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Energy Shifts and Relative Intensities of K X-Rays Produced by Swift Heavy Ions†

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Abstract

We have measured the energy shifts of K x-rays produced by 5 MeV/amu He, C, O, and Ne ions, and by 10 MeV/amu C ions, on targets of Fe, Co, Zr, and Sn; additional data for Ti and Fe have been obtained using O ion energies from 0.625 MeV/amu to 2.84 MeV/amu. In the case of the 5 MeV/amu and 10 MeV/amu data, it was found that for each target there exists a nearly linear relationship between the observed energy shift and the stopping power for the bombarding ion. The projectile energy dependence of the shift for Ti and Fe targets obtained from the O data shows a form similar to that predicted for the L-ionization cross section. The observed K_β/K_α ratios show the same type of simple systematics, suggesting that these multiple ionization phenomena can be quantitatively explained within the framework of existing Coulomb excitation theories.

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Introduction

Weak satellites associated with K-lines were first discovered by Siegbahn and Stenström,[1] and an explanation involving multiple inner shell ionization was proposed by Wentzel.[2] More recently the observation of energy shifts in the K_β lines of Cu and Ni induced by 15-MeV oxygen bombardment has been reported by Richard et al.[3] using a Si(Li) detector for measuring the x-ray energies. They ascribe the shifts to multiple inner shell ionization, an explanation which has been verified by Bragg spectrometer experiments in which the measured lines were resolved into satellites corresponding to 1, 2, 3, etc. vacancies in the L-shell.[4,5,6] Other measurements on K energy shifts have been made[7,8] for a variety of targets at several projectile energies. The available data suggest that the energy shift in the K-lines is related to the L-ionization cross section as predicted by the Plane Wave Born Approximation[9] (PWBA) or the Binary Encounter Model[10] (BEM). Similar measurements have been made by Der et al.[11] for the L x-rays of Ni, Cu, and Zn induced by bombardment with 8-MeV and 20-MeV oxygen ions. They noted shifts in L-satellite energies which decreased as the projectile energy increased and they pointed out that this is consistent with the expected decrease in cross section for the production of M-vacancies when the projectile velocity is greater than the electronic orbital velocity.

If the phenomenon of multiple ionization can be described in terms of simple Coulomb excitation, it is to be expected that a) the energy shift observed at a given ion velocity should be proportional to Z^2, where Z is the projectile charge; b) that for a given projectile the shift at sufficiently high energies should be inversely proportional to the projectile energy, in the same way as the electronic stopping power; and c) that an energy shift due

primarily to L-vacancies would be a maximum in the region of $E/\lambda U_L \approx 1$, where E is the projectile energy, λ is the ratio of projectile mass to the electron mass, and U_L is an average binding energy for L electrons in the target. The aim of the present experiment was to check these predictions.

Experimental

Beams of 5 MeV/amu He, C, O, and Ne ions from the Oak Ridge Isochronous Cyclotron were used with targets of Fe, Co, Zr, and Sn to study the projectile dependence of the K x-ray energy shifts. The same targets were also bombarded with 10 MeV/amu C ions. The dependence of the shifts on the projectile energy was investigated using 0.625 MeV/amu to 2.84 MeV/amu O beams from the Oak Ridge Tandem Van de Graaff accelerator on targets of Ti and Fe. A similar experimental method was used on both accelerators and has been described elsewhere,[12] so that only a brief outline will be given here.

The targets were placed normal to the beam, and the x-rays viewed by a 12 mm^2 Si(Li) detector with an overall system resolution of 250 eV at 6 keV. For the cyclotron (Tandem) experiment the detector was placed at an angle of 150° (43°) to the beam direction, 50 cms (12 cms) from the target. X-rays from the target were viewed directly through the 0.0125 mm Be window of the detector for the cyclotron runs, while at the Tandem an additional 0.0125 cm Mylar window was employed. The beam currents were such that the count rate was always less than 1000 cps, and generally less than 100 cps.

Except for Ti, the targets were self-supporting foils, whose thicknesses were measured by the Rutherford scattering of 5 MeV/amu projectiles at an angle of 11.25°. The Ti target was evaporated onto a 40 $\mu g/cm^2$ carbon backing, the thickness being measured with a deposition gauge. Table I shows the targets and their measured thickness. The estimated uncertainty in the measurements is 10%.

