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Optical absorption from polarons in a diatomic polymer

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The optical absorptions associated with polarons and bipolarons in a diatomic polymer $(A=B)_x$ are studied within the continuum limit. Analytic expressions are obtained and compared with previously known limiting cases. Our results clarify greatly the role of symmetry in determining the relative strengths of these absorptions.

Several recent experiments[1-3] on polythiophene have focused renewed attention on the optical-absorption properties of polaronlike excitations in conducting polymers. In particular, strong intragap absorption features are observed in both photoinduced photoabsorption (PA) and in optical absorption from doped samples. In PA[3] and in moderately doped (≥ 4 mol %)[1,2] samples, two peaks are clearly observed, whereas for lightly doped (≤ 2.5 mol %)[1,2] samples, a third peak occurs at a frequency intermediate between the other two.

Importantly, both this pattern of peaks and its change with doping are exactly what one would expect on the basis of prior theoretical calculations of polaron absorption[4] within the continuum limit of the Su-Schrieffer-Heeger—(SSH)-like model[5] appropriate to a conducting polymer (like polythiophene) with a nondegenerate[6] ground state. For low doping, where single polarons are expected to be dominant excitations, the two localized electronic levels (at $\pm\omega_0$, $\omega_0<\Delta_0$) associated with a polaron lead to three distinct intragap ($\omega<2\Delta_0$) contributions to $\alpha(\omega)$, the optical absorption:[4] (1) $\alpha^{(1)}(\omega)$, a δ-function transition between the two localized levels occurring at $\omega=2\omega_0$, (2) $\alpha^{(2)}(\omega)$, an absorption which begins at $\omega=\Delta_0-\omega_0$ and involves transitions between the valence band and the $-\omega_0$ level (for hole polarons) or between the $+\omega_0$ level and the conduction band (for electron polarons), and (3) $\alpha^{(3)}(\omega)$, an absorption which begins at $\omega=\Delta_0+\omega_0$ and involves transitions between the valence band and the $+\omega_0$ level or between the $-\omega_0$ level and the conduction band. For moderate doping and in the relaxed PA, bipolarons are expected (on the basis of energetic arguments) to be the dominant excitation in polythiophene and related polymers. Although bipolarons also have two localized electronic levels (at $\pm\omega_0'$, and $\omega_0'<\omega_0$ for the same bare parameters), the occupancy of these levels (fully occupied for electron bipolarons, empty for hole bipolarons) precludes the δ-function transition at $\omega=2\omega_0'$, and one is left with two intragap transitions corresponding to $\alpha^{(2)}$ and $\alpha^{(3)}$. Thus the theoretical and experimental pictures are in good agreement on the number of intragap absorptions and their change with doping. Unfortunately, there is clearly one area of disagreement between the experiments and the theory based on an SSH-like model, namely, the theory predicts a striking difference between the two intragap absorptions involving localized-level-to-continuum transitions, with $\alpha^{(2)}$ (the lower-energy absorption) having a square-root *singularity* at its absorption edge ($\omega=\Delta_0-\omega_0$), but $\alpha^{(3)}$ (the higher-energy absorption) having a square root *vanishing* at its absorption edge ($\omega=\Delta_0+\omega_0$). Accordingly, the integrated intensity of the $\alpha^{(2)}$ absorption is expected (theoretically) to be much greater than that from $\alpha^{(3)}$, whereas experimentally one observes roughly equal integrated intensities.[1-3] It is therefore of great importance to understand in detail the basis of this theoretical prediction and to see what additional physical effects might modify it in real systems.

In the original theoretical calculations,[4] it was noted that the strong difference between $\alpha^{(2)}$ and $\alpha^{(3)}$ arose as a consequence of the symmetries of the electronic wave functions for *trans*- and *cis*-$(\mathrm{CH})_x$ and related polymers.[7] In this paper we study a model system—the diatomic polymer, $(A=B)_x$[8]—in which these symmetries are not present and establish that for $(A=B)_x$ both $\alpha^{(2)}$ and $\alpha^{(3)}$ do indeed have square-root singularities at their absorption edges. The existence of analytic forms[9] for the electronic wave functions for the $(A=B)_x$ system allows us to present analytic expressions for $\alpha^{(2)}$ and $\alpha^{(3)}$, which permit direct comparisons with the two previously known limiting cases of $(\mathrm{CH})_x$-like systems (when the A-B site energy difference is zero) and of widely separated kink-antikink pairs in an $(A=B)_x$ system.[10] We note that, apart from clarifying considerably the nature of polaron optical absorption in conducting polymers, our study of the $(A=B)_x$ system is important in its own right. In particular, although no clearcut physical realization of $(A=B)_x$ exists yet among the conducting polymers, the $(A=B)_x$ model is relevant[11,12] for the mixed-stack charge transfer salts (e.g., tetrathiafulvalene Chloranil), which exhibit the very interesting neutral-ionic transition.

