Reply to “Comment on ‘Test of the Stark-effect theory using photoionization microscopy’ ”

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Reply to "Comment on 'Test of the Stark-effect theory using photoionization microscopy' "

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Abstract

The frame-transformation (FT) theory which describes the dynamics of nonhydrogenic atoms in an external uniform electric field was tested for the process of photoionization in a Stark field by Zhao *et al.* [Phys. Rev. A **86**, 053413 (2012)]. Differential cross sections from the FT theory were found to be inconsistent with those from the fully quantum-mechanical coupled-channel theory. The discrepancy was attributed to the frame transformation of irregular wave functions. In a recent investigation, Giannakeas *et al.* [Phys. Rev. A xx, xxxxxx (2015)] draw a different conclusion. They show that the FT theory generates irregular wave functions in good agreement with exact solutions for low angular momenta, although an obvious disagreement is seen for high angular momenta. We performed test calculations for numerous Stark states, and found that our original conclusion remains valid, namely the Fano-Harmin frame-transformation for the irregular wave function is inaccurate.

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The frame-transformation (FT) theory of the Stark effect developed by Fano [1] and Harmin [2] turned out to be a very efficient tool for investigation of photoabsorption spectra of atoms in the presence of a Stark field. It was not our intention to denounce this theory in our previous publication [3]. Rather, we tried to caution the AMO community about limitations and inaccuracies of this theory which were first noticed long before us by Stevens *et al.* [4]. In this Reply to the Comment [5], we will first point out the fundamental reasons for these inaccuracies. Then we will respond to the critique presented in the Comment.

The frame transformation for the irregular solution of the Schrödinger equation in the presence of the Stark field is based on the claim that the spherical and parabolic Green's functions are identical in the Coulomb region. However, because the Green's function is determined not only by the local behavior of the Hamiltonian, but also by the asymptotic behavior of the solutions at large distances, this claim is generally not correct. Consider, for example, one-dimensional motion of an electron with an energy E in a uniform electric field $\mathscr{F}$ corresponding to the potential $-\mathscr{F}x$. The Green's function corresponding to the outgoing-wave boundary conditions is (a.u. are used throughout the paper),

$$G_E(x_1, x_2) = \frac{2\pi}{(2\mathscr{F})^{1/3}} \mathrm{Ai}\left[-\xi(x_<)\right] \mathrm{Ci}\left[-\xi(x_>)\right] \tag{1}$$

where $x_< = \min(x_1, x_2)$, $x_> = \max(x_1, x_2)$, $\xi = (x + E/\mathscr{F})\,(2\mathscr{F})^{1/3}$, and $\mathrm{Ci} = \mathrm{Bi} + i\mathrm{Ai}$ with Ai and Bi being standard Airy functions [6].

In the limit $\mathscr{F} \to 0$ and $x_2 > x_1$, we obtain

$$G_E(x_1, x_2) = \frac{1}{ik}\left[\exp(ik(x_2 - x_1)) - \exp(2i\phi + ik(x_2 + x_1))\right], \tag{2}$$

where $k = (2E)^{1/2}$, $\phi = (2E)^{3/2}/(3\mathscr{F})$. Instead of one wave proportional to $\exp[ik|x_2 - x_1|]$ that we have for $\mathscr{F} = 0$, we obtain *two* waves propagating from left to right. This happens because even for a weak field the wave propagating to the left will be eventually reflected. The same occurs in the three-dimensional case, but now the amplitude of the reflected wave, propagating parallel to the field, will be proportional to $\mathscr{F}/E$ in the region where $\mathscr{F}r \ll E$ [7]. Although, because of the three-dimensional spreading of the outgoing wave, this coefficient goes to 0 if $\mathscr{F} \to 0$, it is still important if E is small, or wherever $\mathscr{F}r > E$. Numerical calculations with the exact Green's function for a three-dimensional particle in a uniform field [8] confirm these observations. Similarly, the effect is important in the Coulomb-Stark problem if the energy is close to the top of the Coulomb-Stark barrier. More generally, the effect is important in all problems when rescattering effects should be included.