Table I
Measured Target Thicknesses

Target	Ti	Fe	Co	Zr	Sn
Thickness ($\mu g/cm^2$)	100	428	936	580	689

The energy calibration of the system was measured using a pulser in conjunction with standard x-ray sources of ^{55}Fe, ^{57}Co, and ^{241}Am. In regions remote from the calibration energies the absolute accuracy was estimated to be ± 10 eV; near to these energies this improved to ± 2 eV. The detector efficiency calibration was made using the same standard sources. Interpolation to other energies was made using a theoretical curve calculated from the detector characteristics supplied by the manufacturer and the x-ray cross section compilation of McMaster et al.,[13] and adjusted in absolute magnitude to fit the data points.

Except for the K_β lines from Zr and Sn, peak positions and areas were extracted from the spectra by a computer code which assumed the peaks to be Gaussian in form and the background to be either linear or quadratic in channel number. Although these assumptions, particularly the first, are not necessarily justified, such a self-consistent analysis does provide reliable estimates of relative areas and positions. In the case of the K_β lines of Zr and Sn, the peak areas were extracted by hand and no attempt was made to estimate the average peak positions due to the relatively poor statistics coupled with the fact that the K_{β_1} and K_{β_2} components were partially resolved.

K_β/K_α ratios for Fe and Sn were estimated from the peak areas and corrections applied for the energy dependence of the detector efficiency, for absorption in the Mylar window where appropriate, and for self-absorption in the target, taking

into account the fact that the large energy shifts were generally sufficient to raise the K_β energy above the K absorption edge.

Results and Discussion

The measured energy shifts induced by 5 MeV/amu He, Co, O, and Ne ions and 10 MeV/amu C ions are shown in Fig. 1, plotted against the stopping power of the target material for the appropriate projectile. An almost linear relationship is evident for each of the x-ray lines considered. In this energy region the electron stopping power is roughly proportional to MZ^2/E, where M, Z, and E are the projectile mass, charge, and energy respectively. If we accept the explanation that the shifts are mainly due to L-vacancies produced at the same time as a K-vacancy, then the linear behavior for the 5 MeV/amu data points merely reflects the Z^2 scaling law for L-ionization cross sections, while the fit of the 10 MeV/amu C point implies that the L-ionization cross section varies as 1/E. The 1/E dependence holds for projectile velocities larger than the electron orbital velocity, a condition which may be expressed in the notation of Garcia[10] as:

$$E/\lambda U_L > 1, \qquad (1)$$

where λ is the ratio of the projectile mass to the electron mass, and U_L is the average binding energy of the L-shell. The value of $E/\lambda U_L$ for 5 MeV/amu particles on Fe, Co, Zr, and Sn is shown in Table II. Condition (1) is seen to be satisfied for all targets except Sn, and indeed for this case the 10 MeV/amu data point does not follow the trend defined by the 5 MeV/amu data.

Table II

$E/\lambda U_L$ for $E = 5$ MeV/amu

Target	Fe	Co	Zr	Sn
$E/\lambda U_L$	3.7	3.3	1.2	0.66

The K_α and K_β shifts for Ti and Fe which were measured as a function of the energy of the bombarding O ion are shown in Fig. 2, plotted against $E/\lambda U_L$. The solid lines have been drawn to guide the eye. The form of the energy dependence is similar to that of the L-ionization cross sections predicted by the PWBA or the BEM, except that the maximum occurs at $E/\lambda U_L \approx 1.3$, rather higher than expected. This probably reflects the fact that the projectiles have, on the average, impact parameters smaller than the Bohr radius of the L-shell as they must also have produced a K-vacancy. One effect of such small impact parameters is to increase the effective nuclear charge seen by L-electrons,[14] resulting in an effective increase in U_L.

The influence of M-vacancies on the energy shifts, at least for the Fe K_α lines, has been shown to be small.[6,15] Relativistic Hartree-Fock-Slater calculations by T. A. Carlson[15] indicate that the average shift per L-vacancy is approximately 25 eV, 55 eV, and 65 eV for the Fe K_α, Fe K_β, and Sn K_α lines respectively. Thus the X x-ray shifts induced by the 5 MeV/amu Ne ions correspond to an average of two or three L-vacancies for Fe, dropping to approximately one L-vacancy in the case of Sn.

