Comment on "Classical description of H(1*s*) and H*(*n*=2) for cross-section calculations relevant to charge-exchange diagnostics"

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(Received 19 October 2015; published 29 June 2016)

Cariatore *et al.* [Phys. Rev. A **91**, 042709 (2015)] have introduced a modification of the classical trajectory Monte Carlo (CTMC) method, specially conceived to provide an accurate representation of charge-exchange processes between highly charged ions and H(1*s*), H*($n=2$). We point out that this new CTMC treatment is based on nonstable initial distributions for H*($n=2$) targets and an improper description of the H(1*s*) target.

DOI: 10.1103/PhysRevA.93.066701

The classical trajectory Monte Carlo (CTMC) method [1] is particularly useful for calculating electron capture and ionization cross sections in collisions of multicharged ions with hydrogen at energies above 25 keV/u, and it can provide the partial cross sections for populating high-lying energy levels, required in plasma diagnostics [2]. The original method [1] employs a microcanonical initial distribution, which has a constant energy and is obtained from a set of five random parameters, all uniformly distributed and independent; this ensures that the distribution is independent of time (see, e.g., [3]). However, this treatment has an important drawback: although the initial momentum density for H(1*s*) is exact, the radial distribution exhibits a nonphysical cut-off [at $r = 2.0$ a.u. for H(1*s*)]. Several authors [4–6], have suggested the use of improved initial distributions, which approximately fit both momentum and position quantal densities. It was found [7] that all these alternatives lead to practically identical electron capture cross sections. In particular, the hydrogenic distribution [4], named hydrogenic-E-distribution (HED) in [8], is constructed as a linear combination with constant coefficients of microcanonical distributions with different energies, E_k. Since the microcanonical distributions are stable, the HED is also time independent. The coefficients of the combination are obtained [4,9,10] by imposing that the average energy is equal to that of the corresponding quantum level, E_n. It is also ensured that the distributions included in the combination fulfill the conditions [11,12]:

$$[(n-1/2)(n-1)n]^{1/3} < n_c \leqslant [(n+1/2)(n+1)n]^{1/3}, \qquad \frac{l}{n} < \frac{l_c}{n_c} \leqslant \frac{l+1}{n}, \tag{1}$$

where, in atomic units, $n_c^2 = -\frac{Z^2}{2E_k}$ and l_c is the classical value of the electronic angular momentum. These conditions permit one to divide the phase space into adjacent nonoverlaping bins associated to the quantum numbers n,l. In practice, the use of the HED and other continuum distributions [5,6] improves the total cross sections with respect to the microcanonical calculation for electron capture and ionization processes, and n-partial electron capture cross sections for relatively large n (see [7]).

Cariatore *et al.* [8] have calculated electron capture cross sections for collisions of C^{6+}, N^{7+}, and O^{8+} with H($n = 1,2$) applying the CTMC method with an alternative initial distribution for H($n = 1,2$) targets, called hydrogenic-Z-distribution (HZD), which follows the spirit of the HED [4]. The authors claim that the HZD provides a more accurate representation of the target system than both microcanonical and hydrogenic-E ensembles. In the HZD treatment, the ion-H collision is represented by an ensemble of collisions with fictitious hydrogenic atoms with different nuclear charges (Z_k) and with the ionization energy of the H atom in the corresponding initial quantum level E_n. In practice, the HZD is constructed by imposing the quantal momentum distribution $\rho^{Q}(p;n,l)$. The radial HZD is a linear combination with coefficients α_k of distributions $\rho_k(r;Z_k,E_n)$, where the values of α_k are chosen by a least-squares fitting of the quantal radial distribution.

In this Comment we discuss the validity of the HZD procedure starting with the particular case of H(2*s*), where the authors claim that they are able to construct an initial classical distribution with a corresponding quantal momentum density $\rho^{Q}(p;2s)$, which has a node at $p = 0.5$ a.u. and a minimum in the radial density near the node at $r = 2.0$ a.u. of the quantal distribution (see Fig. 1). We have repeated the procedure described in [8], we have imposed the quantal momentum distribution, and obtained a set of distributions $\rho_k(r;Z_k,E_2)$ with $E_2 = -0.125$ hartree. The linear combination of those distributions with the coefficients α_k of Ref. [8] leads to the results plotted in Fig. 1, which are identical to those of [8]. In order to study the stability in time of this initial H(2*s*) distribution, we have integrated the Hamilton equations for the systems with charges Z_k, in absence of the projectile, and the time evolution of the radial and momentum densities are shown in Fig. 1. It is clear from this illustration that the HZD is not stable. Moreover, the node of the momentum distribution, which is the most astonishing fact of the illustration of [8], quickly disappears. Similarly, it is also found that the HZD for H(2*p*) plotted in Fig. 1 of Ref. [8] is not stable in time.

