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Comment on "Intensity-dependent ionization potentials for H and He in intense laser beams"*†

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It is argued that a straightforward physical interpretation of the intensity-dependent ionization potentials computed by Choi, Henneberger, and Sanders is not possible.

In a recent article in this journal, Choi, Henneberger, and Sanders[1] have reported ground-state energies for one and two electron systems in the presence of a potential which is purported to account for a portion of the interaction between a linearly polarized laser field and the given atomic system. The potential was derived according to a method proposed by Henneberger,[2] a method perhaps best described as a strong-coupling method. The authors have interpreted the difference in energy between the ionization limit and the calculated ground-state energy as an intensity-dependent ionization potential for the system. In the examples studied these ionization energies are less than those in the absence of the laser field, tending to zero as the field strength becomes enormous. Similar though quantitatively different results have been found for elliptically polarized light.[3] Though the authors caution that no direct comparison to experiment is possible until the newly computed ground-state wave functions are used to compute the transition probabilities to the continuum, the hope is plainly entertained (and stated more explicitly elsewhere[4]) that dividing the intensity-dependent ionization energy by the photon energy should yield an "effective number" of photons needed to ionize the atom.

It is the purpose of this Comment to show that any straightforward interpretation of either the intensity-dependent ionization potentials, or the transition probabilities computed according to the Henneberger scheme,[2] is at best misleading. The arguments presented are arguments of principle based on the simple model of a single atom interacting with a given classical external radiation field (as assumed in Ref. 2). These observations do not preclude the possibility that sensible physical approximations involving portions of the actual physical system not included in the model (as, e.g., the relaxation times of a gas) might lead one to conclude that the calculated quantities do in fact correspond to experimental reality. Because we have not been able to think of any such argument, we believe that it is prudent to view the results of Choi, Henneberger, and Sanders[1] with due caution until reasoning to the contrary is presented.

Our argument is as follows. With Henneberger,[2] we consider for simplicity a one-electron atom with infinitely heavy nucleus bound by a potential V_0 and interacting with an external classical radiation field specified by a vector potential $\vec{A}(t)$. In the Schrödinger picture the Hamiltonian H of the problem is given by $H = H_0 + V_S(t)$ where H_0 and V_S are given by ($\hbar = 1$)

$$H_0 = (2m)^{-1}\vec{p}^2 + V_0, \tag{1}$$

$$V_S(t) = -e(mc)^{-1}\vec{A}(t)\cdot\vec{p} + e^2(2mc^2)^{-1}\vec{A}^2(t). \tag{2}$$

We proceed to the interaction picture in the usual fashion and define the conventional time-evolution operator $U_I(t_1, t_2)$ by the rule

$$\psi_I(t_1) = U_I(t_1, t_2)\psi_I(t_2) \tag{3}$$

where ψ_I is the interaction-picture wave function. If the interaction $V_S(t)$ is turned on at $t = t_i$, then the probability that a transition has occurred from an initial Heisenberg state $|i\rangle$ ($H_0|i\rangle = E_i|i\rangle$) to a final Heisenberg state $|f\rangle$ is given by

$$P_{i\to f}(t, t_i) = |\langle f|U_I(t, t_i)|i\rangle|^2. \tag{4}$$

We want to emphasize that it is the quantity indicated in Eq. (4) that must be calculated according to conventional quantum-mechanical interpretation. Our whole point is that the Henneberger method does not calculate the matrix elements of U_I. This will become apparent below.

The Henneberger method consists of making a time-dependent unitary transformation $\Omega_H(t, t_0)$ to a new picture (which we denote as the Henneberger picture) according to the rule

$$\psi_S(t) = \Omega_H(t, t_0)\psi_H(t). \tag{5}$$

(Here and throughout this paper ψ_H is the wave function in the Henneberger picture, not the Heisenberg picture. Heisenberg states of H_0 are unlabeled.) The particular transformation chosen by Henneberger is given by

$$\Omega_H(t,t_0)=\exp[(ie/mc)\vec{\alpha}\cdot\vec{p}]\exp[-(ie^2/2mc^2)\gamma], \tag{6}$$

where

$$\vec{\alpha}=\int_{t_0}^{t}\vec{A}(t')\,dt' \tag{7}$$

$$\gamma=\int_{t_0}^{t}\vec{A}^2(t')\,dt'. \tag{8}$$

This particular transformation has the property that in the dipole approximation for $\vec{A}(t)$ (which we henceforth assume), ψ_H represents a free electron when $V_0=0$. Thus, if V_S may be considered large compared to V_0, we may effectively generate some sort of strong-coupling expansion.