Burch and Richard[6] have published calculations showing the influence of L- and M-vacancies on the K_β/K_α ratio for Fe. They show that the presence of vacancies in the L-shell and 3p subshell has a marked effect on the ratio, as

might be expected from the simple assumption that the K_β/K_α ratio is proportional to the 3p subshell population and inversely proportional to the number of electrons in the L-shell. Our measurements of the Fe K_β/K_α ratio, shown in Fig. 3, show very similar systematics to the energy-shift data, at least for $E/\lambda U_L \gtrsim 1$. This indicates that the dominating influence is again the production of L-vacancies, and suggests a rather small probability for the production of vacancies in the 3p subshell. From the calculations in Ref. 6, we estimate the K_β/K_α ratio for 32.5-MeV O bombardment of Fe ($E/\lambda U_L = 1.4$) to correspond to an average of $\lesssim 1$ 3p vacancy, if the energy-shift data is interpreted to correspond to two to three L-vacancies. For $E/\lambda U_L > 1$, the influence of 3p vacancies is expected to remain small, as both L and M ionization cross sections will scale roughly as MZ^2/E (or dE/dx). However, for $E/\lambda U_L < 1$, the relative importance of M-vacancies is expected to increase, as the M ionization cross section increases, while the L ionization cross section decreases rapidly. This effect would reach a maximum in the region of $E/\lambda U_M \gtrsim 1$ ($E/\lambda U_L = 0.07$), where U_M is the ionization energy of the 3p subshell, and might result in a K_β/K_α ratio rather lower than the value obtained from an unperturbed atom. A hint of such behavior is evident in the Fe data for low O bombarding energies.

Tin K_β/K_α ratios were obtained only with the 5 MeV/amu and 10 MeV/amu projectiles, and these results showed no MZ^2/E dependence. The average of the five data points is 0.240 ± 0.015 close to the normal value of 0.2226 estimated by Hansen et al.[16] This probably indicates that the effect due to the production of L-vacancies is approximately canceled by that due to M-vacancies in this energy range.

In summary, both the energy shifts and the K_β/K_α ratios reported in the present work show similar systematic behavior with respect to the energy and

charge of the incident projectile. These systematics can be qualitatively understood within the framework of existing theories. If such theories are formulated to extract the impact parameter dependence of the ionization cross sections, as has been done in the case of Au L x-rays by Hansteen *et al.*,[17] it seems likely that a quantitative description of multiple-ionization phenomena may be achieved.

References

1. M. Siegbahn and W. Stenström, Phys. Zeit, 17, 48 (1916).
2. G. Wentzel, Ann. d. Phys. 66, 437 (1921).
3. Patrick Richard, I. L. Morgan, T. Furuta, and D. Burch, Phys. Rev. Letters 23, 1009 (1969).
4. Patrick Richard, D. Burch, and D. McCrary, Bull. Am. Phys. Soc. 15, 1349 (1970); D. Burch and P. Richard, Bull. Am. Phys. Soc. 16, 74 (1971).
5. A. R. Knudson, D. J. Nagel, P. G. Burkhalter, and K. L. Dunning, Phys. Rev. Letters 26, 1149 (1971).
6. D. Burch, Patrick Richard, and R. L. Blake, Phys. Rev. Letters 26, 1355 (1971).
7. D. Burch and Patrick Richard, Phys. Rev. Letters 25, 983 (1970).
8. G. A. Bissinger and S. M. Shafroth, to be published in The Physical Review A.
9. E. Merzbacher and H. W. Lewis, Encyclopedia of Physics (Springer-Verlag, Berlin, 1958) Vol. 34, p. 166.
10. J. D. Garcia, Phys. Rev. A1, 280 (1970) and Phys. Rev. A4, 955 (1971).
11. R. C. Der, R. J. Fortner, T. M. Kavanagh, J. M. Khan, and J. D. Garcia, Phys. Letters 36A, 239 (1971).
12. A. van der Woude, M. J. Saltmarsh, C. A. Ludemann, R. L. Hahn, and E. Eichler, submitted to Physical Review Letters.
13. W. H. McMaster, N. Kerr Del Grande, J. H. Mallett, and J. H. Hubbell, University of California Report UCRL-50174, Sec. II, Rev. 1 (1969).
14. G. Basbas, W. Brandt, and R. Laubert, Phys. Letters 34A, 277 (1971); George Basbas, Werner Brandt, Roman Laubert, Anthony Ratkowski, and Arthur Schwarzschild, Phys. Rev. Letters 27, 171 (1971).

15. T. A. Carlson, private communication.

16. J. S. Hansen, H. U. Freund, and R. W. Fink, Nucl. Phys. A142, 604 (1970).

17. J. M. Hansteen and O. P. Mosebekk, Z. Physik 234, 281 (1970).

Figure Captions

Figure 1. The K x-ray energy shifts measured with 5 MeV/amu He, C, O, and Ne projectiles, and 10 MeV/amu C projectiles. The data are shown as a function of the stopping power of the projectile in the target material.

