Deciphering the coalescence behavior of Coulomb-Schrödinger atomic wave functions from operator product expansion

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We revisit the coalescence behavior of the atomic Schrödinger wave functions from the angle of operator product expansion (OPE) within the nonrelativistic Coulomb-Schrödinger effective field theory. We take the electron-nucleus coalescence as an explicit example to demonstrate our formalism, where the celebrated Kato’s cusp condition can be easily reproduced. An exact OPE relation is rigorously proved to all orders in perturbation theory. Our approach can be readily extended to ascertain the multi-particle coalescence behaviors of atomic wave functions, as well as to take relativistic effects into account.

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Introduction. Quantum Electrodynamics (QED) is rightfully the underlying theory that reigns all phenomena in atomic physics and quantum chemistry. Nevertheless, for the sake of unraveling the property of atoms composed of a heavy nucleus plus $N$ electrons, it looks practically intractable to directly invoke the relativistic field-theoretical machinery, rather it turns to be extremely efficient to start from the nonrelativistic Hamiltonian entailing Coulombic interactions:

$$H_{\text{Coul}} = - \sum_{i=1}^{N} \frac{\nabla_i^2}{2m} - \sum_{i=1}^{N} \frac{Z \alpha}{r_i} + \sum_{j=1}^{N} \frac{\alpha}{r_{ij}}, \quad (1)$$

where the electron mass is labeled by $m$, the nucleus and electron carry electric charges of $Ze$ and $-e$, respectively. $r_i$ signifies the distance between the $i$-th electron and the nucleus, $r_{ij}$ denotes the distance between the $i$-th and $j$-th electrons. Designing accurate numerical algorithms to solve the resulting Schrödinger equation for a multi-electron atom, $H_{\text{Coul}} \Psi = E \Psi$, was actively pursued shortly after the birth of Quantum Mechanics nearly a century ago, yet still occupies the central stage of the atomic physics until today [1].

Atoms are QED bound states formed by a static heavy nucleus together with some slowly-moving electrons. In the modern tenet, these electrically-neutral bound states are believed to be best tackled by the effective field theory (EFT) dubbed nonrelativistic QED (NRQED) [2], which directly descends from QED by integrating out relativistic quantum fluctuations. The notable merit of the EFT approach is to expedite the systematic inclusion of relativistic corrections for various atomic and molecular properties [3–5].

The motif of this work is to demonstrate a remarkable merit of the field-theoretical approach over the traditional Schrödinger equation in portraying the short-distance properties of atomic wave function. Concretely speaking, in this work we are interested in understanding the coalescence behavior of atomic wave functions, that is, what kind of universal behavior the wave function would exhibit when some electrons spatially approach each other, or approach the nucleus. The knowledge about the correct coalescence behavior of wave functions are important, since they provide important constraints on the profiles of trial functions. Better trial functions help to enhance the accuracy of the predicted atomic energy spectrum and various reaction rates. We aim to offer a fresh look at this problem from a field-theoretical angle.

First we give a brief account of the history. To our knowledge, the electron-nucleus coalescence behavior of the hydrogen-like atom with general orbital angular momentum quantum number was first addressed by Löwdin in 1954 [6]. Subsequently, the $S$-wave two-particle coalescence behavior in any multi-electron atom was summarized by Kato in 1957 [7],

$$\left. \frac{\partial \Psi}{\partial r_{12}} \right|_{r_{12}=0} = \gamma \Psi(r_{12} = 0), \quad (2)$$

with $\gamma = \frac{mZ\alpha}{2}$ for two-electron coalescence, and $\gamma = -mZ\alpha$ for an electron coinciding with the nucleus. [7] is often called Kato’s cusp condition. In the 60s, the cusp condition was extended to the hydrogen molecule [8, 9]. Recently, the three-particle coalescence behavior for atomic and molecular wave functions have been addressed by Fournaris et al. [10]. To date nothing is known about four or more particle coalescence behavior of atomic wave functions.

Most preceding work in this area heavily relies upon complicated mathematics in seeking approximate solutions of eigen-wave functions of [1]. which is not so illuminating. In this work, we attempt to provide an alternative perspective, i.e., these universal coalescence behaviors can actually be best understood within the nonrelativistic EFT, by invoking the powerful operator product expansion (OPE) technique.

