Reply to "Comment on 'Complete Keldysh theory and its limiting cases'"

H. R. Reiss

Department of Physics, The American University, Washington, D.C. 20016-8058 (Received 7 February 1991; revised manuscript received 9 October 1991)

Milonni [preceding Comment, Phys. Rev. A 45, 2138 (1992)] raises objections to my work [Phys. Rev. A 42, 1476 (1990)] on Keldysh-like methods in photoionization. His results are shown to follow from procedures that are inappropriate to the strong-field environment of the Keldysh method. These procedures lead to physical inconsistencies. My technique is verified to be rigorous and unique.

PACS number(s): 32.80.Rm, 33.80.Rv, 03.65.Ca

The basic objection raised by Milonni [1] relates to the role of A^2 in Keldysh-like theories applied in the nonrelativistic, dipole approximation. This matter can be resolved in fundamental and unambiguous terms. The key element is that Keldysh theory is formulated with the time-reversed S matrix [2,3] rather than the more familiar direct-time S matrix. The exact, direct-time S matrix [4] or transition amplitude is

$$(S-1)_{fi} = -i \int dt (\Phi_f, H_I \Psi_i) , \qquad (1)$$

where Φ is a noninteracting "reference" state satisfying the Schrödinger equation $(i\partial_t - H_0)\Phi = 0$, Ψ is a fully interacting state which satisfies the Schrödinger equation $(i\partial_t - H_0 - H_I)\Psi = 0$, and H_I represents the effect of the laser field. (Units with $\hbar = c = 1$ are used). For strongfield photoionization, the heart of the problem is to find a way to treat Ψ_i , which is a bound state in a strong electromagnetic field. The field will importantly distort the properties of a simple Φ_i bound state. This problem is profoundly difficult. The "secret" of Keldysh-like methods is to circumvent the difficulty by using instead the time-reversed S matrix:

$$(S-1)_{fi} = -i \int dt (\Psi_f, H_I \Phi_i) . \tag{2}$$

Like Eq. (1), this is an exact expression if the Φ , Ψ states are known exactly. However, in Eq. (2), the initial bound state is required only in its simple Φ_i form, with no field present. This can be treated accurately. The completely interacting state Ψ_f relates to the electron after ionization. For a sufficiently strong field, this will be dominated by the laser field. In that case, the Volkov solution is a good approximation. An analogy is the elementary textbook example of single-photon ionization from the atomic ground state, where, for sufficiently high energy of the photon, one can neglect the Coulomb influence on the energetic outgoing electron. For very strong laser fields satisfying the Keldysh-method validity conditions, most outgoing electrons will be very energetic, in some cases reaching kilo-electron-volts of energy.

The core conclusions of Milonni are in conflict with Eq. (2). He faults the Keldysh method because it "ignores completely the effect of the field on the initial bound state. In particular, it ignores any field-induced shifts of bound levels. . . " [1]. These criticisms would be appropriate were the Keldysh method based on Eq. (1). It is not. It is based on Eq. (2). The strength of the method flows from the fact that it is not necessary (and not correct) to include effects of the field on the initial bound state.

The technique employed by Milonni [5] and others [6,7] is removal of the A^2 term by a contact transformation applied to a factor $\exp(i\alpha \mathbf{A}^2 t)$ in the state Ψ_f . In the dipole approximation, A^2 is a function only of t. From the Fermi golden rule, a time-dependent exponential in a matrix element vanishes in the absolute square called for in the golden rule, and so has no physical consequences. That is a weak-field procedure applied to the strong-field Keldysh problem. Its defect can be seen from Eq. (2). Only Ψ_f in Eq. (2) contains $\exp(i\alpha \mathbf{A}^2 t)$; Φ_i does not. The factor $\exp(i\alpha \mathbf{A}^2 t)$ represents an energy, and the time integral in Eq. (2) gives an energyconserving δ function. The removal of $\exp(i\alpha \mathbf{A}^2 t)$ represents a shift of energy which is unphysical, and can be extremely large. This term can be 10⁵ eV in some experiments. An alternative view is to observe that A^2 removal alters the zero of energy to which Ψ_f refers, while not doing so for Φ_i . The removal of A^2 does have physical consequences. It cannot be removed from Eq. (2).

Another approach to the matter is to rederive the Fermi golden rule from first principles for the strong-field problem. This is done in Appendix B of Ref. [2]. The strong-field golden rule manifests directly the consequences of the A^2 term in the energy-conservation condition.

Equation (2) reveals the nature of the Keldysh approximation. Atomic information occurs primarily in the initial state Φ_i . Equation (2) contains field-dependent quantities only in H_I and in Ψ_f , where they are retained as completely as possible by using the Volkov solution. Residual Coulomb effects in Ψ_f of Eq. (2) become less important as the field strength increases, so the Keldysh approximation approaches exactness as field strength increases. The Keldysh method is thus a strong-field method. Of course, as explicit correction terms show [2,3], corrections will depend on the field. Milonni concludes [5] that the Keldysh method is a weak-field method based on inappropriate manipulations of the A^2

Milonni dismisses the fact that my relativistic deriva-

tion [2] exactly sustains the nonrelativistic result attained earlier [3]. The smooth connection of the relativistic and nonrelativistic Keldysh theories is fatal for the Milonni position. There is no possibility of removing relativistic A^2 terms, where A depends on the coordinate \mathbf{r} . The A^2 terms can contribute kilo-electron-volts and more of energy, as well as the well-known "double-frequency" terms that are fundamental in the strong-field free-electron [8] and plasma physics [9] problems. In the nonrelativistic limit, these phenomena remain enormously important. Physical continuity demands that these terms cannot abruptly disappear. Milonni's A^2 manipulations have these huge terms vanish when nonrelativistic procedures become permissible.

