Comment on "Complete Keldysh theory and its limiting cases"

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(Received 29 August 1990)

Reiss [Phys. Rev. A **42**, 1476 (1990)] has argued that Volkov states exist unambiguously only in relativistic theory, and that $\mathbf{A}^2$ must be retained in strong-field theory even after the nonrelativistic and dipole approximations are made. We show that these arguments and their conclusions are incorrect.

PACS number(s): 32.80.Rm, 33.80.Rv

The Keldysh approximation (KA) [1] was originally formulated within the $\mathbf{r}\cdot\mathbf{E}$ form of interaction in the nonrelativistic and dipole approximations (NRD), and was later reformulated under the same approximations by Reiss [2] in terms of the $\mathbf{A}\cdot\mathbf{p}+\mathbf{A}^2$ form. The latter was criticized because, *within the NRD used by Reiss,* the $\mathbf{A}^2$ term can be eliminated by a simple contact transformation, whereas in Reiss's analysis it was found to play a major physical role [3–5]. Reiss subsequently claimed that one must begin with relativistic theory and then perform a nonrelativistic limit in order to resurrect the results of the earlier formulation [6]. Reiss also arrives at the conclusion that a Volkov state exists unambiguously only in relativistic theory.

Before addressing these points, I wish to note that, to my knowledge, no one denies that the KA "remains a valuable benchmark" [5] in strong-field ionization theory. A modified version of the KA was recently proposed [5] and found to work quite well in a model problem [7]. At issue, in addition to Reiss's version, is the accuracy of the KA. Whereas Keldysh noted that the KA differs from standard perturbation theory only in its use of a Volkov state rather than a final state unperturbed by the field [1], Reiss claims that "the field interaction has been completely retained everywhere that it originally appeared in the exact S matrix" and that "the approximation ... should improve in accuracy" as the field becomes stronger [6].

I wish to take issue with three claims in Reiss's paper: (1) that $\mathbf{A}^2$ must be retained even after the NRD, (2) that the KA fully retains the field interaction appearing in the exact S matrix, and (3) that Volkov states exist unambiguously only relativistically.

The $\mathbf{A}^2$ term, of course, cannot in general be transformed away, especially when the field cannot be accurately treated semiclassically. A recent example where the $\mathbf{A}^2$ term is crucial may be found in Ref. [8]. But when the field may be treated as classically prescribed and when the NRD are good approximations, it is very well known that $\mathbf{A}^2$ is of no physical consequence [9].

This issue is an old one. It was shown by Kibble *et al.* [10] that the $\mathbf{A}^2$ term is canceled in a low-frequency approximation by the second-order contribution from the $\mathbf{A}\cdot\mathbf{p}$ interaction. This explained the null result of experiments to observe this shift. It must be emphasized that this cancellation does not depend on the NRD. The same result does obtain within the NRD, however [11]. In Ref. [5] it was shown that "the famous ponderomotive potential" that Reiss claims to obtain in his approach is only *approximately* equal to $\mathbf{A}^2$ in magnitude, at low frequencies, and furthermore that it follows without the $\mathbf{A}^2$ term in the Hamiltonian. All these results are in contrast to Reiss's analysis, where the ponderomotive shift is *exactly* $\mathbf{A}^2$ at all field frequencies.

The issue here is also closely connected with Reiss's claim that in his version of the KA "the field interaction has been retained everywhere that it appeared in the exact S matrix". It was shown in Ref. [5] that the KA ignores completely the effect of the field on the initial bound state. In particular, it ignores any field-induced shifts of bound levels; this is why Reiss does not find the cancellation mentioned in the preceding paragraph. This feature of the KA was recognized by Keldysh [1] and in a brief literature search the author has found two other papers where precisely this point about the KA is noted [12,13].

It is not difficult to see just how the perturbation of the initial atomic bound state by the field arises in the S-matrix formalism. The transition amplitude in the time-reversed form employed by Reiss is

$$A_{fi}(t)=\delta_{fi}-\frac{1}{\hbar}\int_{-\infty}^{t}dt'\langle f|U^{\dagger}(t')H_I(t')\overline{U}_0(t')|i\rangle\ , \quad (1)$$

where H_I is the $\mathbf{A}\cdot\mathbf{p}+\mathbf{A}^2$ interaction, U is the exact time-evolution operator for the full interaction, and $\overline{U}_0$ is the evolution operator in the absence of any applied field. Equation (1) is equivalent to Reiss's equation (3.4). I employ the time-evolution operator in order to establish contact with earlier analyses [4,5] whose notation is followed here. Note that $\overline{U}_0(t')|i\rangle=|i\rangle e^{-iE_it'}$ is what Reiss calls a "reference state ... free of the transition-causing interaction $[H_I]$" whereas $U(t')|f\rangle$ is an exact, "fully interacting" state.

Consider a Dyson expansion of U for approximating the exact expression (1). If we replace $U(t)$ by the evolution operator $U_0(t)$ corresponding to the full Hamiltonian minus the binding potential, then (1) reduces exactly to the KA [4,5]. However, the next order of iteration, using the binding potential as the perturbation, is not independent of U_0:

$$U(t)-U_0(t)=-\frac{i}{\hbar}\int_{-\infty}^{t}dt'U_0^{\dagger}(t')VU_0(t')+\cdots\ . \quad (2)$$

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Thus *corrections to the KA involve field-dependent terms.* If we keep the Volkov state $U_0(t')|f\rangle$ as our "final state," then the corrections (2) to the KA act as further perturbations [in addition to the interaction H_I appearing in (1)] *on the bound state.* Thus in the KA it is not true that "the field interaction has been retained everywhere that it appeared in the exact S matrix."

