exciton interactions. (a) Multiple scattering processes: If the state $|k_0q|$ is real the scattering represents an Orbach process and if $(k_0q)$ is a virtual state then it is a Raman process in which $W_+ = W^-$. In either case the exciton coherence is lost. (b) Single phonon absorption process with no change in the exciton or phonon wavevector. No loss in the exciton coherence results.

Figure 2: Schematic representation of one dimension dispersionless phonon branches and exciton bands. $|00\rangle$ represents the ground state, $|0q\rangle$ is the lowest optic phonon branch, $|k0\rangle$ represents the phononless exciton band while $|kq\rangle$ represents the phonon-exciton band states. Finally $|k_00\rangle$ and $|k_0q\rangle$ are the $k=0$ states in the excited exciton and exciton-phonon band respectively.

Figure 3: Schematic representation of the absorption and emission of a phonon by the exciton $|k_00\rangle$ state. The absorption rate is given by the $W_+$ while the emission rate is given by $W_-$. $\tau$ is the lifetime in the state $|k_0q\rangle$. Such a process exchange averages the electric dipole transition moment $\langle 00|\mu(t)|k_00\rangle$ at a frequency $\omega_0$ with $\langle 0q|\mu(t)|k_0q\rangle$ at a frequency $\omega_0 + \delta \omega$. $\delta \omega$ is given by the difference of the phonon energy in excited state ($\epsilon'_i$) and ground state ($\epsilon_i$).
OPTICAL DEPHASING ($T_2$) PROCESSES
FOR ELECTRIC DIPOLE TRANSITIONS TO $k=0$

(a) Multiple Phonon Scattering: Loss of Exciton Coherence

\[ k_0 + q \xrightarrow{W_+} (k_0q) \]
\[ (k_0q) \xrightarrow{W'} k' + q' \]

(b) Phonon-Absorption: No Loss of Exciton Coherence

\[ k_0 + q \xrightarrow{W_+} (k_0q) \]

Figure 1

XBL774-5292
Exciton-Phonon Band

\( \epsilon_1' \)

Exciton Band

\( \epsilon (k_0) \)

\( \langle k_0q \rangle \)

Ground State Optic Branch

\( \epsilon_1 \)

\( \epsilon_0 = 0 \)

\( \langle k_{00}l \rangle \)

-\( \pi/a \) to +\( \pi/a \)

Exciton and Phonon Wavevectors

Figure 2
Figure 3
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