Reply to "Comment on 'Gamma-ray spectra from low-energy positron annihilation processes in molecules' "

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In reply to the Comment of Green *et al.* [Phys. Rev. A **95**, 036701 (2017).] on our paper [Phys. Rev. A **94**, 052709 (2016)], we reconfirm that all the conclusions are based on the observation and the comparisons of the theoretical and experimental data. One criticism of Green *et al.* [Phys. Rev. A **95**, 036701 (2017).] concerns the positrophilic electrons and the inner valence electrons. The inner valence electrons or positrophilic electrons show most agreeable widths with the corresponding experimental measurements due to their narrowest momentum distributions for all 59 molecules. However, we agree with the criticism of Green *et al.* [Phys. Rev. A **95**, 036701 (2017).] and reconfirm that this agreement does not represent the dominance of the inner valence in the annihilation process. In this Reply, we will clarify the difference between agreement and dominance and illustrate with some figures. Another criticism is about the approximation used in our paper. We emphasize that the averaged discrepancy of 34.2% for these molecules of the theoretical γ-ray spectra from the experimental measurements is due to the neglect of the positron-electron correlations, vibrational couplings, virtual-state formation, even tunneling of core electrons not the neglect of the positron wave function. In this Reply, we will show, even in this zero-order approximation, these positron-induced effects in the electron-positron annihilation process of molecules can also be analyzed with more corrections and explanations.

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One criticism of Green *et al.* [1] concerns the approximation used in our paper [2]. The calculations are based on the low-energy plane-wave positron approximation which the positron wave function is set to unity, i.e., neglected. This approximation is reasonable because the experiments for the present 59 molecules are performed at room temperature ("the momentum of the annihilating pairs is typically dominated by the momenta of the electrons" [3]). The relative average deviations $\delta = \frac{\sum_i (W_{T_i} - W_{E_i})/W_{T_i}}{59} \times 100\%$ between these 59 theoretical (W_{T_i}) and the experimental (W_{E_i}) widths of γ-ray spectra (in Table V of our paper [2]) are $\delta_T = 34.2\%$ and $\delta_V = 29.4\%$ for total and valence electrons, respectively. Which agrees well with the well-known conclusion that only about 70% agreement will be achieved under this approximation [4]. The positron-induced suppression [4], virtual-state formation [5], vibrational coupling [6], and even exchange-assisted tunneling [7] and core electron enhancement [8] have chiefly been considered to explain this discrepancy. The understanding of these positron annihilation enhancement effects in the low-energy region depends on the accurate positron wave function which is still difficult to obtain in the molecular systems. Our paper shows [2], even in this zero-order approximation, these positron-induced effects in the electron-positron annihilation process of molecules can also be analyzed from these statistical deviations of different kinds of electrons.

According to this criticism, however, this zero-order approximation indeed could cause some misunderstanding of the widths of the γ-ray spectra listed in Table V of our paper [2]. More explanations are needed to interpret these data to avoid misunderstanding. We numbered the molecules having experimental measurements (from Table V of our paper) in Table I and drew the corresponding widths (from Table V of our paper) in Figs. 1 and 2, respectively. One thing should be mentioned that, as shown in Fig. 1, all the predicted widths of the γ-ray spectra for total and valence electrons are wider than the available experimental measurements for all these molecules. The variation of the widths for both the valence and the experiments of all these molecules shows a similar role. In the experimental condition, the typical kinetic energy of these thermalized positrons is $\frac{3}{2}kT = 0.04$ eV [3]. This energy is small enough, compared with the kinetic energies of bound electrons in molecules. The thermalized positrons almost have no opportunities to encounter and annihilate with core electrons. Hence, the experiments might have measured the γ-ray spectra of the valence electrons in molecules [3]. Only about 4.8% ($\delta_T - \delta_V$) contributions or enhancements are from the core electrons.

More attention should be paid to the zero-order approximation-induced huge core electrons, about 43%, 73%, and 40% enhancements, respectively, for krypton, xenon, and carbon tetrabromide when comparing the total and valence electrons as shown in Fig. 1. In these three molecules (the down-arrow indicated in Fig. 1), most of the electrons are distributed in the core orbitals (in Table VII of our paper [2]). Hence, the present approximation for these molecules will mislead readers. Even if $3d$ of krypton, carbon tetrabromide and $4d$ of xenon atomic electrons were considered as valence orbitals, these outermost d electrons will tunnel and annihilate with positrons. The widths of d-plus-other-valence electrons ("valence + d") are 3.38, 2.98, and 3.36 keV for krypton, xenon, and carbon tetrabromide, respectively. The widths of the rest of the core electrons in these molecules are corrected to 11.25, 11.99, and 11.65 keV and are still extremely big. The approximation obviously overestimates the contribution of the core electrons and widely enhances the γ-ray spectra dramatically. If these three molecules are excluded in the statistics, the enhancements from the core electrons decrease to about 3.9%. This value is more reasonable.

