Observation of the Mn $K\alpha\alpha$ x rays in the electron-capture decay of ^{55}Fe

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$K\alpha\alpha$ x rays have been observed for the first time in the electron-capture decay of ^{55}Fe. These x rays are due to the two-electron–one-photon transition to the K shell doubly ionized through K-shell shakeoff and shakeup during the K-electron capture decay. The measured $K\alpha\alpha$ x-ray energy is 11 907 ± 20 eV and the intensity ratio of $K\alpha\alpha$ x rays to $K\alpha$ x rays is $(5.5 \pm 1.2) \times 10^{-8}$. The results are discussed and compared with theoretical calculations and other measurements.

I. INTRODUCTION

In collisions of heavy ions with heavy target atoms, highly excited atoms with several inner-shell vacancies can be produced through the excitation mechanisms of molecular level crossings and Coulomb excitation.[1] These processes can create two vacancies in the K shell of one atom. The double K vacancies are usually filled by the independent transitions of two electrons, accompanied by the emission of two photons or Auger electrons. An alternative deexcitation mode of the double K-vacancy state is the correlated jump of two electrons into the empty K shell, with only one photon emitted that carries the total transition energy, approximately equal to twice the $K\alpha$ x-ray energy. Wölfli *et al.* first observed x rays due to the two-electron–one-photon transition in Al-Al, O-Ca, Ca-Ca, Fe-Fe, Fe-Ni, and Ni-Ni collisions.[2] The transitions to the empty K shell have also been investigated for other atomic systems.[3-7]

The double K-vacancy states produced in the ion-atom collisions are not pure; L, M, and other outer-shell vacancies are also created by multiple ionizations with a high probability. Since it is difficult to know exactly the initial atomic configurations of the double K-vacancy states in the collision experiments, some ambiguities are usually accompanied in the comparison between experimental results and theoretical predictions. Rather pure double K-vacancy states can be produced through K-shell shakeoff and shakeup during K-electron capture decay or K-shell internal conversion[8]; a sudden change of electronic charge distribution during K capture or K conversion can eject the other K-shell electron into the continuum with a very small probability, e.g., $10^{-4} \sim 10^{-5}$ per decay for $Z \simeq 50$. Because of very little multiple ionization in the K-shakeoff and -shakeup processes, the initial configurations of the double K-vacancy states thus produced can be expected to be very simple, compared with those produced in the collision experiments.

The hypersatellite $K\alpha^h$ x ray due to the $1s^{-2} \rightarrow 1s^{-1}2p^{-1}$ transition was first discovered in the radiation following radioactive decay by K capture and K conversion.[9] No evidence of the two-electron–one-photon transition has, however, so far been observed in radioactive decay. We report here on a measurement of $K\alpha\alpha$ x rays due to the $1s^{-2} \rightarrow 2s^{-1}2p^{-1}$ transition, which follows K-shell shakeoff and shakeup during K-electron capture decay of ^{55}Fe.

II. MEASUREMENTS

Carrier-free ^{55}Fe was produced by the reaction ^{55}Mn$(p,n)^{55}$Fe. The ^{54}Mn impurity contained in the original source provided by NEN (New England Nuclear, USA) was removed by the anion exchange resin technique. The purified ^{55}Fe solution was absorbed in a 4-mm square filter paper, which was attached to a 6-mm thick Lucite plate. Two sources, 11 MBq and 110 MBq, were prepared for the x-ray measurements. The ^{54}Mn contamination in these sources was found to be less than 10^{-7} disintegrations per K capture of ^{55}Fe, as determined by high-statistics γ spectrometry.

