Direct observation of the double Auger decay of a *K* hole

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The double Auger (DA) decay of a *K* hole has been observed directly by detecting the two emitted electrons in coincidence. The hole was created in ^{37}Cl following the electron-capture decay of ^{37}Ar. The probability of DA decay, per Auger decay, with the two electrons both having an energy greater than 250 eV was found to be $3.7\pm0.2\%$. The DA probability was found to decrease exponentially as the energy partitioning between the two electrons changed from the asymmetric case ($E_1 \gg E_2$) to the symmetric case ($E_1 \approx E_2$). The DA probability accounts for the bulk of the intensity of high charge states previously measured in ^{37}Cl. [S1050-2947(96)50506-8]

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An inner-shell vacancy in a light atom is usually filled by an Auger transition—a nonradiative process in which one higher electron fills the vacancy and another is ejected with an energy equal to the transition energy. In a certain fraction of the Auger decays, however, another electron is shaken off, with the *two* electrons sharing the transition energy. This process is known as double Auger (DA) decay. In the independent (i.e., single) particle model of the atom DA decay is forbidden; its occurrence, therefore, is due to many-body correlations in the wave function, and the study of the process provides means for testing theoretical models that incorporate such correlations. Although evidence for the process was first reported 30 years ago [1], most measurements to date, like the first one, deduce the probability of DA decay only indirectly, from the charge distribution of photoionized atoms [2–6]. There are very few measurements of the energy distribution of the ejected electrons [7] and no coincidence measurements of the two emitted electrons nor of the angular correlation between them.

Mirroring the experimental effort, the various theoretical calculations also concentrate on predicting the final charge-state distribution left by the vacancy [8–12] except for the calculation by Amusia *et al.* [13], who give the energy distribution and the angular correlation between the two emitted electrons for the $1s^{-1} \rightarrow 2s^{-2}2p^{-1}$ transition in Ne. Because the final charge-state distribution produced in filling a deep vacancy is the result of a cascade that increases in complexity as the initial hole multiplies and propagates to higher shells, extracting accurate values for the probability of DA decay from this distribution is difficult. It seems preferable to test the theoretical calculations directly, by measuring the probability of the DA decay of a specific hole and the energy distribution of the two emitted electrons. In this Rapid Communication we report such a measurement for the DA decay of the *K* hole in ^{37}Cl produced following the electron capture (EC) decay of ^{37}Ar. The nucleus ^{37}Ar decays (with a half-life of 35.0 days) by capturing a 1*s* electron 90.2% of the time, a 2*s* electron 8.7% of the time, and a 3*s* electron the remaining 1.1% of the time [14]. Thus the dominant EC decay channel leaves the daughter ^{37}Cl atom in a *neutral* charge state, and with the electrons having the configuration of a 1*s* hole in Ar.

The apparatus used for this experiment is part of a setup for an ongoing study of the recoil spectra of ^{37}Cl ions following the EC decay of ^{37}Ar [15]. For the work described here, the ^{37}Ar was produced via the ^{40}Ca(n,α)^{37}Ar reaction by irradiating 0.6 g of Ca metal (99.99% elemental purity) for one week at Brookhaven National Laboratory's High Flux Beam Reactor. The resulting activity of the ^{37}Ar was about 40 mCi. The ^{37}Ar was released from the Ca metal by heating the sample in vacuum to a temperature of 950 °C. Non-inert contaminants were removed from the released gas using a Ti sublimation pump, and the residual gas captured by a molecular sieve at liquid-nitrogen temperature. A γ-ray spectrum of the adsorbed gas showed an internal bremsstrahlung spectrum with an end-point energy of 814 keV (the *Q* value of the ^{37}Ar decay [14]); no other γ-ray lines were observed above the normal room background. The Ar gas was released by warming the molecular sieve to a temperature of ≈ -120 °C and then leaked via a leak valve into an ultra-high-vacuum (UHV) chamber with a base pressure of 2×10^{-10} torr. A quadrupole mass spectrometer in the UHV system showed only masses 2 (H_2), 4 (He), and 37 (^{37}Ar) above background levels. The ^{37}Ar gas was adsorbed on a graphite substrate that was cooled to 16 K and collimated by a 5-mm-diam aperture just in front of it. The activity of the visible area of the sample was 2.0 MBq (54 μCi), corresponding to a coverage of 0.016 monolayer.