In the Henneberger picture, the evolution of the system is governed by a new Hamiltonian H_H given by

$$H_H=\Omega_H^{-1}H\Omega_H+i\dot{\Omega}_H^{-1}\Omega_H. \tag{9}$$

For the Hamiltonian given by Eqs. (1) and (2), and the transformation (6), H_H is explicitly given by

$$H_H=(2m)^{-1}\vec{p}^{\,2}+V_0(\vec{r}+\vec{\alpha}). \tag{10}$$

We write this as $H_H=H_H^0+V_H(t)$, where H_H^0 is the time-independent portion of H_H. Henneberger suggests that we use H_H to compute transition probabilities because H_H must represent the same physics as H. In particular, the scheme consists in finding eigenstates of H_H^0 and using $V_H(t)$ as the perturbation. Defining a Henneberger interaction picture in the obvious fashion, this leads to the calculation of the time-evolution operator U_{HI} in the Henneberger interaction picture. The following relation between U_I and U_{HI} is easily established:

$$U_I(t_1,t_2)=\Gamma(t_1,t_0)U_{HI}(t_1,t_2)\Gamma^{-1}(t_2,t_0). \tag{11}$$

In Eq. (11), Γ is given by

$$\Gamma(t_1,t_0)=\exp(iH_0t_1)\Omega_H(t_1,t_0)\exp(-iH_H^0t_1). \tag{12}$$

Equation (11) is the key to our discussion, for it shows that U_I and U_{HI} are not the same operators. We explore the ramifications of this identity below.

Let us begin with an interaction potential $\vec{A}(t)$ which vanishes for $t<t_i$ [or if we wish to let $t_i\to-\infty$, $\lim_{t_i\to-\infty}A(t_i)=0$]. This corresponds to the actual experimental situation. In this event, t_0 in Eqs. (6)–(8) is taken to be t_i, so that $\Omega_H(t,t_i)=\underline{1}$ for $t<t_i$. This is sensible because Ω_H is intended to account for the interaction with $\vec{A}(t)$ and there is no interaction for $t<t_i$. It follows that $H_H^0=H_0$. This is true because for $t<t_i$, $H_H=H=H_0$, so that the only part of the Hamiltonian which is constant throughout all time is H_0. (If there were some other constant Hamiltonian H_0', then it would be constant for $t<t_i$ as well, because by definition it is constant.) Thus, observers in both the Schrödinger picture and the Henneberger picture would agree upon the states which are to be used for calculating transition probabilities, the Heisenberg states of H_0. But by Eq. (11), U_{HI} and U_I are not the same, so that these two observers would disagree on the transition probability.

To make this clear we consider the concrete example of a one-dimensional harmonic oscillator, with the polarization of $\vec{A}$ in the direction of the oscillator. For this case we have $V_0=\frac{1}{2}m\Omega x^2$, Ω being the natural oscillator frequency. We allow a general wave packet $\vec{A}=\hat{e}_x f(t)$, but require $f(t)\to 0$ as $t\to-\infty$. The transition probability from the ground state $|0\rangle$ at $t=-\infty$ to an excited state $|n\rangle$ is easily calculated to be[5]

$$P_{0\to n}(t)=(\nu^n/n!)e^{-\nu}, \tag{13}$$

where

$$\nu=\frac{e^2\Omega}{2mc^2}\left|\int_{-\infty}^{t}dt'\,f(t')e^{-i\Omega t'}\right|^2. \tag{14}$$

