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High-resolution state-selective study of transfer with excitation in the $F^{8+} + H_2$ system

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In a recent publication [M. Schulz *et al.*, Phys. Rev. Lett. **62**, 1738 (1989)] experimental evidence for a new process, two-electron transfer with excitation, was reported. In this paper we describe a higher-resolution version of the experiment, in which two-electron transfer with excitation was not observed.

In recent years transfer with excitation (TE), an ionatom collision process in which a projectile ion has an electron excited and additionally captures a target electron, has been studied extensively by several groups for a number of collision systems.¹ When the projectile electron is excited by the transferred target electron, the process is an energy resonant one, and can be likened to dielectronic recombination. In this case the process is referred to as RTE, or resonant transfer with excitation. Much of the interest in TE is due to this correlated twoelectron channel. The width and shape of the resonance reflect the target electron's Compton profile, or the projection of the electron's momentum distribution on the collision axis. When the projectile electron is excited by the target nucleus, the TE process is not energy resonant and is referred to as NTE. Under the impulse approximation the NTE cross section should look like

$$\sigma_{\rm NTE} = \int 2\pi P_{\rm cap}(b) P_{\rm ex}(b) b \, db \,, \tag{1}$$

where P_{ex} is the probability for the excitation of the projectile by the target nucleus, and P_{cap} is the probability for capture of the target electron. Therefore, qualitatively, the NTE cross section should initially rise with increasing projectile energies, following the trend exhibited by P_{ex} , and then drop rapidly as the projectile energy increases, following the trend exhibited by P_{cap} . This is, in fact, the observed NTE dependence on projectile energy.²

A third type of TE has been proposed³ in which the projectile electron is excited by one target electron, followed by the capture of a second target electron in the same collision. This process is known as 2e TE, and while not energy resonant, would be expected to exhibit a threshold effect, much the same as is seen in electron-impact excitation. The process is really just another form of NTE, except that P_{ex} now refers to electron-impact excitation, implying that the threshold would lie at higher projectile energies than for NTE.

Recently, some evidence for 2e TE was reported by Schulz et al.⁴ In their experiment they investigated the TE process by measuring KLL, KLM, KLN, and KLO Auger electron production as a function of projectile energy for the $F^{8+} + H_2$ collision system. The experiment was performed at low resolution since TE was the only possible mechanism for Auger electron production. (Double capture was found to be negligible and in any case would give rise to Auger electrons at very different energies from those measured.) Also in that experiment, the Auger electrons were detected at 9.6° in the laboratory frame.

The evidence for 2e TE was based on a small rise in the *KLL* cross section at approximately the expected threshold energy, as well as on the apparent shifting of the maximum in the TE cross section from the expected RTE resonance energy toward higher projectile energies for the *KLM*, *KLN*, and *KLO* cross sections.

Although electron-impact excitation has been shown to be a viable process in ion atom collisions,⁵ the magnitude of the reported 2e TE process nevertheless seemed surprisingly large since, as mentioned, the probability for electron capture falls very rapidly with increasing projectile energy. Indeed, in the case of S¹³⁺ + He, where NTE has been observed² to be non-negligible at low projectile energies, the NTE cross section dropped to nonobservable levels at projectile energies comparable to those at the calculated 2e TE thresholds.

A possible explanation for the reported 2e TE cross section is that, while the large RTE structure might be assumed to be coming exclusively from the $(2p^2)^1D$ line,⁶ 2e TE could arise via all of the available intermediate states. In order to test this hypothesis an experiment has been performed here, in which the individual *KLL* lines were resolved.

Our experimental setup has been described elsewhere.⁷ Briefly, a tandem, parallel-plate electron spectrometer was used to measure the TE cross section at 0° (laboratory frame) for the $F^{8+}+H_2$ collision system. Electrons were decelerated between the two stages of the spectrometer in order to achieve a resolution of 1.5-2.0 eV. This was sufficient to clearly resolve all the *KLL* lines with the exception of the pair $(2p^2)^{1}D$ and $(2s2p)^{1}P$, which are separated by approximately 1.7 eV.⁸ A 10-cm-long gas cell was used, and an MKS Baratron system maintained the gas pressure at a constant 20 mTorr throughout the duration of the experiment. The F^{8+} ions were provided by the J. R. Macdonald Laboratory's EN tandem Van de Graaff accelerator. Line assignments were based on the predictions of structure calculations.⁸

Figure 1 shows representative electron spectra taken at various projectile energies. The spectra have been transformed to the rest frame of the projectile. The predicted locations of the relevant lines are indicated. The smooth curves are best fits of the data to sums of Fano line shapes. Special care was taken to obtain data with good statistics, especially in the region of the expected 2e TE

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FIG. 1. Doubly differential projectile electron spectra for 18, 20, 22, 26, 28, and 30 MeV $F^{8+}+H_2$. The spectra have been transformed to the rest frame of the projectile. The lines are identified as indicated in the text.

threshold. As can be seen, there is virtually no contribution to TE from any line other than the $(2p^2)^1D$ and perhaps the $(2s2p)^1P$ which, as already stated, is not clearly resolvable. Even at the projectile energies near the expected threshold for 2e TE (approximately 29 MeV for F^{8+}), there is no indication of contribution from any but the 1D line.

In Fig. 2 the differential TE cross section is plotted as a function of projectile energy for the sum of the $(2p^2)^1D$ and $(2s2p)^1P$ lines. The error bars represent total relative error, consisting of statistical errors added in quadrature to estimates of uncertainties of fit, including background subtraction and the fits to the Fano line shapes. Not shown is the estimated 50% absolute uncertainty in the cross sections. The solid curve is the corresponding theoretical prediction⁹ for the differential RTE cross section at 0°, multiplied by 0.69.

The data of Schulz *et al.*,⁴ along with their statistical error bars, are also displayed in Fig. 2. They have been multiplied by 9 in order to facilitate comparisons with our data. One notes, however, that the data of Schulz *et al.* do not display the expected H₂ Compton profile, and the factor of 9 used here to provide a fit to our data was rather arbitrary. Recent theoretical⁹ and experimental¹⁰ work indicate that the differential RTE cross section should be



FIG. 2. Differential transfer with excitation (TE) cross sections, measured at 0° (solid circles), and multiplied by 4π to facilitate comparisons with other reported TE measurements. The solid curve is the theoretically predicted result, scaled to experiment as discussed in the text. The open circles connected by the dashed line represent the data of Schulz *et al.* (Ref. 4), scaled to the results of this work. See the text for an explanation of the error bars on both sets of data.

somewhat smaller at 9.6° than at 0° , however we have no explanation for the magnitude of the discrepancy found.

In a high-resolution experiment designed to shed some light on the origins of the unexpectedly large 2e TE cross sections, no evidence for the process was found in the KLL Auger spectra. This is contrary to the measurements of Schulz et al.,⁴ who observe "a second rather small maximum ... at 29 MeV," in σ_{KLL} . Furthermore, the highresolution measurement reported here is a more sensitive test for 2e TE, since, like NTE (but unlike RTE), one would expect contributions to all of the individual KLL lines rather than just the $(2p^2)^1D$ observed in this experiment.

It was also determined that the assumption⁴ that RTE could be characterized through examination of the *KLL* peak (without resolution sufficient to resolve the individual *KLL* Auger lines) was valid for the system of $F^{8+} + H_2$. This is a useful result since it facilitates other studies of this collision system such as the impact parameter or angular dependence of RTE, or the study of RTE with higher-Z, hydrogenlike ions.

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