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Bipolaron formation in polar solids

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Abstract : By a combination of canonical transformation and perturbation it has been possible to go beyond the Pekar approximation to predict the feasibility of a single centre bipolaron. Alternative criterion on $\eta = \varepsilon_\infty/\varepsilon$ has been obtained from lowest order calculations.

Keywords : Bipolaron, canonical transformation, Landau-Pekar method

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The complex of a pair of electrons bound by the lattice distortion was defined by Pekar (Pekar 1963), who at that time found the optical bipolaron to be energetically unfavourable as compared to two separate polarons. Recently due to its relevance to semiconductor technology (Anderson 1975) and particularly high T_c super conductivity (Chakraverty and Ranninger 1985, Emin 1989) a somewhat vigorous revival of interest in bipolaron has been witnessed. Almost all the treatments are single centre bipolarons (like the helium atom) except Vinitskii's (Vinitskii and Gitterman 1961) which is a Heitler-London approach (like the hydrogen molecule). Path integral calculations (Verbist *et al* 1991, Hiramoto and Toyozawa 1985, Sil *et al* 1991) support the formation of bipolaron for some $\eta \leq \eta_c$ where $\eta = \varepsilon_\infty/\varepsilon$ is the ratio of the high frequency and static dielectric constants of the material. But the Landau-Pekar (without explicit correlation factors) ansatz of Pekar fails to favour a bipolaron as has also been very recently pointed out (Suprun and Moizhes 1982, Sil *et al* 1991) because the LP ansatz is based on the effective hamiltonian H_{LP} which differs from the total hamiltonian by a remainder term $H\text{-}H_{LP}$ which ought to be taken into account in some approximate way. We thus show, for the first time, in this brief communication that going beyond the Pekar approximation by including the effects of this remainder term perturbatively, which is fairly simple to take into account in the lowest orders, one indeed gets a bipolaron.

The hamiltonian of two electrons in a polar lattice is given by (in Fröhlich units)

$$\tilde{H} = p_1^2 + p_2^2 + \mu/r_{12} + \sum_{\vec{q}} (\xi_{\vec{q}\vec{r}_1} b_{\vec{q}} + \text{hc}) + \sum_{\vec{q}} (\xi_{\vec{q}\vec{r}_2} b_{\vec{q}} + \text{hc}) + \sum_{\vec{q}} b^+_{\vec{q}} b_{\vec{q}} \quad (1)$$

Here $$\mu = \frac{e^2 u}{\hbar\omega\varepsilon_\infty} = \frac{2\alpha}{1-\eta}$$

$$\Delta E^{(1)} + \Delta E^{(2)} < 0$$

$$\Delta E^{(1)} = 2\alpha \left(\frac{1}{1-\eta} - 1 \right) \frac{5}{8} \lambda \tag{18}$$

Eq. (17) contains a plethora of terms, but for the purpose of this brief note we shall only quote the results from a few such which already establishes (18). All the others when included will enrich the criterion, which will be reported in a detailed version of this paper later on. Taking only the matrix elements

$$\begin{aligned}
&< 1s,\ 1s,\ O \left| H_{\text{int}} \right| 1s,\ 2s,\ O >, < 1s,\ 1s,\ O \left| H_{\text{int}} \right| 2s,\ 1s,\ O > \\
&< 1s,\ 1s,\ O \left| H_{\text{int}} \right| 2s,\ 2s,\ O >, < 1s,\ 1s,\ O \left| H_{\text{int}} \right| 2p,\ 2p,\ O > \\
&< 1s,\ 1s,\ O \left| H_{\text{int}} \right| 1s,\ 3s,\ O >, < 1s,\ 1s,\ O \left| H_{\text{int}} \right| 3s,\ 1s,\ O > \\
&< 1s,\ 1s,\ O \left| H_{\text{int}} \right| 3s,\ 3s,\ O >
\end{aligned} \tag{19}$$

for calculating the partial contribution from (17), we get after some algebra,

$$\Delta E^{(1)} + \Delta E^{(2)} = -\alpha^2 \left(Ax^2 - Bx + C \right) \tag{20}$$

where $A = 0.1303204$, $B = 0.6492814$, $C = 0.5537308$ and $x = \dfrac{1}{1-\eta}$

Put in (18) this leads already in these lowest order corrections a respectable criterion for the bipolaron formation for $\eta \leq 0.084$.

In conclusion we have shown here that the single centre bipolaron formation, in the framework of the strong coupling LP approximation, is feasible if the remainder term, neglected in Pekar's (Pekar 1963) negative result, is taken into account. Taking only the matrix elements (19) we demonstrate this possibility by obtaining the energy correction (20) and the consequent restriction on η ($\eta \leq 0.084$). Correlation need not be introduced artificially in this approach, it being built in the formalism. A more detailed version of this work will be reported shortly.

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