Comment on "Complete Fragmentation Pattern for Two-Step Double Photoionization in Xenon"

In a recent Letter [1], Kämmerling and Schmidt presented experimental values for the three complex amplitudes for photoionization of the $4d_{5/2}$ subshell of xenon at photon energy 94.5 eV. To determine these amplitudes, they measured the angular correlation between the photoelectron and the $N_5O_{23}O_{23}$ 1S_0 Auger electron emitted in the decay of the residual Xe⁺ ion. This measurement was combined with measurements of the $4d_{5/2}$ partial cross section, $\sigma(4d_{5/2}) = 12.2(1.5)$ Mb, of the photoelectron angular distribution asymmetry parameter, $\beta(4d_{5/2}) = 0.35(1)$, and of the noncoincident alignment parameter, $\mathcal{A}_{20}(\frac{5}{2}) = -0.230(15)$. Kämmerling and Schmidt report the values

$$d_1 = 0.138(22), \quad d_2 = 0.131(17), \quad d_3 = 0.474(59), \\ \Delta_1 - \Delta_2 = \mp 0.10(10), \quad \Delta_2 - \Delta_3 = \pm 1.35(10), \quad (1)$$

where the parameters d_k and Δ_k for k = 1,2,3 are the magnitudes (a.u.) and phases (radians) of the complex amplitudes for the channels $4d_{5/2} \rightarrow p_{3/2}$, $4d_{5/2} \rightarrow f_{5/2}$, and $4d_{5/2} \rightarrow f_{7/2}$, respectively.

We calculate these same three amplitudes in the relativistic random-phase approximation (RRPA) [2] and obtain

$$d_1 = 0.121, \quad d_2 = 0.122, \quad d_3 = 0.558, \\ \Delta_1 - \Delta_2 = -0.522, \quad \Delta_2 - \Delta_3 = 0.014.$$
(2)

The RRPA calculation coupled the thirteen open channels originating from the photoionization of the $5p_{3/2}$, $5p_{1/2}$, $5s_{1/2}$, $4d_{5/2}$, and $4d_{3/2}$ subshells of xenon. These theoretical amplitudes lead to the values $\sigma(4d_{5/2}) = 15.6$ Mb and $\beta(4d_{5/2}) = 0.26$ [3]. Comparing the experimental amplitudes from Eq. (1) with the theoretical amplitudes in Eq. (2), one sees that whereas the magnitudes are in approximate agreement, the phases disagree seriously.

In Fig. 1, we plot the theoretical amplitudes in the photon energy range 85-105 eV, and in Fig. 2, we plot the relative phases in the same energy range. Because of the rapid rate of change of the theoretical phase difference



FIG. 1. Magnitudes of the $4d_{5/2}$ photoionization amplitudes as a function of photon energy.



FIG. 2. Relative phases of the $4d_{5/2}$ photoionization amplitudes as a function of photon energy.

 $\Delta_1 - \Delta_2$, the discrepancy for this quantity could possibly be accounted for by correlation corrections of the type reported in Refs. [4,5]. Such an explanation could not account for the discrepancy in $\Delta_2 - \Delta_3$, however, since the theoretical phase difference between the channels $4d_{5/2} \rightarrow f_{5/2}$ and $4d_{5/2} \rightarrow f_{7/2}$ nearly vanishes at all energies in the interval considered, reflecting the expected nonrelativistic degeneracy of the phases of these two channels. It appears, therefore, that the experimental values of the phase differences are in error by a considerably larger margin than reported in Ref. [1]. In this regard, it should be mentioned that a similar discrepancy between experiment and theory is found for the phase difference between the channels $5p_{3/2} \rightarrow d_{5/2}$ and $5p_{3/2} \rightarrow d_{3/2}$ in the photoionization of the $5p_{3/2}$ subshell of xenon. The experimental phase difference for photon energies in the range 15-30 eV was found to be approximately $\pi/2$ [6], while the theoretical phase difference in this energy range is close to 0.

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