OPE was originally formulated by Wilson in 1969 [11]. Shortly after it was applied to account for the scaling violation observed in deep-inelastic scattering experiment, and played a vital role in establishing QCD as the fundamental theory.
of strong interaction. OPE has also served a crucial element in the influential QCD sum rules to extract nonperturbative hadron properties \cite{12}.

Besides its ubiquitous applications in high energy physics, OPE also proves to be useful in the realm of atomic physics. It was Lepage who first clarify the implication of renormalization and OPE in nonrelativistic quantum mechanics \cite{13}. In 2008, Braaten and Platter applied the OPE technique to prove the Tan relation for the unitary Fermi gas with a zero-range interaction \cite{14}. Recently Hofmann et al. used OPE to understand the coalescence behavior of electrons within the jellium model \cite{15}.

In this work, we apply the OPE to deduce the coalescence behaviors of atomic wave functions. Unlike \cite{14,15}, the nonrelativistic effective theory we start with has a direct connection to QED. We will also present a rigorous proof based on perturbation theory. Our work can be further improved by including more fields in the operator product, as well as including relativistic corrections.

**Non-relativistic Effective Field Theory for Atoms.** An atom is characterized by the following well-separated scales: \(mv^2 \ll m \ll M_N\), with \(v \sim \alpha \ll 1\) designating the typical velocity of electrons. All aspects of atomic physics can be adequately described by the following effective lagrangian:

\[
\mathcal{L}_{\text{atom}} = \mathcal{L}_{\text{Maxwell}} + \mathcal{L}_{\text{NRQED}} + \mathcal{L}_{\text{HNET}},
\]

where

\[
\mathcal{L}_{\text{Maxwell}} = -\frac{1}{4} F_{\mu \nu} F^{\mu \nu} + \cdots,
\]

\[
\mathcal{L}_{\text{NRQED}} = \psi^\dagger \left(iD_0 + \frac{\nabla^2}{2m}\right) \psi + N^i iD_0 N + \frac{1}{2} (\nabla A^0)^2,
\]

and

\[
\mathcal{L}_{\text{HNET}} = N^i iD_0 N + \cdots,
\]

where \(\psi\) is the Pauli spinor field that annihilates an electron, \(N\) is the Dirac spinor field that annihilates a heavy nucleus at rest. \(D_\mu = \partial_\mu - i e A_\mu\) acts on electron field, \(D_\mu = \partial_\mu + i Z e A_\mu\) acts on nucleus field. The nonrelativistic electron field is described by NRQED \cite{2}, whereas the nucleus, whose role is solely providing a static electric source, is treated in the *heavy nucleus effective theory* (HNET), in a fashion analogous to the heavy quark effective theory (HQET) \cite{16,17}. Here the nucleus is approximated by an infinitely heavy, structureless point charge, and we do not concern about the immaterial effect due to the magnetic moment of nucleus. Note \cite{3} only retains those operators at lowest order in \(v\) and \(1/M_N\).

Eq. (3) is manifestly gauge invariant. However, a common practice to tackle nonrelativistic charged system is to impose the Coulomb gauge \(\nabla \cdot A = 0\), where the instantaneous Coulomb photon can be cleanly separated from the dynamic transverse photons. Since the \(A\) is suppressed with respect to \(A^0\) in NRQED power counting, it is a controlled approximation to drop all occurrences of \(A\). Eq. (3) then essentially reduces into the following nonrelativistic Schrödinger field theory:

\[
\mathcal{L}_{\text{Coul-Schr}} = \psi^\dagger \left(iD_0 + \frac{\nabla^2}{2m}\right) \psi + N^i iD_0 N + \frac{1}{2} (\nabla A^0)^2,
\]

where the last term is responsible for the instantaneous Coulomb potential. This theory may be dubbed Coulomb-Schrödinger effective lagrangian. Obviously, the effect due to dynamical photon, such as Lamb shift \cite{4}, will be inaccessible in \cite{5}. Since the spin degree of freedom decouples in the nonrelativistic limit, for simplicity we will replace the spinor fields \(\psi\) and \(N\) in \cite{5} by complex scalar fields henceforth.

**Operator Product Expansion in Coulomb-Schrödinger EFT.** In order to deduce the coalescence behavior of Coulomb-Schrödinger wave function of an atom, we are motivated to examine how the product of the electron field \(\psi\) and nucleus field \(N\) scales in short-distance, since in the field-theoretical context, the wave functions can be viewed as the product of a string of spatially-nonlocal, yet equal-time \(\psi\)-fields and the \(N\)-field, sandwiched between the vacuum and the bound state \cite{18}.