The x rays from the sample source were measured with a Si(Li) detector with a sensitive volume of 80 mm^2 by 5 mm deep, provided with a 25-μm Be window. The detector resolution was 210 eV full width at half maximum (FWHM) at 5.9 keV. The sample sources fixed on the 6-mm thick Lucite absorber were mounted directly on the detector, which was shielded with 2-mm nylon, 3-mm aluminum, 3-mm brass, and 10-mm lead in order to reduce the natural γ-ray background from the surroundings. Two separate measurements were performed; the first run with the 11-MBq ^{55}Fe source for 71 days, and the second run with the 110-MBq ^{55}Fe source for 81 days. The counting rate was less than 10 sec^{-1} for 1 to 20-keV photons. The drift of the peak position during the measurements was less than

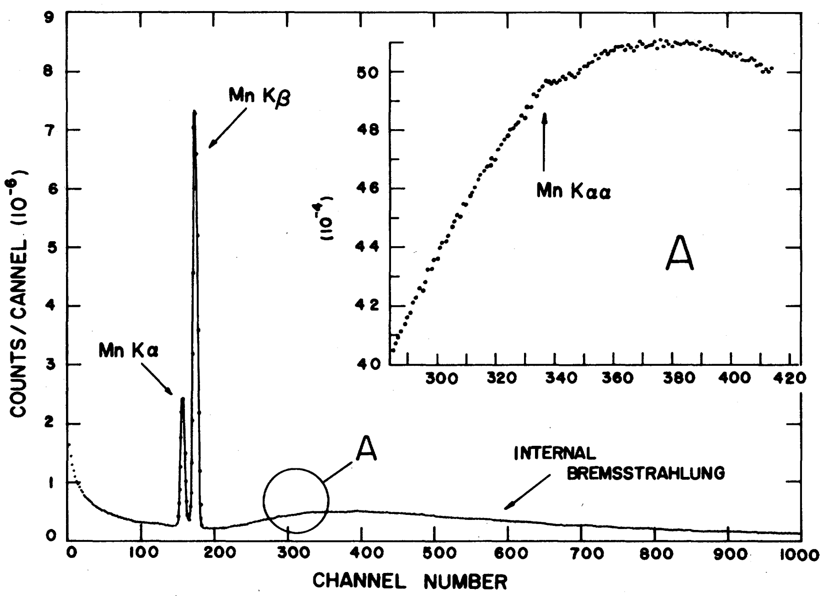

FIG. 1. Photon spectrum from ^{55}Fe measured with an 80-mm^2 by 5-mm-thick Si(Li) detector (resolution 210 eV at 5.9 keV). A 6-mm thick Lucite absorber changes the intensity ratio between Mn $K\alpha$ and $K\beta$ x rays. The continuum background is caused by the internal bremsstrahlung emitted during electron capture decay. The line denoted by Mn $K\alpha\alpha$ in the insert A is due to the correlated two-electron transition.

10 eV for 6.5-keV Mn $K\beta$ x ray.

The Mn $K\alpha\alpha$ x rays were observed in both measurements. The photon spectrum obtained in the second measurement is shown in Fig. 1. The spectrum mainly consists of two big peaks due to Mn $K\alpha$ and $K\beta$ x rays and the continuum due to internal bremsstrahlung. A small bump appears at about twice the $K\alpha$ x-ray energy [Fig. 1(a)]. The intensity of K x rays from the ^{55}Fe source was reduced by a factor of 5×10^{-6} by the 6-mm thick Lucite absorber, so that the counting rate of the $K\alpha$ x rays from the 110-MBq source was less than 2 sec^{-1}. Consequently, the bump cannot be due to the accidental coincidence of $K\alpha$ x rays. Furthermore, no bump was found near twice the $K\alpha$ x-ray energy in the background spectrum obtained by a separate long-run measurement (20 days) without the ^{55}Fe source. Thus, it is confirmed that the bump is due to the Mn $K\alpha\alpha$ line caused by the two-electron–one-photon transition to the doubly ionized K shell. The $K\alpha\beta$ x ray with an energy ~0.7 keV higher than the $K\alpha\alpha$ x-ray energy could not be found in the present measurement, probably because of its lower intensity compared with the large internal bremsstrahlung background.