To begin our analysis we recall that in the adiabatic continuum limit the Hamiltonian for the $(A=B)_x$ system can be written as[8]

$$H=\sum_{\sigma}\int dx\left[\psi_\sigma(x)\left[-iv_F\sigma_1\frac{\partial}{\partial x}-\sigma_2\Delta(x)+\delta\sigma_3\right]\psi_\sigma(x)+\frac{K}{2}\Delta^2(x)\right]. \qquad (1)$$

Here the σ_i are the usual Pauli matrices, $\Delta(x)$ describes the (adiabatic) phonons, and $\psi_\sigma(x)$ is, for each spin σ $(=\pm\frac{1}{2})$, the two-component field describing the electron. The parameter δ (Ref. 13) represents the difference in site energies between the A and B moieties in the dimer. Standard manipulations[6,8,14] lead to the equations for the electronic wave functions of energy ε_n, $\psi^{+}_{n,\sigma}=(A^{*}_{n,\sigma}, B^{*}_{n,\sigma})$ of the form

$$(\varepsilon_n-\delta)A_{n,\sigma}=\left[-iv_F\frac{\partial}{\partial x}+i\Delta(x)\right]B_{n,\sigma}\ , \tag{2a}$$

$$(\varepsilon_n+\delta)B_{n,\sigma}=\left[-iv_F\frac{\partial}{\partial x}-i\Delta(x)\right]A_{n,\sigma}\ , \tag{2b}$$

and to the self-consistent equation for the gap parameter

$$\Delta(x)=-\left[\frac{i4}{K}\right]\sum_{n,\sigma}{}'[A^{*}_{n,\sigma}(x)B_{n,\sigma}(x)-A_{n,\sigma}(x)B^{*}_{n,\sigma}(x)]\ , \tag{2c}$$

where the prime indicates a sum over all occupied states.[14] The polaron solution[9] to Eqs. (2) is described by a gap parameter

$$\Delta_p(x)=\Delta-\kappa_0 v_F\{\tanh[\kappa_0(x+x_0)]-\tanh[\kappa_0(x-x_0)]\}\ , \tag{3}$$

where $\tanh(2\kappa_0 x_0)=(\kappa_0 v_F/\Delta)$ and $\kappa_0 v_F\equiv(\Delta_0^2-\omega_0^2)^{1/2}$, with $\Delta_0^2\equiv\Delta^2+\delta^2$. The corresponding electronic wave functions are found by solving (2a) and (2b) with $\Delta(x)$ given by (3). Solving these equations[9,14] we find that the spectrum consists of two localized levels at $\pm\omega_0$, and continua with $\varepsilon(k)=+\omega(k)\equiv(\Delta_0^2+k^2v_F^2)^{1/2}$ (conduction band) and $\varepsilon(k)=-\omega(k)$ (valence band). The wave function (A_0, B_0) for the electronic level at $+\omega_0$ is

$$A_0=i[\kappa_0(\omega_0+\delta)/4\omega_0]^{1/2}\operatorname{sech}[\kappa_0(x-x_0)]\ , \tag{4a}$$

$$B_0=[\kappa_0(\omega_0-\delta)/4\omega_0]^{1/2}\operatorname{sech}[\kappa_0(x+x_0)]\ , \tag{4b}$$

and that for the level at $-\omega_0$ satisfies $A_{-0}=B_0(x_0\rightarrow-x_0)$, $B_{-0}=A_0(x_0\rightarrow-x_0)$. For the negative-energy continuum, one has wave functions (A_{-k}, B_{-k}),

$$A_{-k}=N_k e^{ikx}[\omega(k)-\delta]\{k+i\kappa_0\tanh[\kappa_0(x-x_0)]\}\ , \tag{5a}$$

$$B_{-k}=N_k e^{ikx}(\Delta+ikv_F)\{ik-\kappa_0\tanh[\kappa_0(x+x_0)]\}\ , \tag{5b}$$

while for the positive-energy continuum one has (A_k, B_k),

$$A_k=N_k e^{ikx}(\Delta-ikv_F)\{ik-\kappa_0\tanh[\kappa_0(x-x_0)]\} \tag{6a}$$

and

$$B_k=N_k e^{ikx}[\omega(k)-\delta]\{k+i\kappa_0\tanh[\kappa_0(x+x_0)]\}\ . \tag{6b}$$