Figure 2. The K x-ray energy shifts as a function of the O bombarding energy. The errors indicated at $E/\lambda U_L = 0$ reflect the uncertainty in the absolute energy calibration.

Figure 3. The Fe K_β/K_α intensity ratios. The lower curve shows the 5 MeV/amu and 10 MeV/amu data as a function of the stopping power of the incident ion in the target material. The upper curve shows the ratio as a function of bombarding energy for oxygen projectiles. The value at $dE/dx = 0$ and $E/\lambda U_L = 0$ is taken from Ref. 16.

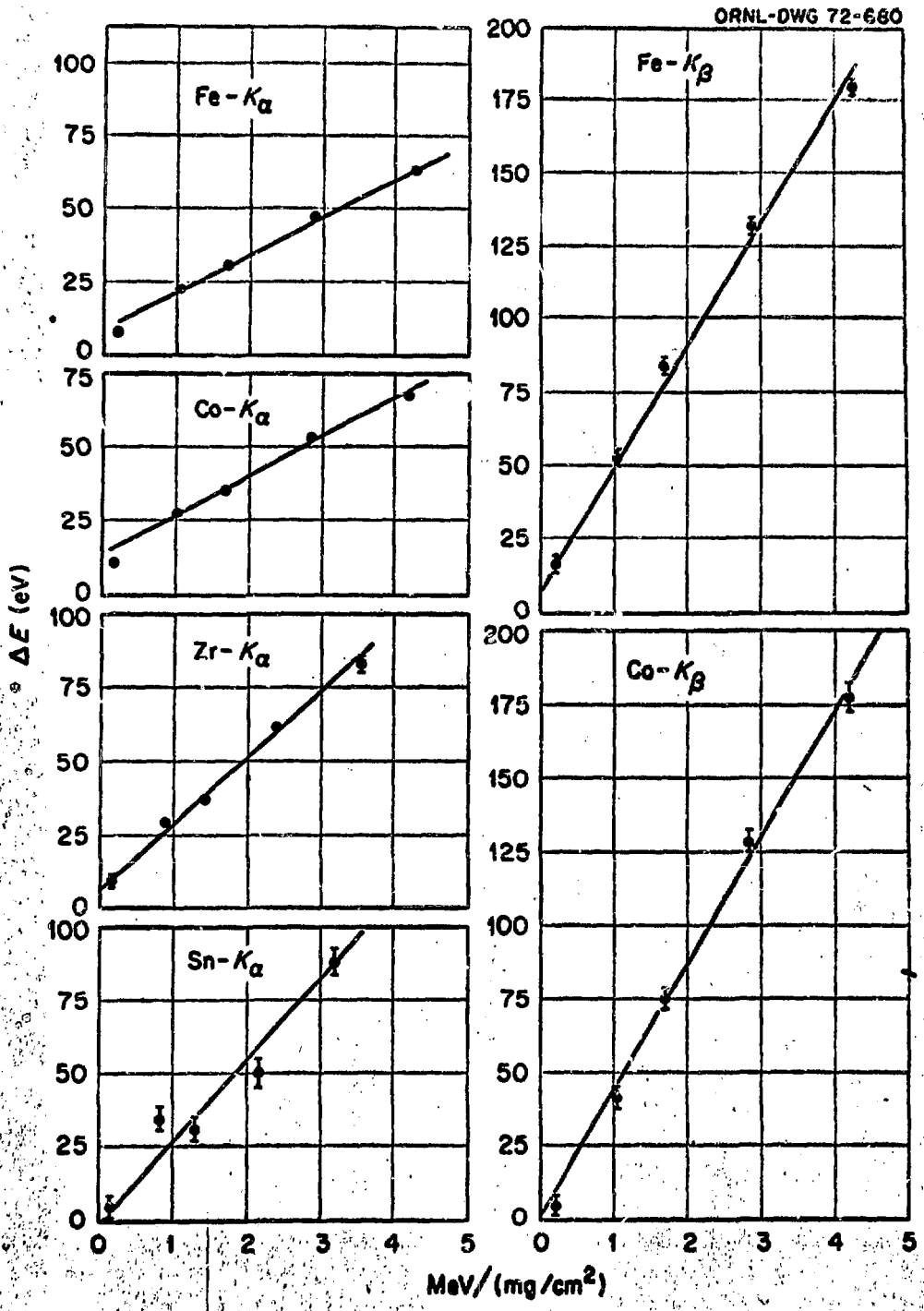
ORNL-DWG 72-680
Fe-K_α
Co-K_α
Zr-K_α
Sn-K_α
Fe-K_β
Co-K_β
ΔE (eV)
MeV/(mg/cm^2)

