longer neglect relativistic corrections to the Landau orbitals. From the condition $\hbar\omega_c = mc^2$ the upper limit for the magnetic field is estimated to be of the order of 4.14×10^{13} G.

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Energies and Relative Intensities of $K\alpha\alpha$ X-Ray Transitions

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$K\alpha\alpha$ absolute energies and energy shifts relative to twice the $K\alpha$ energy have been measured in ion-atom collisions at MeV energies in the region from $Z=12$ to 26. For $Z=12$ to 22 good agreement is obtained with Hartree-Fock energy calculations assuming an $E1$ transition with two L-shell vacancies in the initial state. $K\alpha\alpha$ to $K\alpha$ intensity ratios are also presented.

The study of transitions in which two electrons both change to more tightly bound inner-shell orbitals and the energy is carried off by a single photon has an interesting history. Such cooperative transitions received extensive theoretical attention in early attempts to understand the energy of x-ray satellite lines,[1-3] and then were neglected for several decades. The recent observation by Wölfli *et al.*[4] of cooperative $K\alpha\alpha$ x-ray transitions (resulting from the filling of two K-shell vacancies by two L-shell electrons) from Fe and Ni in high-energy heavy-ion collisions has caused a resurgence of interest in this area.[5-11] These transitions provide a new testing ground for determining the adequacy of present calculational methods for obtaining transition energies and rates.

Final configurations associated with the observed $K\alpha\alpha$ transitions were not initially identified by Wölfli *et al.*[4] Calculations indicated that the observed energy was in agreement with an $E1$ transition $[(1s)^{-2}\to(2s)^{-1}(2p)^{-1}]$, but the $E2$ transition $[(1s)^{-2}\to(2p)^{-2}]$ was predicted to be dominant[8] in highly stripped ions. This Letter presents the first extensive comparison of the measured and calculated $K\alpha\alpha$ energies (over the region $12\leq Z\leq 26$), together with the measured intensities of the $K\alpha\alpha$ transition relative to the $K\alpha$ transition.

In the present experiment ions accelerated by a 5-MV Van de Graaff accelerator were used to bombard thick targets. The energy of the ions was generally between 3 and 3.5 MeV, although in two cases doubly ionized beams of 7.0 MeV were also used. X rays emitted at 90° to the incident beam were measured with a Si(Li) detector of 200-eV resolution at 6 keV. In obtaining the $K\alpha\alpha$ spectra, sufficiently thick Be and Al absorbers were used to reduce the transmission of the corresponding $K\alpha$ radiation to about 10^{-8} in order to avoid any possibility of pulse pileup which might obscure or simulate the $K\alpha\alpha$ line. These absorbers resulted in about 10% transmission for the $K\alpha\alpha$ radiation. Spectra were also measured with the absorbers removed in order to obtain the yield of $K\alpha$ x rays. Energy calibrations were obtained by using Fe^{55} and Am^{241} sources and also by bombarding various targets with proton and helium beams. When helium beams were used, corrections were made to account for the presence of enhanced satellite lines.[12]

Figure 1 shows the spectrum obtained from 3.5-MeV Ar^+ ions incident on a Ca target. A 0.0076-cm Al absorber was used in addition to

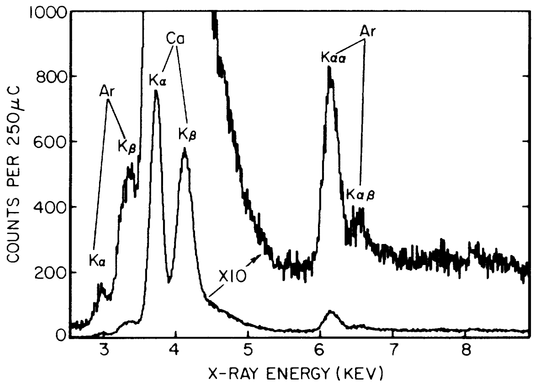

FIG. 1. X-ray spectrum obtained with 3.5-MeV Ar^+ ions incident on a Ca target. The same spectrum is also shown with the vertical gain increased by a factor of 10 to show the $K\alpha\alpha$ and $K\alpha\beta$ lines more clearly.