Incidentally, the $\mathbf{A}^2$ contributions from these corrections can be summed exactly in the NRD, and the result is the exact cancellation of the $\mathbf{A}^2$ shift in the Volkov state. Reiss argues, incorrectly I believe, on the basis of photoionization "boundary conditions," that $\mathbf{A}^2$ can appear correctly only in a fully interacting state, not a reference state, and that therefore previous NRD arguments that $\mathbf{A}^2$ contributes the same shift to initial and final states, and can be removed by a simple contact transformation, are "not permissible". This is wrong. The exact transition matrix element $\langle f|U^\dagger(t)|i\rangle$, to which (1) is equivalent, of course involves a reference state $|i\rangle$ and a fully interacting state $U(t)|f\rangle$. And the reference state of course is unaffected by the field. But $\langle f|U^\dagger(t)|i\rangle$ is just the amplitude at time t for the *evolving* interacting state to be found in the reference state of interest. The evolution operator of the interacting state is governed by the Schrödinger equation, and the contact transformation can be performed on this equation without any reference whatsoever to the reference state. Put another way, the effect of $\mathbf{A}^2$ on the fully interacting state can be pulled outside the exact transition matrix element in the NRD and, since it represents only a phase, has no physical consequences. With regard to the "boundary conditions," note that the fact that there may be no field present when a measurement is made [6] is irrelevant to this argument: since the Schrödinger equation involves a derivative, it is the *integral over time* of $\mathbf{A}^2$ that matters, not $\mathbf{A}^2$ itself.

Reiss acknowledges that the $\mathbf{A}^2$ term does cancel in the problem considered by Kroll and Watson [14] and states that this is acceptable in that problem because of the particular initial and final states involved. This is inconsistent with his statement earlier in the paper that the contact transformation (employed by Kroll and Watson and many others) is "not permissible" because the S matrix involves the overlap of an unperturbed reference state with a fully interacting state. Furthermore, his claims conflict with the well-established Kroll-Watson theory on at least two other counts. First, Kroll and Watson eliminated $\mathbf{A}^2$ in the NRD immediately, before ever having to specify initial and final states. Second, they used nonrelativistic Volkov states, which Reiss claims exist unambiguously only in relativistic theory. I wish now to focus attention on the latter claim.

Reiss states that he has "found first that the Volkov solution, upon which the KA depends, exists unambiguously only in the relativistic case." This "finding" is actually only an assertion in his paper: ". . . a Volkov solution must describe a free particle. It must therefore be applicable over many wavelengths. It is not correct to restrict the motion to a small fraction of a wavelength in the original equation of motion." Suppose that the nonrelativistic approximation is valid, and consider the Heisenberg equation of motion for an electron in a plane-wave field:

$$m\ddot{\mathbf{r}}=e\mathbf{E}_0e^{-i(\omega t-\mathbf{k}\cdot\mathbf{r})} . \qquad (3)$$

Since $\mathbf{k}\cdot\mathbf{E}_0=0$, $\mathbf{k}\cdot\dot{\mathbf{r}}(t)$ is constant: the motion in the direction of field propagation, the direction in which there are spatial variations on the scale of a wavelength, is unaffected by the field in the nonrelativistic approximation. Once the nonrelativistic approximation is made, there is no need to assume a dipole approximation. Contrary to Reiss's assertion, Eq. (3) is perfectly valid *over many wavelengths.* This fully justifies the use of a nonrelativistic Volkov solution for an unbound electron.

Regarding the numerical results presented by Reiss, it is not surprising that a model (in this case Reiss's SFA) in which $\mathbf{A}^2$ appears, correctly or not, will yield very different results depending on whether or not $\mathbf{A}^2$ is included.

[1] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys—JETP **20**, 1307 (1965)].

[2] H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).

[3] H. S. Antunes Neto and L. Davidovich, Phys. Rev. Lett. **53**, 2238 (1984).

[4] P. W. Milonni, Phys. Rev. A **38**, 2682 (1988).

[5] P. W. Milonni and J. R. Ackerhalt, Phys. Rev. A **39**, 1139 (1989).

[6] H. R. Reiss, Phys. Rev. A **42**, 1476 (1990).

[7] J. Parker and C. R. Stroud Jr., Phys. Rev. A **40**, 5651 (1989).

[8] P. W. Milonni, R. J. Cook, and J. R. Ackerhalt, Phys. Rev. A **40**, 3764 (1989).

[9] For level shifts the $\mathbf{A}^2$ term is inconsequential in the NRD even when the field is quantized. See, for instance, J. J. Sakurai, *Advanced Quantum Mechanics* (Addison-Wesley, Reading, MA, 1967), Problem 2-11. For similar reasons $\mathbf{A}^2$ does not contribute to the Raman effect because a change of state is involved, whereas it does contribute to the Kramers-Heisenberg formula because there no change of state is assumed.

[10] T. W. B. Kibble, A. Salam, and J. Strathdee, Nucl. Phys. B **96**, 255 (1955).

[11] L. W. Pan, L. Armstrong, Jr., and J. H. Eberly, J. Opt. Soc. Am. B **3**, 1319 (1986). See also Ref. [5] for a generalization pertaining to the Stark shift.

[12] W. C. Henneberger, Phys. Rev. Lett. **21**, 838 (1968).

[13] R.F. O'Connell, Phys. Rev. A **12**, 1132 (1975).

[14] N. M. Kroll and K. M. Watson, Phys. Rev. A **8**, 804 (1973).