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# Comment on "Electron-impact excitation of the $B\ ^1\Sigma_u^+$ and $C\ ^1\Pi_u$ electronic states of $H_2$"

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In a recent paper [Kato *et al.*, Phys. Rev. A **77**, 062708 (2008)], optical oscillator strengths for the Lyman and Werner transitions of $H_2$, extrapolated from the authors' experimental generalized oscillator strengths, are compared with a suite of other values, several of which have been incorrectly derived from the literature. These significant errors are corrected and it is suggested that claiming the status of an "optical" oscillator strength for the extrapolated generalized oscillator strength associated with electron energy-loss spectra at 100–200-eV impact energy is somewhat questionable.

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In a recent experimental study of electron-impact excitation of the $B\ ^1\Sigma_u^+$ and $C\ ^1\Pi_u$ electronic states of $H_2$, primarily concerned with the determination of differential and integral cross sections from electron-energy-loss spectra (EELS) recorded at impact energies in the range 40–200 eV, Kato *et al.* [1] compare "optical" oscillator strengths (OOSs) extrapolated from their generalized oscillator strength (GOS) measurements with a variety of other determinations. Unfortunately, the OOS values attributed to some of the other experimental studies in their Table III are incorrect. It appears that they simply summed the vibrational oscillator strengths $f_{v'0}$ determined by the other workers, *regardless of the particular number of* $(v',0)$ *bands measured*, to obtain their OOSs for comparison, necessarily resulting in underestimates, some of which are very severe.

In Table I, we compare the experimental $B\ ^1\Sigma_u^+ – X\ ^1\Sigma_g^+$ and $C\ ^1\Pi_u – X\ ^1\Sigma_g^+$ OOSs of Ref. [1] with the results of several benchmark studies [2–5], representing a range of experimental and theoretical techniques, having taken care to compare like with like. The tabulated OOSs represent the sum over all significant vibrational bands, i.e., $f_0 = \Sigma_{v'} f_{v'0}$, with maximum $v' = 36$ and 13, respectively, for the Lyman and Werner transitions. Missing bands in the experimental studies [2,4,5] have been corrected for, on a relative basis, by using the vibrational dependences implied by the theoretical oscillator strengths of Allison and Dalgarno [3] (estimated ~6% uncertainty), which are based on the very accurate internuclear-distance-dependent *ab initio* electronic transition moments of Wolniewicz [6]. The OOSs in Table I exceed the corresponding values in Table III of Ref. [1] by up to 250%.

An excellent and detailed discussion of the relative merits of the various experimental oscillator-strength determinations has been given by Chan *et al.* [5]. Briefly, the only *absolute* experimental oscillator strengths are those of Lewis *et al.* [4,7,8], measured with rotational resolution using extreme-ultraviolet (EUV) photoabsorption spectroscopy, the usual problems of line saturation taken care of by means of a curve-of-growth analysis. The estimated uncertainty in these summed optical measurements is ~6%, mainly caused by the uncertainty in the absorbing column density of $H_2$. The OOSs of Geiger and Schmoranzer were determined using extremely high-energy, forward-scattering EELS, reliant on indirect calibration against elastic scattering cross sections, with a corresponding uncertainty of 15%. Finally, the OOSs of Chan *et al.* [5] are from high-energy EELS experiments with negligible momentum transfer ($K$), in which the cross section is calibrated indirectly using the Thomas-Reiche-Kuhn sum rule, leading to an estimated uncertainty in the summed oscillator strengths of ~5%. Under the particular scattering conditions of these two EELS experiments, both are expected to yield oscillator strengths representative of the optical limit. In summary, if we ignore, for a moment, the results of Ref. [1], there is excellent agreement on the OOS for the Lyman transition, with the extreme $f_0$ values in Table I differing by only 4%, well within the combined uncertainty estimates. In the case of the Werner transition, the extreme values differ by ~20%, at the edge of the combined uncertainties.

However, the extrapolated GOS values of Ref. [1] in Table I, with quoted uncertainties of ~20%, lie 22% and 30% below the means of the other OOSs for the Lyman and Werner transitions, 0.310 and 0.325, respectively. Thus, there must still be some doubt regarding the usefulness of analytically extrapolating a GOS vs $K^2$ curve to $K^2=0$ and claiming the result as an "optical" value, particularly for impact energies as low as 100–200 eV. Indeed, Fig. 3 of Ref. [1] shows that their extrapolated GOSs at 200 eV are systematically ~4% higher than the 100-eV values, albeit well within their uncertainty estimates. Similar discrepancies have been found recently in the case of $N_2$, where the extrapolated GOS for the $b\ ^1\Pi_u – X\ ^1\Sigma_g^+(0)$ transition at 100-eV impact energy was

TABLE I. Representative Lyman- and Werner-band electronic oscillator strengths for $H_2$, determined by a variety of techniques. The tabulated values represent $f_0 = \Sigma_{v'} f_{v'0}$.

| Technique | Ref. | $f_0\,[B\text{-}X]$ | $f_0\,[C\text{-}X]$ |
|---|---|---|---|
| EELS, 100–200 eV | [1] | 0.241 | 0.226 |
| EELS, 3 keV | [5] | 0.310 | 0.351 |
| EELS, 34 keV | [2] | 0.302 | 0.287 |
| Optical, EUV spectroscopy | [4] | 0.315 | 0.304 |
| Theory, *ab initio* | [3,6] | 0.311 | 0.356 |

only 72 ± 20 % of the accurately known optical value [9].

In conclusion, optical oscillator strengths are most often best known from a variety of other experimental techniques, specifically photoabsorption spectroscopy and/or the "dipole (*e*,*e*)" method of Chan *et al.* [5]. In our view, the usefulness of extrapolating the GOS curve to zero momentum transfer in lower-impact-energy EELS experiments is mainly as a self-consistency check to reveal any serious systematic errors in the experimental differential cross sections, rather than as a *determination* of an OOS.

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