

INFLUENCE OF THE NONADIABATIC TERMS ON  
THE ENERGY OF THE EXCITONIC MOLECULES

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Abstract

We suggested a new method for calculating energy levels of excitonic molecules. Previous variational calculations did not account nonadiabatic corrections due to the nuclear motion and because of that results for the binding energy of excitonic molecules are much smaller than experimental values. Accounting nonadiabatic terms we obtained quantitative agreement with experiment.

1. INTRODUCTION

The excitonic molecules are the simplest systems of four particles (two negative and two positive) with Coulomb interaction, for which in a wide range of electron ( $m_e$ ) to hole ( $m_h$ ) mass ratio,  $\sigma = m_e/m_h$ , the adiabatic condition is not well satisfied.

Previous variational calculations [1,2] usually neglected this fact and obtained poor agreement with experimental results [3].

In Refs. 4 and 5 it was shown that a consistent inclusion of nonadiabatic corrections produced a noticeable increase in the binding energies of excitonic molecules (biexcitons).

The best theoretical results for the ground-state energies of biexcitons were obtained using the Green's function Monte Carlo method (GFMC) [6]. For all  $\sigma$  values these energies are significantly lower than variational estimates and in favourable agreement with experimental results [3].

In Ref. 7 the method of calculating isotope effects in diatomic molecules is developed. In our work [8] we showed how the method of Ref. 7 could be modified in order to give satisfactory results for the energy levels of biexcitons.

In the present paper we perform more detailed calculations of binding energies of biexcitons on basis of method of Ref. 8. The obtained results are compared with best previous theoretical calculations [ 6 ].

## 2. METHOD OF SOLUTION AND RESULTS

When the adiabatic basis is applied to two-electron diatomic molecule, one gets a differential equation for the binding energy,  $\tilde{E}_{\nu k}$ , of the system (details of the method can be found in Refs. 7 and 8):

$$\left[ -\frac{1}{2M} \frac{d^2}{dR^2} + \frac{K(K+1)}{2MR^2} + \tilde{E}_0(R) + \frac{1}{2M} V(R) - \tilde{E}_{\nu k} \right] \chi_{\nu k}(R) = 0 \quad (1)$$

(System of atomic units is used:  $e = m_e = \hbar = 1$ ).

Here  $R$  is the distance between positively charged particles  $\underline{a}$  and  $\underline{b}$ ,  $M$  is reduced mass of the same particles:  $M^{-1} = M_a^{-1} + M_b^{-1}$  and  $\chi_{\nu k}(R)$  is the radial part of the molecular wave function, which corresponds to rovibrational state  $(\nu k)$ . Let us note that for excitonic molecules  $M_a = M_b = m_e$ .

Here the following notations are introduced:

$$\frac{1}{M} = \frac{1}{M} - \frac{2}{M^2} \left( \delta^2 + \frac{k^2}{4} \right) \quad (2)$$

$$\tilde{E}_0(R) = E_0(R) + 1,$$

where:  $k = \frac{M_b - M_a}{M_c + M_a}$ ,  $E_0(R)$  is the ground state potential energy curve of hydrogen molecule [ 9, 10 ],  $V(R)$  is the effective potential, which describes nonadiabatic terms (Eq. (12) in Ref. 8).

In Ref. 7 the single parameter  $\delta^2$  was determined by using the experimentally measured rovibronic transition energies of  $H_2$  molecule.

For excitonic molecules all Coulomb centers have comparable effective masses. Therefore, the nonadiabatic terms cannot be taken into account by perturbation theory, as has been usually done when one calculates properties of ordinary molecules with  $\mathcal{G} \lesssim 10^{-3}$ . So, one could not expect to get appropriate binding energies of excitonic molecular system from Eq. (1) with the single  $\delta^2$  value for the whole region  $0 < \mathcal{G} \leq 1$

Because of that we have recently described excitonic molecules by means of Eq. (1), but with several values of parameter  $\gamma^*$  [ 8 ] . In this manner we practically used parameter  $\gamma^* = \gamma^*(\zeta)$  , as a scaling parameter which changes properly the effective potential  $\tilde{\mathcal{E}}_0(R) + \frac{1}{2H} V(R)$

In our previous work [ 8 ] we obtained good overall agreement with respective GFMC results for binding energies of biexcitons.

In the present work we did much more careful investigation of binding energies of excitonic molecules as a function of electron to hole mass ratio  $\zeta$

Our results for molecular binding energies  $\mathcal{E} = -\tilde{\mathcal{E}}_{00} = 2E_a - E$  (the energy which is required to dissociate the biexciton into two noninteracting excitons) as a function of  $\zeta$  are given in Table 1 and Fig. 1.

Fig. 1.

