Some fundamental aspects of the modelling of interaction between atoms and photons with emphasis on ultrastrong coupling and the Dicke criticality

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Origin of standard models of quantum optics
QED of electric dipoles in Coulomb (minimal-coupling) gauge

Charges in EM field in Coulomb gauge ($\nabla \cdot \mathbf{A} \equiv 0$):

$$H = \sum_{\alpha} \frac{1}{2m_{\alpha}} \left[ p_{\alpha} - q_{\alpha}A(r_{\alpha}) \right]^2 + V_{\text{Coulomb}} + H_{\text{field}}$$

Dipole approximation

- $V_{\text{Coulomb}}$ split into intra- & interatomic ($\Rightarrow$ dipole–dipole) parts
- Long-wavelength approximation: $A(r_{\alpha}) \to A(r_A)$

$$H_{\text{ED}} = \sum_{A} \left[ H_A - u p_A A(r_A) + v A^2(r_A) + \sum_{B} V_{\text{Coulomb}}^{\text{dipole-dipole}}(\|r_A - r_B\|) \right] + H_{\text{field}}$$

With single-atom Hamiltonian $H_A = \sum_{\alpha \in A} \left( \frac{p_{\alpha}^2}{2m_{\alpha}} + \frac{q_{\alpha}}{8\pi\varepsilon_0} \sum_{\beta \in A, \beta \neq \alpha} \frac{q_{\beta}}{|r_{\alpha} - r_{\beta}|} \right)$
Origin of standard models of quantum optics

Further necessary approximations

Neglect of $A^2$ & dipole-dipole term in limit of small charge density

Atomic two-level approximation $\Rightarrow$ atomic operator $\sigma_A \equiv |g\rangle_A \langle e|_A$
$\Rightarrow$ collective operator $S \equiv \sum_A \sigma_A$ (spin algebra)

Single-mode (+ rotating-wave) approximation for field $\Rightarrow$ field operator $a$

\[
\begin{align*}
\left\{H_{\text{Dicke}} \quad H_{\text{Tavis–Cummings}}\right\} &= \omega_A S_z + \left\{\frac{\gamma}{\sqrt{N}} (a + a^\dagger) S_x \quad \frac{\gamma}{\sqrt{N}} (a S^\dagger + a^\dagger S)\right\} + \omega_C a^\dagger a \\
\Rightarrow & \quad \left\{\begin{array}{l} 2\text{nd order phase transition} \\
1\text{st order phase transition}\end{array}\right.
\end{align*}
\]

Neglected terms

1. $A^2$ term: basis of „no-go“ argumentation
2. dipole-dipole interaction term: usually not considered in this respect

Proposition: 2 exactly cancels 1 (in arbitrary geometry)
Historical overview, motivation

1954 Dicke model
   ▶ Electric dipoles in an EM mode

1973 Superradiant phase transition in Dicke model
   ▶ Hepp–Lieb; Wang–Hioe
   ▶ Condition: ultrastrong atom-field coupling

1975–2010 “No-go” statement for phase transition – several versions
   ▶ Rzazewski–Wodkiewicz; Aharonov–Knight; etc.
   ▶ Presence of $A^2$ term eradicates phase transition

1982 $A^2$ term is compensated by dipole-dipole term
   ▶ Power–Thirunamachandran

2010 Dicke model & superradiant phase transition realized
   ▶ Esslinger group, ETH Zürich (excitations of BEC in cavity field)

2010– ultrastrong atom-field coupling realized
   ▶ 2D electron gas in semiconductor heterostructures
   ▶ hybrid circuit-CQED systems

2012–’14 Elimination of $A^2$ term in general
   ⇒ refutation of „no-go” argumentations
   ▶ Vukics–Grießer–Domokos
Electrodynamics in Coulomb gauge
in arbitrary domains bounded by perfect conductors

