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Atomic Readjustment to an Inner-Shell Vacancy: Manganese K X-Ray Emission Spectra from an Fe^{55} K-Capture Source and from the Bulk Metal*

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A comparison is made between manganese $K\alpha_{1,2}$ and $K\beta_{1,3}$ x-ray emission lines with initial states produced (a) by electron bombardment (EB) of a metal anode at 13 and 15 keV and (b) by K capture (KC) in $Fe.^{55}$ The lines in each case have, within experimental accuracy, the same energy position, width, asymmetry, and relative intensity. The complex $K\beta'$ secondary structure on the low-energy side of $K\beta_{1,3}$ is also independent of the means of production of the initial state. It is argued that K capture leads to an essentially pure, single-hole, relaxed ground state and that the probability of multiple hole production is negligibly small. From the comparison of the spectra, it is concluded that upon the production of a K hole by a low-energy knockout process, the state formed with highest probability is the ground state for the system with a K hole, and that this state is formed in a time short compared to its lifetime. Differences between KC and EB spectra would exhibit the effects of either incomplete readjustment and/or multiple hole production. Various mechanisms leading to multiple hole production are qualitatively discussed in terms of previous experimental observations of emission and absorption spectra.

I. INTRODUCTION

THE problem of atomic readjustment following a change in effective nuclear charge has been extensively discussed.[1–3] By accurately comparing the K-emission spectra obtained from an Fe^{55} K-capture (KC) source with similar spectra obtained by electron bombardment (EB) of a manganese target this experiment focuses attention on atomic readjustment which may occur during K ionization by a knockout process. The probability of producing states of multiple excitation and ionization by readjustment is discussed in terms of secondary features observed in emission and absorption spectra. The primary features are conventionally interpreted as dipole transitions in which only one electron changes its quantum numbers.

II. THEORY

States produced by KC are discussed first, then EB effects are outlined and, finally, some comments on previous observations of atomic readjustment are made.

Accurate spectrometric measurements of the x-ray emission from some KC sources have been reported.[4] These show that after an atom of atomic number $Z+1$ undergoes K capture, the x rays emitted are characteristic of atomic number Z. Several observations on Fe^{55} have been made with low-energy–resolution instruments.[5]

For Fe^{55}, KC leads to an essentially pure state in which there is a hole in the K shell and the rest of the atom is in its ground state for a manganese nucleus and a hole in the K shell. The probability that states of multiple atomic excitation and ionization are formed is extremely small.[1] This is explained by noting that KC does not significantly change the charge seen by the configuration outside the K shell because the screening initially provided by the captured $1s$ electron is essentially replaced by the decrease in nuclear charge. There remains the possibility of forming a state with two K holes, but calculations[1] and observations[5] show that, for present purposes, this state can be neglected. It then seems reasonable to assume that the x-ray emission observed when the hole is filled is characteristic of the transition from an almost stationary, pure, K state.

The situation for EB is much different. An electron from the K shell is removed *through* the system.[6] There is a large perturbation which may allow mixing of singly ionized states with states of multiple excitation and ionization. An exact time-dependent treatment of the electron-electron interaction of both the incident and ejected electrons with the entire configuration is too complex. It is, however, useful to describe the interaction by several distinct mechanisms which lead to states of multiple excitation and ionization with probabilities which can, in principle, be calculated. Since the system is observable only before and after the interaction, there is no way to distinguish between the mechanisms by experiment.

States of multiple excitation and ionization may be produced directly by the incident particle, but the probability for such an event is very small except at resonance energies. The discussion will now be restricted to the case where the incident particle produces only one hole directly.

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[1] H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953).

[2] J. S. Levinger, J. Phys. Radium **16**, 536 (1955); Phys. Rev. **90**, 11 (1953).

[3] J. N. Bahcall, Phys. Rev. **129**, 2683 (1962).

[4] E. N. Hatch, Z. Physik **177**, 337 (1964).

[5] For example, see F. T. Porter and H. P. Holz, Phys. Rev. **89**, 938 (1953).

[6] For the experiments described here, the incident electron has an energy 2 to 3 times the K binding energy.