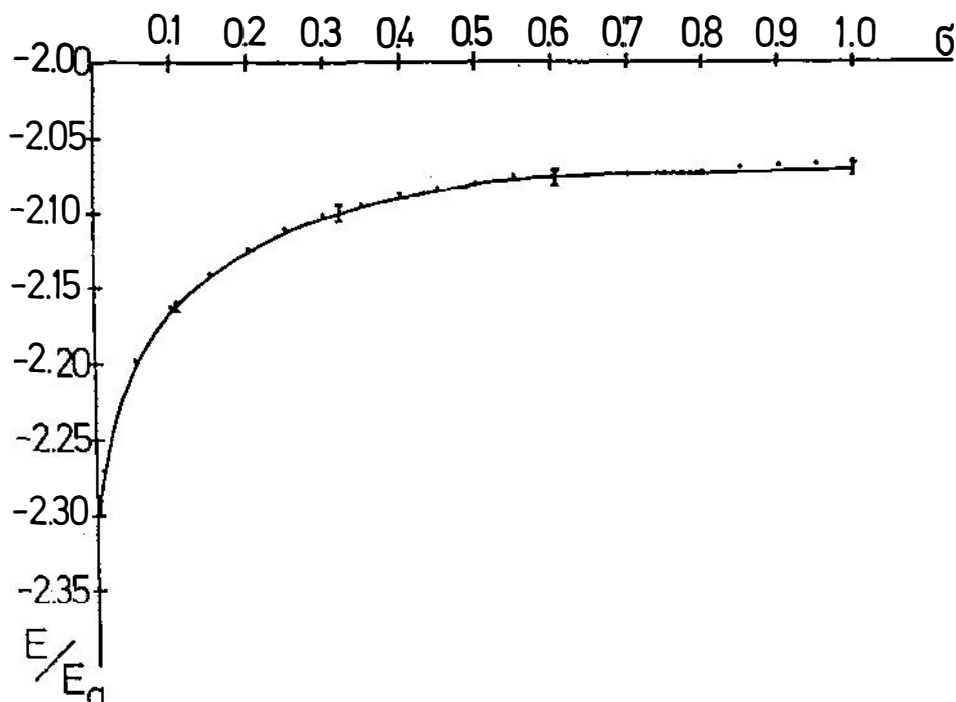


Table 1. The binding energy  $\epsilon = -\tilde{E}_\infty = 2E_a - E$  [eV] of biexcitons as a function of the electron-hole mass ratio  $\mathcal{G}$  and the scaling parameter  $\mathcal{J}^*$  ( $|E_a(\sigma)| = 13.605/(1+\sigma)$ ,  $M = 1/2\mathcal{G}$ ,  $b = 0$ ).

$\mathcal{G}$	$\mathcal{J}^*$	$\epsilon$	$\epsilon/ E_a $
0.005	0.7	3.9499	-2.2918
0.01	0.7	3.6405	-2.2703
0.05	0.7	2.5565	-2.1973
0.1	0.7	2.0131	-2.1628
0.15	0.6	1.6732	-2.1414
0.2	0.525	1.4381	-2.1269
0.25	0.44	1.1982	-2.1101
0.3	0.4	1.0585	-2.1011
0.35	0.37	0.9570	-2.0950
0.4	0.34	0.8557	-2.0881
0.45	0.32	0.7973	-2.0850
0.5	0.3	0.7362	-2.0812
0.55	0.28	0.6656	-2.0758
0.6	0.27	0.6656	-2.0783
0.65	0.255	0.6183	-2.0750
0.7	0.2425	0.5853	-2.0731
0.75	0.2325	0.5729	-2.0737
0.8	0.222	0.5457	-2.0722
0.85	0.212	0.5146	-2.0700
0.9	0.2025	0.4791	-2.0669
0.95	0.195	0.4677	-2.0670
1.0	0.188	0.4575	-2.0673

In Fig. 1 ground state energies of excitonic molecules as a function of electron-hole mass ratio  $\mathcal{G}$  are present. The curve shows GFMC results [ 6 ] , while the points represent our results. Here energies are represented in excitonic Rydbergs,  $E_a$  .

Let us note that the scaling parameter  $\mathcal{J}^*$  , for the arbitrary value of electron-hole mass ratio  $\mathcal{G}$  , can be obtained by means of Table 1. and some simple, linear or quadratic, interpolation procedure.

### 3. CONCLUSIONS

In conclusion it can be said that the method used in this paper gives much better results than the best variational [ 1 ] and adiabatic [ 4,5 ] calculations for biexcitons.

Our method is very simple and it gives results which are now as good as GFMC results [ 6 ] . But GFMC is not so suitable and economical, especially for fermionic systems [ 11 ] .

There are some other advantages of our method. Besides ground state binding energies and wave functions, we can obtain from our numerical program, without any changes, other excited rovibronic energy levels and respective wave functions. These wave functions can be used for the calculation of lifetimes of excitonic molecules.

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