- Domain $\mathcal{D}$ – boundary $\partial \mathcal{D}$ – physical vector fields $L^2(\mathcal{D}, \mathbb{R}^3)$
- finite-energy electrodynamics (no point charges)
- Gauge & boundary condition: $\nabla \cdot \mathbf{A} = 0$, $\mathbf{A} \times \mathbf{n}|_{\partial \mathcal{D}} = 0 \Rightarrow \mathbf{A} \in L^2_0(\mathcal{D}, \mathbb{R}^3)$
- Helmholtz–Hodge decomposition:
  \[
  L^2 = \text{ker}(\text{div}) \oplus \text{ran}(\text{grad}_0) \oplus \text{ran}(\text{curl}) \Rightarrow L^2_0 = \text{ran}(\text{grad}_0) \oplus \text{ker}(\text{div}_0)
  \]

- $\mathbb{H}_2$ – cohomological fields
  - $\dim(\mathbb{H}_2) = b_2 = \# \text{ (components of } \partial \mathcal{D}) - \# \text{ (components of } \mathcal{D})$
  - e.g. Fabry–Pérot resonator: $\dim(\mathbb{H}_2) = 1$

- Full gauge fixing: $U|_{\partial \mathcal{D}} = 0 \Rightarrow U \in \text{dom}(\text{grad}_0)$, $\mathbf{A} \in \text{ker}(\text{div}_0)$
- Depending on all these

\[
H = \sum_\alpha \frac{1}{2m_\alpha} \left[ p_\alpha - q_\alpha A(r_\alpha) \right]^2 + \frac{\varepsilon_0}{2} \int_\mathcal{D} dV \left( \nabla U \right)^2 + \frac{\varepsilon_0}{2} \int_\mathcal{D} dV \left[ \left( \frac{\Pi}{\varepsilon_0} \right)^2 + c^2 \left( \nabla \times \mathbf{A} \right)^2 \right]
\]

- Mode expansion: $\nabla \times \nabla \times \varphi_\lambda = \frac{\omega_\lambda^2}{c^2} \varphi_\lambda$, with $\varphi_\lambda \in \text{ker}(\text{div}_0)$
  - $\omega_\lambda = 0 \iff \varphi_\lambda \in \mathbb{H}_2$
Hamiltonian to electric-dipole order …
… and general problems of Coulomb-gauge description

Electric dipole approximation

\[
H_{ED} = \sum_A \left[ H_A - u p_A \cdot A(r_A) + v A^2(r_A) + V_{\text{dipole-self Coulomb}}(r_A) + \sum_B V_{\text{dipole-dipole Coulomb}}(r_A, r_B) \right] + H_{\text{field}}
\]

Problems

1. Atomic canonical & kinetic momenta differ

2. Presence of \( A^2 \) term
   2.1 Photon-pair creation & annihilation
   2.2 Intercouples all modes (including 0 frequency)
      ▶ single-mode approximation?
      ▶ photon-picture?

3. Electrostatic problem
   3.1 \( V_{\text{dipole-dipole Coulomb}} \) modified by boundaries
   3.2 \( V_{\text{dipole-self Coulomb}} \) as new term
Generalized orthogonal projector decomposition

$$\text{id}_{L^2_0} = \mathcal{Q} + \mathcal{R}$$

With $\mathcal{Q} : L^2_0 \rightarrow \text{ran}(\text{grad}_0), \quad \mathcal{R} : L^2_0 \rightarrow \text{ker}(\text{div}_0)$

- $\mathcal{R}$ projector related to gauge and boundary condition, e.g. $A = \mathcal{R}A$

(Explicit expressions:

- $\forall \mathbf{v} \in L^2 \quad (\mathcal{Q}\mathbf{v})(\mathbf{r}) = -\nabla \int_{\mathcal{D}} d\mathbf{V}' \cdot (\nabla' \cdot \mathbf{v}(\mathbf{r}')) G(\mathbf{r}, \mathbf{r}')$

- $\mathcal{R} = \sum_{\lambda} \varphi_{\lambda} \otimes \varphi_{\lambda}$)

In free space

- $\dim(\mathcal{H}_2) = 0 \quad \Rightarrow \quad \text{ran}(\text{grad}) = \text{ker}(\text{curl}), \quad \text{ran}(\text{curl}) = \text{ker}(\text{div})$