We start from the product of a single \(\psi\) field and a \(N\) field. One may tentatively guess that, in the small \(x\) limit, the operator product can be expanded as follows,

\[
\psi(x) N(0) = |\psi(0)| N(0) + x \cdot |\nabla N(0)| + \cdots,
\]

which is nothing but the Taylor expansion of \(\psi(x)\) around the origin. Note these two field operators are defined in equal time \(t = 0\). The first operator in the right-hand side carries \(S\)-wave quantum number, the second is of \(P\)-wave type, and the ellipsis represent those irreducible-spherical-tensor operators carrying two or more gradients. We stress again that, it is crucial to introduce the HNET field to fulfill a valid OPE relation.

Incorporating the Coulomb interaction will modify the naive expectation of (6). The highlight of this work is, as we will prove shortly, there exists an exact OPE relation in the EFT defined by (5):

\[
\psi(x) N(0) = (1 - m Z a |x|) |\psi(0)| + (1 - m Z a |x|/2) x \cdot |\nabla N(0)| + \cdots,
\]

where \(\ldots\) is used to denote the renormalized composite operator.

Before moving on, it is worth emphasizing some significant difference between renormalizable and nonrenormalizable (effective) theories on application of OPE. In the former case, the distance \(|x|\) between two operators could literally tend to 0, and the radiative corrections to each Wilson coefficient in the naive OPE series bears the form of \(\ln^a |x|\). By contrast, since (5) only has a limited range of applicability, the smallest distance of \(|x|\) one can probe is of order \(1/m\), the inverse of the UV cutoff of the Coulomb-Schrödinger EFT. The additional corrections to each Wilson coefficient can be linear in \(|x|\), balanced by the Bohr radius \(a_0 = (m Z a)^{-1}\).
The OPE in the momentum space turns out to be also useful:

\[
\psi(q)N(0) = \int d^3x e^{-i\mathbf{q} \cdot \mathbf{x}} \psi(x) N(0) = \frac{8\pi Z\alpha m}{q^4} - \frac{16\pi Z\alpha m}{q^6} q \cdot [\nabla \psi N](0) + \cdots .
\]  

(8)

This expansion is valid provided that \(1/\alpha_0 \ll |q| \lesssim m\). With the aid of the following rudimentary Fourier integrals,

\[
\int \frac{d^3q}{(2\pi)^3} e^{i\mathbf{q} \cdot \mathbf{x}} = \frac{1}{8\pi} \delta(\mathbf{x}),
\]

(9a)

\[
\int \frac{d^3q}{(2\pi)^3} e^{i\mathbf{q} \cdot \mathbf{x}} - \frac{i}{q^6} q = -\frac{i}{32\pi} \delta(\mathbf{x}),
\]

(9b)

one readily recovers the coordinate-space OPE (7). Note that one has to subtract the contribution arising from the local composite operators, to sweep IR divergences encountered in Fourier transform (20).

**Proof of Operator Product Expansion in Coulomb-Schrödinger field theory.** The goal is to prove the momentum-space version of OPE, (8), to all orders in Ze. The proof here is similar in spirit, but technically considerably simpler, than that for the renormalizable quantum field theories [19, 20]. Moreover, to our knowledge, essentially in a nonperturbative fashion. Our proof is entirely based on perturbation theory. Moreover, to our knowledge, this is the first time that the HNET field is introduced to define an OPE relation.

Operator product expansion is an operator equation, which is insensitive to the presence of other fields or external states. We take this freedom to investigate the following connected Green functions:

\[
\Gamma(q, p, E \equiv k^0 + p^0) = \int d^4y d^4z e^{-i\mathbf{p} \cdot \mathbf{y} - i\mathbf{k} \cdot \mathbf{z}} \langle 0 \mid \{\bar{\psi}(\mathbf{q})N(0)\psi^\dagger(\mathbf{y})N^\dagger(\mathbf{z})\} \mid 0 \rangle_{\text{amp}},
\]

(10a)

\[
\Gamma_S(p, E) = \int d^4y d^4z e^{-i\mathbf{p} \cdot \mathbf{y} - i\mathbf{k} \cdot \mathbf{z}} \langle 0 \mid \{\bar{\psi}(\mathbf{q})N(0)\psi^\dagger(\mathbf{y})N^\dagger(\mathbf{z})\} \mid 0 \rangle_{\text{amp}},
\]

(10b)

\[
\Gamma_P(p, E) = \int d^4y d^4z e^{-i\mathbf{p} \cdot \mathbf{y} - i\mathbf{k} \cdot \mathbf{z}} \langle 0 \mid \{\nabla \psi N(0)\psi^\dagger(\mathbf{y})N^\dagger(\mathbf{z})\} \mid 0 \rangle_{\text{amp}},
\]

(10c)

where \(p^\mu, k^\mu\) can be chosen as arbitrary 4-momenta that are much smaller than \(m\). The subscript “amp” in (10) implies that the external propagators carrying soft momenta \(p\) and \(k\) get amputated.