Two microchannel plate (MCP) detectors, each with a diameter of 2.5 cm, were placed at $-30°$ and 30° from the normal to the substrate, at a distance of 6.5 cm from it. Each detector had three screens, each with 90% transmission, in front of it. The first and last screens were grounded; a negative bias *V* applied to the middle screen restricted the MCP to detecting electrons with energy $E \geq eV/\sin^2\theta$, where θ is the angle of incidence of the electron on the screen ($\theta = 90°$ being normal to the screen). A time-to-amplitude converter (TAC) was started on a signal from one of the detectors and stopped on a cable-delayed signal from the other detector. A PC-based data acquisition system recorded the TAC spectrum for coincidence events as well as the singles counts in each detector.

Figure 1 shows the count rate in one of the detectors as a function of a (negative) screen retarding voltage between 50

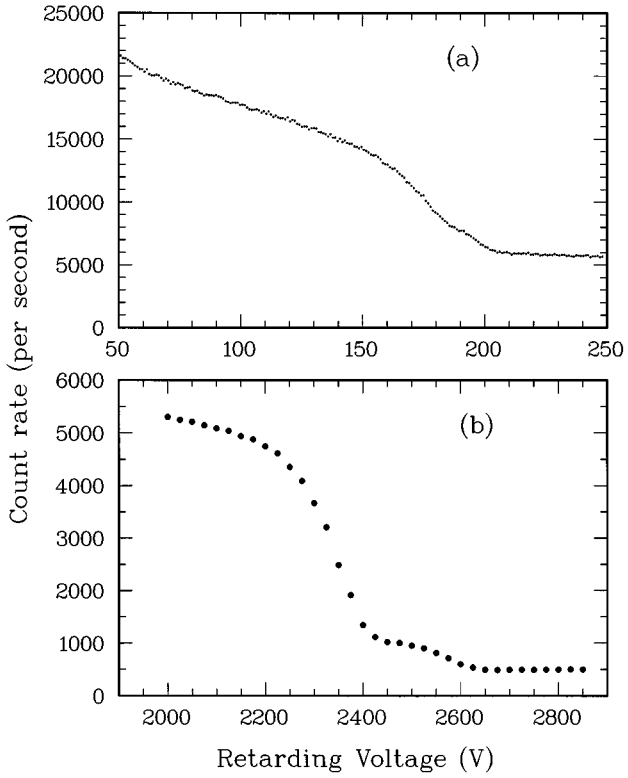

FIG. 1. Retarding field spectra of Auger electrons emitted following the EC decay of ^{37}Ar in the region of (a) L Augers and (b) K Augers.