- $\mathcal{Q} = \delta_\parallel, \quad \mathcal{R} = \delta_\perp$
Canonical transformation

generalized Power–Zineau–Woolley transformation

Type-2 generator function (P at this point an arbitrary vector):

\[ G_2 \equiv \int_\mathcal{D} d\mathcal{V} \ A \cdot (\Pi' + \mathcal{R}P) + \sum_\alpha \mathbf{r}_\alpha \cdot \mathbf{p}'_\alpha \]

\[ \Rightarrow \text{displacement of momenta:} \]

\[ \Pi = \frac{\delta G_2}{\delta \mathbf{A}} = \Pi' + \mathcal{R}P, \quad \mathbf{p}_\alpha = \frac{\partial G_2}{\partial \mathbf{r}_\alpha} = \mathbf{p}'_\alpha + \frac{\partial}{\partial \mathbf{r}_\alpha} \int_\mathcal{D} d\mathcal{V} \ A \cdot \mathbf{P}. \]

Transformed Hamiltonian:

\[ H' = \sum_\alpha \frac{1}{2m_\alpha} \left[ \mathbf{p}'_\alpha + \frac{\partial}{\partial \mathbf{r}_\alpha} \int_\mathcal{D} d\mathcal{V} \ A \cdot \mathbf{P} - q_\alpha A(\mathbf{r}_\alpha) \right]^2 \]

\[ + \frac{\varepsilon_0}{2} \int_\mathcal{D} d\mathcal{V} (\nabla U)^2 + \frac{\varepsilon_0}{2} \int_\mathcal{D} d\mathcal{V} \left[ \left( \frac{\Pi' + \mathcal{R}P}{\varepsilon_0} \right)^2 + c^2 (\nabla \times \mathbf{A})^2 \right] \]

Imposed condition:

\[ \varepsilon_0 \nabla U = \mathfrak{Q} \mathbf{P} \iff \nabla \cdot \mathbf{P} = -\rho \quad \Rightarrow \text{interpretation of P: polarization density} \]
Electric dipole approximation in new picture

Condition to other orthogonal component of $\mathbf{P}$

$$\frac{\partial}{\partial r_\alpha} \int_D d\mathbf{V} \mathbf{A} \cdot \mathcal{R}\mathbf{P} = q_\alpha \mathbf{A}(r_\alpha)$$

$\Rightarrow$ simplified Hamiltonian

$$H' = \sum_\alpha \frac{p'_\alpha}{2m_\alpha} + \frac{1}{2\varepsilon_0} \int_D d\mathbf{V} \mathbf{P}^2 - \frac{1}{\varepsilon_0} \int_D d\mathbf{V} \mathbf{D} \cdot \mathbf{P} + H'_\text{field}$$

with $\mathbf{D} \equiv \varepsilon_0 \mathbf{E} + \mathbf{P}$ ($= -\mathbf{\Pi}'$), and $\mathbf{D} = \mathcal{R}\mathbf{D}$

$\Rightarrow$ Cf. no “free” charges (that would not be described by $\mathbf{P}$)

Condition fulfilled approximately by separated atomic dipoles

$$\mathbf{P} = \sum_A \mathbf{P}_A \quad \Rightarrow \quad \int_D d\mathbf{V} \mathbf{P}^2 = \sum_A \int_D d\mathbf{V} \mathbf{P}_A^2$$

$$\mathbf{P}_A(\mathbf{r}) = \mathbf{d}_A \delta_< (\mathbf{r} - \mathbf{r}_A) \quad \text{“long-wavelength delta function”}$$
Result

Hamiltonian in new picture in electric dipole order

\[ H'_{\text{ED}} = \sum_A \left( H'_A - d_A \cdot \frac{D(r_A)}{\varepsilon_0} \right) + H'_{\text{field}} \]

with single-atom Hamiltonian

\[ H'_A = \sum_{\alpha \in A} \frac{p'_\alpha^2}{2m_\alpha} + \frac{1}{2\varepsilon_0} \int_{\text{supp}(P_A)} dV P_A^2 \]

Formally equivalent to free-space

- if \( \text{supp}(P_A) \cap \partial D = \emptyset \) then also quantitatively

Quantization

\( D \) purely “transverse” field (\( D = \mathcal{R} \mathcal{D} \)):

\[ D = -\Pi' = i \sum_{\lambda} \sqrt{\frac{\hbar \varepsilon_0 \omega_\lambda}{2V}} \left( \varphi_\lambda a_\lambda - \varphi^*_\lambda a_\lambda^\dagger \right) + \sum_{\lambda} \varphi_\lambda p_\lambda \]