In order to further explain [13] the instability of the initial distributions of Cariatore *et al.* we have expressed them analytically as

$$\rho_k(r,p;Z_k,n,l) = f_{nl}^k(p)\delta(r - r_0(p)), \tag{2}$$

where $r_0(p) = Z_k(-E_n + p^2/2)^{-1}$ and the function $f_{nl}^k(p)$ is chosen in order to fulfill

$$4\pi p^2 \int \rho_k(r,p;Z_k,n,l)d\boldsymbol{r} = \rho^{Q}(p;n,l). \tag{3}$$

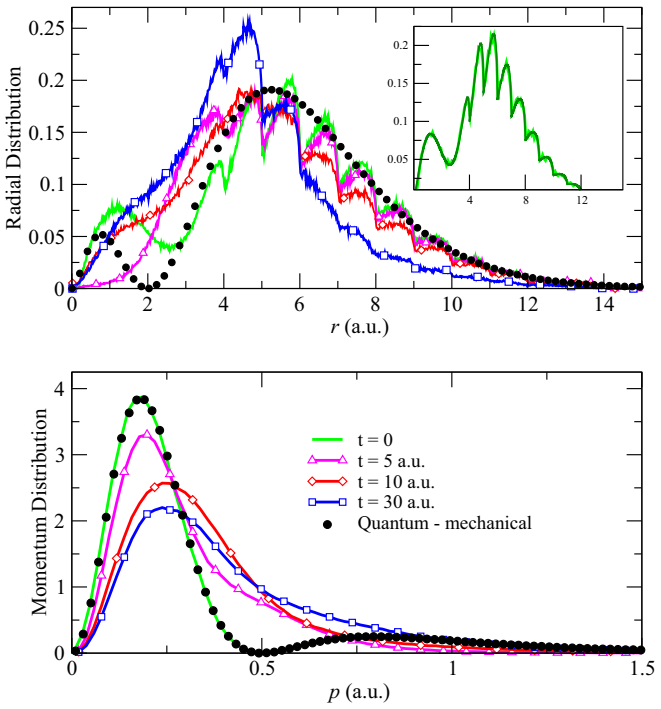

FIG. 1. Time evolution of radial (upper panel) and momentum (bottom panel) densities for the H(2s) initial distribution of Ref. [8], compared to the corresponding quantum-mechanical densities. In the inset of the upper panel we compare the numerical initial radial density with that calculated as a superposition of the radial densities from Eq. (2) (smooth line).

We have checked that the corresponding radial distributions are identical to the numerical ones (see the inset in the upper panel of Fig. 1). The distribution $\rho_k(r,p;Z_k,n,l)$ can be related to the microcanonical distribution by using the relationship (see, e.g., Ref. [14])

$$\begin{aligned}\delta(H-E_n) &= \delta\left(\frac{p^2}{2}-\frac{Z_k}{r}-E_n\right)\\ &= \delta(r-r_0)\left|\frac{\partial\left(\frac{p^2}{2}-\frac{Z_k}{r}-E_n\right)}{\partial r}\bigg|_{r_0}\right|^{-1}\\ &= \frac{r_0^2}{Z_k}\delta(r-r_0),\end{aligned} \tag{4}$$

which yields the expression

$$\rho_k(r,p;Z_k,n,l) = F_{nl}(p)\,\rho_{\rm M}(r,p;Z_k,E_n), \tag{5}$$

where $\rho_{\rm M}(r,p;Z_k,E_n) = K\delta(H-E_n)$ is the microcanonical distribution and $F_{nl}(p) = f_{nl}^k(p)K^{-1}r_0^{-2}Z_k$. In order to analyze the stability of this distribution we calculate the Poisson bracket $[\rho_k,H]$:

$$[\rho_k,H] = \frac{\partial\rho_k}{\partial\boldsymbol{r}}\frac{\partial H}{\partial\boldsymbol{p}} - \frac{\partial\rho_k}{\partial\boldsymbol{p}}\frac{\partial H}{\partial\boldsymbol{r}}. \tag{6}$$