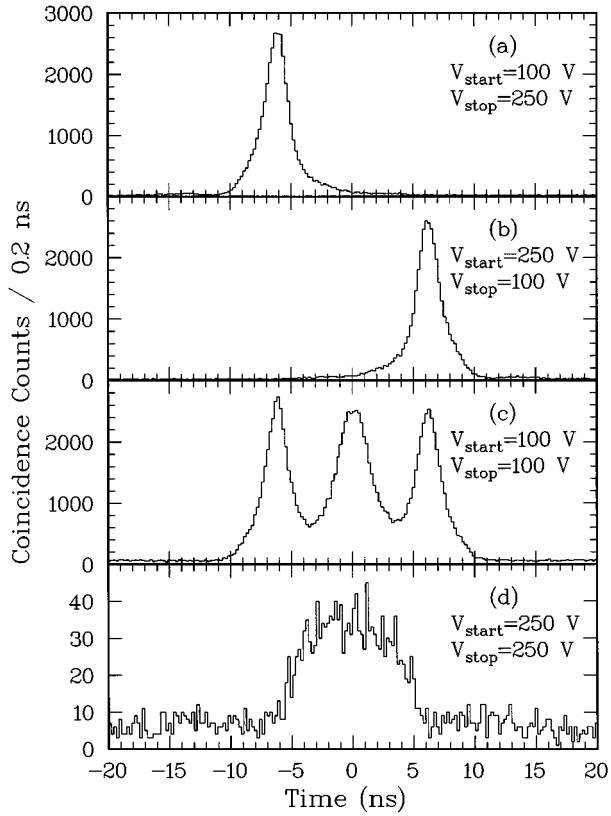

FIG. 2. Time coincidence spectra between two MCP detectors with screen biases of (a) $V_{\text{start}}=100$ V, $V_{\text{stop}}=250$ V, (b) $V_{\text{start}}=250$ V, $V_{\text{stop}}=100$ V, (c) $V_{\text{start}}=100$ V, $V_{\text{stop}}=100$ V, and (d) $V_{\text{start}}=250$ V, $V_{\text{stop}}=250$ V. The origin of each of the peaks is explained in the text.

and 250 V [Fig. 1(a)] and between 2000 and 2900 V [Fig. 1(b)]. As expected, the count rate drops when the retarding voltage increases past 200 V, thus preventing LMM Auger electrons ($E\approx 175$ eV) from entering the detector [16], drops again when the KLL Augers are retarded (at 2400 V), and finally drops to background levels when the KLM Augers are retarded (at 2600 V).

Figure 2(a) shows the time (TAC) spectrum for coincidences between a start detector with a retarding bias of 100 V and a stop detector with a retarding bias of 250 V. The peak corresponds to coincidences between the (slow) LMM electrons detected in the start MCP and the (fast) KLL or KLM electrons detected in the stop MCP. The spectrum was collected over a period of 1000 s. Figure 2(b) shows a coincidence spectrum with the biases on the two MCPs swapped, and now, as expected, the peak is shifted to later times because the fast K electron is detected in the start MCP and the slow L electron is detected in the stop MCP. When both detectors have retarding voltages of 100 V (thus allowing each to detect both K and L Auger electrons) one observes [Fig. 2(c)] both the early (L_{start}-K_{stop}) peak and the late (K_{start}-L_{stop}) peak, as well as a ''prompt'' peak due to coincidences between the two LMM Auger electrons that follow the KLL transition.

Figure 2(d) shows a coincidence spectrum, again collected for 1000 s, with the retarding voltage on both detectors set at 250 V, thus preventing LMM Auger electrons from entering either detector. Electron-electron coincidences would not be expected from a normal Auger decay for these retarding voltages. For the DA decay of the K hole, however, the two emitted electrons share the total available energy of about 2200 eV (for a $KLLL$ transition), and it is possible for the two electrons to have more than 250 eV each. The time spread reflected in the spectrum shown in Fig. 2(d) is consistent with that expected from two electrons sharing 2200 eV of energy; the shortest time corresponds to starting on a 250-eV electron and stopping on a $\approx$1950-eV electron and the longest time to starting on a $\approx$1950-eV electron and stopping on a 250-eV electron. [The time resolution is not sufficient to resolve $KLLL$ transitions from $KLLM$ transitions; i.e., the time spread observed in Fig. 2(d) is also consistent with coincidences between the 250-eV and 2150-eV electrons, which could be emitted in a $KLLM$ transition.] The flat background in Fig. 2(d) is due to accidental coincidences between two K Augers.

Aside from DA decay, the following processes could also result in coincidences between the two detectors: (1) Double K-shell ionization during electron capture [17]. This process results in two vacancies in the K shell, leading to coincidences between the two K Auger electrons that fill the double vacancy, as well as between the ejected electron and one of the K Auger electrons. The probability of double K-shell ionization during the electron capture decay of ^{37}Ar is $(3.7\pm 0.9)\times 10^{-4}$ per K capture [18]. (2) Coincidences between a K Auger electron and an x ray or a UV photon from a subsequent atomic transition. (3) Coincidences between a fast secondary electron generated in the

graphite substrate by a K Auger electron and the backscattered primary electron. The contribution of processes (1) and (2) was deduced from the number of coincidences observed with a retarding voltage of 1500 V on each detector. At these retarding voltages there should be no coincidences from DA electrons, since the maximum energy available to the two DA electrons is 2800 eV. The rate of coincidences that was measured with both retarding voltages set at 1500 V was of the same order as that expected from double K-shell ionization. From this rate we deduce that coincidences due to processes (1) and (2) constitute only 2% of the coincidences observed at a retarding voltage of 250 V [Fig. 2(d)]. To estimate the rate of coincidences due to process (3) we performed a Monte Carlo simulation [19,20] using the methods of Ref. [19] and the Gryzinsky cross section [21] for fast secondary electron production from graphite. The Monte Carlo calculation gave a rate of coincidence due to secondary electron production that was 6% of the measured coincidence rate. The Gryzinsky model gives the largest secondary production cross section [19] and hence the figure of 6% is probably an overestimate.