0.0038 cm of Be. Slightly to the right of the center are the Ar $K\alpha\alpha$ and $K\alpha\beta$ peaks. The $K\alpha\beta$ x ray is produced when one L and one M electron simultaneously fill the double K vacancy. Peak positions were determined by the use of a light pen operating on computer-generated oscilloscope displays of the measured spectra. Table I presents the measured $K\alpha\alpha$ energies together with the collision system and projectile energies used in each measurement. With regard to the present measurements, several points are worth mentioning. Three cases involve the observation of $K\alpha\alpha$ x rays from target atoms, namely Mg, Cl, and Ca. In the remainder of the cases the $K\alpha\alpha$ x rays were emitted by the projectile. In all cases $K\alpha\alpha$ x rays were observed only from the lighter of the two collision partners. The uncertainty of the $K\alpha\alpha$ energy is about 7 eV for Mg and increases to about 10 eV for Fe. As indicated by the third column from the left, it was found empirically that the energies of the $K\alpha\alpha$ x rays excited by 3-to 3.5-MeV collisions could be represented to an accuracy of about 0.5% by the expression $20.0\,Z(Z-1)$.

Of course the energy of the $K\alpha\alpha$ x ray will depend on the electronic configuration of the emitting atom. This dependence on the state of the atom is somewhat reduced if one calculates the energy shift of the $K\alpha\alpha$ x ray relative to two times the corresponding $K\alpha$ energy. Our results for this quantity are plotted in Fig. 2 together with the data of Wölfli *et al.*[4] Also shown are the results of Hartree-Fock calculations[13] of the energy shift for both the $(1s)^{-2} \rightarrow (2p)^{-2}$ ($n=6$) $E2$ $K\alpha\alpha$ transition and the $(1s)^{-2} \rightarrow (2s)^{-1}(2p)^{-1}$ ($n=6$) and $(1s)^{-2}(2p)^{-2} \rightarrow (2s)^{-1}(2p)^{-3}$ ($n=4$) $E1$ $K\alpha\alpha$ transitions. (Here n is the number of $2p$ electrons present in the initial state.) It should be noted that it

TABLE I. Measured energies of the projectile and target $K\alpha\alpha$ x rays for the indicated elements and collision systems. The third column indicates that the measured $K\alpha\alpha$ energies are represented to an accuracy of about 0.5% by the expression $20.0Z(Z-1)$.

Z	$K\alpha\alpha$ energy (eV)	$K\alpha\alpha$ energy / $Z(Z-1)$	Collision system
12 (Mg)	2655	20.11	3.0-MeV Al^+ on Mg
13 (Al)	3119	19.99	3.0-MeV Al^+ on Si
16 (S)	4799	20.00	3.5-MeV S^+ on KCl
	4826	···	7.0-MeV S^{++} on KCl
17 (Cl)	5418	19.92	3.5-MeV Ar^+ on KCl
18 (Ar)	6128	20.03	3.5-MeV Ar^+ on Ca
	6158	···	7.0-MeV Ar^{++} on Ca
20 (Ca)	7638	20.09	3.3-MeV Ti^+ on Ca
22 (Ti)	9273	20.07	3.3-MeV Ti^+ on Mn
26 (Fe)	13 025	20.04	3.5-MeV Fe^+ on Co and Ni
	13 048[a]	···	41.5-MeV Fe on Fe
28 (Ni)	15 228[a]	···	40.0-MeV Ni on Ni

[a]From Ref. 4.

