# Response to "Comment on 'Comparison of the $k \cdot p$ and the direct diagonalization approaches for describing the electronic structure of quantum dots'" [Appl. Phys. Lett. 73, 1155 (1998)]

Cite as: Appl. Phys. Lett. **73**, 1157 (1998); https://doi.org/10.1063/1.122155
Submitted: 10 June 1998 • Accepted: 23 June 1998 • Published Online: 27 August 1998

**Huaxiang Fu, Lin-Wang Wang and Alex Zunger**



# Response to ''Comment on 'Comparison of the $k\cdot p$ and the direct diagonalization approaches for describing the electronic structure of quantum dots' '' [Appl. Phys. Lett. 73, 1155 (1998)]

Huaxiang Fu, Lin-Wang Wang, and Alex Zunger[a)]
*National Renewable Energy Laboratory, Golden, Colorado 80401*

(Received 10 June 1998; accepted for publication 23 June 1998)

[S0003-6951(98)02234-7]

Efros and Rosen[1] (hereafter denoted as ER) raise three issues (indicated by italics in the following) regarding our Letter.[2] Our reply follows:

*(i) That the effective-mass approximation (EMA) has been spectacularly successful in explaining experiments for dots.*

Our work[2–4] had shown that because the EMA builds into the Hamiltonian the correct physical symmetry of the system, it can achieve good agreement with experiment via judicious selection[5–7] of its parameters. It thus provides a useful *representation* and a practical *fitting scheme*. However, good agreement with experiment does not always imply good theory. This is true in the present case for two reasons: (1) some of the parameters (e.g., $E_p$ and $f$) of the multiband EMA are not uniquely determined physical observables in their own rights, but have meaning only in the context of a given, highly simplified model. Since the $8\times8$ $k\cdot p$ model is correct only to the second order, the nonparabolicity of the real bulk bands can be only *partially* represented by using the $k\cdot p$ parameter $E_p$. Other sources of nonparabolicity remain unspecified. Thus, the derived $E_p$ value depends on which aspect of the nonparabolicity one wants to describe. For example, $E_p$ defined[1] from the wavefunction momentum $|\langle\psi_s|\hat{P}_z|\psi_z\rangle|^2$ need not equal $E_p$ derived from fitting the coefficients of the wave-vector $k^4$ of the real conduction band dispersion[8] near $\Gamma$. Furthermore, these two $E_p$'s need not agree with $E_p$ obtained by fitting the diamagnetic exciton Landau level.[9] Consequently, the values of $E_p$ extracted from experiment have an intrinsic scatter even though the measurement precision could be high. For bulk InP, for example, the experimental literature gives $E_p=20.6$ eV (Ref. 9, the value favored by ER), $17\pm1$ (Ref. 10), 16.7 (Ref. 11), and 16.6 eV (Ref. 12). However, as shown[1] by ER, changing the input $E_p$ from 20.6 to 18.0 eV changes *completely* the EMA result (including the order of states). Thus, the ''predicted'' physics of EMA depends sensitively on the parameters that unfortunately have an *intrinsic scatter*. This highlights the intrinsic nontransferability of $E_p$, thus the limited reliability of using $E_p$ to *predict* the quantum dot physics from $k\cdot p$. (2) In some cases[5–7] the EMA parameters were adjusted to the experiments they claim to explain theoretically (i.e., directly to quantum dot data). Because of this practice, and because different experiments yield inherently different $E_p$'s, the EMA cannot examine the legitimacy of either its successes or its failures.

*(ii) That the $8\times8$ $k\cdot p$ produces very different results if one changes the input parameters from $E_p=20.6$ eV, $f=-1.1$ to $E_p=18.0$ eV, $f=-0.19$.*

The fact that in the EMA, rather small change in the input parameter (equal or smaller than the scatter in the literature values[9–12]) alters the results radically should be a concern to the users of this approach. We do not recommend the use of the $k\cdot p$.