The strategy is to verify that, order by order in Ze, the large \(|q|\) behavior of the Green function \(\Gamma\) does possess the following factorized structure:

\[
\Gamma(q, p, E) \xrightarrow{nZ\alpha \approx q \lesssim m} \frac{8\pi m Z\alpha}{q^4} \Gamma_S(p, E)
\]

\[
- \frac{i16\pi m Z\alpha}{q^6} q \cdot \Gamma_P(p, E) + \cdots .
\]

(11)

Due to the specific causal structure of the propagators associated with electron, nucleus, and Coulomb photon,

\[
D_e(q^0, q) = \frac{i}{q^0 - \frac{q^2}{2m} + i\epsilon}, \quad D_N(q^0) = \frac{i}{q^0 + i\epsilon}, \quad D_C(q) = \frac{i}{q^2},
\]

only those Coulomb ladder diagrams (crossed ladder, self-energy and vertex-correction type diagrams yield null results) contribute to \(\Gamma, \Gamma_S\), and \(\Gamma_P\), as the virtue of nonrelativistic Coulomb-Schrödinger field theory. One can express \(\Gamma = \sum_{\alpha} \Gamma^{(\alpha)}\), with \(n\) signaling the number of exchanged Coulomb ladders (the number of loops is then \(n - 1\)). This topology is pictured in Fig. 1 with single and double solid lines representing the electron and nucleus, respectively. \(\Gamma_S\) and \(\Gamma_P\) also bear the same ladder structures, thereby one can introduce \(\Gamma^{(\alpha)}\) and \(\Gamma_P^{(\alpha)}\) in a similar fashion. It is worth mentioning that, to all orders in Ze, the Green functions \(\Gamma, \Gamma_S\), and \(\Gamma_P\) are UV finite.

Explicitly, the \(n\)-ladder contribution to \(\Gamma\) reads

\[
\Gamma^{(\alpha)} = (Ze^2)^p \int \frac{d^4l_0}{2\pi} \prod_{i=1}^{n-1} \frac{d^4l_i}{(2\pi)^d} \left( \prod_{i=1}^{n-1} D_e(l_i) D_N(E - l_i^0) D_C(l_i - l_{i-1}) \right)
\]

\[
\times \left( \prod_{i=1}^{n-1} \frac{d^4l_i}{(2\pi)^d} \left( \prod_{i=1}^{n-1} D_e(l_i) D_N(E - l_i^0) D_C(l_i - l_{i-1}) \right) \right)
\]

\[
\times D_C(l_n - l_{n-1}) \cdot q,
\]

(12)

where \(l_0 \equiv p\). Note in the final expression, the upper-right HNET propagator in Fig. 1 disappears after integration over \(q^0\), as a consequence of Fourier-transforming the equal-time product of the \(\psi\) and \(N\) fields in (10a). Interestingly, the hard momentum \(q\) injected from the upper-left electron line exits through the upper-right HNET leg, which is however of null impact since the HNET propagator is insensitive to the residual nucleus three-momentum.
The OPE relation holds only for the leading operators for each partial wave, just like the leading \(S\), \(P\)-wave operators specified in (7). The argument is as follows. To the second order in operator expansion, we encounter the local operator \(\nabla_1 \nabla_2 \phi N\), which can be decomposed into a \(S\)-wave and a \(D\)-wave piece. The \(S\)-wave operator \(\nabla_1 \nabla_2 \phi N\) turns out to be power UV-divergent upon implementing the loop corrections, which becomes ill-defined and invalidates the above inductive proof of diagrammatic factorization of OPE. Were this operator allowed to emerge, it would make a contribution to \(\Gamma\) asymptotically \(\propto 1/q^6\). When Fourier-transformed to the coordinate space, one would inevitably encounter insurmountable IR divergence in the limit \(q \to 0\), which appears to be unacceptable.

