Comment II on "Topological angular momentum in electron exchange excitation of a single atom"

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A recent article by Williams *et al.* [Phys. Rev. A **85**, 022701 (2012)] highlights a discrepancy between experiment and theory for the linear light polarization P_2 measured after impact excitation of zinc atoms by a spin-polarized electron beam. The claim is made that current collision theories must be modified by including a geometric (Berry) phase in the calculations in order to reproduce the experimental data for Zn and similar data from the Münster group for Hg. We show that the *e*-Hg data can be qualitatively reproduced by our fully relativistic *B*-spline *R*-matrix approach *without* any further modification.

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A serious discrepancy between experimental data and theoretical predictions was recently reported [1] for spin-polarized electron-impact excitation of the $(4s5s)\,^3S_1$ state in Zn atoms. The linear light polarization P_2, measured for optical decays to the $(4s4p)\,^3P_{0,1,2}$ states with a photon detector aligned along the direction of the spin polarization P_e of the incident electron beam, was found to be significantly (nearly 10% for the final state $^3P_0$) different from zero, whereas, all available numerical calculations predicted an effect of less than 0.01% in the cascade-free region just above the excitation threshold. In 1982, Bartschat and Blum [2] predicted a zero result for P_2/P_e if the following assumptions were valid: (i) Relativistic effects, both for the projectile-target interaction and in the target structure description alone, can be neglected; (ii) the orbital electronic angular momentum L and the spin S of the excited target state are well defined, i.e., configuration interaction with terms of different L's and S's is negligible. The $(4s5s)\,^3S_1$ state in Zn seems to be a very good candidate for such a case, and Zn is sufficiently light that spin-orbit effects during the excitation process are likely small. Hence, very small absolute values of P_2/P_e were expected, and the experimental results reported in Ref. [1] came as a major surprise.

After discussing the P_2/P_e discrepancy between theory and experiment once again for *e*-Zn excitation in their subsequent paper [3], to which this Comment is directed, Williams *et al.* conclude with the firm recommendation: "The task remains for theory to include a topological nondynamical phase." No suggestion, however, is made how this might or should be performed. We are not aware of any electron-atom collision theory that includes such a phase. The above statement about "theory," however, seems to imply a view that something is missing. It is worth pointing out, therefore, that the most successful *ab initio* theories, especially those based on some variant of the close-coupling expansion for the projectile-target collision system, employ properly antisymmetrized wave functions for all electrons in the system rather than the *alternative* formulation proposed by Berry and Robbins [4]. Note that Berry and Robbins themselves emphasize the *alternative* character of their approach. In Sec. 7 (p. 1784) of their paper, they state that ". . . this [their formulation] quantum mechanics leads to the same physics (e.g., the exclusion principle) as more conventional formulations, ..."

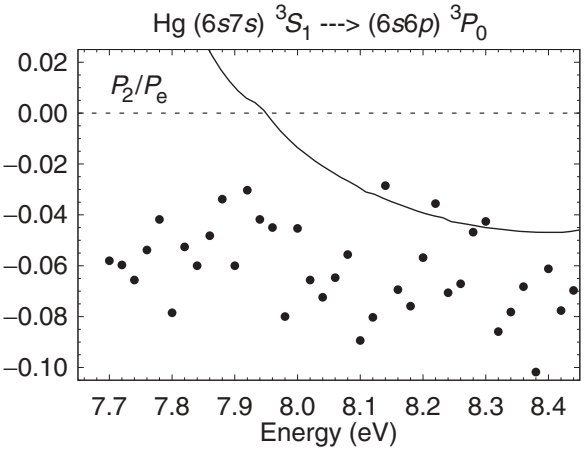

FIG. 1. P_2/P_e for spin-polarized electron-impact excitation of the $(6s7s)\,^3S_1$ state in Hg with subsequent optical decay to the $(6s6p)\,^3P_0$ state. The experimental data of Goeke [5] are compared with the DBSR prediction based on the model described in Ref. [6].

We have no doubt that tremendous experimental effort and attention to detail were exercised in generating the experimental *e*-Zn results. As elaborated on in Refs. [1] and [3], the data appear to be consistent against all cross checks performed to date and, hence, remain a mystery that we cannot explain at the present time. Figure 1, however, shows that the experimental data, obtained in Münster for the *e*-Hg collision system nearly 30 years ago [5], which were used in Ref. [3] as further supporting evidence for the need to include a Berry-type phase, can, at least, be qualitatively reproduced by the Dirac-based *B*-spline *R*-matrix (DBSR) model described in Ref. [6] *without* making any further changes to the method or the accompanying computer code. As seen in the figure, we obtain absolute values of up to nearly 5% for *e*-Hg in the cascade-free region.

We are currently investigating the dependence of these predictions on the details of the model, in particular, the description of the target structure. These calculations are extremely demanding and computationally expensive. Hence, we cannot yet pinpoint an unambiguous reason for the differences in the theoretical results for Zn and Hg. A promising candidate is a significant admixture (about 7% in the present DBSR model) of what would correspond, in a semirelativistic model,

to a $(6p^2)\,^3P_1$ term in the target description of the $(6s7s)\,^3S_1$ state in Hg, whereas, the respective $(4p^2)\,^3P_1$ admixture to the $(4s5s)\,^3S_1$ state is very small in our structure calculation for Zn. Note that such a $^3P_1$ term would indeed violate the conditions outlined by Bartschat and Blum [2] and, hence, would allow for a nonvanishing value of P_2/P_e.

We strongly believe that the discrepancy between theory and experiment for P_2/P_e in Zn [1] deserves further investigations on both the theoretical and the experimental fronts. It seems highly advisable to independently repeat the e-Zn experiment, as unlikely as a technical problem might appear to our experimental colleagues. At the same time, further investigation of the computational model is needed. Furthermore, while performed with a state-of-the-art apparatus nearly 30 years ago, the Münster group never published their P_2/P_e data for this particular process in Hg. Hence, we recommend checking those data as well, using the best available experimental methods today. In particular, the finite-energy resolution might partly explain the major discrepancies in Fig. 1 between the experimental data and the DBSR predictions in the near-threshold region where the cross section and, hence, the signal, is very small. Other targets, such as Cd (poisonous and, hence, experimentally challenging but certainly not impossible) or other quasi-two-electron systems (Yb or alkaline-earth metals) come to mind. Although we remain open to the possibility of something still being missing in our, and presumably other current theoretical models for e-Zn collisions, we are convinced that the resolution of the discrepancy does not, and should not, involve modifying our approach by introducing the Berry phase in an *ab initio* formulation that already accounts for the Pauli principle to the full extent.

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