The inaccuracy of the irregular solution in the FT theory also appears due to its nonuniformity in the spherical angle θ. The derivation of the irregular solution for the FT theory is based on comparison of the Green's functions for $r < r'$ and $\eta < \eta'$, where η is one of the parabolic coordinates, $\eta = r(1 - \cos\theta)$. It is easy to see that these two conditions are not equivalent. There is a range of θ and θ', where $\eta > \eta'$, even if $r < r'$. This nonuniformity was noticed by Stevens *et al.* [4], who pointed out that the left side of the frame-transformed equation is phase shifted isotropically, while the right side is phase shifted on the negative z axis, but not on the positive z axis ($\eta = 0$). This deficiency 'heals' quickly off the z axis, but may be a limiting factor in precision calculations. We will illustrate below that such a nonuniformity leads to incorrect irregular solutions in the FT theory.

We performed test calculations for numerous Stark states and found the obvious nonuniformity. Here we demonstrate this effect through two FT calculations for $m = 0$ and $m = 1$ Stark states. The same Stark state with $E = -54.1078$ cm^{-1} and $\mathscr{F} = 3590$ V/cm, as reported in Ref. [3], is selected. We checked the FT irregular wave function for $m = 0$, where a remarkable difference between differential cross sections from the FT theory and coupled-channel theory is observed in Ref. [3]. Figure 1 shows the exact irregular Coulomb function compared to the FT functions for different values of the spherical angle θ with $m = \ell = 0$, where ℓ is the orbital angular momentum quantum number. The function should be independent of θ for the $\ell = 0$ case. However, it is seen from this figure that the FT function strongly depends on θ, and a relatively good agreement appears only at $\theta = 150°$.

Figure 2 shows the exact irregular Coulomb function compared to the FT functions for different values of the spherical angle θ with $m = \ell = 1$. It is obvious that the FT function is substantially different from the exact irregular Coulomb function for small θ. This discrepancy becomes small with increasing θ and the excellent agreement is seen at $\theta = 150°$. However, the discrepancy is noticeable even at $\theta = 170°$.

Our results in Figs. 1 and 2 reveal the limitation of the FT theory for the low orbital angular momenta. For higher angular momenta, such a limitation is also seen in Fig. 3 of Giannakeas *et al.* [5] where a remarkable difference between the exact and FT irregular functions is shown for the $\ell = 6$ case. Although this significant inaccuracy should not influence the photoelectron currents due to negligible quantum defects for $\ell = 6$, as stressed in Ref. [5], it provides a caution mark — a further check of the FT theory is essential. With

regard to the convergence in n_1, we agree with Giannakeas *et al.* [5] that in our previous calculation for $\theta = 150°$, we failed to include sufficient number of terms to guarantee the convergence for the irregular FT wavefunction. Our new calculation for $\theta = 150°$ shows a good agreement between the exact and the FT wavefunctions similar to that shown in Fig. 2 of Ref. [5]. Even so, it must be pointed out that for this Stark state, the difference between the exact and FT irregular wave functions is found to vary with the spherical angles in our calculation, as shown in Figs. 1 and 2.

There are extra issues arising in connection with this calculation. First, Giannakeas *et al.* claim that all n_1 corresponding to $\beta < 1$ should be included. According to our calculation, for $m = 1$ and chosen values of $\mathscr{F}$ and E, $\beta(n_1 = 24) = 0.95973$ and $\beta(n_1 = 25) = 1.0011$, so it is not clear to us why Giannakeas *et al.* included the $n_1 = 25$ state in their calculations. Second, as Giannakeas *et al.* point out, there is no guarantee that the expansion is convergent even if all n_1 corresponding to $\beta < 1$ are included. However, it is unclear how the additional summation with the inclusion of terms corresponding to $\beta > 1$ is possible. In this case the equation in the η coordinate contains the repulsive Coulomb term $(\beta - 1)/\eta$, and it is unclear if the FT procedure is possible at all in this case since there is no region in the Coulomb zone where the wavefunction is oscillating.