III. DATA ANALYSIS

The spectrum from the measurement with the 110-MBq source was used for the data analysis, because of its better statistics. The peak position of the $K\alpha\alpha$ x ray and its intensity were determined by a nonlinear least-squares fit. The peak shape was assumed to be a Gaussian with exponential tails. The peak width and tail parameters were derived from a fit to the spectrum of Np Ll x rays [Fig. 2(a)], whose energy (11 890 eV) (Ref. 10) is very near that of the Mn $K\alpha\alpha$ x rays. The Mn $K\alpha\alpha$ x-ray spectrum was fitted [Fig. 2(b)] to determine the peak position and the peak height; parameters for the peak shape were assumed from the Np Ll spectrum and the background was taken to be a cubic function. The energy of the Mn $K\alpha\alpha$ x ray was calibrated by comparison with observed spectra of L x rays from ^{241}Am, ^{207}Bi, and ^{195}Au. The energies of x rays used in the calibration were taken from Ref. 10: Np Ll (11 890 eV), Pb Ll (9184.5 eV), Pb $L\beta1$ (12 613.7 eV), Pb $L\gamma1$ (14 764.4 eV), Pt Ll (8268 eV), Pt $L\beta1$ (11 070.7 eV), and Pt $L\gamma1$ (12 942.0 eV). The Mn $K\alpha\alpha$ x-ray energy thus determined is

$$E_{\alpha\alpha}=11\,907\pm20 \text{ eV}. \quad (1)$$

The ratio R of the $K\alpha\alpha$ x-ray intensity to the $K\alpha$ γ-ray intensity is

$$R=(N_{\alpha\alpha}/N_\alpha)(A_\alpha/A_{\alpha\alpha})(D_\alpha/D_{\alpha\alpha}). \quad (2)$$

where N_α is the total number of counts of $K\alpha$ x rays during the measurement, and $N_{\alpha\alpha}$ is the total number of counts of $K\alpha\alpha$ x rays. The ratio $N_{\alpha\alpha}/N_\alpha$ was estimated as $(3.1\pm0.6)\times10^{-3}$. The

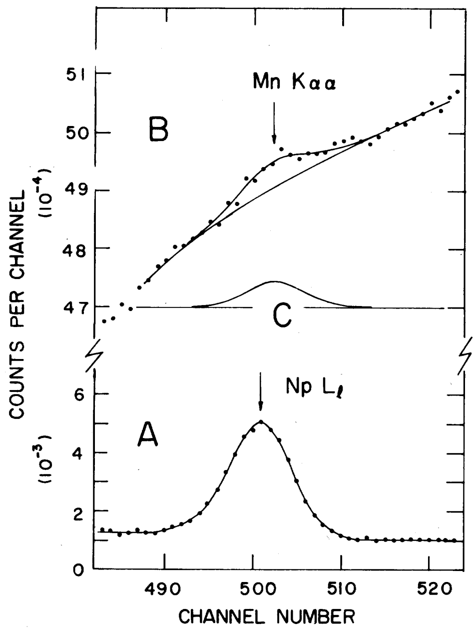

FIG. 2. Results of the nonlinear least-squares fits. The spectrum A is the Np Ll x ray from ^{241}Am, which was used to determine the line shape of the Mn $K\alpha\alpha$ peak. Figure B shows the decomposition of the measured spectrum into Mn $K\alpha\alpha$ peak and background; the peak shape was assumed to be given by a Gaussian with exponential tails, while the background was assumed to be a cubic function. Curve C is the decomposed peak of the Mn $K\alpha\alpha$ x ray.

