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1981 J. Phys. A: Math. Gen. 14 L129

(http://iopscience.iop.org/0305-4470/14/5/006)

J. Phys. A: Math. Gen. **14** (1981) L129–L132. Printed in Great Britain

**LETTER TO THE EDITOR**

# Estimates of the $A^2$-term in the interaction of matter with radiation

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Received 2 February 1981

**Abstract.** If the energy of a molecule coupled to the quantised radiation field is made an extremum with respect to products of arbitrary molecular states and coherent field states, then there results a nonlinear Schrödinger equation for the molecule from which bounds for the energy contribution of the $A^2$-term are derived. It is shown in what sense this contribution can be expected to be small under rather general circumstances.

Recent non-perturbative investigations of both stationary and thermal equilibrium states of matter interacting with radiation, mostly restricted to finitely many field modes, have led to an extensive debate over justification, if any, of the neglect of the so-called $A^2$-term in the fundamental Hamiltonian (for the case of two-level molecules, see e.g. Orszag (1977), Knight *et al* (1978), van Hemmen (1980); for treatments without two-level truncation, but with the electric dipole approximation, see Woolley (1976), Bialynicki-Birula and Rzążewski (1979)). Arguments for this simplification range from perturbation theory to selection rules to canonical transformations.

To state the problem in full generality, consider (all in au) the molecular Hamiltonian

$$h=\sum_{\nu=1}^{N}(2m_\nu)^{-1}|\boldsymbol{p}_\nu|^2+\sum_{\substack{\nu,\mu=1\\ \nu<\mu}}^{N} z_\nu z_\mu|\boldsymbol{q}_\nu-\boldsymbol{q}_\mu|^{-1}$$

with masses $m_\nu$, charges $z_\nu$, position and momentum operators $\boldsymbol{q}_\nu$ and $\boldsymbol{p}_\nu$ ($\nu=1,\ldots,N$). In the Coulomb gauge, the Hamiltonian for the molecule coupled to the transverse part of the electromagnetic field (quantised in a cube of length $L$) reads

$$\begin{aligned}H^{(L)}=&\sum_{\nu=1}^{N}(2m_\nu)^{-1}:|\boldsymbol{p}_\nu\otimes 1-z_\nu\boldsymbol{A}^{(L)}(\boldsymbol{q}_\nu)|^2:\\&+\sum_{\substack{\nu,\mu=1\\ \nu<\mu}}^{N} z_\nu z_\mu|\boldsymbol{q}_\nu-\boldsymbol{q}_\mu|^{-1}\otimes 1+1\otimes\sum_{n=1}^{\infty}c|\boldsymbol{k}_n^{(L)}|b_n^*b_n\\=&H'^{(L)}+\sum_{\nu=1}^{N}(2m_\nu)^{-1}:|z_\nu\mathbf{A}^{(L)}(\boldsymbol{q}_\nu)|^2:,\end{aligned}$$

$$\boldsymbol{A}^{(L)}(\boldsymbol{q}_\nu)=\sum_{n=1}^{\infty}\boldsymbol{\lambda}_n^{(L)}\,[\exp(\mathrm{i}\boldsymbol{k}_n^{(L)}\cdot\boldsymbol{q}_\nu)\otimes b_n+\exp(-\mathrm{i}\boldsymbol{k}_n^{(L)}\cdot\boldsymbol{q}_\nu)\otimes b_n^*],$$

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0305-4470/81/050129+04\$01.50  

and acts formally in $L^2(\mathbb{R}^{3N})\otimes\mathcal{H}$. Here, $\mathcal{H}$ is the ('complete') infinite tensor product of the one-photon Hilbert space; : : means normal order of the photon creation and annihilation operators $b_n^*$, $b_n$ $(n=1,2,\ldots)$; $c$ is the velocity of light; and $\{(\boldsymbol{\lambda}_n^{(L)}, \boldsymbol{k}_n^{(L)})\}_{n=1}^{\infty}$ is an enumeration of

$$\{((cL^2|\boldsymbol{n}|)^{-1/2}\boldsymbol{e}_{\boldsymbol{n},l}, (2\pi/L)\boldsymbol{n})\}_{\substack{\boldsymbol{n}\in\mathbb{Z}^3\setminus\{\boldsymbol{0}\}\\ l=1,2}}$$

where the polarisation vectors $\boldsymbol{e}_{\boldsymbol{n},l}\in\mathbb{R}^3$ satisfy $\boldsymbol{e}_{\boldsymbol{n},l}\cdot\boldsymbol{e}_{\boldsymbol{n},l'}=\delta_{l,l'}$and $\boldsymbol{e}_{\boldsymbol{n},l}\cdot\boldsymbol{n}=0$ $(l, l'=1,2;$ $\boldsymbol{n}\in\mathbb{Z}^3\setminus\{\boldsymbol{0}\})$. So $c\boldsymbol{A}^{(L)}(\ )$ is the (transverse) vector potential. (Note that no two-level, dipole, or finite-number-of-modes approximation is invoked. Neither is the Coulomb potential in $h$ essential for what follows.) The question then is under what conditions the difference $H^{(L)}-H'^{(L)}$ is negligible. Of special interest is the infrared limit $L\to\infty$, which is often taken in conjunction with $N\to\infty$ such that $N/L^3=\text{constant}$.