Next, consider an atom where a K hole is produced at time zero. The Hamiltonian of the system relaxes from H_0 to H_1 during the time of electron ejection τ_h. States of multiple excitation and ionization, i.e., KL and KM are possible as a result of the change in potential. These are discussed in terms of two limiting cases.[7] If the electron travels slowly enough through the system such that $\partial H/\partial t$ is small enough between H_0 and H_1, the wave function distorts gradually and no multiple hole states are formed (adiabatic approximation). This approximation is valid when $[h/(E_{KS}-E_K)^2]\partial H/\partial t \ll 1$, where E_{KS} and E_K are the energies of the KS and K states and where $S=K, L, M$ and so forth. On the other hand, if the change in Hamiltonian is instantaneous, multiple hole states may be formed by electron shake-off (sudden approximation). This approximation is valid when $[(E_{KS}-E_K)/h]\tau_n \ll 1$, where τ_n is the time of transit of the ejected electron past the n shell, where $n=1, 2, 3$, etc. Under ordinary conditions of EB this restriction may not be very well satisfied and a large change in wave function (on the order of $\exp[-i(E_{KS}-E_K)\tau_n/h]$) is required in order to mix in states with multiple holes. These states will then have a relatively small amplitude which should be a strong function of energy just above the K edge.

In either case, if the lifetime of the K state τ_K is much larger than τ_h the wave functions for the excited state (the initial state for K emission) will satisfy H_1. KS states, when they occur, can be observed as they decay and produce secondary (satellite) lines on the high-energy side of the primary (parent) K emission line. The spectrum of the primary (parent) K lines is not affected by $\partial H/\partial t$ if $\tau_K \gg \tau_h$.

There remains the further possibility of line broadening over the range of energies transferred in electron-electron interactions. This effect is significant when the energies of the transiting electron and the bound electron are comparable. If the process of filling the hole begins during the time of ejection, then any energy exchange between the two electrons will, by energy conservation, show up as a gain or loss in the energy of the emitted photon. Each shell will contribute broadening up to the binding energy of an electron in that shell. This process becomes indistinguishable from the sudden approximation when a second electron is ejected either before or during the time the hole is filled. If $\tau_K \gg \tau_n$ and no electron is ejected, the wave function of the bound electron merely deforms reversibly during the time of transit and no line broadening should occur since no transitions take place during the time of distortion. A few remarks concerning observation of the effects discussed are necessary. First absorption, then emission spectra are discussed. When a photon is absorbed, the system makes a transition from its ground state to some allowed excited state. This may involve more than one electron changing its quantum numbers. In K absorption, the lowest excited state is formed when a $1s$ electron is placed in the first of a series of allowed, unoccupied, bound orbitals. Each member of the series may be populated as the energy of the incident photon is increased. If the electron is given enough energy to escape the system, it reaches one of a continuum of allowed orbitals. The K-absorption spectrum can be characterized as a series of resonance absorption lines followed by a continuum.

The lowest state of multiple excitation occurs when both a $1s$ electron and the outer electron with the least binding energy are placed in the first two of a series of allowed, unoccupied, bound orbitals. The absorption structure for this process is superposed on the single excitation-ionization spectrum as a series of secondary resonance lines and additional continuum. Since the probability for a multiple process is low, it may not always be observable against a high background absorption. The absorbed photon must have had enough energy to produce the multiple process, but there is no way of specifying the mechanism.

X-ray emission is observed when the excited state decays.[8] If the excited state is a sharply defined, single K-hole state then the primary emission line is sharp. Other electrons may be excited or ejected as the K hole is filled and a quantum of lower energy than the primary x ray is emitted. The low-energy secondary reflects the allowed transitions of the second electron.

When a multiple hole state, i.e., KM, decays, it is unlikely that both holes are filled at once since the lifetime of a K hole is very much shorter than that of an M hole. A typical primary transition $K \rightarrow M$ would then have the transition $KM \rightarrow M^2$ as a secondary. The multiple hole transition emission line comes at only slightly higher energy than the primary line since only one hole decays. These lines may not be distinguishable from secondaries arising from electron-electron interactions as the hole is being filled.

The emission from the KC source is assumed to be the decay of a pure K state. Secondary emission structure can be attributed either to interactions as the hole is filled or to the configuration of the final state after emission. It is useful to review some of the present experimental knowledge in terms of the discussion above.