Here,

$$(N_k)^{-2}=4\pi\omega(k)[\omega(k)-\delta](\kappa_0^2+k^2)\ .$$

In conventions consistent[15] with those used in the Hamiltonian in Eq. (1), the optical-absorption coefficient is given formally by

$$\alpha(\omega)=\frac{1}{\omega}\sum_{1,2}|\langle\varepsilon_1|\sigma_1|\varepsilon_2\rangle|^2\delta(\omega-\varepsilon_1-\varepsilon_2)\ . \tag{7}$$

Using the forms of the wave function above and performing the relevant spatial integrals as described in Ref. 4, we find that the valence-band to localized-level matrix elements have the forms, in an obvious notation,

$$\begin{aligned}\langle-\omega_0|\sigma_1|-\omega(k)\rangle=&\left[\frac{\kappa_0}{4\omega_0}\right]^{1/2}N_k\left[\frac{\pi}{\kappa_0 v_F}\right]\operatorname{sech}\left[\frac{\pi k}{2\kappa_0}\right]\\&\times\{[\omega(k)-\delta](\omega_0+\delta)^{1/2}[(\omega_0^2-\delta^2)^{1/2}e^{+ikx_0}-(\Delta+ikv_F)e^{-ikx_0}]\\&+(\Delta+ikv_F)(\omega_0-\delta)^{1/2}[(\omega_0^2-\delta^2)^{1/2}e^{-ikx_0}-(\Delta-ikv_F)e^{+ikx_0}]\}\end{aligned} \tag{8a}$$

and

$$\begin{aligned}\langle+\omega_0|\sigma_1|-\omega(k)\rangle=i&\left[\frac{\kappa_0}{4\omega_0}\right]^{1/2}N_k\left[\frac{\pi}{\kappa_0 v_F}\right]\operatorname{sech}\left[\frac{\pi k}{2\kappa_0}\right]\\&\times\{(\omega(k)-\delta)(\omega_0-\delta)^{1/2}[(\omega_0^2-\delta^2)^{1/2}e^{ikx_0}-(\Delta+ikv_F)e^{-ikx_0}]\\&-(\Delta+ikv_F)(\omega_0+\delta)^{1/2}[(\omega_0^2-\delta^2)^{1/2}e^{-ikx_0}-(\Delta-ikv_F)e^{+ikx_0}]\}\end{aligned} \tag{8b}$$

Hence, taking the modulus squared and using the δ function in (7) to perform the integral over k at fixed ω, we find that the $\alpha^{(2)}$, which corresponds to the $-\omega(k)\rightarrow-\omega_0$ transition, is given by

$$\alpha^{(2)}(\omega)=\frac{\pi}{8}\frac{\omega}{\omega_0^2 v_F}\frac{1}{(\kappa_0^2+K^2)}\frac{1}{K}\operatorname{sech}^2\left[\frac{\pi}{2}\frac{K}{\kappa_0}\right][(\omega_0^2-\delta^2)^{1/2}(\Delta\cos(2Kx_0)+Kv_F\sin(2Kx_0))+\delta^2+\omega_0(\omega_0+\omega)]\ , \tag{9a}$$

where $Kv_F=[(\omega+\omega_0)^2-\Delta_0^2]^{1/2}$, whereas $\alpha^{(3)}$, corresponding to the $-\omega(k)\rightarrow+\omega_0$ transition, is given by[16]

$$\alpha^{(3)}(\omega)=\frac{\pi}{8}\frac{\omega}{\omega_0^2 v_F}\frac{1}{[\kappa_0^2+(K')^2]}\frac{1}{K'}\operatorname{sech}^2\left[\frac{\pi}{2}\frac{K'}{\kappa_0}\right]\{(\omega-\omega_0)\omega_0-\delta^2-(\omega_0^2-\delta^2)^{1/2}[\Delta\cos(2K'x_0)+K'v_F\sin(2K'x_0)]\}\ , \tag{9b}$$

where $K'v_F=[(\omega-\omega_0)^2-\Delta_0^2]^{1/2}$.

