SUPPORTING INFORMATION FOR

Probing the presence and absence of metal-fullerene electron transfer reactions in helium nanodroplets by deflection measurements

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I. C_{60}Yb ion mass spectra

Fig. S1 shows the mass spectrum of ions produced by 70 eV electron impact ionization of helium nanodroplets doped with C_{60} and Yb. As described in the main text, the deflection profile was acquired by setting the mass spectrometer to the position of the $C_{60}Yb^+$ ion peak.

Although the figure shows that water, a ubiquitous contaminant in vacuum systems, is also present in the beam, the $C_{60}Yb^+$ peak is water-free. Furthermore, the relative intensity of the C_{60}-water complexes changes when the nanodroplet beam is collimated for passage through the deflecting field, implying that a significant fraction of the water is picked up by the nanodroplets in the course of their subsequent free flight toward the detector, where it cannot alter the deflection. Consequently, the measured dipole moment of the embedded C_{60}Yb system is not appreciably modified by water molecules.

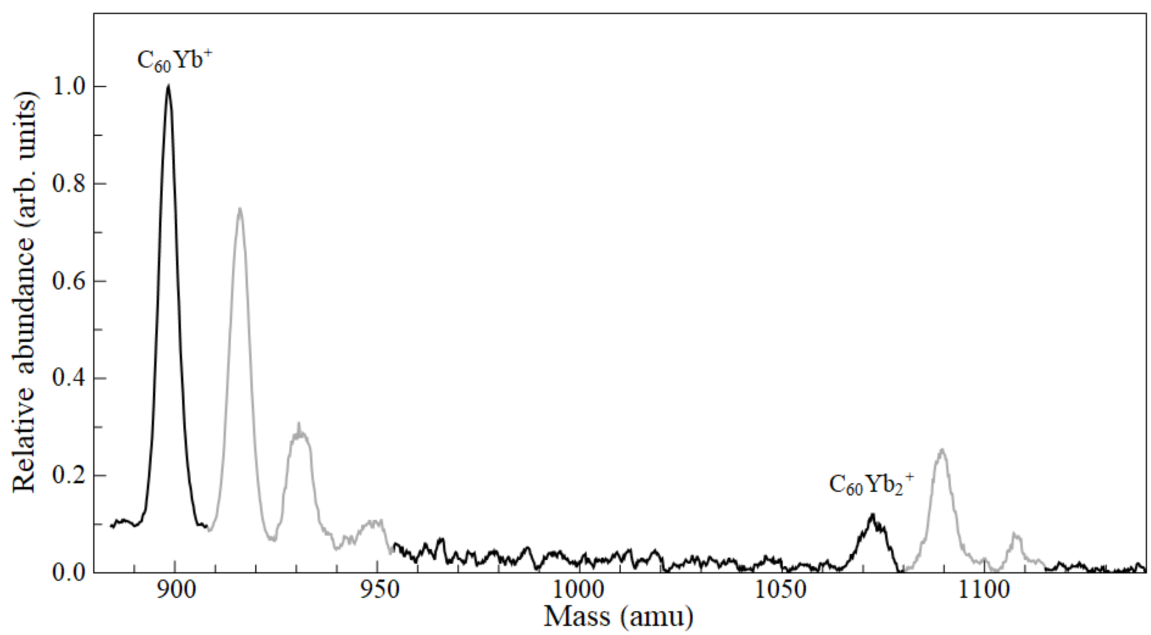

Figure S1. Mass spectra of $C_{60}Yb^+$ ions. The gray peaks are $C_{60}Yb_n$-water complexes. The isotopic composition of both C_{60} and Yb contributes to the peak widths.

II. $C_{60}Yb$ dipole and error estimation

As described in Section 2 of the main text, the dipole moment of the embedded polar complex is found from the best match of a Monte Carlo simulation of the deflection process to the measured deflection profile. Fig. S2 compares three simulated profiles with the data for nanodroplets containing the $C_{60}Yb$ complex. The deflection data were obtained by setting the mass spectrometer to the $C_{60}Yb^+$ peak shown in Fig. S1.