Giannakeas *et al.* compared the differential cross sections calculated using the FT theory and the time-dependent quantum-mechanical method for the Stark state with $E = -62$ cm^{-1} and $\mathscr{F} = 3590$ V/cm and obtained excellent agreement. Unfortunately, we did not find the details of the time-dependent calculation and how the non-Coulombic features of the atomic field were included. In the comment, the time-dependent calculation is refereed to Ref. [9] which in turn is refereed to Ref. [10] where the calculation is believed to be performed for *hydrogen* rather than multielectron atoms. For the Stark state with $E = -62$ cm^{-1} and $\mathscr{F} = 3590$ V/cm, we have checked FT irregular wave functions for low orbit angular momenta and $m = 0$ and 1, and found very similar disagreement as in Figs. 1 and 2. Why the poor transformation of the irregular wave functions produces excellent agreement of differential cross sections shown in Fig. 1 of Giannakeas *et al.* remains unexplained.

In addition to the case $E = -62$ cm^{-1}, we also checked FT irregular wave functions at $E = -77.1926$ cm^{-1}, -54.1078 cm^{-1}, and $E = -41$ cm^{-1} for $m = 0$ and 1, and found that the differences between the exact and FT irregular wave functions for all these energies are consistent with the differences of differential cross sections shown in Fig. 1 of Ref. [3].

For example, for the four energies with $m = 1$ and two energies $E = -41$ cm^{-1} and -62 cm^{-1} with $m = 0$, the differences between the exact and FT irregular wave functions are far smaller, but not negligible, than those at $E = -77.1926$ cm^{-1} and -54.1078 cm^{-1} with $m = 0$. This may explain the existing differences of differential cross sections shown in Fig. 1 of Ref. [3].

Once again, we recognize the success of the FT theory which was employed in many calculations in the past, but there are evidences suggesting this theory should be used with some cautions because of inaccuracies originated from the derivation of the FT equation for the irregular solution. In fact, a recent precise spectroscopic measurement of the Stark spectrum of neon was not reproduced by the FT theory [11]. A significant difference has been found there. This indicates the necessity of reexamination of the FT theory. Our work [3] is an effort in this direction. Finally, we welcome this kind of comments, because they stimulate discussions leading to a better understanding of the problem.

Acknowledgments

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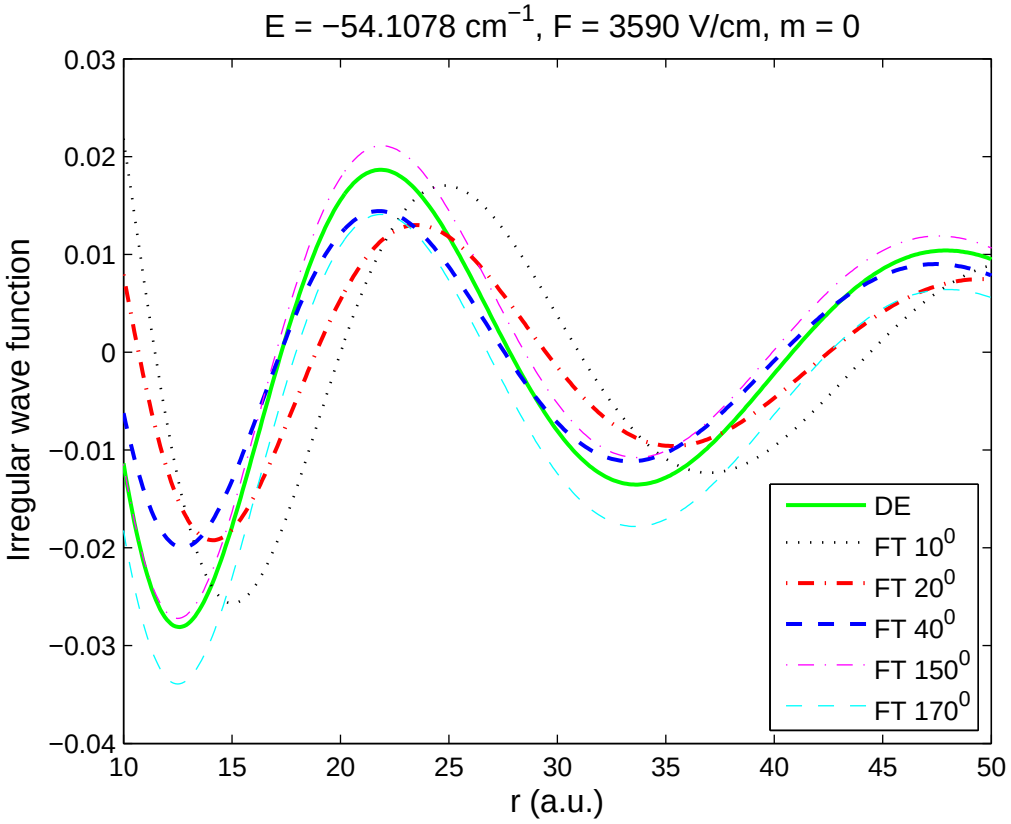