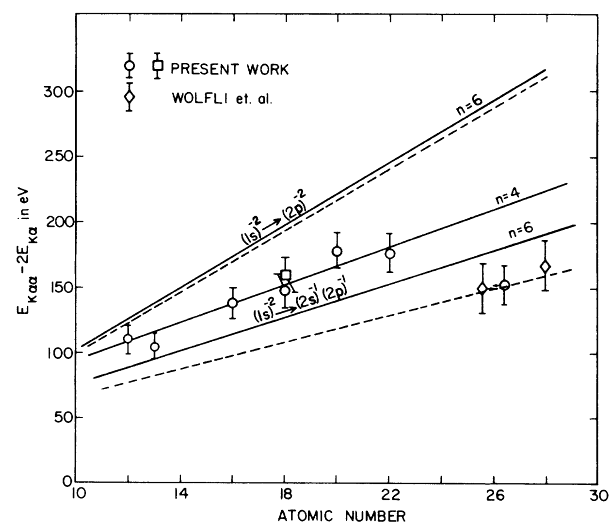

FIG. 2. Predicted and measured $K\alpha\alpha$ energy shifts. The solid lines represent $K\alpha\alpha$ energy shifts predicted by Hartree-Fock calculations. The top solid line assumes an $E2$ $K\alpha\alpha$ transition, while the lower pair of solid lines assumes an $E1$ transition with a full L shell ($n=6$) and with two $2p$ vacancies ($n=4$) in the initial state. The dashed lines are the shifts for $n=6$ with multiplet splitting taken into account. The circles represent data obtained with 3.0- to 3.5-MeV projectiles; the square was obtained with a 7.0-MeV projectile, and the diamonds are the measurements of Wölfli *et al.* (Ref. 4) at 40 MeV.

has previously been shown that the Slater exchange approximation is inadequate for the calculation of transition energies when double K-shell vacancies are involved.[14] The $E2$ energy shifts are relatively insensitive to the presence of vacancies in the L and M shells, but as is shown the removal of two $2p$ electrons ($n = 4$ curve) increases the $E1$ energy shifts by about 15%. The solid curves in Fig. 2 are derived from Hartree-Fock calculations for particular configurations and thus represent averages over the various multiplets that may be produced from a particular configuration. The two dashed curves were derived by taking into consideration the fact that only singlet final states are allowed for $n = 6$ if one assumes that L-S coupling is valid throughout this range of atomic numbers. The appropriate multiplet energies were calculated for Mg and Fe; the dashed curves are linear interpolations between the Mg and Fe energy shifts ($E_{K\alpha\alpha} - 2E_{K\alpha}$). For the $n = 4$ $E1$ transition, both the initial $(1s)^{-2}(2p)^{-2}$ and the final $(2s)^{-1}(2p)^{-3}$ states are split into several multiplets. Without knowing the transition rates between these various initial- and final-state multiplets it is difficult to determine a properly weighted energy-shift curve corresponding to the $n = 4$ case. However, if one assumes equal transition rates for all allowed transitions ($^3P \rightarrow {}^3S^0$, $^3P^0$, and $^3D^0$; $^1D \rightarrow {}^1P^0$, and $^1D^0$; and $^1S \rightarrow {}^1P^0$), and statistical population of the initial states, the resulting shift lies about 10 eV below the $n = 4$ curve at $Z = 26$. Most of the data points are in agreement with calculations for an $E1$ transition with two L-shell vacancies in the initial state. Olsen and Moore[15] have observed that essentially the same distribution of L-shell vacancies accompanies both double and single K-shell vacancy production. The present result is therefore in reasonable agreement with the number of L-shell vacancies observed in high-resolution measurements of $K\alpha$ spectra at these ion energies.[16,17] However, the data at $Z = 26$ and 28 agree with energy-shift calculations which assume $E1$ transitions, L-S coupling, and no L-shell vacancies in the initial state. This agreement of the 40-MeV data[4] with a calculation assuming no L-shell vacancies contradicts the data obtained by Jundt and Nagel[18] from 60-MeV Ni-Ni collisions which indicate that on the average about three L-shell vacancies accompany the production of a K-shell vacancy. Relativistic effects cannot explain this discrepancy, because the calculations by Hodge[9] show that the inclusion of relativistic effects produces only a small reduction in the energy shifts, about 6 eV for Fe and 8 eV for Ni.