factor $A_{\alpha\alpha}$ and A_α are the transmission of $K\alpha\alpha$ and $K\alpha$ x rays through the 6-mm thick Lucite absorber, respectively. The transmission factor A_α was easily determined using Mn $K\alpha$ x rays from the ^{55}Fe source with and without the Lucite absorber. Np Ll x rays from ^{241}Am source were used to determine $A_{\alpha\alpha}$. We found $A_\alpha/A_{\alpha\alpha} = (2.1 \pm 0.1)\times 10^{-5}$. The factors D_α and $D_{\alpha\alpha}$ are overall detection efficiencies without the Lucite absorber for $K\alpha$ x rays and $K\alpha\alpha$ x rays, respectively. These efficiencies were not determined in the present work; the ratio of $D_\alpha/D_{\alpha\alpha}$ was deduced from Fig. 9 in Ref. 11 as 0.85 ± 0.10; the error was estimated rather conservatively. The $K\alpha\alpha/K\alpha$ x-ray intensity ratio was thus found to be

$$R = (5.5 \pm 1.2)\times 10^{-8}. \tag{3}$$

IV. DISCUSSION

As indicated,[12-14] the $K\alpha\alpha$ x rays arise from two-electron-one-photon $E1$ transitions ($1s^{-2} \to 2s^{-1}2p^{-1}$) into an empty K shell, leaving two holes in the L shell. Denoting the total energy of the atomic state λ as $B(\lambda)$, the $K\alpha\alpha$ x-ray energy $E_{\alpha\alpha}$ is given by

$$E_{\alpha\alpha} = B(1s^{-2}) - B(2s^{-1}2p^{-1}). \tag{4}$$

Åberg and Tulkki[15] and independently Briançon[16] have recently calculated the $K\alpha\alpha$ x-ray energy with the aid of the multiconfiguration Dirac-Fock (MCDF) code of Desclaux.[17,18] The allowed $E1$ transition in the pure L-S coupling scheme is only $1s^{-2}(^1S) \to 2s^{-1}2p^{-1}(^1P)$. In the intermediate coupling scheme, the ^3P component is mixed with the ^1P state through the spin-orbit interaction. The results by the MCDF calculations are listed in Table I; both calculations include the Breit correction and the QED correction for electron self-energy and vacuum polarization. According to the intermediate coupling analysis by Åberg and Tulkki, the intensity of the high-energy component ^3P is about 3% of the ^1P intensity. Thus, we can compare the measured $E_{\alpha\alpha}$ with the energy of the ^1P component, neglecting the ^3P component. As seen in Table I, the measured energy agrees fairly with both predictions for the ^1P component.

According to the combination principle, the $K\alpha\alpha$ x-ray energy of ^1P component is expressed by the other measurable quantities as

$$E_{\alpha\alpha}(^1P) = E^h_{\alpha 2} + E_{\alpha 4} - E(KL_2L_3) + E(KL_1L_2). \tag{5}$$

In the expression, $E^h_{\alpha 2}$ is the energy of the hypersatellite $K\alpha^h_2$ x ray emitted through the $1s^{-2} \to 1s^{-1}2p^{-1}(^1P)$ transition. The $K\alpha^h_2$ x rays induced by the K-shell double photoionization were first measured by Keski-Rehkonen *et al.* with a plane crystal Bragg spectrometer.[19] Their result for

TABLE I. Experimental and theoretical $K\alpha\alpha$ x-ray energies.

Components	Expt. energy present work	$E_{\alpha\alpha}$ (eV) Calculated energies Åberg & Tulkki	Briançon[a]	Energy deduced from Eq. (5)
^1P	11 907 ± 20	11 930	11 935	11 935 ± 4
^3P		11 964	11 969	

[a] Corrected for the solid-state effect.