It is straightforward to verify that for $\delta\neq0$, both $\alpha^{(2)}$ and $\alpha^{(3)}$ exhibit square-root singularities at their respective absorption edges. Specifically, as $K\rightarrow0$,

$$\alpha^{(2)}\omega\rightarrow\frac{\pi\omega}{8\omega_0^3\kappa_0}\frac{1}{K}[(\omega_0^2-\delta^2)^{1/2}\Delta+\delta^2+\omega_0(\omega+\omega_0)]\ , \tag{10a}$$

whereas $K'\rightarrow0$,

$$\alpha^{(3)}(\omega)\rightarrow\frac{\pi\omega}{8\omega_0^3\kappa_0}\frac{1}{K'}[(\omega-\omega_0)\omega_0-\delta^2-\Delta(\omega_0^2-\delta^2)^{1/2}]\ . \tag{10b}$$

Note that for $\delta\neq0$, both absorptions show square-root singularities, but for $\delta=0$, we see—recalling that for $\delta=0$, $\Delta=\Delta_0$—that $\alpha^{(3)}$ does revert to the square-root vanishing found in Ref. 4.

More generally, for arbitrary ω, one can write both $\alpha^{(2)}$ and $\alpha^{(3)}$ as their $\delta=0$ limits—which we shall call $\alpha_{\mathrm{FBC}}^{(2)}$ and $\alpha_{\mathrm{FBC}}^{(3)}$ (Ref. 16) (FBC stands for Fesser, Bishop, and Campbell)—plus the δ-dependent term arising from the $(A=B)_x$ site energy difference. Explicitly, $\alpha^{(2)}(u)=\alpha_{\mathrm{FBC}}^{(2)}(u)+\tilde{\alpha}^{(2)}(u)$, with

$$\tilde{\alpha}^{(2)}(\omega)=\frac{\pi}{8}\frac{\omega}{\omega_0 v_F}\left[\frac{1}{\kappa_0^2+K^2}\right]\frac{1}{K}\operatorname{sech}^2\left[\frac{\pi}{2}\frac{K}{\kappa_0}\right]$$
$$\times\left\{\frac{\delta^2}{\omega_0}+\Delta_0\cos(2Kx_0)\left[\left[1-\frac{\delta^2}{\omega_0^2}\right]^{1/2}\left[1-\frac{\delta^2}{\Delta_0^2}\right]^{1/2}-1\right]+Kv_F\sin(2Kx_0)\left[\left[1-\frac{\delta^2}{\omega_0^2}\right]^{1/2}-1\right]\right\} \tag{11a}$$

and $\alpha^{(3)}(\omega)=\alpha_{\mathrm{FBC}}^{(3)}+\tilde{\alpha}^{(3)}(u)$ (Ref. 16), with

$$\tilde{\alpha}^{(3)}(u)=\frac{-\pi}{8}\frac{\omega}{\omega_0 v_F}\left[\frac{1}{\kappa_0^2+(K')^2}\right]\frac{1}{K'}\operatorname{sech}^2\left[\frac{\pi}{2}\frac{K'}{\kappa_0}\right]$$
$$\times\left\{\frac{\delta^2}{\omega_0}+\Delta_0\cos(2K'x_0)\left[\left[1-\frac{\delta^2}{\omega_0^2}\right]^{1/2}\left[1-\frac{\delta^2}{\Delta_0^2}\right]^{1/2}-1\right]+K'v_F\sin(2K'x_0)\left[\left[1-\frac{\delta^2}{\omega_0^2}\right]^{1/2}-1\right]\right\}\ . \tag{11b}$$

To gain further insight into the development of the singularity in $\tilde{\alpha}^{(3)}$, we take the limit $K'\rightarrow0$ in Eq. (11b), and, expanding to leading (nonvanishing) order in δ, obtain

$$\tilde{\alpha}^{(3)}(\omega)\sim\frac{\pi}{8}\frac{\omega}{\omega_0^2\kappa_0}\frac{1}{K'}(\tfrac{3}{2}-\sqrt{2})\frac{\delta^2}{\Delta_0}\ . \tag{12a}$$

Thus the square-root singularity at $\omega=\omega_0+\Delta_0$ appears with strength proportional to δ^2. Interestingly, one finds that, as $K\rightarrow0$,

$$\tilde{\alpha}^{(2)}(\omega)\sim-\frac{\pi}{8}\frac{\omega}{\omega_0^2\kappa_0}\frac{1}{K}(\tfrac{3}{2}-\sqrt{2})\frac{\delta^2}{\Delta_0}\ , \tag{12b}$$

so that the increase in the intensity of $\alpha^{(3)}$ is compensated by a decrease in $\alpha^{(2)}$. More generally, using the full forms of $\tilde{\alpha}^{(2)}$ and $\tilde{\alpha}^{(3)}$, one can show, by a simple change of variable, that their combined contribution to the optical sum rule cancels, so that the total integrated intensity of $\alpha^{(2)}+\alpha^{(3)}$ is the same (for given ω_0) as that for $(\mathrm{CH})_x$:[4] the $(A=B)_x$ site energy difference δ does not affect the total intragap intensity for bipolaron absorption.

