Comment on "Experimental Study of Multiple Scattering in X-Ray-Absorption Near-Edge Structure"

In a recent Letter Bunker and Stern[1] reported an experimental study of multiple scattering (MS) in x-ray-absorption near-edge structure (XANES) and extended x-ray-absorption fine structure (EXAFS). MS events were partitioned into two classes, those for which an intervening atom lies approximately collinearly with the excited atom and the backscattering atom (type-1 MS), and all other types of events (type-2 MS). Only if type-2 MS is measurable does XANES provide structural information inacessible to EXAFS. Bunker and Stern concluded that type-1 MS is important throughout the XANES regime while type-2 is important only if bond distances are $\lesssim 1.6$ Å. In particular, the authors attribute the failure of the curved-wave single-scattering (SS) approximation[2] for the O K-edge XANES in NiO to the presence only of type-1 MS. We demonstrate in this Comment that beyond approximately 20 eV above the O K edge in NiO, type-1 MS is indeed dominant, while at lower energies neglect of type-2 MS produces *experimentally distinguishable* differences in the calculated XANES.

Our calculations were performed with an updated version of the original XANES code,[3] which is based upon a cluster method and includes all MS contributions. The cluster is divided into concentric shells of atoms centered around the excited atom, and the MS equations are solved first within each shell and then between the shells themselves and the central atom. By choosing the shells to be the individual coordination shells of neighbors of the central oxygen atom we obtain a natural distinction between type-1 (*inter*shell) and type-2 (*intra*shell) MS and the importance of each may be addressed separately.

In Fig. 1(a) we have compared the calculated O K-edge XANES in NiO including full MS with spectra calculated using (1) the curved-wave SS approximation and (2) the approximation suggested by Bunker and Stern (full intershell MS but no intrashell MS). Our calculational parameters are similar to those of Ref. 2, and we include a 1-eV lifetime broadening of the photoelectron. We see that the discrepancies between the SS and full MS calculations beyond 20 eV above threshold are removed by including intershell MS, though serious differences remain at lower energies. Inclusion of intrashell MS only within the second (oxygen) coordination shell yields a spectrum that reproduces the full MS calculation over the entire range of energies [Fig. 1(b)]. We have compared our calculations with the measured[4] O K-edge XANES in NiO in Fig. 1(b) to demonstrate the observability of type-2 MS. The structure in the absorption spectrum within 5 eV of the edge remains a matter of some controversy and shall not be addressed here.

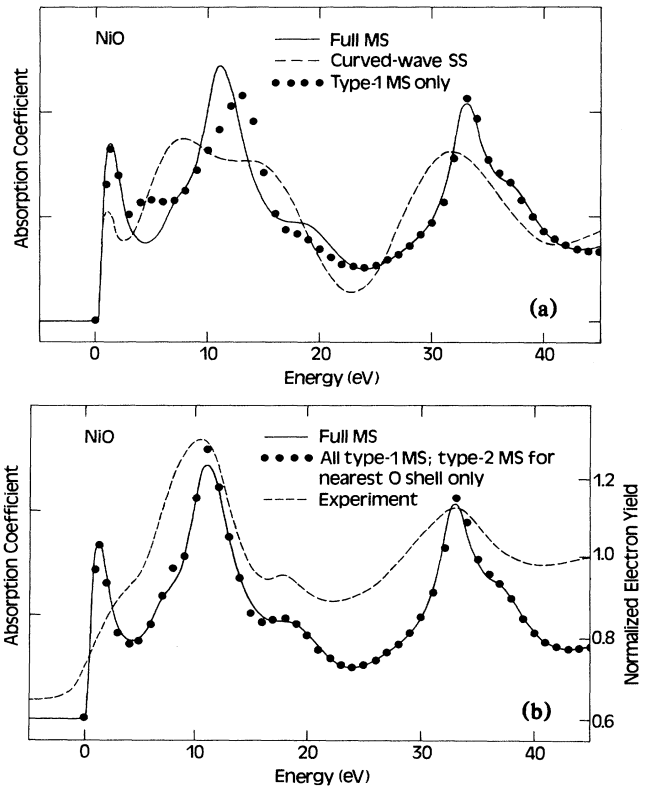

FIG. 1. Comparison of calculated O K edge XANES including full MS with calculations using the indicated approximations (left scale) and with experimental data (right scale).

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