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01 Apr 1989

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Michael Schulz Missouri University of Science and Technology, schulz@mst.edu

J. P. Giese

J. K. Swenson

Sheldon Datz

et. al. For a complete list of authors, see https://scholarsmine.mst.edu/phys\_facwork/1316

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## **Recommended Citation**

M. Schulz and J. P. Giese and J. K. Swenson and S. Datz and P. F. Dittner and H. F. Krause and H. Schone and C. R. Vane and M. Benhenni and S. M. Shafroth, "Electron-Electron Interactions in Transfer and Excitation in  $F^{8+} \rightarrow_2$  Collisions," *Physical Review Letters*, vol. 62, no. 15, pp. 1738-1741, American Physical Society (APS), Apr 1989.

The definitive version is available at https://doi.org/10.1103/PhysRevLett.62.1738

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## Electron-Electron Interactions in Transfer and Excitation in $F^{8+} \rightarrow H_2$ Collisions

M. Schulz, <sup>(a)</sup> J. P. Giese, J. K. Swenson, S. Datz, P. F. Dittner, H. F. Krause, H. Schöne, C. R. Vane,

M. Benhenni,<sup>(b)</sup> and S. M. Shafroth<sup>(b)</sup>

Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

(Received 19 September 1988)

We have measured projectile Auger electrons emitted after collisions of H-like F with  $H_2$ . The cross sections for emission of *KLL*, *KLM*, *KLN*, and *KLO* Auger electrons show maxima as a function of the projectile energy. One maximum in the *KLL* emission cross section is due to resonant transfer and excitation. A second maximum in the cross section for *KLL* emission as well as the maxima in the emission cross section for the higher-*n* Auger electrons are attributed to a new transfer and excitation process. This involves excitation of a projectile electron by one target electron accompanied by the capture of a second target electron.

PACS numbers: 34.50.Gb, 34.70.+e

Transfer and excitation (TE) processes have been studied intensively in the last several years.<sup>1-11</sup> In these processes, a target electron is captured by the projectile and, simultaneously, a projectile electron is excited. Originally, the interest was focused on resonant transfer and excitation (RTE). In RTE, the projectile electron is excited by an interaction with the captured target electron, which is initially weakly bound (quasifree). RTE is analogous to dielectronic recombination (DR), where a truly free electron recombines with the projectile via the excitation of projectile electron.

In ion-atom collisions, the presence of the target nucleus opens an additional reaction channel that can populate states indistinguishable from those populated by RTE.<sup>10</sup> This process, nonresonant transfer and excitation (NTE), in which the excitation of the projectile electron by the target nucleus is independent of the capture process, was found to be the dominant TE process at low projectile energies.<sup>11</sup>

Recently, TE processes were studied for Li-like F colliding with  $H_2$ .<sup>4</sup> In the measured cross section, two maxima were observed in the projectile energy dependence. One maximum was attributed to RTE. It was not clear, however, that the second maximum was also due to RTE. Hahn and McLaughlin<sup>12</sup> proposed that this second maximum is due to a new TE process, which was designated two electron transfer and excitation (2*e* TE).<sup>4</sup> In the 2*e* TE process, a collision with a target electron excites the projectile electron and, again as in NTE, a second independent electron is captured into the projectile. For this process, however, the threshold lies at the projectile electron excitation energy, i.e., at the *upper* edge of RTE processes.

In this Letter, we present evidence for 2e TE which was obtained by measuring projectile Auger electron spectra for H-like F colliding with H<sub>2</sub>. It is shown that at high projectile energies, as well as for high-*n* states, 2eTE is the dominant TE process.

H-like projectiles were chosen because of some impor-

tant advantages over the Li-like ions often used in the past<sup>1,3-5</sup>: (i) The interaction of the electrons, which form the doubly excited states with the spectator electrons, introduces in Li-like ions a much larger multiplicity of states than in H-like ions (Li- or H-like refers to the initial charge state). This makes Li-like ions a rather complex system for theoretical interpretation. (ii) For H-like projectiles, both electrons involved in an Auger process are necessarily those which populate the doubly excited states. For Li-like ions, this is not always the case since an electron that is only a spectator in the formation of the doubly excited state can be involved in the Auger decay. Therefore, for H-like ions the Auger electrons can be directly related to the population of a specific doubly excited state. (iii) For H-like ions, an Auger electron can only be emitted after the formation of a doubly excited state via a transfer and excitation process or via double capture. Since double capture should be negligible for the H<sub>2</sub> target used here, every projectile Auger electron is a signature of a TE process. Therefore, the very good energy resolution required for Li-like ions is not necessary. This, in turn, makes it possible to measure a broad range of electron energies, i.e., a whole Rydberg series, simultaneously.