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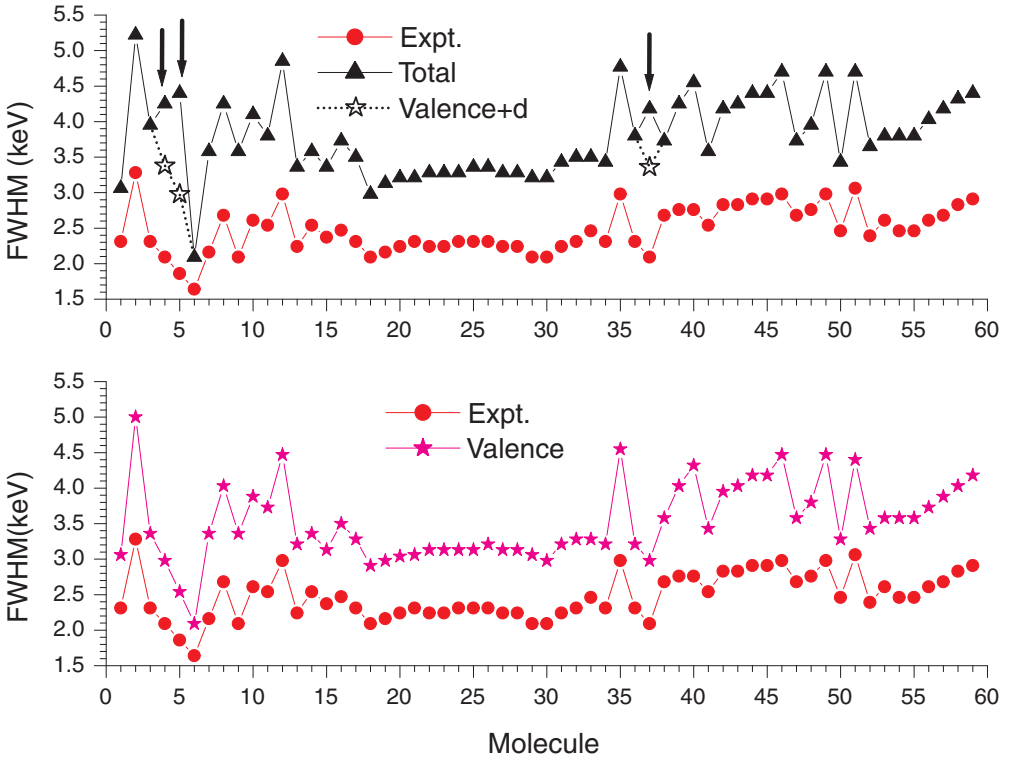

FIG. 1. The comparison of the γ-ray linewidths of the experimental and theoretical total and valence electrons for all molecules, respectively.

The other criticism concerns the positrophilic electrons and the inner valence electrons. These concepts are based on the observations and comparisons between the zero-order calculations and the experiments. The inner valence electrons show most agreeable widths with the experimental data ($\delta_I = 9.6\%$) due to their narrowest momentum distributions as shown