The transition probability in the Henneberger picture $P_{i\to f}^{H}$ has the same form as in Eq. (13) with the substitution $\nu\to\nu_H$, where

$$\nu_H=\frac{e^2\Omega^3}{2mc^2}\left|\int_{-\infty}^{t}dt'\,\alpha(t')e^{-i\Omega t'}\right|^2 \tag{15}$$

and $\alpha(t)$ is given by Eq. (7). Inspection of Eqs. (14) and (15) shows that ν and ν_H are not the same. We are forced to conclude that the matrix elements of U_{HI} are not the transition amplitudes we are seeking. Instead, they are related to the transition amplitudes by taking matrix elements of Eq. (11), a fact which may be explicitly verified for the case of the harmonic oscillator.

What are the matrix elements of U_{HI}? They are simply the transition amplitudes in the Henneberger picture. The use of U_{HI} for calculating transition probabilities requires the validity of making observations in the Henneberger picture. This is our primary conclusion. However, because Henneberger appears to have shown in Ref. 2 that perturbation theory in the new picture (in powers of e) yields the same results as conventional perturbation theory, we wish to make several more points.

The reader might expect that since the Henneberger and Schrödinger pictures agree for $t<t_i$, that they should agree again when the interaction is turned off at $t=t_f$. This would require $\Gamma(t_f,t_0)=\underline{1}$ and establish the identity of $U_I(t_f,t_i)$ and $U_{HI}(t_f,t_i)$. Unfortunately Ω_H depends on the time integral of the interaction $\vec{\alpha}(t)$. Only in the most fortuitous of circumstances could one expect $\vec{\alpha}(t_f)=0$. Thus,

U_I and U_{HI} are not equal even after the interaction is turned off. In our concrete example we may see this by performing an integration by parts on Eq. (15) to yield

$$\nu_H = \frac{e^2\Omega}{2mc^2}\left|\left[\int_{-\infty}^{t} dt' f(t')e^{-i\Omega t'}\right] - \alpha(t)e^{-i\Omega t}\right|^2 . \tag{16}$$

If the second term inside the absolute value symbol were absent, then ν_H would yield the correct transition amplitude. For $t > t_f$, this term becomes $\alpha(t_f)e^{-i\Omega t}$ which does not take on a definite limit as $t \to +\infty$. That is, ν_H becomes singular in this limit. Notice that this does not happen to ν, which is perfectly well behaved for a smooth wave packet which tends to zero as $t \to +\infty$. Of course, if we arbitrarily set these unwanted singular terms to zero, we reproduce the correct answer.

The appearance of singular terms as $t \to +\infty$ (even with well-behaved wave packets) is a general feature of the interaction in the Henneberger picture. This is easily seen from Eq. (10), because $\alpha(t)$, which specifies the interaction, does not tend to zero as $t \to +\infty$. These terms manifest themselves in a powers-of-e perturbation theory in the following fashion. Noting that Ω_H depends upon e, we may expand both sides of Eq. (11) in powers of e and establish the relation between the powers-of-e expansions of U_I and U_{HI}. Denoting the power of e for the relevant quantity by a superscript (n), we find the first term yields $U_I^{(1)} = U_{HI}^{(1)} + \Gamma^{(1)}$. The second term gives $U_I^{(2)} = U_{HI}^{(2)} + \Gamma^{(1)}U_{HI}^{(1)} + \Gamma^{(2)}$, while the general term gives $U_I^{(n)} = U_{HI}^{(n)} +$ (terms involving $\Gamma^{(1)}, \ldots, \Gamma^{(n)}$). For finite times and well-behaved wave packets, these extra terms involving $\Gamma^{(1)}, \ldots, \Gamma^{(n)}$ are invariably present and nonzero. As $t \to +\infty$, both $U_{HI}^{(n)}$ and these extra terms become singular, though their sum (which is $U_I^{(n)}$) is well behaved. Reversing the emphasis, the above relation tells us that $U_{HI}^{(n)} = U_I^{(n)} +$ (extra terms). This means that we can always algebraically rearrange $U_{HI}^{(n)}$ to look like $U_I^{(n)}$ plus extra terms. If these extra terms are ignored because they become singular at $t \to +\infty$, we have then "proved" the equivalence of the two perturbation series. The above remarks may be explicitly verified for our concrete example.