- Single mode of \( D \) is in general composed of all the modes of \( A \)
- But: in new picture, 0 frequency modes of \( D \) not coupled directly to the rest
Nature of single-atom Hamiltonian

\[ H = \sum_{\alpha} \frac{p_{\alpha}^2}{2m_{\alpha}} + \text{[magnetic term]} + \frac{1}{2\varepsilon_0} \int_{\mathcal{D}} d\mathcal{V} \, P^2 - \frac{1}{\varepsilon_0} \int_{\mathcal{D}} d\mathcal{V} \, \mathbf{D} \cdot \mathbf{P} + H_{\text{field}} \]

⇒ if \( P = \sum_A P_A \) with non-overlapping \( P_A \)s,

\[ H_A = \sum_{\alpha \in A} \frac{p_{\alpha}^2}{2m_{\alpha}} + \frac{1}{2\varepsilon_0} \int_{\text{supp}(P_A)} d\mathcal{V} \, P_A^2 \]

P field for point charges

Power's form (Power & Zienau, Phil. Trans. R. Soc. A 251:427 [1959])

\[ P(r) = \sum_{\alpha} q_{\alpha} r_{\alpha} \int_{0}^{1} du \, \delta(r - ur_{\alpha}) \equiv \sum_{\alpha} q_{\alpha} r_{\alpha} \, C_{r_{\alpha}}(r) = \sum_{A} P_A(r) \]

Problem: potential term contains distribution squared
Proposed solution

Notice that (due to $P^\parallel = -\varepsilon_0 E^\parallel$)

$$V_A = \int d\mathcal{V} P_A^2 = \int d\mathcal{V} \left( P_A^\parallel \right)^2 + \int d\mathcal{V} \left( P_A^\perp \right)^2 \equiv V_{\text{Coulomb},A} + \Delta V_A$$

$P^\perp$ not determined directly by the charges
⇒ freedom of choice, Power form is over-determined

Restrictions on $P^\perp$

I. Elimination of the A-square term

$$\frac{\partial}{\partial r_\alpha} \int d\mathcal{V} P^\perp \cdot A \overset{!}{=} q_\alpha A(r_\alpha) \approx q_\alpha A(r_A), \quad \forall \alpha \in A$$

II. Elimination of the P-square term

$$P_A(r) \simeq d_A \delta_<(r - r_A)$$

III. Respecting atomic physics of the last century

$\Delta V_A$ is only a perturbation to $V_{\text{Coulomb},A}$
Appropriate choice of $P^\perp$ in dipole order

$$P_A^\perp(r) = \delta^\perp(r - r_A) \, d_A$$

Truncated transverse delta function

$$\tilde{\delta}^\perp(k) = \frac{1}{(2\pi)^3} \left( \text{id}_{\mathbb{R}^3} - \frac{k \circ k}{k^2} \right) \frac{k_M^2}{k^2 + k_M^2}$$

In real space

$$P_A^\perp(r) = \frac{\eta(r)}{4\pi r^3} \left[ \frac{3 \,(r \cdot d_A) \, r}{r^2} - d_A \right]$$

with $\eta(r) = 1 - \left( 1 + k_M r + \frac{k_M^2 r^2}{2} \right) e^{-k_M r}$

$\Rightarrow$ condition II. fulfilled in dipole order, if inter-atomic distance $\gtrsim k_M^{-1}$
Nature of the cutoff

No renormalization parameter, but part of the freedom of choice of $P^\perp$

$\Rightarrow$ physical meaning: “intimacy zone” of atoms

(standard models of QO valid if atoms do not penetrate each other’s intimacy zone)

Limits of $k_M$

Lower limit from condition I.

$$\int d\mathcal{V} \frac{\partial P^\perp}{\partial r_\alpha} A = q_\alpha \int d^3 k \frac{k_M^2}{k^2 + k_M^2} \sum_\epsilon [\alpha_\epsilon(k) + \text{c.c.}] \equiv q_\alpha A < (0) \approx q_\alpha A(0)$$

$\Rightarrow k_M \gg k_{\text{optical}}$

Upper limit from condition III.