Since $\rho_{\rm M}$ is a function of H, $[\rho_{\rm M},H]=0$ and, using (5), we obtain

$$[\rho_k,H] = \frac{\boldsymbol{r}\cdot\boldsymbol{p}}{rp}\,\rho_{\rm M}\,\frac{dF_{nl}}{dp}\,\frac{dV_k}{dr}, \tag{7}$$

with $V_k=-Z_k/r$. This Poisson bracket does not vanish unless $\frac{dF_{nl}}{dp}=0$ and, using the Liouville equation:

$$\frac{\partial\rho_k}{\partial t} = -[\rho_k,H] \neq 0. \tag{8}$$

Although the distribution (2) is not stationary for excited states, the HZD for H(1s) is stable as a consequence of the fact that the microcanonical momentum distribution is identical to the the quantum-mechanical one [1]

$$\rho_k(r,p;Z_k,1,0) = \rho_{\rm M}(r,p;Z_k,E_1). \tag{9}$$

Therefore, $F_{10}=1$, and the HZD for H(1s) is a linear combination of microcanonical distributions and it is stable in time. However, the procedure of Ref. [8] leads to difficulties for applying the binning procedure of Eqs. (1), applied to obtain the partial cross sections, but unlike the case of the HED treatment, high values of n_c are used in the HZD. In fact, since $n_c = Z_k$ for each nuclear charge, one-third of the total number of trajectories belong to excited bins ($n_c > 1.442$). Analogously, there is a sizable number of trajectories with angular momentum $l_c > 1$. This implies that HZD results for ion-H(1s) collisions do not correspond strictly speaking to that of a classical H(1s) target.

CTMC calculations are relevant because they provide the electron capture cross section in collisions of highly charged ions with H, needed in plasma diagnostics. From this practical point of view, Cariatore *et al.* argue that the use of the HZD notably improves the CTMC results by comparing them with the available quantum-mechanical calculations. Since the HZDs for excited H are not stable, we do not consider these results and we discuss the cross sections for electron capture in ion-H(1s) collisions. In this respect, it is worth mentioning that Cariatore *et al.* do not compare their results with existing theoretical and experimental results; in particular, with the work of Jorge *et al.* [15,16] for C^{6+}, $N^{7+}+H(n=1,2)$ collisions. The comparison of total electron capture cross sections, not presented in Ref. [8], is shown in Fig. 2, where one can note that the results of [8] are very similar to those obtained with the microcanonical distribution, and a better agreement is found between the CTMC results of [15] and the close-coupling calculations of [17], and also with previous theoretical [18,19] and experimental results [20,21], not included in the figure for clarity.

Cariatore *et al.* based the justification of the HZD on the improvement of partial electron capture cross sections with respect to those calculated using other initial distributions. We show in Fig. 3 similar comparisons for $C^{6+}+H(1s)$, including our previous results [15]. In Ref. [8], n-partial cross sections are tabulated at three energies: 10, 50, and 100 keV/u. The collision energy $E=10$ keV/u is probably too low for using the classical treatment, and it was not included in [15]. However, this illustration is employed by Cariatore *et al.* to conclude that the HED is not appropriate. To further analyze this point, we include in Fig. 3 our unpublished results using this distribution to show that the calculation of Cariatore *et al.* with the HED is probably not correct at this energy.

At $E=50$ keV/u, the HZD cross sections reproduce the microcanonical ones for $n\leqslant 4$, which agree with the quantal results [17], obtained for a slightly lower collision energy, $E=45$ keV/u. On the other hand, for high n, relevant to