An effort was made to determine the $K\alpha$ hypersatellite intensity for Ar incident on Ca in order to obtain a branching ratio for two of the radiative decay modes of the double K-shell vacancy, namely, one- and two-L-electron transitions accompanied by emission of a single photon. In this effort a flat crystal spectrometer was used to obtain the resolution necessary to separate the $K\alpha$ hypersatellite from the $K\alpha$ and $K\beta$ lines. The branching ratio obtained was 1.0×10^{-3} with an uncertainty of about a factor of 2. Calculations by Richard *et al.*[7] predict a value of 0.76×10^{-3}. The difficulty of this measurement due to the low $K\alpha$ hypersatellite intensity deterred us from performing additional measurements of this type for other collision systems which were expected to be less favorable. However, it was practical to measure the ratio of $K\alpha\alpha$ to $K\alpha$ intensity, and these results are presented in Fig. 3. The ratio measured for a given set of collision partners is represented by the atomic number difference between the two collision partners. The observation that the largest intensity ratio for emission from an atom with atomic number Z is obtained in collisions with atoms of atomic number Z or $Z+1$ is consistent with the assumption of a molecular orbital mechanism for K-shell vacancy production.[19] These ratios have been obtained by cor-

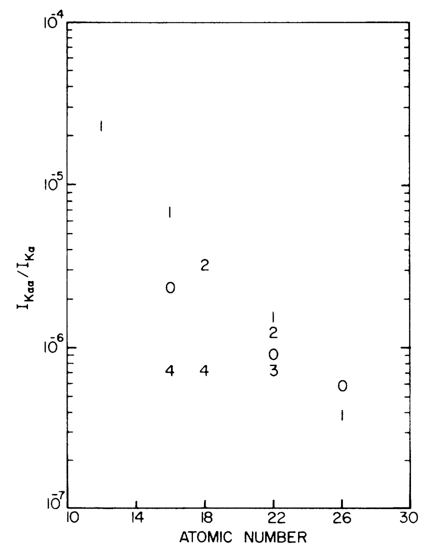

FIG. 3. Measured ratios of $K\alpha\alpha$ intensity with respect to $K\alpha$ intensity for various collision systems are indicated by numerals corresponding to $|Z_1 - Z_2|$. The atomic number of the emitting atom, which is always the lighter collision partner, is given along the abscissa.

recting for absorption in the Be and Al absorbers by using the absorption coefficients of Viegele *et al.*[20] and Short and Tabock,[21] respectively. Corrections for detector efficiency were performed using information supplied by the manufacturer and were small. The accuracy of the intensity ratios is estimated to be about 25%.

The ratio of $K\alpha\alpha$ intensity to $K\alpha$ intensity varies from 2.3×10^{-5} to 3.8×10^{-7} over the range of atomic number studied. The intensity ratio decreases with increasing atomic number, as would be expected, since the K-shell ionization probability decreases with increasing atomic number. Our measurement on Ar, combined with Wölfli's measurements[4] on Fe and Ni, indicates that the branching ratio is also decreasing with increasing atomic number, consistent with the calculations of Richard *et al.*[7]

In conclusion, $K\alpha\alpha$ energies and energy shifts relative to two times the $K\alpha$ energy, as well as intensity ratios of $K\alpha\alpha$ to $K\alpha$ in the region from $Z=12$ to 26, have been measured. These energy shift measurements establish that the $K\alpha\alpha$ transitions observed in this study are $E1$ transitions with approximately two L-shell vacancies present in the initial state for $12 \leq Z \leq 22$. The relatively small energy shifts observed for $Z=26$ and 28 are not presently understood and suggest that additional measurements in the region $Z>22$ would be worthwhile.

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