Figure 1

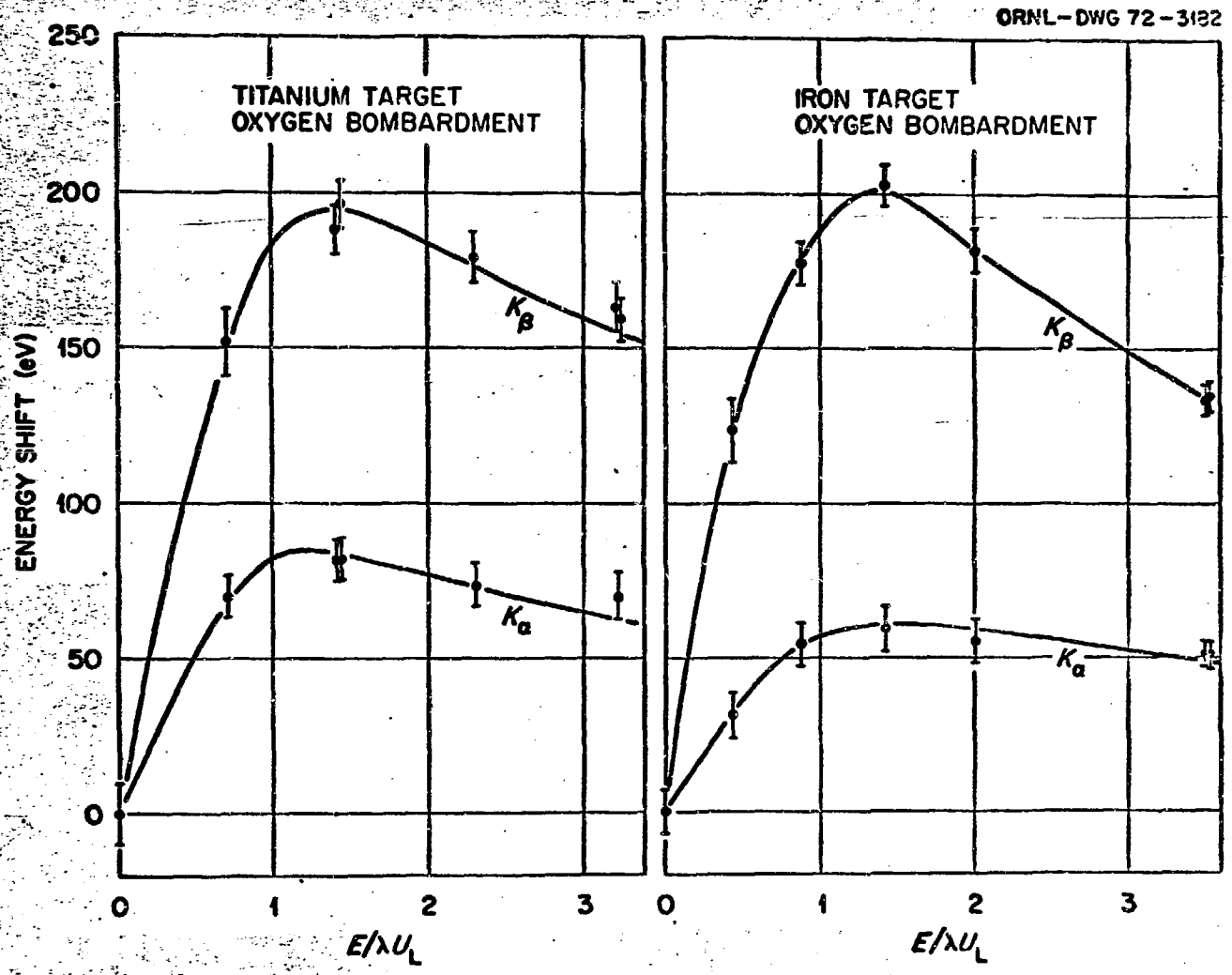
ORNL-DWG 72-3132
TITANIUM TARGET
OXYGEN BOMBARDMENT
IRON TARGET
OXYGEN BOMBARDMENT
ENERGY SHIFT (eV)
250
200
150
100
50
0
K_β
K_α
0
1
2
3
$E/\lambda U_L$