The experiment was performed at the EN Tandem of Oak Ridge National Laboratory. H-like F beams at energies between 17 and 33 MeV passed through a differentially-pumped H<sub>2</sub> gas target. The target pressure was 15 mTorr and the effective length of the gas cell was 12.5 mm. The electrons produced here were decelerated by a factor of 2 by a high voltage applied between the first and second pumping stage of the gas cell. Electrons emitted at an angle of 9.6° with respect to the beam axis were energy analyzed by a two-stage 30° electron spectrometer. The electrons were then detected by a position-sensitive microchannel plate detector (MCP). The electron spectrometer and the MCP are described in detail elsewhere.<sup>13</sup> The ion beam was collected in a Faraday cup for normalization.

In Fig. 1, we show electron spectra taken for the  $F^{8+}$ - $H_2$  system at collision energies of 21, 25, and 31 MeV. In these spectra, the monotonic background, which arises mainly from direct ionization of the H<sub>2</sub> target, was subtracted and the electron energies are given in the projectile frame. It should be noted that the Li-like Auger lines representing double capture are not observed using the  $H_2$  target, although they were clearly observed when a Ne target was used. The He-like KLL, KLM, KLN, and KLO groups, labeled 1-4, respectively, are observable in Fig. 1. The n distribution of the observed Auger electrons is quite sensitive to the projectile energy. With increasing collision energy, the n distribution shifts to higher n's. At 31 MeV, a broadband can be seen which extends up to the KLn series limit (827 eV) and which is therefore attributed to high-n KLn Auger electrons. The KLO group (4) can barely be resolved from that distribution. In Fig. 2, the F Auger electron production cross sections  $\sigma_{KLn}$  (n = L, M, N, O) for KLL, KLM, KLN, and KLO transitions are plotted versus the projectile energy. The error bars are statistical (including background subtraction) errors only. The systematic errors are of the order of 50%. In  $\sigma_{KLL}$ , a pronounced maximum can be

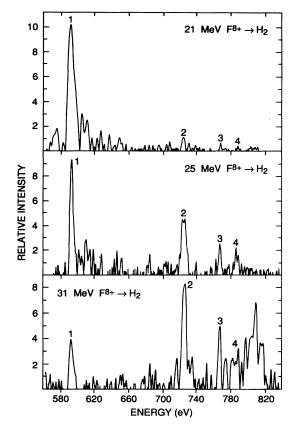


FIG. 1. Electron spectra for 21, 25, and 31 MeV  $F^{8+} \rightarrow H_2$ . The energy scale is in the rest frame of the projectile. The labeled Auger groups are as follows: 1, *KLL*; 2, *KLM*; 3, *KLN*; and 4, *KLO*.

seen around 21 MeV. A second rather small maximum is observable at 29 MeV. For all the other transitions (*KLM*, *KLN*, and *KLO*), the cross sections have a maximum at the same projectile energy of 29 MeV.

The arrows in Fig. 2 indicate the RTE resonance energy for the corresponding Auger transition. Additionally, the *KLn* RTE Auger series limit is shown ( $KL \infty$ ). The position of the first maximum in  $\sigma_{KLL}$  is in very good agreement with the *KLL* RTE resonance energy. Also the shape of the maximum is in accordance with the expected resonance shape of RTE.

For all the other transitions the positions of the maxima of the cross sections do not agree with the resonance energy of the corresponding state. The maxima for these transitions and the second maximum in  $\sigma_{KLL}$  are all at the series limit for KLn RTE Auger resonances or at even slightly higher energies. This means that the main contribution to these transitions is *not* due to a population of the corresponding doubly excited states by RTE and a subsequent direct Auger transition to the ground state. In principle, there is the possibility that a doubly excited state with one of the electrons in a very-high-*n* state was populated by RTE which then decayed radia-

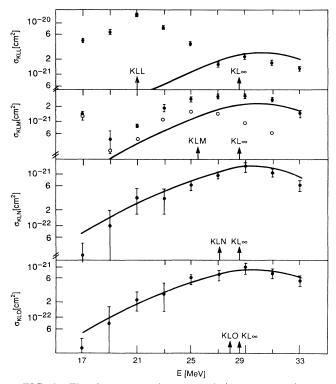


FIG. 2. Fluorine Auger electron emission cross sections vs projectile energy for *KLL*, *KLM*, *KLN*, and *KLO* transitions. The solid curves are estimated 2*e* TE cross sections based on Eq. (1). The open circles in  $\sigma_{KLM}$  are the differences between the measured cross sections and the estimated 2*e* TE cross sections. The arrows indicate the RTE resonance energies of the corresponding doubly excited states.

