Comment on “Near-Field Electron Energy Loss Spectroscopy of Nanoparticles”

In a recent article [1] Cohen et al. found a more fashionable name for a technique called many years ago [2] “aloof beam energy loss spectroscopy” and already by that time usefully employed in surface studies [3] and in scanning transmission electron microscopy (STEM) [4]. The related emission of radiation emitted from a surface excited by an external electron beam was observed earlier still by Smith and Purcell [5]. Thanks to subsequent STEM experiments and related theoretical investigations for planar [6], spherical [7], and other surfaces [8], most of the questions raised in [1] have been thoroughly understood for over a decade. In this light, some more specific comments can be made.

(1) The authors’ explanation of the tendency for the excitation of lower losses $\omega$ to fall off more slowly with impact parameter $b$ is simply a restatement of the well-known [9] involvement of the parameter $ab/\nu$ in all such Coulomb interactions, where $\nu$ is the electron velocity and $\gamma$ is the relativistic factor.

(2) When the fast electron travels outside and parallel to the planar surface of a semi-infinite dielectric, the impact parameter dependence of the loss spectrum (showing a slight concave curvature in a log intensity plot) is theoretically [10] and experimentally [6] reasonably well described by the function $K_0(2ab/\nu)$. Relativistic modifications to this result have been calculated and measured [11].

(3) Contrary to the authors’ belief, the “plasmonic” or collective modes are not in any way favored over single electron excitation in aloof beam spectroscopy. The $\omega$ dependence noted above is the main factor, and the only other one is the change (in the nonrelativistic regime) in the characteristic excitation function from $\text{Im}\{-1/\epsilon(\omega)\}$ for a penetrating beam to $\text{Im}\{-1/[1 + \epsilon(\omega)]\}$ for an external beam [6,12].

(4) The excitation theory in which the fast electron is treated as a classical charge and which the authors criticize as inadequate has in fact been extremely successful in quantitatively fitting experimental data [6,11,12]. Provided that a small momentum recoil term can be ignored, this model has been shown [13] to be exact in cases where all of the inelastic scattering is collected. Should it be necessary for some reason to employ a quantal model of the fast electron, localized descriptions based on a superposition of plane waves [14] or on a more general self-energy formalism [15] have already been developed for this purpose. The highly localized initial state of the fast electron can be expressed in terms of any other basis functions, including box states. Such a description is, however, inappropriate for the final states since, to an excellent approximation, the detector sums incoherently over points in the far-field region and so defines these to be plane waves. Although the highly focused fast electron propagates in a longitudinal magnetic lens field of about 1 T, the oscillator functions, which constitute a more appropriate basis for this situation [16], are in fact closely similar to plane waves in the small interaction region.

(5) The advantage of an aloof beam over a penetrating beam in generating less specimen damage for a given amount of data collected in the low loss region has already been noted [12] and is likely to be particularly significant in cases where structural damage follows only after core excitation and an Auger process.

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