It is known from measurements on x-ray spectra that the lifetime of a K hole is in the range 10^{-14} to 10^{-16}

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[7] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed., pp. 213–220; D. Bohm, *Quantum Theory* (Prentice-Hall, Inc., New York, 1951), pp. 496–509.

[8] Difficulties might be expected for the case of solids where the ejected electron is removed to the continuum but still remains within the solid. Deslattes (Ref. 11) studied the $K\beta_{1,3}$ emission spectra from an isoelectronic series of argon-like atoms. When spectra from solids containing Cl^- and K^+ are compared with those from Ar, they all show the same features which vary in a smooth way with atomic number. It may be assumed that solid-state effects do not dominate when the final state after emission is an inner hole.

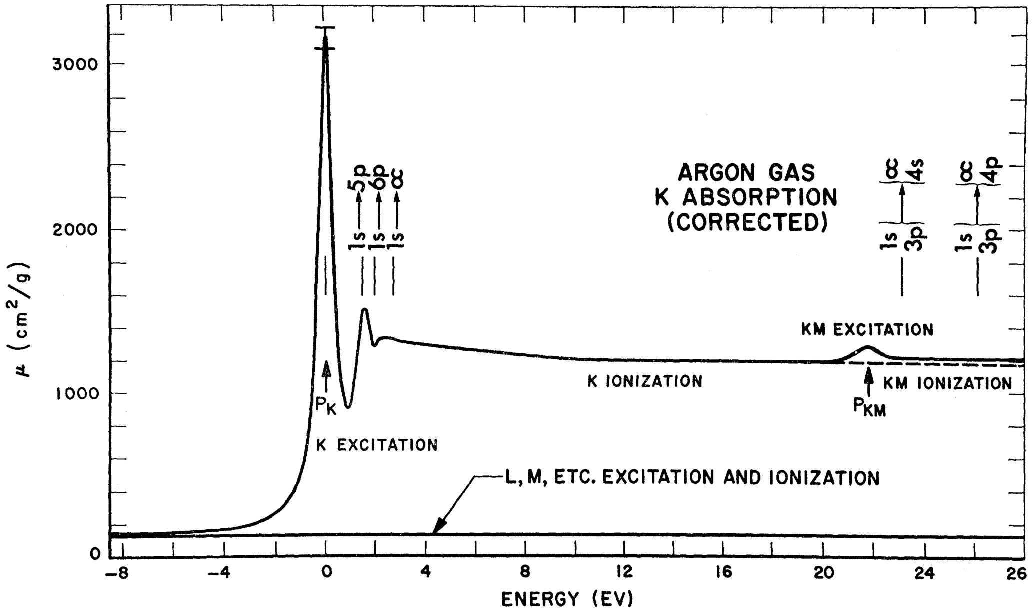

FIG. 1. The argon K absorption spectrum taken from Ref. 12. The energy levels shown above the curves are taken from the potassium I and II series and are adjusted to make the potassium $4s \rightarrow 4p$ line coincide with the argon $1s \rightarrow 4p$ line (P_K). The zero of energy is arbitrarily taken at P_K. See Table I.

sec.[9,10] These spectra do not give the relaxation lifetime directly and it is only through comparison with theory that the extent of relaxation can be known. Unfortunately, most x-ray spectra from solids or other multiatom systems are complicated by effects which tend to make a meaningful comparison difficult. Monatomic gases are the best choice, but only a few of them can be studied with suitable accuracy because either the spectral region of interest is experimentally inconvenient or the energy width of the inner state limits the energy resolution. Argon represents a suitable compromise and both its K emission[11] and absorption[12,13] have been studied.

The argon K-absorption edge (Fig. 1) is characterized by a series of lines $\{1s \rightarrow np, n \geq 4\}$ (single-electron transitions), and two other series: $\{1s; 3p_{3/2} \rightarrow ns; n'p\}$ and $\{1s; 3p_{1/2} \rightarrow ns; n'p, n \geq 4, n' \geq 4\}$ (two-electron transitions). Watanabe[14] has made a 7-parameter fit to the data shown in the initial part of the curve (Fig. 1) and concluded on statistical grounds that the resonance absorption lines have a Lorentzian shape, which implies a transition to a pure state having a single K hole and no further excitation. Bagus[15] finds excellent agreement between an SCF (self-consistent field) calculation with relaxation and the experimental value of the argon K ionization limit ($1s \rightarrow \infty$) reported by Brogren.[16] In this case, the electron is ejected with barely enough energy to reach the continuum and it travels through the atom rather slowly, allowing for a smooth relaxation.