tively to a lower-lying doubly excited state. The decay of this latter state could then lead to a *KLL*, *KLM*, *KLN*, or *KLO* Auger electron. Indeed, a maximum would be expected near the *KLn* series limit for such processes. However, this is an upper limit and if these high-*n* states were populated, lower-*n* states should certainly also contribute. Thus, the sum of all the resonances should lead to a maximum at energies lower than the *KLn* series limit energy (*KL* $\infty$ ) rather than at higher energies as observed in the data. Furthermore, the radiative decay rate to a lower doubly excited state is much smaller than the rate for the direct Auger transition to the ground state, so that the branching ratio should favor the direct Auger transition to the ground state.

We also rule out the possibility that the maxima at 29 MeV are due to higher RTE series resonances. If, for example, a KMM state was populated by RTE, there could in principle be a radiative transition to the L shell followed by a KLM Auger transition. However, the radiative cascade transition should again be very slow compared to the direct Auger transition.

Finally, the maxima at 29 MeV cannot be explained by NTE. It is known that the cross sections for this process have a maximum at much lower projectile energies than the *KLL* RTE resonance.<sup>4,10,11,14</sup> Therefore, its contribution cannot be larger than it is at the lowest projectile energy (17 MeV) and can certainly not lead to a maximum at energies as high as 29 MeV.

We propose that the process of 2e TE is contributing to the Auger emission cross sections. If the binding of the target electron that excites the projectile electron in 2e TE is neglected, then 2e TE should have a threshold energy which is equal to the excitation energy in the projectile's rest frame. For a K to L excitation, this threshold energy is in the electron's rest frame identical with the projectile energy for the KLn RTE series limit (KL $\infty$ ) at 29 MeV. Because the capture cross section decreases strongly with increasing projectile energy, the 2e TE cross section should have a maximum slightly above the threshold energy. This is in agreement with the measured KLM, KLN, and KLO cross sections and the second maximum in  $\sigma_{KLL}$ .

We can estimate the projectile energy dependence of 2e TE. If the transfer and excitation processes in 2e TE are considered as completely uncorrelated, then

$$\sigma_{2e\,\mathrm{TE}} = \int 2\pi P_{\mathrm{cap}}(b) P_{\mathrm{ex}}(b) b \, db \,, \tag{1}$$

where b is the impact parameter and  $P_{\rm cap}$  and  $P_{\rm ex}$  are the capture probabilities and the excitation probabilities by a bound electron. This formulation is equivalent to that used for NTE cross-section calculations,<sup>15</sup> except that for NTE  $P_{\rm ex}$  would be the excitation probability by the nucleus rather than that for a bound electron. For the capture process, we assume that the projectile energy dependence and the *n* distribution of  $P_{\rm cap}(b)$  is not very sensitive to the impact parameter. We, therefore, calculations

late  $P_{cap}$  at zero impact parameter with the Oppenheimer-Brinkman-Kramers approximation for capture to the L, M, N, and O shell.  $P_{cap}$  can then be taken out of the integral. The integral is now just the excitation cross section  $\sigma_{ex}$ . We estimate  $\sigma_{ex}$  within the impulse approximation, i.e., we use excitation cross sections by free electrons  $\sigma'_{ex}$  and fold them with the momentum distribution of the target electron. We only consider K to L excitation since the energies studied here are below the K to Mexcitation threshold and even above this threshold the cross sections for K to M excitation is smaller than K to L excitation. The absolute magnitude of  $\sigma_{2e \text{ TE}}$  we obtain by fitting  $\sigma_{2e \text{ TE}}$  (with  $\sigma'_{ex}$  as the only free parameter) with the measured cross section for KLN Auger emission at 29 MeV. Above the threshold  $\sigma'_{ex}$  should drop very slowly with increasing energy. Here, we used the  $\sigma'_{ex}$  calculated by Bhatia and Temkin<sup>16</sup> for He-like F normalized to the fitted cross section at the threshold. For all the other projectile energies and transitions,  $\sigma_{2e \text{ TE}}$  is calculated with the same  $\sigma'_{ex}$ .