Mn $K\alpha_2^h$ x ray is 6142 ± 3 eV, which agrees well with MCDF calculations.[19,20] The second term of Eq. (5), $E_{\alpha 4}$, is the energy of the satellite x ray due to the $1s^{-1}2p^{-1}(^1P) \to 2p^{-2}(^1D)$ transition. The shift of the satellite x ray relative to the $K\alpha_1$ x-ray energy was measured accurately by Parratt.[21] Using the shift by Parratt and the $K\alpha_1$ x-ray energy taken from Bearden,[10] we obtain $E_{\alpha 4} = 5929$ eV. In Eq. (5), $E(KL_2L_3)$ and $E(KL_1L_2)$ indicate the energies of electrons emitted by the Auger transitions, $1s^{-1} \to 2p^{-2}(^1D)$ and $1s^{-1} \to 2p^{-2}(^1P)$, respectively. These energies were determined from the KLL Auger spectrum of Mn measured for the electron capture decay of ^{55}Fe: $E(KL_1L_2) = 5068 \pm 2$ eV and $E(KL_2L_3) = 5204 \pm 2$ eV.[22] Thus, we obtain $11\,935 \pm 4$ eV for the $K\alpha\alpha$ x-ray energy deduced from Eq. (5). As seen in Table I, an agreement between this value and results by the MCDF calculations is excellent. The origin of a small difference (~20 eV) between the present experimental result and the value from Eq. (5) is not clear. A refined measurement with higher statistics and resolution may be desirable.

The double K-vacancy state decays through various modes, being expressed formally as

$$(\omega_{K\alpha}^h + A_{K\alpha}^h) + (\omega_{K\beta}^h + A_{K\beta}^h) + (\omega_{K\alpha\alpha} + A_{K\alpha\alpha}) + (\omega_{K\alpha\beta} + A_{K\alpha\beta}) + \cdots = 1 , \quad (6)$$

where ω and A indicate fluorescence and Auger yields, respectively, and each term on the right-hand side corresponds to the $(1s^{-2} \to 1s^{-1}2p^{-1})$, $(1s^{-2} \to 1s^{-1}3p^{-1})$, $(1s^{-2} \to 2s^{-1}2p^{-1})$, and $(1s^{-2} \to 3s^{-1}2p^{-1}$ or $2s^{-1}3p^{-1})$ transitions, respectively. A branching ratio Q, which is defined as a ratio of the $K\alpha^h$ x-ray emission rate to the $K\alpha\alpha$ x-ray emission rate, i.e.,

$$Q \equiv \omega_{K\alpha}^h / \omega_{K\alpha\alpha} , \quad (7)$$

was studied in previous ion-atom collision experiments.[5,6] Using the ratio Q, the ratio R of Eq. (2) is expressed as

$$R = P_{KK} \cdot \omega_{K\alpha\alpha} / \omega_{K\alpha} = (P_{KK}/Q) \cdot (\omega_{K\alpha}^h / \omega_{K\alpha}) , \quad (8)$$

where P_{KK} is the double K-vacancy creation probability per K-capture decay and $\omega_{K\alpha}$ is the fluorescence yield for $K\alpha$ x rays. The probability P_{KK} has been calculated from the theory of the K-shell shakeoff and shakeup during K-electron capture,[23,24] as $\sim 8 \times 10^{-5}$. The ratio Q has been estimated under different assumptions.[14,25-28] Previous calculations indicate that a probable Q value for Mn may be 2000 ~ 5000. Assuming $\omega_{K\alpha}^h / \omega_{K\alpha} = 1$, the ratio R is then predicted to be in the region from 2×10^{-8} to 4×10^{-8}, which agrees qualitatively with the present experimental value. Refined calculations for P_{KK} and Q are clearly necessary.

In summary, the $K\alpha\alpha$ x ray due to the two-electron–one-photon $E1$ transition has been observed for the first time in the electron-capture decay of ^{55}Fe. The measured x-ray energy agrees fairly well with the MCDF calculation for the $1s^{-2} \to 2s^{-1}2p^{-1}$ transition. The measured ratio of the $K\alpha\alpha$ x-ray intensity to the $K\alpha$ x-ray intensity has been qualitatively explained as the product of the double K-vacancy creation probability P_{KK} and the ratio $1/Q$ of $K\alpha\alpha$ x ray to $K\alpha^h$ x-ray emission probabilities.

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