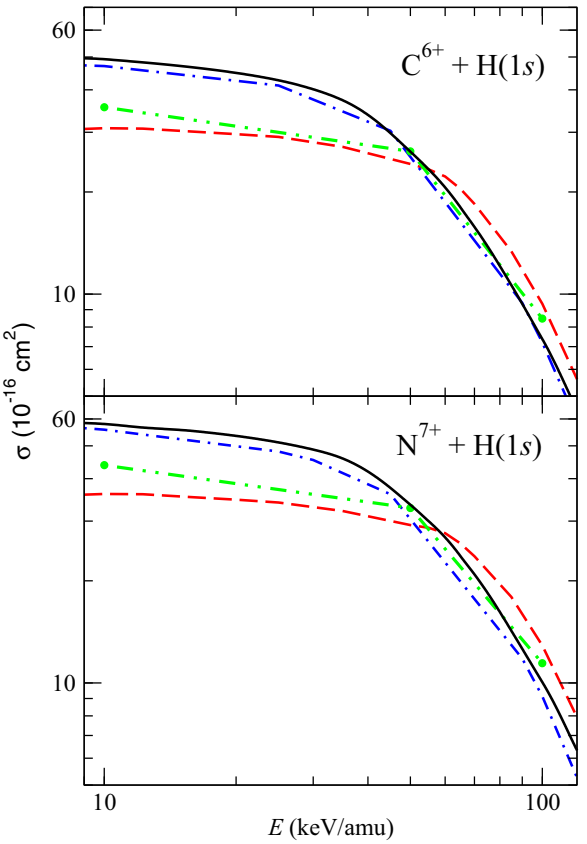

FIG. 2. Total cross sections for electron capture in C^{6+} and N^{7+} collisions with H(1s), as functions of the collision energy. CTMC calculations with different initial distributions: —, HED [15]; _ _ _, microcanonical [15]; ● — · · · —●, HZD [8]; _ · _ · _, quantum-mechanical calculation [17].

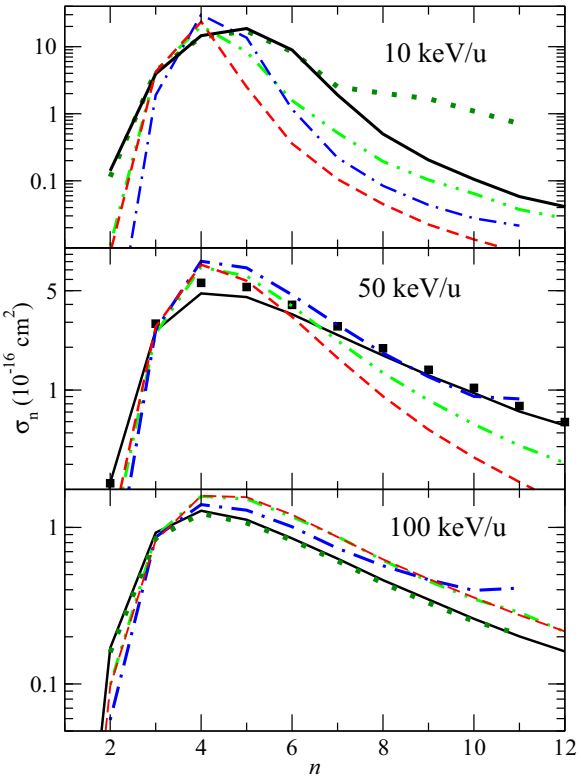

FIG. 3. n-partial cross sections for electron capture in C^{6+} + H(1s) collisions. —, HED calculation [15]; (■ ■ ■), HED calculation [8]; _ _ _, microcanonical result; _ · _ · _, HZD result [8]; _ · · _ · · _, quantum-mechanical calculation [17]. The quantum-mechanical results in the middle panel are the cross sections at a slightly different energy, $E = 45$ keV/u, and for comparison, the HED cross sections [15] are shown at the same energy (■).

charge exchange recombination spectroscopy diagnostics, one observes good agreement between quantal and HED results, while the HZD calculation clearly underestimates the partial cross sections. The comparison of the partial cross sections at $E = 100$ keV/u clearly shows that the HZD leads to results which are practically identical to the microcanonical ones. The HED yields partial cross sections below the quantal calculation of Ref. [17], but, at these collision energies, the partial cross sections from the atomic close-coupling calculation of Ref. [17] probably include overpopulation from the ionizing flux, as indicated by the increase of the partial cross section for $n > 9$. This limitation of the atomic-basis close-coupling calculation was previously discussed in Ref. [15] and is mentioned in the conclusions of Ref. [8].

To summarize, the CTMC treatment introduced by Cariatore *et al.* is not correct for collisions with neither excited nor ground-state H targets. The use of the HZD for H(1s) precludes the application of the usual partition of the phase space for the initial state. In practice, the improvement of the total (not illustrated in [8]) and partial electron capture cross sections is questionable. In general, the HZD electron capture results are close to the microcanonical ones, and the ionization cross section, which is more sensitive to the quality of the initial distribution is not presented. For excited state H targets the hydrogenic-Z distribution is not stable and cannot be employed.

This work has been partially supported by Ministerio de Economía y Competitividad (Spain) (Projects No. ENE2011-28200 and No. ENE2014-5432-R).

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