A second significant check on our results is the limit $\delta\rightarrow\Delta_0/\sqrt{2}$, at which point the $(A=B)_x$ polaron "destabilizes"[9] to a kink-antikink pair, the optical absorption of which in $(A=B)_x$ has previously been studied.[10] Recalling that as $\delta\rightarrow\Delta_0/\sqrt{2}$ we also have $\delta\rightarrow\omega_0$, we see immediately that

$$\alpha^{(2)}(\omega)\sim\frac{\pi v_F}{8\omega_0}\frac{1}{K}\operatorname{sech}^2\left[\frac{\pi}{2}\frac{K}{\kappa_0}\right] \tag{13}$$

and $\alpha^{(3)}$ approaches the same form with K' replacing K. This is the previously calculated result.[10]

Although, in view of their dominance over a wide range of experimental conditions, we have focused on the two intragap transitions associated with bipolarons, the δ-function absorption—called $\alpha^{(1)}$ above—found for the single polaron also shows interesting structure. Using the wave functions from Eq. (4), we find that

$$\langle+\omega_0|\sigma_1|-\omega_0\rangle=2x_0(\omega_0^2-\delta^2)^{1/2}/\kappa_0 v_F\ ,$$

so that, in our normalization,[15]

$$\alpha^{(1)}(\omega)=\frac{2x_0^2}{\omega_0^3}(\omega_0^2-\delta^2)\delta(\omega-2\omega_0)\ , \tag{14}$$

where the last term is the δ function enforcing energy conservation. Note that, in the limit $\delta\rightarrow0$, we recover the $(\mathrm{CH})_x$ result,[4] whereas for $\delta\rightarrow\omega_0$ $(=\Delta_0/\sqrt{2})$, this absorption vanishes, as one would expect since the localized levels are being created by two infinitely separated entities and hence should have no overlap.

Although our results have been derived explicitly in the context of an SSH-like model for a diatomic polymer, through an interesting connection recently discovered[17] they may have relevance to the case of conventional conducting polymers—both with and without degenerate ground states—when Coulomb interactions effects are taken into account. Specifically, if one adds to the SSH-like model describing polythiophene on-site (U) and nearest-neighbor (V) Coulomb repulsions and if one treats these interactions in the unrestricted Hartree-Fock (UHF) approximation, one finds that the equations determining the electronic wave functions have exactly the same structure[17,18] as those for the $(A=B)_x$ system. The only difference is that for the $(A=B)_x$ system the parameters—for example, the site energy difference δ—are given, whereas for the UHF case they must be determined self-consistently. Since the UHF treatment still leads to single-particle electron energy levels, the optical absorption can be calculated[17] exactly as in the pure SSH-like case. In view of the connection to the $(A=B)_x$ system, it is clear that this calculation should lead to the appearance of a square-root singularity in $\alpha^{(3)}$ and hence to more equal integrated intensities for the two intragap bipolaron absorptions. This is precisely the recently derived result.[17] Although there remain legitimate questions[19] about the applicability of HF methods to correlated quasi-one-dimensional systems, it may be the case that the nearly equal intensities observed for the two intragap bipolaron absorptions in polythiophene[1-3] are due to the effects of Coulomb interactions neglected in the SSH-like theories but included in the UHF treatment.

In summary, we have presented explicit analytic expressions for the intragap optical absorptions associated polarons in a diatomic polymer. Our results confirm the important role of the symmetries of the electronic wave functions and provide explicit comparisons with previously known limits.

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[14] See, for example, D. K. Campbell and A. R. Bishop, Nucl. Phys. B **200**, 297 (1982) for details.

[15] Note the difference in conventions, including normalization, from Ref. 4. This change was made for more direct comparison with Refs. 8—10.

[16] Readers checking the details of the correspondence between the $(A=B)_x$ system as $\delta \rightarrow 0$ and the previous $(CH)_x$ results should be aware of an unfortunate misprint in Ref. 4. Equation (3.10) of that reference simply repeats the Eq. (3.9) for $\alpha^{(2)}$; the correct expression for $\alpha^{(3)}$ is given by the $\delta=0$ limit of Eq. (9b) here.

[17] U. Sum, K. Fesser, and H. Büttner (unpublished). These authors' present figures, rather than analytic expressions, indicating the nature of the absorptions $\alpha^{(2)}$ and $\alpha^{(3)}$. As far as can be checked, the results of our two groups are entirely consistent.

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