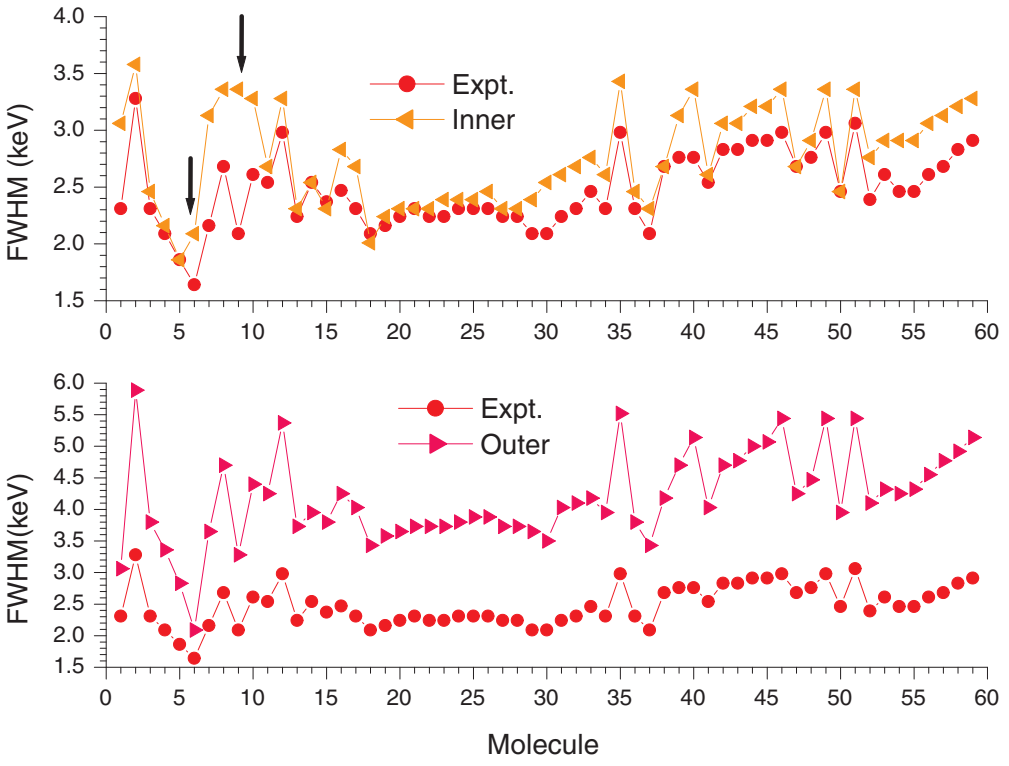

FIG. 2. The comparison of the γ-ray linewidths of the experimental and theoretical inner and outer valence electrons, respectively, for all molecules.

TABLE I. The sequence numbers of the 59 molecules.

No.	Molecule	No.	Molecule	No.	Molecule
1	Helium	2	Neon	3	Argon
4	**Krypton**	**5**	**Xenon**	6	Hydrogen
7	Nitrogen	8	Oxygen	9	Carbon monoxide
10	Carbon dioxide	11	Water	12	Sulfur hexafluoride
13	Ammonia	14	Methanol	15	Tetraethylsilane
16	Nitrobenzene	17	Pyridine	18	Methane
19	Ethane	20	Propane	21	Butane
22	Pentane	23	Hexane	24	Nonane
25	Dodecane	26	Cyclohexane	27	2-Methylbutane
28	2,2-Dimethylpropane	29	Ethylene	30	Acetylene
31	Benzene	32	Naphthalene	33	Anthracene
34	Toluene	35	Carbon tetrafluoride	36	Carbon tetrachloride
37	**Carbon tetrabromide**	38	Methyl fluoride	39	Difluoromethane
40	Trifluoromethane	41	Fluoroethane	42	1,1,1-Trifluoroethane
43	1,1,2-Trifluoroethane	44	1,1,1,2-Tetrafluoroethane	45	1,1,2,2-Tetrafluoroethane
46	Hexafluoroethane	47	2,2-Difluoropropane	48	1,1,1-Trifluoropropane
49	Perfluoropropane	50	1-Fluorohexane	51	Perfluorohexane
52	Fluorobenzene	53	1,2-Difluorobenzene	54	1,3-Difluorobenzene
55	1,4-Difluorobenzene	56	1,2,4-Trifluorobenzene	57	1,2,4,5-Tetrafluorobenzene
58	Pertafluorobenzene	59	Hexafluorobenzene		

in Fig. 2. The relative average deviations between experiments and outer valence electrons are $\delta_O = 40.2\%$. However, as the criticism pointed out, without considering the accurate positron wave function and the correlations between electron and positron, the seemingly strong correlation of the inner valence electrons with the experimental annihilation process cannot be confirmed indeed. From our paper, good agreement between the inner valence electrons and the experiments does not mean the dominant or positrophilic electrons in the annihilation process under the present approximation. This misunderstanding only can be corrected by considering the accurate positron wave functions. In addition, a typographical error has been found in Table VII of our paper. $\frac{a}{b}1a_{1g}^2$ should be $1a_{1g}^2$. The readers should be reminded to notice it. We will work on the positron-induced effects to confirm or correct this in the following study under higher-order approximation.

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