With respect to the relativistic nature of the Volkov solution, one can use the nonrelativistic limit of it in the free-electron problem if electron transport in the direction of propagation in a wave period is small compared to a wavelength. For the bound-state problem, the relevant quantity is the field-induced transport of the electron as compared to the atomic radius. In strong-field environments, the component in the propagation direction of the "figure-8" motion [10] of the electron in the laser field can exceed the Bohr radius in conditions otherwise non-relativistic. This can be examined only with relativistic forms. Milonni explores the relativistic case with his Eq. (3), which discards the magnetic force, so it is not relativistic. Had he made it relativistic, he would have rediscovered the figure-8 solution.

Milonni makes reference to papers that actually support my position. He mentions the work of Kroll and Watson [11], who evaluate the problem describable by the S matrix [12]

$$(S-1)_{fi} = -i \int dt (\Psi_f^F, V \Psi_i^F) , \qquad (3)$$

where the two states Ψ^F are Volkov solutions and V is the atomic potential. In contrast to Eqs. (1) or (2), a contact transformation on the states in Eq. (3) is balanced, both energy references are treated the same, and no problem arises. The Kroll and Watson procedure is entirely consistent with mine.

Another anomalous reference by Milonni is to a 25-

year-old speculation [13] that the mass shift of the freeelectron problem might be manifested in a bound system, and would be amplified by the use of very low (microwave) frequencies, a strategy now widely employed. In analyzing why experiments did not reveal the mass shift, Kibble et al. [14] emphasized the interchangeability (above first order) of $\mathbf{A} \cdot \mathbf{p}$ and \mathbf{A}^2 terms. This is one of the basic points made in my paper [2], and precludes the Milonni treatment of \mathbf{A}^2 .

In addition to the above considerations, the following list summarizes some established physical properties, each of which is incompatible with the conclusions of Milonni [1,5]:

- (a) The strong-field, low-frequency limit of Keldysh-like methods gives tunneling. Tunneling transition rate behavior is $\exp(-\alpha/F)$, where F is electric-field strength. This cannot arise in a weak-field theory, as Milonni concludes the Keldysh theory to be.
- (b) The SFA of Ref. [2] predicts "stabilization" [3,15], characterized by an eventual decline in transition rate as the intensity increases. This is incompatible with weak-field behavior.
- (c) Keldysh-like techniques are the methods of choice in fitting multiphoton, multiple-ionization experiments. These experiments can have electron pondermotive energy in the laser field of 10³ and 10⁴ times the atomic binding energy. These are not weak-field experiments.
- (d) In particular, the SFA, when employed with appropriate atomic wave functions, gives excellent agreement [16] with no adjustable parameters with very-strong-field multiple-ionization experiments.

To summarize my method [2,3], Eq. (2) is exact, and the starting point. The leading approximation is to replace Ψ_f by the Volkov solution Ψ_f^{Volkov} . This is the Keldysh approximation. Each element in it is well defined and unique. If one wishes formal correction terms, they are available [2,3] as an expansion in powers of the binding potential. This is also unique (as are all power series), and well behaved since the Volkov propagators in the expansion are well behaved [17]. The relativistic derivation of this method in Ref. [2] is rigorous. Milonni's challenge to it is based on qualitative considerations which are shown above not to be correct.

^[1] P. W. Milonni, preceding paper, Phys. Rev. A 45, 2138 (1992).

^[2] H. R. Reiss, Phys. Rev. A 42, 1476 (1990).

^[3] H. R. Reiss, Phys. Rev. A 22, 1786 (1980); H. R. Reiss, in Atoms in Strong Fields, edited by C. A. Nicolaides, C. W. Clark, and M. H. Nayfeh (Plenum, New York, 1990).

^[4] J. D. Bjorken and S. D. Drell, Relativistic Quantum Mechanics (McGraw-Hill, New York, 1964).

^[5] P. W. Milonni, Phys. Rev. A 38, 2682 (1988); P. W. Milonni and J. R. Ackerhalt, *ibid*. 39, 1139 (1989).

^[6] H. S. Antunes Neto and L. Davidovich, Phys. Rev. Lett. 53, 2238 (1984).

^[7] M. H. Mittleman, Phys. Rev. A 40, 463 (1989).

^[8] For example, see J. H. Eberly, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1967), Vol. 7, p. 361.

^[9] See P. Sprangle, E. Esarey, and A. Ting, Phys. Rev. A 41, 4463 (1990).

^[10] E. S. Sarachik and G. T. Schappert, Phys. Rev. D 1, 2738 (1970).

^[11] N. M. Kroll and K. M. Watson, Phys. Rev. A 8, 804 (1973).

^[12] N. K. Rahman, Phys. Rev. A 10, 440 (1974).

^[13] H. R. Reiss, Phys. Rev. Lett. 17, 1162 (1966).

^[14] T. W. B. Kibble, A. Salam, and J. Strathdee, Nucl. Phys. B 96, 255 (1975).

^[15] H. R. Reiss, Bull. Am. Phys. Soc. 36, 1249 (1991); and unpublished.

^[16] R. S. Bardfield and H. R. Reiss, Bull. Am. Phys. Soc. 36, 1249 (1991), and unpublished.

^[17] H. R. Reiss and J. H. Eberly, Phys. Rev. 151, 1058 (1966).