Figure 2

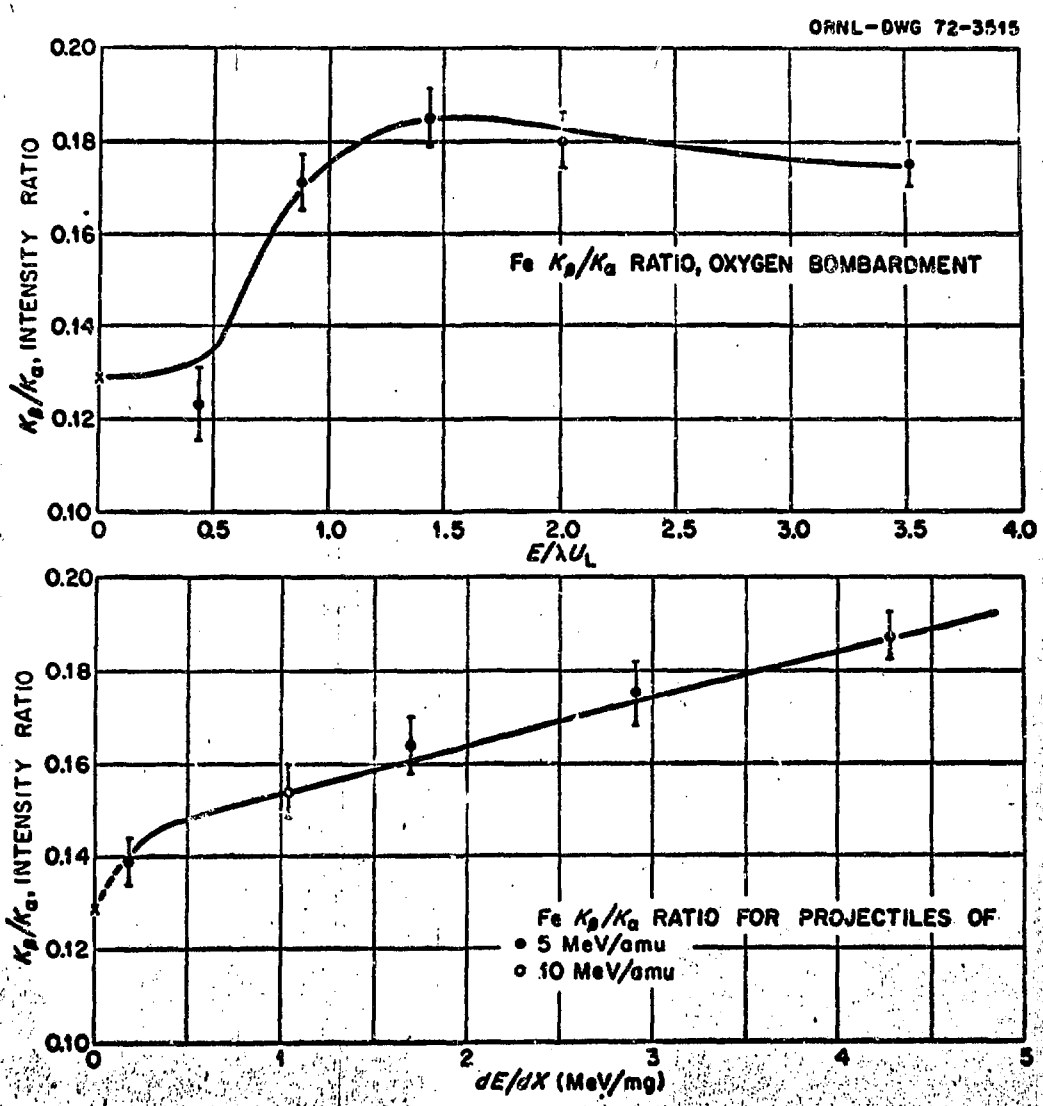
ORNL-DWG 72-3515
0.20
0.18
0.16
0.14
0.12
0.10
K_β/K_α, INTENSITY RATIO
Fe K_β/K_α RATIO, OXYGEN BOMBARDMENT
0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
$E/\lambda U_L$
0.20
0.18
0.16
0.14
0.12
0.10
K_β/K_α, INTENSITY RATIO
Fe K_β/K_α RATIO FOR PROJECTILES OF
• 5 MeV/amu
o 10 MeV/amu
0
1
2
3
4
5
dE/dX (MeV/mg)

Figure 3