*(iii) That we should change our pseudopotential, in an attempt to fit their $E_p=20.6$ eV value.*

The letter[2] being commented on tests the commonly used[5,13] $6\times6$ $k\cdot p$ approach, a formalism in which the value of $E_p$ does not enter (it does[3] in $8\times8$ $k\cdot p$). While our calculated[3] $E_p=2|\langle\psi_s|\hat{P}_z|\psi_z\rangle|^2=15$ eV is close to most of the ''experimental'' values,[10–12] the aim of our work[2,3] is not to derive a new $E_p$ value, to be added to the long list of the existing values. Instead, we aim to compare the predictions of two approaches—the $k\cdot p$ and the direct diagonalization method (DDM)—*starting from a common physical input* (the bulk band structure). Given that the two approaches tested have equivalent ''input,'' and that one (the DDM) is systematically converged by including many bands while the other is hard-wired to only four, six, or eight bands, the disagreement between two methods does mean a failure of the EMA. Our side-by-side comparison shows that the EMA for dots sometimes gives incorrect level ordering[2] and even omits some levels.[3]

As to the value of $E_p=15$ eV used in our $8\times8$ $k\cdot p$ calculation[3] we note the following:

(a) The real criterion for selecting $E_p$ should be to obtain a good fit to the real bulk band structure in those parts of the Brillouin zone (BZ) that matter for small dots. Our pseudopotential calculation[14] (not $k\cdot p$) produces, by design, an excellent fit to the measured bulk bands *throughout* the BZ, and therefore, needs no adjustment.

(b) If one wants to improve the pseudopotential by fitting the definition of[15] $E_p\equiv2|\langle\psi_s|\partial\hat{H}/\partial k_z|\psi_z\rangle|^2$, one can only achieve this by improving the bulk wave functions $|\psi_s\rangle$ and $|\psi_z\rangle$. However, by our construction of the pseudopotential,[14] there is no need to further improve the wave functions, because they already have 99.7% overlap with the local-density approximation wave functions, which are usually very accurate compared with many-body wave functions.

(c) Our work clearly showed[2] that the wave functions of

[a)]Electronic mail: alex_zunger@nrel.gov

 

dots have large contributions *away* from bulk $\Gamma$. Thus, any theory that works only near $\Gamma$ (e.g., EMA) is insufficient, regardless of the numerical value of $E_p$.

[1] Al. L. Efros and M. Rosen, Appl. Phys. Lett. **73**, 1155 (1998).
[2] H. Fu, L. W. Wang, and A. Zunger, Appl. Phys. Lett. **71**, 3433 (1997).
[3] H. Fu, L. W. Wang, and A. Zunger, Phys. Rev. B **57**, 9971 (1998).
[4] A. Zunger, MRS Bull. **23**, 35 (1998).
[5] M. Nirmal, D. J. Norris, M. Kuno, M. G. Bawendi, Al. L. Efros, and M. Rosen, Phys. Rev. Lett. **75**, 3728 (1995).
[6] Al. L. Efros, V. A. Kharchenko, and M. Rosen, Solid State Commun. **93**, 281 (1995).
[7] O. Wind, F. Gindell, and U. Waggon, J. Lumin. **72-74**, 300 (1997).
[8] D. J. Norris and M. G. Bawendi, Phys. Rev. B **53**, 16 338 (1996).
[9] S. I. Kokhanovski, Yu. M. Makushenko, R. P. Seisyan, Al. A. Efros, T. V. Yazeva, and M. A. Abdullaev, Sov. Phys. Solid State **33**, 967 (1991).
[10] P. Rochon and E. Fortin, Phys. Rev. B **12**, 5803 (1975).
[11] L. G. Shantharama, A. R. Adams, C. N. Ahmad, and R. J. Nicholas, J. Phys. C **17**, 4429 (1984).
[12] T. Nakashima, C. Hamaguchi, J. Komento, and M. Ozeki, J. Phys. Soc. Jpn. **54**, 725 (1985).
[13] T. Richard, P. Lefebvre, H. Mathieu, and J. Allegre, Phys. Rev. B **53**, 7287 (1996).
[14] H. Fu and A. Zunger, Phys. Rev. B **55**, 1642 (1997).
[15] The correct form is $E_P = 2|\langle \psi_s | \partial \hat{H} / \partial k_z | \psi_z \rangle|^2$. It differs slightly (by 7%) from $2|\langle \psi_s | \hat{P}_z | \psi_z \rangle|^2 = 15$ eV since the Hamiltonian $H$ contains a nonlocal pseudopotential component.