If relaxation is assumed, then the energy difference between two excited states of argon with configurations $1s^1 2s^2 2p^6 3s^2 3p^6 np$ and $1s^1 2s^2 2p^6 3s^2 3p^6 n'p$ is closely approximated by that of potassium with configurations $1s^2 2s^2 2p^6 3s^2 3p^6 np$ and $1s^2 2s^2 2p^6 3s^2 3p^6 n'p$ (see Table I).

Two-hole, i.e. KM, states which can be correlated with the potassium II energy levels are observed[12,13] (Fig. 1). They are formed with low probability either by a direct process or by the interaction of the ejected electron with an M-shell electron.

Further evidence for multiple ionization (KM) is provided by Deslattes's[11] study of the satellites accompanying argon $K\beta_{1,3}$ emission. The relative satellite

TABLE I. Energy levels with respect to the potassium-I ionization limit.[a]

Potassium I		Potassium II	
Level	Energy (eV)	Level	Energy (eV)
$4p$	−2.7	$4s$	20.2
$5p$	−1.3	$4p$	23.2
$6p$	−0.7	limit	31.8
limit	0.0		

[a] Data from C. E. Moore, Nat'l. Bur. Std. (U. S.) Circ. **467**, Vol. I (1949).

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[9] L. G. Parratt, Rev. Mod. Phys. **31**, 616 (1959).

[10] M. A. Blokhin, *X-Ray Spectroscopy* (Hindustan Publishing Corporation, Delhi, 1962), pp. 515 and 516.

[11] R. D. Deslattes, Phys. Rev. **133**, A390 (1964); **133**, A399 (1964).

[12] H. W. Schnopper, Phys. Rev. **131**, 2558 (1963).

[13] C. Bonnelle and F. Wuilleumier, Compt. Rend. **256**, 5106 (1963).

[14] T. Watanabe, Phys. Rev. **139**, A1747 (1965).

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[16] G. Brogren, Nova Acta Regiae Soc. Sci. Upsaliensis **14**, No. 4 (1948).

emission intensity is larger than the relative intensity of the multiple hole states observed in absorption. As discussed below, it is expected that the probability of *KM* ionization be energy-dependent and that other mechanisms, i.e., shake-off, lead to *KM* ionization. Therefore, the results need not be inconsistent.

If the various radiative and nonradiative (Auger) channels for decay of an excited state are examined, it is possible to calculate the relative abundances of the final charged ions. Experiments and calculations on rare-gas atoms lead to an excess number of highly charged ions which cannot be explained by single-electron-emission processes.[17,18] In particular, Ne ions of charge greater than 2 are observed, but definitely ruled out by any single process. These results are explained partially in terms of multiple-electron photoelectric processes and partially in terms of multiple-electron Auger processes.[19,20]

The *KL-LLL* Auger spectrum has been observed from gaseous Ne bombarded by a 5.7-keV electron beam.[21] The high intensity of lines from states of double initial ionization is in agreement with shake-off calculations.[22]

The abundance of ions attributed to shake-off processes is relatively constant for photoelectron energies where the sudden approximation is valid. The probability for shake-off falls rather rapidly to zero as the incident photon energy approaches the threshold energy for the multiple process. For Ne, the shake-off process seems to be fully operative for values of $(E_{KS}-E_K)\tau_n/h$ below about 0.2 which is significant because this is a weaker restriction than usually applied for the sudden approximation.

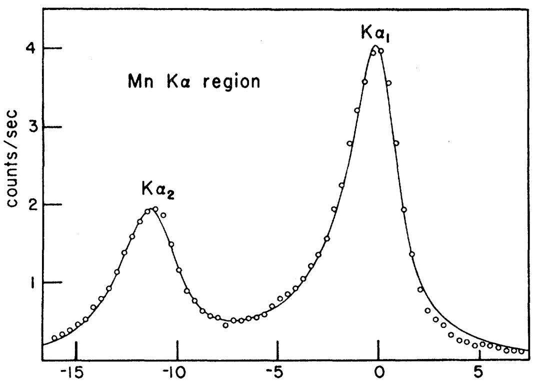

FIG. 2. The Mn $K\alpha$ spectra recorded on a two-crystal spectrometer from a K-capture source (Fe^{55}) (solid line) and from electron bombardment of bulk manganese (open circles). Both curves are normalized to the same area. The intensity scale refers to the K-capture data. See Table II.