This Letter substantiates some recent announcements (Pfeifer 1980b, Davis 1980) by developing an answer for a Hartree-type ansatz for the eigenstates of $H^{(L)}$, for $L\to\infty$ and fixed $N$. Focus on such molecule $\otimes$ field factorisation seems justified both on mathematical and conceptual grounds (Spohn 1980, Pfeifer 1980a, Primas 1981).

We need some preparations.

*Definitions.* For $p\in[1,\infty]$, let $\boldsymbol{L}^p(\mathbb{R}^3)=L^p(\mathbb{R}^3)\oplus L^p(\mathbb{R}^3)\oplus L^p(\mathbb{R}^3)$. In $\boldsymbol{L}^2(\mathbb{R}^3)$, define the unitary Fourier transform $F$, the self-adjoint Laplacian $\Delta$, and the orthogonal projector $T$ onto the transverse functions, by

$$(F\boldsymbol{f})(\boldsymbol{k})=\int_{\mathbb{R}^3}\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\boldsymbol{f}(\boldsymbol{r})\,\mathrm{d}(\mathbf{r}),\qquad (F\Delta\boldsymbol{f})(\boldsymbol{k})=-|\boldsymbol{k}|^2(F\boldsymbol{f})(\boldsymbol{k}),$$

$$(FT\boldsymbol{f})(\boldsymbol{k})=\boldsymbol{k}\times((F\boldsymbol{f})(\boldsymbol{k})\times\boldsymbol{k})|\boldsymbol{k}|^{-2},$$

$(\boldsymbol{k}\in\mathbb{R}^3)$ for all suitable $\boldsymbol{f}\in\boldsymbol{L}^2(\mathbb{R}^3)$. For $\varphi\in\mathcal{D}(h)$ ($\mathcal{D}(\ )$ denotes the domain), define the electrical current density $\boldsymbol{j}_\varphi$ and the even part of the mass-weighted charge density, $w_\varphi$, by

$$\boldsymbol{j}_\varphi(\boldsymbol{r})=\Big\langle\varphi\Big|\tfrac{1}{2}\sum_{\nu=1}^{N}(z_\nu/m_\nu)[\delta(\boldsymbol{r}-\boldsymbol{q}_\nu)\boldsymbol{p}_\nu+\boldsymbol{p}_\nu\delta(\boldsymbol{r}-\boldsymbol{q}_\nu)]\varphi\Big\rangle,$$

$$w_\varphi(\boldsymbol{r})=\Big\langle\varphi\Big|\tfrac{1}{2}\sum_{\nu=1}^{N}(z_\nu^2/m_\nu)[\delta(\boldsymbol{r}-\boldsymbol{q}_\nu)+\delta(\boldsymbol{r}+\boldsymbol{q}_\nu)]\varphi\Big\rangle,$$

$(\boldsymbol{r}\in\mathbb{R}^3)$; $W_\varphi$ is the self-adjoint operator of multiplication (in $\boldsymbol{L}^2(\mathbb{R}^3)$) by $w_\varphi$. Finally, for self-adjoint operators $A$ and $B$ we write $0\leqslant A\leqslant B$ if both are non-negative and satisfy $\mathcal{D}(B^{1/2})\subset\mathcal{D}(A^{1/2})$ with $\|A^{1/2}f\|\leqslant\|B^{1/2}f\|$ for all $f\in\mathcal{D}(B^{1/2})$.

*Proposition 1.* If $\varphi\in\mathcal{D}(h)$, then: $\boldsymbol{j}_\varphi\in\boldsymbol{L}^p(\mathbb{R}^3)$ for $p\in[1,2]$; $\boldsymbol{j}_\varphi\in\mathcal{D}((-\Delta)^{-1/2})$; $w_\varphi\in L^p(\mathbb{R}^3)$ for $p\in[1,3]$.

*Proof.* This follows from $\mathcal{D}(h)=\mathcal{D}(\Sigma_{\nu=1}^{N}|\boldsymbol{p}_\nu|^2)$, Hölder estimates, standard Sobolev embeddings and simple fall-off properties of the Fourier transform.