Of course, Henneberger is not suggesting that we do a powers-of-e expansion of U_{HI}, but rather that V_H be used as the perturbation, the hope being that this series converges more rapidly than the V_S perturbation series. However, to yield easily interpretable answers, Eq. (11) must be used to bring these approximations for U_{HI} back to the Schrödinger picture, thus giving an alternate expansion for U_I. A particular example of a circumstance in which such a strong-coupling expansion is very sensible is noted below. Our point is simply that the matrix elements of U_{HI} are not to be interpreted in any obvious fashion as the physical transition amplitudes. This lack of direct experimental interpretation also holds for the energy shifts in the Henneberger picture. This is made clear below in the context of an example.

We saw above that for a packeted radiation field, no energy shifts were introduced into the discussion. It is natural however, to consider the limit where the packet becomes purely sinusoidal. This causes no particular difficulty so long as we remain in finite time, but does raise problems in infinite-time. (This can be seen in our harmonic oscillator example, where $P_{0\to n}$ becomes badly defined in this limit). Physically, this means that it is necessary to work in finite times, and then prescribe some method (e.g., relaxation-time averaging) for obtaining long-time results. In this circumstance, it is natural to define the "constant" part H_H^0 of the Hamiltonian H_H as the time average of the Hamiltonian. This need not be the same as H_0, as the interaction term need not have vanishing time average. This was the procedure used by Henneberger[2] to derive the potential used by Choi, Henneberger, and Sanders.[1] That the energy levels of H_H^0 are not directly interpretable may be seen from the exactly soluble example of a two-level atom interacting with a circularly polarized radiation field.[6] (The harmonic oscillator example is not particularly illuminating because all the levels are shifted by the same amount.)

The Hamiltonian for this problem is $H = H_0 + V$, where

$$H_0 = \tfrac{1}{2}\lambda\sigma_3, \quad V = \tfrac{1}{2}\beta(\sigma_1 \cos\omega t + \sigma_2 \sin\omega t), \tag{17}$$

and the σ_i's are the standard Pauli matrices. The exact solution to this problem shows a single resonance at $\omega = \lambda$. To carry out the Henneberger program, we must define the analog of Eq. (6) for the present system. The analog of an unbound particle is to take $\lambda = 0$. If $\lambda = 0$, one easily verifies that the operator Ω_H given by

$$\Omega_H(t) = \exp(-\tfrac{1}{2}i\omega t\sigma_3)\exp[-\tfrac{1}{2}it(\beta\sigma_1 - \omega\sigma_3)] \tag{18}$$

carries the Schrödinger wave function to one which is constant in time, i.e., a free spinor, and hence qualifies as a Henneberger transformation. Carrying out the steps indicated in Eq. (9), and extracting the time-averaged portion of H_H, we find that H_H^0 and V_H are given by

$$H_H^0 = \frac{\epsilon}{2}\left(\frac{\omega}{\theta}\sigma_3 - \frac{\beta}{\theta}\sigma_1\right), \tag{19}$$

$$V_H = \frac{\lambda}{2}\left[\left(\frac{\beta^2}{\theta^2}\cos\theta t\right)\sigma_3 + \left(\frac{\omega\beta}{\theta^2}\cos\theta t\right)\sigma_1 + \left(\frac{\beta}{\theta}\sin\theta t\right)\sigma_2\right], \tag{20}$$

where

$$\epsilon = \lambda\omega/\theta, \tag{21}$$

$$\theta = +(\omega^2 + \beta^2)^{1/2}. \tag{22}$$

The eigenvalues of H_H^0 are given by $E_\pm = \pm\frac{1}{2}\epsilon$ being the "shifted energies" for this system. The driving frequency has also been shifted from ω to θ, and we might expect the system to have a resonance when $\epsilon = \theta$, or $\omega_\pm = \frac{1}{2}\lambda\{1 \pm [1 - 4(\beta/\lambda)^2]^{1/2}\}$. We would thus speak of a "shift" in the resonance frequency of the system which depends on the intensity β. Unfortunately, we know that the exact solution is resonant at $\omega = \lambda$ for all intensities, so that any such interpretation is incorrect.