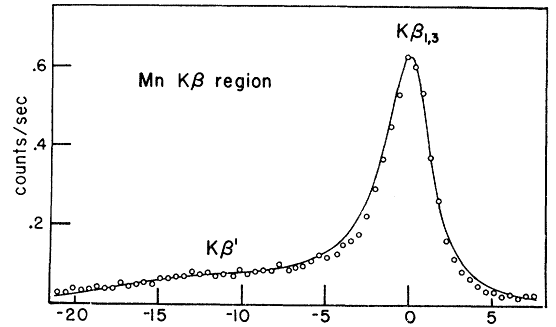

FIG. 3. The Mn $K\beta$ spectra recorded on a two-crystal spectrometer from a K-capture source (Fe^{55}) (solid line) and from electron bombardment of bulk manganese (open circles). Both curves are normalized to the same area. The intensity scale refers to the K-capture data.

III. EXPERIMENT

A direct observation of the relative magnitudes of the relaxation time τ_h and the K-state lifetime τ_K is available in the comparison of manganese $K\alpha_{1,2}$ and $K\beta_{1,3}$ x-ray emission spectra from sources with the initial K states produced (a) by electron bombardment (EB) of a Mn anode and (b) by K capture (KC) in Fe^{55}. If $\tau_K \gg \tau_h$ then relaxation will be essentially complete and the KC and EB single-hole K spectra should be identical. Secondary lines which are due to multiple processes may be observed in the EB spectrum when they occur with sufficient probability.

The choice of sources was made for a number of reasons:

(1) The product after K capture, Mn^{55}, is formed in its nuclear ground state.

(2) The wavelengths of the K emission lines lie in the region of conventional x-ray spectroscopy.

(3) There is no background radiation (i.e., electrons or positrons) from the nucleus which accompanies the emission of x rays.

(4) A source of Fe^{55} of sufficient activity was available.

The electron excitation spectra have been described previously.[23,24] A pure bulk manganese target was used for recording the $K\alpha_{1,2}$ lines (15 keV) and an *in situ* evaporated anode was used for the $K\beta_{1,3}$ lines (13 keV). The Fe^{55} source was produced from neutron activated Fe^{54} evaporated on a platinum strip. Its age was approximately one half-life (2.9 yr) at the time of use and its activity approximately 1 Ci. The data were recorded either on a two-crystal vacuum-spectrometer[25] equipped

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[17] T. A. Carlson and M. O. Krause, Phys. Rev. **137**, A1655 (1965).

[18] T. A. Carlson and M. O. Krause, Phys. Rev. **140**, A1057 (1965).

[19] T. A. Carlson and M. O. Krause, Phys. Rev. Letters **14**, 390 (1965).

[20] M. Wolfsberg and M. L. Perlman, Phys. Rev. **99**, 1833 (1955).

[21] H. Korber and W. Melhorn, Z. Physik **191**, 217 (1966).

[22] T. A. Carlson (private communication).

[23] L. G. Parratt, Phys. Rev. **45**, 364 (1934).

[24] H. W. Schnopper, M. S. thesis, Cornell University, 1958 (unpublished).

[25] H. W. Schnopper (to be published).