For hydrogen $\Delta U_{\text{hydrogen}} = \frac{e^2 k_M^3}{24\pi \varepsilon_0} r^2 \Rightarrow E_{1s}^{(1)} = (k_M a_0)^3 \text{ Ry}$

$\Rightarrow k_M^3 \ll (a_0)^{-3}$

The two can be simultaneously met since

$$k_{\text{optical}} \approx k_A = \frac{3\alpha}{8} \frac{1}{a_0}$$
Consequences on feasibility of ultrastrong coupling

Figure of merit

$$\mathcal{F} \equiv \frac{Ng^2}{\omega \omega_A}$$

Expressed in two forms:

With atomic-resonance quality factor

$$\mathcal{F} = \frac{N}{V} \lambda_A^3 \frac{3}{8\pi^2} \frac{1}{Q}$$

With atomic size

$$\mathcal{F} = \frac{N}{V} 16\pi a_0^3$$

From $k_M$ upper limit, the intimacy zone can be minimized to $k_M^{-1} \approx 2 \ a_0$

$\Rightarrow$ ultimate limit:

$$\mathcal{F} \lesssim 1$$

@ $2 \ a_0$ inter-atomic distance – electron-orbit delocalization

$\Rightarrow$ molecule formation, solidification
Dicke critical vs. solid-state density

From the condition of Dicke criticality, $\mathcal{F} = 1$

<table>
<thead>
<tr>
<th></th>
<th>$n_{\text{solid}}$ [$10^{27}/\text{m}^3$]</th>
<th>$n_{\text{critical}}$ [$10^{27}/\text{m}^3$]</th>
<th>$r_{\text{atom}}$ [pm]</th>
<th>$d_{\text{critical}}$ [pm]</th>
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<tr>
<td>Rb</td>
<td>11</td>
<td>7</td>
<td>248</td>
<td>520</td>
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<tr>
<td>Cs</td>
<td>8.6</td>
<td>5.2</td>
<td>265</td>
<td>580</td>
</tr>
</tbody>
</table>

Conclusion

Dicke criticality $\sim$ solidification

but, the dipolar Hamiltonian gives an oversimplified view on the phase transition

Papers

Vukics–Grießer–Domokos

Elimination of A-square problem  PRA86:053807(2012),
                                    PRL112:073601(2014)

Appendix

\[ H_{\text{field}} = \frac{\varepsilon_0}{2} \int_{\mathcal{D}} d\mathcal{V} \left[ \left( \frac{\mathbf{\Pi}}{\varepsilon_0} \right)^2 + c^2 (\nabla \times \mathbf{A})^2 \right] \]

atomic momentum: \( u \mathbf{p}_A \equiv \sum_\alpha \frac{q_\alpha}{m_\alpha} \mathbf{p}_\alpha \)

\[ u \text{ from commutation relation with } \mathbf{r}_A \text{ depending on latter’s definition} \]

vector potential with transverse modes: \( \mathbf{A} = \sum_\lambda \sqrt{\frac{\hbar}{2\varepsilon_0 V\omega_\lambda}} \left( \varphi_\lambda a_\lambda - \varphi_\lambda^* a_\lambda^\dagger \right) \)

An explicit expression in Fabry–Pérot geometry

\[
\mathcal{R}(\rho) \sim \pi \sum_n e^{i k_n \rho z} \int_0^\infty dk_\perp \frac{k_\perp}{k^2} \left( \begin{array}{ccc}
(2k_n^2 + k_\perp^2) J_0(k_\perp \rho_\perp) + k_\perp^2 J_2(k_\perp \rho_\perp) & 0 & -2ik_nk_\perp J_1(k_\perp \rho_\perp) \\
0 & (2k_n^2 + k_\perp^2) J_0(k_\perp \rho_\perp) - k_\perp^2 J_2(k_\perp \rho_\perp) & 0 \\
-2ik_nk_\perp J_1(k_\perp \rho_\perp) & 0 & 2k_\perp^2 J_0(k_\perp \rho_\perp)
\end{array} \right).
\]