*Proposition 2.* Let $\varphi\in\mathcal{D}(h)$ and put

$$K_\varphi=-c^2(4\pi)^{-1}\Delta+TW_\varphi T,$$

$$a_\varphi=4\pi c^{-2}\min\Big[4(4\pi/3)^{-2/3}\|w_\varphi\|^{\#}_{3/2},\ \tfrac{1}{3}(2/\pi)^{4/3}\|w_\varphi\|_{3/2},\ \Big(6\pi^{-4}\int_{\operatorname{supp} w_\varphi}\mathrm{d}(\boldsymbol{r})\Big)^{2/3}\|w_\varphi\|_\infty\Big]$$

where $\| \|_p$ and $\| \|_p^{\#}$ are the $L^p(\mathbb{R}^3)$-norm and weak $L^p(\mathbb{R}^3)$-'norm'. (Note that $TK_\varphi = K_\varphi T$ and $a_\varphi < \infty$.) Then

$$K_\varphi = K_\varphi^*, \qquad \mathscr{D}(K_\varphi) = \mathscr{D}(\Delta), \qquad K_\varphi^{-1} \text{ exists,}$$
$$0 \le K_0 \le K_\varphi \le (1 + a_\varphi)K_0, \qquad 0 \le (1 + a_\varphi)^{-1}K_0^{-1} \le K_\varphi^{-1} \le K_0^{-1}.$$

*Proof.* Since $w_\varphi \in L^2(\mathbb{R}^3)$ (proposition 1), $K_\varphi = K_\varphi^*$ and $\mathscr{D}(K_\varphi) = \mathscr{D}(\Delta)$ by the Kato–Rellich theorem as usual. Invertibility of $K_\varphi$ follows from $0 \le K_0 \le K_\varphi$ and that of $K_0$. Use uncertainty inequalities (Faris 1978) for $K_\varphi \le (1 + a_\varphi)K_0$. See Kato (1966) for inversion of these operator inequalities.

These preliminaries show that all expressions in the main results are well defined.

*Theorem 1.* (a) For $L \to \infty$, the energy $\langle \varphi \otimes \Phi | H^{(L)}(\varphi \otimes \Phi)\rangle$ is stationary with respect to normalised $\varphi \in \mathscr{D}(h)$ and $\Phi = |\alpha_1, \alpha_2, \ldots\rangle \in \mathscr{H}$, such that $b_n|\alpha_1, \alpha_2, \ldots\rangle = \alpha_n|\alpha_1, \alpha_2, \ldots\rangle$ ($n = 1, 2, \ldots; \alpha_1, \alpha_2, \ldots \in \mathbb{C}$), if and only if $\varphi$ is a stationary point of the functional given by

$$E(\psi) = \langle \psi | h\psi \rangle - \tfrac{1}{2}\|K_\psi^{-1/2} Tj_\psi\|^2 \qquad (\psi \in \mathscr{D}(h); \|\psi\| = 1)$$

(and $\alpha_1, \alpha_2, \ldots$ are chosen in a prescribed, $(\varphi, L)$-dependent manner); in which case

$$\lim_{L \to \infty} \langle \varphi \otimes \Phi | H^{(L)}(\varphi \otimes \Phi)\rangle = E(\varphi).$$

(b) Assertion (a) remains true if $H^{(L)}$ and $E(\;)$ are replaced by $H'^{(L)}$ and $E'(\;)$ where

$$E'(\psi) = \langle \psi | h\psi \rangle - \tfrac{1}{2}\|K_0^{-1/2} Tj_\psi\|^2 \qquad (\psi \in \mathscr{D}(h); \|\psi\| = 1).$$

*Proof.* The energy depends quadratically on the coherent-state arguments $\alpha_1, \alpha_2, \ldots$. For fixed $\varphi$ and $L \to \infty$, the ensuing system of coupled linear equations for $\alpha_1, \alpha_2, \ldots$ can be solved in terms of the solution of the limiting integral equation, which leads to the appearance of $K_\varphi^{-1}$. Insertion in the energy expression yields the stated result.

Thus, within the scope of theorem 1, we can estimate the effective size of the $A^2$-term from the following corollary of proposition 2.

*Theorem 2.* For (normalised) $\varphi \in \mathscr{D}(h)$,

$$0 \le E(\varphi) - E'(\varphi) \le a_\varphi(1 + a_\varphi)^{-1}\tfrac{1}{2}\|K_0^{-1/2} Tj_\varphi\|^2.$$

For example, for low-lying stationary points $\varphi$ of $E(\;)$ one expects that the density $w_\varphi$ is bounded with $\|w_\varphi\|_\infty$ not exceeding a few au for moderate charges $z_\nu$ (high peaks in $\varphi$ for large masses $m_\nu$ will presumably be compensated by the weights $m_\nu^{-1}$ in $w_\varphi$). So even the very crude estimate

$$\begin{aligned} a_\varphi &\le (8/3)(2/\pi)^{1/3} c^{-2} \|w_\varphi\|_{3/2} \\ &\le (8/3)(2/\pi)^{1/3} c^{-2} \left( \sum_{\nu=1}^{N} z_\nu^2 / m_\nu \right)^{2/3} \|w_\varphi\|_\infty^{1/3} \\ &\le 10^{-4} (\text{no of electrons})^{2/3} \|w_\varphi\|_\infty^{1/3} \end{aligned}$$

indicates that, at least for small molecules, the contribution of the $A^2$-term is at most a few percent of the radiative correction $E'(\varphi)-\langle\varphi|h\varphi\rangle$. In any case, the latter is never smaller than the $A^2$-contribution.

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