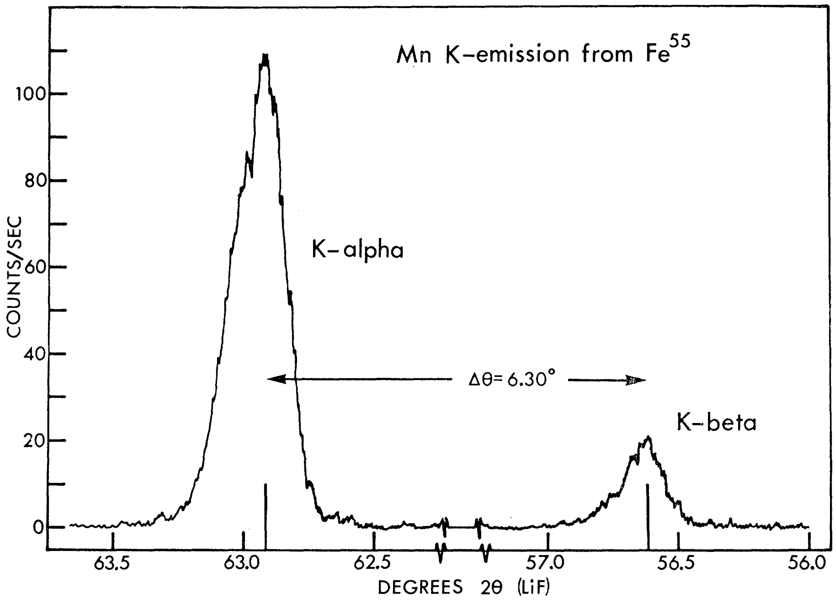

FIG. 4. The Mn $K\alpha$ and $K\beta$ spectra recorded on a single-crystal spectrometer from a K-capture source. Scan speed 0.25 deg/min for 2θ. See Table III.

with calcite crystals ($\lambda/d\lambda \simeq 10^4$) or in one case on a Siemens spectrometer equipped with a LiF crystal and a Soller slit of angular divergence 0.07 deg for 2θ. The The EB $K\alpha_{1,2}$ spectra were recorded using an ionization chamber. All other data were recorded with a P-10 (90% Ar, 10% CH_4) filled proportional counter followed by a linear amplifier with pulse-height discrimination. For the spectra from the Fe^{55} source an energy discrimination window of approximately $\frac{1}{2}$ the width of the pulse-height distribution was used to increase the signal-to-noise ratio. The count rate at the peak of $K\alpha_1$ line was 5 per second and the non-x-ray background was about 1 count in 10 sec.

IV. RESULTS

Figures 2 and 3 show the comparison of the spectra obtained from the Fe^{55} source and by electron bombardment. At least four curves were recorded for each spectrum. The non-x-ray background has been subtracted from all spectra, but no resolving-power corrections[26] have been made. In each figure, both spectra have, within experimental error, the same width, index of asymmetry, and relative intensity. In addition, the $K\alpha_{1,2}$ separation and the $K\beta'$ complex do not appear to depend on the method of K-hole production.

TABLE II. Energies of Mn and Fe K emission lines.[a]

	$K\alpha_1$ (keV)	$K\alpha_2$ (keV)	$K\beta_{1,3}$ (keV)	$K\alpha_1$-$K\alpha_2$ (eV)	$K\beta_{1,3}$-$K\alpha_1$ (eV)
Mn	5.89875	5.88765	6.49045	11.10	591.75
Fe	6.40384	6.39084	7.05798	13.00	654.14

[a] Data from J. A. Bearden, U. S. Atomic Energy Commission Report No. NYO-10586, 1964, p. 28 (unpublished).

Since the EB and KC curves in each figure were recorded at different times, it is not possible to put them on exactly the same absolute energy scale. The spectrometer has a tangent arm mechanism for fine motion and only a coarse circle for Bragg-angle reference. With this limitation it was not possible to measure the absolute Bragg 2θ angle to better than 1 min of arc ($\sim$2 eV). However, the $K\alpha_{1,2}$ separation is definitely that of manganese rather than iron (see Table II).

In order to check further the energy position of the KC spectra with respect to the EB spectra, the Fe^{55} source was mounted on a standard single-crystal spectrometer. The instrument was carefully zeroed and the $K\alpha_{1,2}$ and $K\beta_{1,3}$ lines were run in a single pass. Table III shows the expected values in 2θ Bragg angle for the principal features of the curve shown in Fig. 4. Except for $K\beta'$ no secondary structures with as much as 2% of the intensity of $K\alpha$ were observed in the energy region between $K\alpha$ and $K\beta$. The agreement between the expected and experimental positions is limited only by the ability to read the positions from the curve, i.e., $\pm$0.01 deg for 2θ ($\sim$1 eV). For display purposes, both the EB and KC spectra were normalized to the same area and then adjusted along the energy scale until a good fit was obtained.