The probability of DA decay was obtained from the ratio of coincidence counts obtained at retarding voltages of $(V_{\text{start}}, V_{\text{stop}}) = (250,250)$ V (after subtracting the contributions of the processes discussed above) to the coincidence counts at retarding voltages of (100,250) V. Taking the ratio of coincidence rates removes the dependence on solid angles, screen transmissions, detector efficiencies, and electronic thresholds. The small contribution of DA coincidences to the (100,250) V run was taken into account by assuming that the shape of the energy distribution of the DA electron below 250 eV is the same as that given by Amusia *et al.* (Fig. 5 in Ref. [13]) and that the ratio of $KLLL$ to $KLLM$ to $KLMM$ transitions is the same as that for KLL to KLM to KMM, namely 84.3:15.1:0.6. With these assumptions the probability of DA decay (per Auger transition of the K hole) with the two electrons having energies greater than 250 eV was found to be $(3.65 \pm 0.13)\%$, where the indicated error is statistical only. Allowing the shape of the energy distribution below 250 eV to vary by $\pm 40\%$ from that given by Amusia *et al.* gives a variation of $\pm 5\%$ in the probability of DA decay with the two electrons having energies greater than 250 eV. Taking this variation into account, we adopt $(3.7 \pm 0.2)\%$ for this probability.

The dependence of the DA decay probability on the energy distribution of the two emitted electrons was measured by collecting coincidence spectra similar to that shown in Fig. 2(d), but with the retarding voltages set, sequentially, at (300,300) V, (350,350) V, . . . , (950,950) V. Each run was for 1000 s, except for the (950,950) V run, which was collected for 4.2 h. We monitored the strength of the source and the possible attenuation of electrons due to a buildup of overlayers of residual gas adsorbed on top of the ^{37}Ar atoms by conducting short (100-s) coincidence runs at the same voltages shown in Figs. 2(a)–2(c) at regular intervals. The results of these short runs were all consistent and showed a negligible decrease in the ratio of L to K Augers. The contribution of coincidences due to non-DA processes was accounted for as discussed above. The resulting DA probability is plotted versus the retarding voltage in Fig. 3.

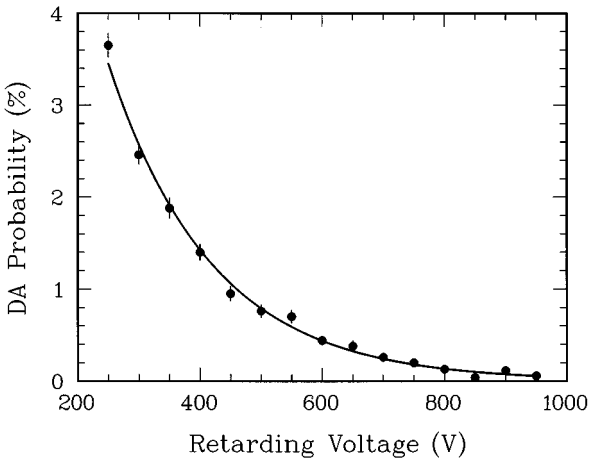

FIG. 3. Double Auger probability as a function of the (equal) retarding voltage on the two screens. The solid line is an empirical fit using an exponential dependence on the retarding voltage.

The solid line in Fig. 3 shows an empirical fit to the probability of DA decay $P_{\text{DA}}(V)$ as a function of the retarding voltage V using the expression $P_{\text{DA}}(V) = P(0)\exp(-V/a)$ with $P(0) = (15.1 \pm 0.8)\%$ and $a = 170 \pm 4$ V. This simple expression seems to give a reasonable representation of the data and indicates that the energy distribution of the electrons emitted in the DA decay (which is proportional to the *derivative* of the data of Fig. 3 with respect to the retarding voltage V) favors a very asymmetric sharing of the transition energy between the two electrons. This result is in qualitative agreement with the calculation of Amusia *et al.* [13] for Ne. For a quantitative comparison of our results with theory, not only is the energy distribution for Cl (having the electronic configuration of Ar with a K hole) required, but also the distribution of the final three holes and the angular distribution of the DA electrons for each energy partitioning and for each final state.