We conclude that, in the Coulomb-Schrödinger EFT defined in (5), the OPE relations encoded in (7), or equivalently, (8), are exact for the leading local operators of each partial wave. There are no further correction to the corresponding Wilson coefficients.

**Application of OPE to Hydrogen-like atoms** The OPE relations (7) and (8) provide a lucid way to understand the universal electron-nucleus coalescence behavior of wave function for any atom. Nevertheless, for the sake of illustration, we choose the simplest and extremely well-known case, e.g., the wave functions of hydrogen-like atoms to test our formalism. The corresponding wave functions can be identified with the following spatially nonlocal matrix element:

\[
\Psi_{nlm}(x) = \langle 0 | \phi(x) N(0) | nlm \rangle, \tag{14}
\]

with \(n, l, m\) refer to radial, orbital, and magnetic quantum numbers, respectively.

Sandwiching (7) between vacuum and the \(S\)-wave/\(P\)-wave states, after some algebra one predicts that the radial wave functions near the origin become approximately

\[
R_{nl}(x) \xrightarrow{1/m, x \ll a_0} R_{nl}(0) \left(1 - \frac{x}{a_0}\right), \tag{15a}
\]

\[
R_{nl}(x) \xrightarrow{1/m, x \ll a_0} x R'_{nl}(0) \left(1 - \frac{x}{2a_0}\right), \tag{15b}
\]

where the shorthand \(x = |x|\) is used. Note (15) is true for any \(n\), regardless of being discrete or continuum label. One can explicitly confirm these relations by expanding the exact hydrogen atom wave functions near the origin. Reassuringly, the universal behavior is found to be violated at order-\(x^2\) for each partial wave, which confirms our anticipation that OPE must break down at this relative order.
A general relation concerning the wave functions near the origin for the hydrogen-like atoms, valid for any \( l \), was given by Löwdin in 1954 \cite{5}:

\[
R_n(l, x) \equiv \frac{x^l}{l!} \frac{d^l R_{nl}(0)}{dx^l} \left[ 1 - \frac{1}{l + 1} \frac{x}{a_0} + O(x/a_0)^2 \right],
\]

which can be recast into an equivalent form,

\[
\frac{d^l R_{nl}(x)}{dx^l} = \frac{d^l R_{nl}(0)}{dx^l} \left[ 1 - \frac{x}{a_0} + O(x/a_0)^2 \right].
\]

Obviously, our OPE predictions for S- and P-wave hydrogen-like atoms, \cite{15}, coincide with Löwdin’s relation \cite{16}.

Eq. \cite{8} can also be applied to predict the universal behavior of momentum-space wave functions in the large momentum limit:

\[
\tilde{R}_{nl}(q) = 2^{l+2} \frac{d^l R_{nl}(0)}{dx^l} \left[ \frac{(2n)^2}{a_0^4 q^{4l+4}} \right] \left[ 1 + O(1/(qa_0)^2) \right],
\]

with \( q \equiv |q| \), and \( n \) can again be either discrete or continuum label. This universal behavior of the bound wave functions in momentum space appears to be first noted in the famous text by Bethe and Salpeter \cite{21}, yet without any physical explanations. One can verify our OPE predictions by directly expanding the exact momentum-space radial wave functions for both bound \cite{22} and continuum \cite{23} states for arbitrary \( l \).

Summary and Outlook. The atomic physics and quantum chemistry are quite mature fields, the central theme of which is to effectively solve the Schrödinger equations for atoms and molecules. Knowing the true coalescence behavior of wave functions provide important guidance for constructing the optimal trial wave functions. While the two-particle and three-particle coalescence behavior are known, it seems to be a formidable task to extend the traditional differential-equation-based approach to infer the four or more particle coalescence behaviors.

This work approaches this old problem from a fresh field-theoretical perspective. Concretely, we have rigorously proved an exact operator product expansion relation within the Coulomb-Schrödinger effective field theory. This OPE relation can naturally explain Kato’s cusp condition. Our approach, based upon systematic Feynman-diagrammatic technique, can be readily generalized to deduce multi-particle coalescence behavior of atomic or molecular wave functions.

Another interesting direction is to incorporate relativistic corrections into our Coulomb-Schrödinger EFT. Interestingly, this theoretical framework appears to offer interesting insight for solving the long-standing puzzle about the divergent behavior of wave function near the origin for the Klein-Gordon, or more realistic Dirac hydrogen atom \cite{24}.

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