TABLE III. LiF 2θ values for Mn and Fe K emission spectra.[a]

	$K\alpha_1$ 2θ (deg)	$K\alpha_2$ 2θ (deg)	$K\beta_{1,3}$ 2θ (deg)	$K\alpha_1$-$K\beta_1$ 2θ (deg)
Mn	62.91	63.01	56.61	6.30
Fe	57.45	57.58	51.72	5.73
Mn-Fe	5.46	5.43	4.89	0.58

[a] Data from M. C. Powers, *X-Ray Fluorescent Spectrometer Conversion Tables* (Philips Electronic Instruments, Mount Vernon, New York, 1960).

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[26] J. O. Porteus, J. Appl. Phys. **33**, 700 (1962).

V. SUMMARY AND CONCLUSIONS

The results presented show, within experimental error, that the primary lines observed in the KC and EB spectra are identical except as noted below. This implies that in both cases the initial and final states for the primary emission process must be the same. In particular, upon the production in manganese of a *K* hole by a low-energy knockout process, the remaining electronic configuration relaxes to a state which is identical to the one formed when Fe^{55} *K* captures and, furthermore, the time of relaxation is short compared with the lifetime of the *K* hole.[27]

Little or no line broadening which can be attributed to inelastic scattering of the ejected electron as it leaves the atom is observed. A difference between the KC and EB linewidths would be a positive indication of this process since it cannot occur in KC. The difference is, however, expected to be small if $\tau_K \gg \tau_h$.

No discrete high-energy lines were observed with EB. Because of the high multiplicity of the optical levels available to the second electron, the multiple hole spectrum might be only a smear on the high-energy side of the emission lines. There seems to be some inconclusive evidence for this in the data.

The probabilities, interpolated from calculations using Hartree-Fock wave functions, for electron shake-off arising from a sudden removal of a *K*-shell electron from a Mn atom are about 10, 4.4, 0.7, 0.8 and 0.1 for the $3d$, $3p$, $3s$, $2p$, and $2s$ shells, respectively.[22] These values are to be regarded as upper limits for high-energy incident particles. The values for the present experiment which would have to include solid-state effects for the outer shells have not been calculated. The lower binding energy of the outer shells in the solid (i.e., $3d$, $4s$) relative to the atom might actually enhance the probability of electron shake-off for the outer shells of a solid. It is unlikely that $3s$, $2p$, and $2s$ shake-off could have been observed in this experiment, but perhaps the difference referred to above is due in part to $3d$ and $3p$ shake-off.

Using the data presented here it is not possible to give a complete interpretation to the complex low-energy secondary structure $K\beta'$ accompanying the $K\beta_{1,3}$ emission line. It is, however, possible to eliminate several proposed mechanisms. Since the structure appears to be identical with either mode of *K*-hole production,[28] it is concluded that $K\beta'$ is to be identified only with the mode of decay[29,30] of the *K* state and the final electronic configuration[31,32] but not with the electronic configuration of the initial state[9,33,34] (since no multiply excited or ionized states are produced by KC). The $K\beta'$ structure is very sensitive to the chemical environment of the emitting atom.[31] The similarity of the iron and manganese conduction bands allows both the KC and EB final multiple hole states to be essentially the same. This similarity would not hold in general in comparing KC and EB sources, i.e., Ar^{37} which undergoes *K* capture giving monatomic Cl^{37} and diatomic chlorine gas.

One final conclusion comes from a comparison of all the results quoted. It appears that the ion formed as a result of an excitation or ionization by a knockout process, whether it be singly or multiply excited, is left with a definite configuration and energy both of which can be determined by experiment.

ACKNOWLEDGMENTS

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[27] The lifetime of a manganese *K* hole is taken to be 10^{-15} sec. References 9 and 10 both give *K*-state widths on the order of 1 eV.

[28] In addition to the data shown here, a photon bombardment spectrum has been recorded by P. E. Best (unpublished data). A two-crystal spectrometer equipped with quartz crystals was used and the data cannot be compared directly until they are corrected for resolving power. No qualitative differences are present between his data and those shown in Fig. 4. In addition, for pure manganese, no high-energy satellites are observed accompanying $K\beta_{1,3}$ (1% of the $K\beta_{1,3}$ intensity could be detected).

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[30] M. A. Blokhin, Zh. Eksperim. i Teor. Fiz. **9**, 1515 (1939).

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[34] M. Sawada, Mem. Coll. Sci. Kyoto Imp. Univ. **15A**, 43 (1932).