We now explore the implications of our measurement for the charge-state distribution of ^{37}Cl following the EC decay of ^{37}Ar. This distribution was measured by Snell and Pleasonton [22] in 1955; they found $6.2 \pm 0.1\%$, $15.7 \pm 0.4\%$, $39.2 \pm 0.2\%$, $26.7 \pm 0.4\%$, $10.0 \pm 0.2\%$, $1.8 \pm 0.1\%$, and $0.4 \pm 0.1\%$, respectively, for the relative abundance of charge states 1–7. Within experimental error, this distribution is essentially the same as that of the K hole produced by photoionization of Ar ($1.8 \pm 1\%$, $11.7 \pm 2\%$, $10.9 \pm 2\%$, $40.4 \pm 4\%$, 24.7 ± 3, and $10.5 \pm 2\%$, for charge states 1–6 [4]), after taking into account the lower fluorescence yield in Cl and the contribution of L_1 primary vacancies produced via $2s$ capture, and noting that the initial charge state produced by photoionization is $+1$, while that produced in EC is neutral. It is difficult to account for the intensity of charge states 5 (10.0%) and 6 (1.8%) in Cl without invoking the DA decay of the K hole. A calculation for normal transitions (using the intensity ratios of the main K Auger lines in Ar of Asplund *et al.* [24] and the branching ratios of Krause [23] for the L transitions) gives only 2.1% for charge state 5 and 0% for charge state 6. Shake-off accompanying the L transitions ($\approx 10\%$ per transition) is not sufficient to explain these charge states either. Indeed, the coincidence measure-

ments of Levin *et al.* [3] show no charge state 6 (5 in Cl) in coincidence with the dominant $KL_{2,3}L_{2,3}$ transition and very little with the weaker $KL_1L_{2,3}$ transition. On the other hand, the DA decay of the *K* hole in Cl will lead primarily to charge states 5 and 6. If we assume that the shape of the energy distribution of the DA electrons below 250 eV is given by the calculation of Amusia *et al.* for Ne, our measured value of 3.7% at (250,250) V extrapolates to 11% for the total DA probability (per *K* Auger transition); if we further assume that the intensities of the DA transitions are roughly 70% for $KL_{2,3}L_{2,3}L_{2,3}$, 15% for $KL_1L_{2,3}L_{2,3}$, 12% for $KL_{2,3}L_{2,3}M$, and 3% for $KL_1L_{2,3}M$, then the intensity of charge state 5 (per EC) is 8.4% and that of charge state 6 is 1.3%. If the energy distribution of the DA electrons extrapolates (below 250 eV) according to the simple exponential form which describes the shape above 250 eV (Fig. 3), the total DA probability would be 15% and the intensity of charge state 5 would be 10.7% and that of charge state 6 would be 1.8%, in good agreement with the measured values. These approximate calculations, which ignore the effect of shake-off accompanying *L* Auger transitions, are only intended to demonstrate that the DA probability which we measure implies that the bulk of the intensity of charge states 5 and 6 observed in Cl is due to the extra *L* hole produced in the DA decay of the *K* hole.

Finally, we would like to point out some of the advantages that the EC decay of adsorbed radioactive atoms offers for the study of DA decay. (1) EC decay produces an almost ''pure'' inner-shell vacancy, thus overcoming problems usually encountered when using external sources, namely the need to resolve charges and electrons produced during the primary vacancy production from those produced by the DA process itself. (2) EC decay offers the possibility of studying the DA decay from the same shell as a function of increasing nuclear charge. Such studies cannot be easily done with current synchrotron light sources, since these do not provide light of sufficient energy and/or intensity to ionize the *K* shell of high *Z* atoms, such as xenon. (3) The localization of the source in a small area allows the study of angular correlations between the emitted electrons. Since the angular correlation depends on the particular final state of the atom, an energy-selective detector (such as a cylindrical mirror analyzer) would be required for each electron to pin down the final state.

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