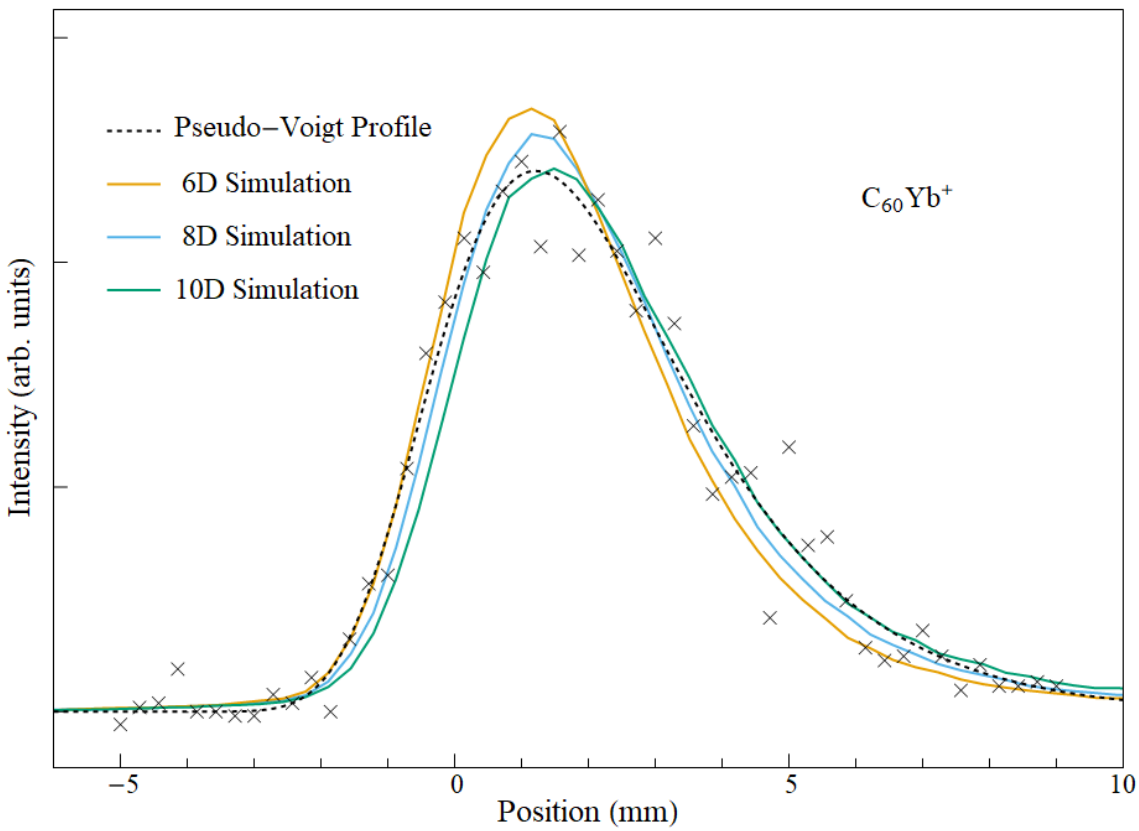

Figure S2. The crosses are beam deflection data points for nanodroplets containing $C_{60}Yb$, and the dashed line is a smoothing fit to these points using an asymmetric pseudo-Voigt function. The colored lines are the results of Monte Carlo simulations assuming three different values of the dipole moment of the complex and a dopant ionization probability parameter $\gamma = 0.12$.

An additional source of uncertainty, mentioned in the main text, is the efficiency of charge transfer from He^+ ions, produced within nanodroplets by electron bombardment in the mass spectrometer ionizer, to the dopant. As described in ref. 12 in the main text, this probability has the form $\exp(-\gamma N^{1/3})$, where N is the number of helium atoms in the nanodroplet and γ lies in the range of ≈0.06–0.12. This range translates into a corresponding uncertainty in the average nanodroplet size and, correspondingly, in the magnitude of the dipole moment. Fig. S.3 shows the quality of the simulation fit to the data for two values of the γ parameter. In both cases, the optimal fit is found for a dipole magnitude of ≈8 D. The uncertainty in this value, based on the analysis illustrated here, is estimated at ±1.5 D.

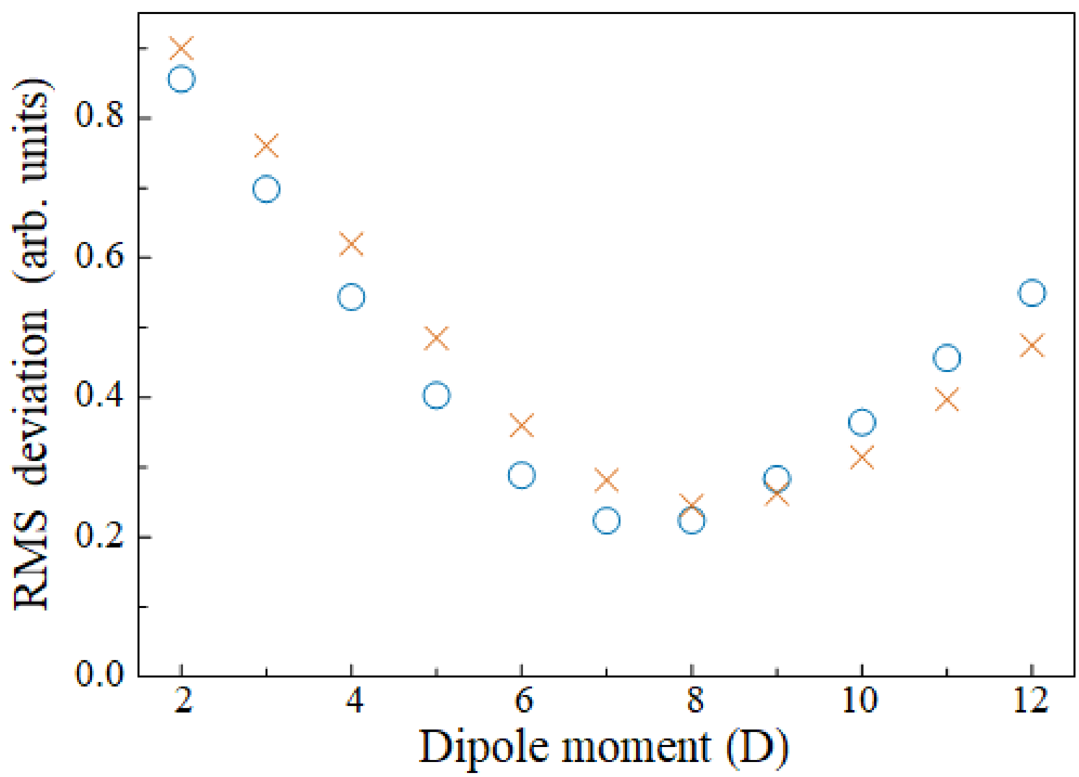

Figure S3. Root mean square deviation between the $C_{60}Yb$ profile data and the deflection simulation as a function of the assumed dipole moment of the complex. Crosses and circles correspond to dopant ionization probability parameters (described in the text) γ=0.06 and γ=0.12, respectively