We put this argument on a firmer footing by computing the first order (in V_H) transition matrix. $U_{HI}^{(1)}$ is easily found to be

$$U_{HI}^{(1)}(t,0) = \frac{\lambda\beta}{2\theta(\epsilon-\theta)}\left[\left(\frac{\omega}{\theta}\sigma_1 + \frac{\beta}{\theta}\sigma_3\right)\sin(\epsilon-\theta)t + \sigma_2[\cos(\epsilon-\theta)t - 1]\right]. \tag{23}$$

In this expression we see the resonance denominator $\epsilon - \theta$ as expected. Following Henneberger, we take the matrix element of $U_{HI}^{(1)}$ between the "shifted" eigenstates $u_\pm(H_H^0 u_\pm = E_\pm u_\pm)$, square, and take a time average to yield the time-averaged transition probability $\bar{P}^H_{-\to+}$ in the Henneberger picture. One finds that $\bar{P}^H_{-\to+}$ is given by

$$\bar{P}^H_{-\to+} = \lambda^2\beta^2/2\theta^2(\epsilon-\theta)^2. \tag{24}$$

It is easy to show that this expression has real poles at $\omega_\pm$ if $(\beta/\lambda)^2 < \frac{1}{4}$, while it maximizes at $\omega = \frac{1}{2}\lambda$ (the real part of $\omega_\pm$) for $(\beta/\lambda)^2 > \frac{1}{4}$. We know of no interpretation which can be attached to this behavior. It can also be shown that taking the matrix element of $U_{HI}^{(1)}$ between the unshifted states, or between an unshifted initial state, and a shifted final state leads to equally uninterpretable transition probabilities.

By contrast, if Eq. (11) is used to return U_{HI} to the Schrödinger picture, one obtains reasonable approximations to the exact Rabi transition probability. The zeroth-order contribution to U_I is obtained from Eq. (11) by taking the zeroth-order contribution to U_{HI} ($U_{HI}^{(0)} = \underline{1}$). Taking the matrix element between unshifted states, squaring, and time averaging, one finds

$$\bar{P}^{(0)}_{-\to+} = \beta^2/2\theta^2 \tag{25}$$

which is simply the Rabi solution evaluated at $\lambda = 0$, as expected. Keeping the first-order term in U_{HI} given by Eq. (23), yields, by the same procedure

$$\bar{P}^{(1)}_{-\to+} = \frac{\beta^2}{2\theta^2}\left(1 - \frac{\lambda\omega}{\theta(\epsilon-\theta)}\right)^2. \tag{26}$$

Although this expression runs into trouble for large λ/β, for small λ/β it provides an excellent strong coupling approximation to the exact Rabi solution. For example, at $\lambda/\beta = 0.1$, Eq. (26) fits the exact solution within 1% over a large range of ω. It also peaks within 1% of the $\omega = \lambda$ resonance. Further terms in U_{HI} may be kept. The resulting terms in U_I give a reasonable strong-coupling expansion ($\lambda/\beta \ll 1$) for the Rabi solution.

It should be mentioned that a similar analysis holds for a two-level system in a plane-polarized field.[7] In this instance detailed calculations have been performed,[8] and compared to experiment in connection with the Hanle effect.[9] The results are in good agreement with experiment, showing that this is a reasonable method of obtaining a strong-coupling solution, so long as one returns to the Schrödinger picture for comparing with experiment.

We must conclude that a straightforward interpretation of level shifts in the Henneberger picture is not possible. This being the case, the utmost caution must be exercised in ascribing any experimental significance to the intensity-dependent ionization potentials calculated by Choi, Henneberger, and Sanders.

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