FIG. 1: (Color online). Comparison of the exact irregular Coulomb function for $m = \ell = 0$, and the FT wavefunction with different values of the spherical angle $\theta = 10°, 20°, 40°, 150°$, and $170°$. DE represents the exact irregular Coulomb function, while FT is the frame-transformed wavefunctions.

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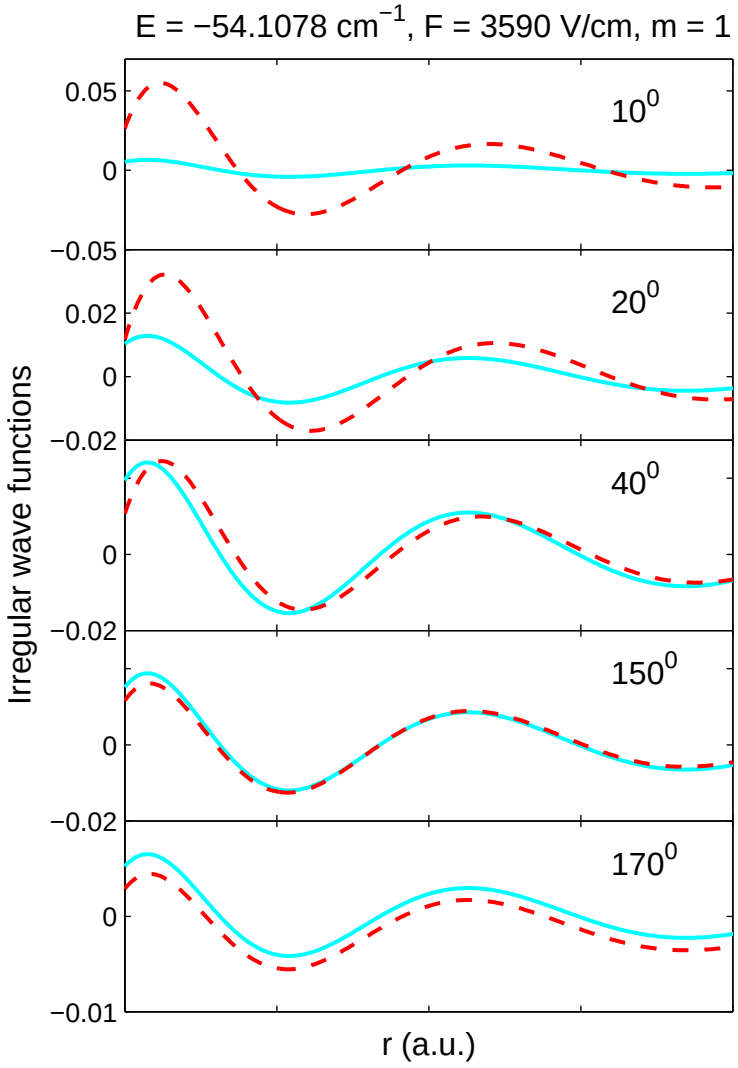

FIG. 2: (Color online). Comparison of the exact and FT irregular Coulomb function for $m = \ell = 1$ with different values of the spherical angle $\theta = 10°, 20°, 40°, 150°$, and $170°$, and $\phi = 0$. The cyan solid curves represent the exact irregular Coulomb function, while the red dashed curves are the frame-transformed wavefunctions.