The estimated relative 2*e* TE cross sections are shown in Fig. 2 as solid curves. The agreement with  $\sigma_{KLN}$  and  $\sigma_{KLO}$  is very good. For the *KLM* transitions,  $\sigma_{2e \text{ TE}}$  is systematically lower than the measured cross sections. Only at high projectile energies does  $\sigma_{2e \text{ TE}}$  approach  $\sigma_{KLM}$ . For the *KLL* transitions, the agreement with the estimated 2*e* TE cross sections is reasonable at energies above 27 MeV. At lower energies, the contributions from RTE Auger to  $\sigma_{KLL}$  dominate.

Relatively strong contributions from RTE Auger might also be expected in  $\sigma_{KLM}$ . If the estimated 2e TE cross sections are subtracted from the data, yielding the open circles in Fig. 2, one indeed obtains a shape that is consistent with the shape of RTE resonances. There is a maximum in these points at about 25 MeV, which is in very good agreement with the *KLM* RTE resonance energy of 25.5 MeV. The height of the maximum is about 1 order of magnitude lower than the first maximum in  $\sigma_{KLL}$ . This may be taken as an indication that the *KLn* RTE Auger cross sections are sharply decreasing with increasing *n*. This is also consistent with  $\sigma_{KLN}$  and  $\sigma_{KLO}$ where no indication of contributions from RTE Auger can be discerned.

It should be emphasized, however, that we can only describe the relative magnitude of  $\sigma_{2e \text{ TE}}$  for the different *n*'s. As far as the absolute magnitude is concerned, one might argue that the cross sections for NTE and 2*e* TE should be comparable at identical velocities. The two processes should not differ for an independent transfer process. As far as the excitation is concerned, one would *expect* the cross section for proton impact to be comparable to the one for free-electron impact. However, it has recently been shown that the cross section for excitation by a bound electron is larger than that for a free electron or a proton. In a calculation by Thumm, Briggs, and Schöller, <sup>17</sup> it was found that the excitation of F<sup>7+</sup> colliding with He was clearly dominated by the

interaction with the He electrons rather than with the He nucleus. Since nuclear excitation is  $\alpha Z^2$ , the dominance of the electronic excitation should even be stronger for the H<sub>2</sub> target used in our experiment. Furthermore, the excitation of  $F^{6+}$  projectiles to the  $(1s 2s 2p)^4 P$  state in collisions with H<sub>2</sub> and He was studied experimentally by Zouros et al.<sup>18</sup> In that work, it was found that near threshold the electronic excitation clearly dominates the nuclear excitation for the H<sub>2</sub> target, whereas for the He target the electronic and nuclear excitation are of comparable magnitude. It should be noted that 2e TE is a much more complex process than excitation by a single particle. The impact-parameter dependence for excitation accompanied by capture is not necessarily the same for a collision with the proton in the H<sub>2</sub> molecule as for a collision with one of the bound electrons, because the bound electron has a different spatial distribution. In Eq. (1), the contribution of  $d\sigma_{cap}(b) = P_{cap}(b)b db$  is identical for both NTE and 2e TE.  $d\sigma_{cap}$  is known to have a maximum at relatively large impact parameters. The main contributions to the total cross section for the process studied here come from the impact-parameter range where this maximum occurs. The relative cross section between NTE and 2e TE are then given by the relative excitation probabilities  $P_{ex}^N$  and  $P_{ex}^e$  for proton and bound electron impact in that impact-parameter range. Because of the spatial distribution of the electrons around the target nucleus, one should expect  $P_{ex}^{e}$  to extend to larger impact parameters than  $P_{ex}^N$  and therefore be larger at impact parameters where  $d\sigma_{cap}$  has a maximum. Therefore, the dominance of 2e TE over NTE near threshold, which is expected from the excitation cross sections from Ref. 17, should be further enhanced by the selection of larger impact parameters due to the capture.

The present data provide strong evidence for 2e TE. This process is the dominant TE process for populating high-*n* states and for all *n* states at projectile energies above the threshold energy. We expect this to be the case in general for collision systems typically used to study TE processes (i.e., light targets). Recently, possible interferences between the indistinguishable processes of NTE and RTE were discussed.<sup>8</sup> The present data show that 2e TE needs to be considered in the discussion of such interference effects. This research was sponsored in part by the U.S. DOE, Office of Basic Energy Sciences, Division of Chemical Sciences under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc. The research of J.P.G. and J.K.S. was supported in part by a program administered by Oak Ridge Associated Universities.

<sup>(a)</sup>Visitor at the Joint Institute for Heavy Ion Research, ORNL, Oak Ridge, TN 37831.

<sup>(b)</sup>Permanent address: University of North Carolina, Department of Physics, Chapel Hill, NC 17514.

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