Title
HIGH RATE X-RAY FLUORESCENCE ANALYSIS BY PULSED EXCITATION

Author
Jaklevic, Joseph M.

Publication Date
1972-03-01

To be published in IEEE
Trans. on Nuclear Science

LBL-504
Preprint

HIGH RATE X-RAY FLUORESCENCE ANALYSIS BY PULSED EXCITATION

Joseph M. Jaklevic, Fred S. Goulding, and Donald A. Landis

March 1972

AEC Contract No. W-7405-eng-48

DISCLAIMER

HIGH RATE X-RAY FLUORESCENCE ANALYSIS BY PULSED EXCITATION *

Joseph M. Jaklevic, Fred S. Goulding and Donald A. Landis

Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

SUMMARY

We describe the application of pulsed X-ray excitation to X-ray fluorescence spectrometry as a method for increasing the output counting rate of the system by a substantial factor. Using a pulsed X-ray tube that is immediately turned off when a signal is detected, and held off during the pulse processing time, it is possible to eliminate the need for a pulse pile-up rejection. We have achieved output counting rates significantly greater than with conventional operation for equivalent shaping networks. No significant degradation in spectrometer resolution was observed at the increased counting rates.

INTRODUCTION

The electronics used in semiconductor detector spectrometers constitute a sophisticated analogue data processing system capable of extracting the maximum amount of useful information from events in the detector. In addition to the shaping amplifier designed for optimum signal/noise performance, modern spectrometers employ base-line restoration and pulse pile-up rejection in order to ensure a minimum amount of resolution degradation at high count rates. Recently, the application of pulsed light feedback to Si(Li) X-ray spectrometers has resulted in improved resolution and count-rate performance.[1]

All of these systems are subject to the same fundamental rate limitation resulting from the finite time interval required to process an event. Due to the random time distribution of events detected in most measurements, there is a large probability at high counting rates of two events occurring within the measurement (or shaping) time of the amplifier. To prevent distortion of the energy information, a pile-up rejector is normally employed to eliminate such events from the output spectrum. This results in a sizeable loss of events at high counting rates and constitutes a particularly severe limitation in low-energy X-ray spectrometers where optimum energy resolution demands the use of long pulse-shaping time-constants.

This counting rate limit becomes a serious problem in analytical applications using X-ray tube excitation where counting rates are currently limited only by the electronics. Since the analysis time per sample is determined directly by the available counting rate, it is possible to speed up analysis only if these limitations can be overcome.

We describe a pulsed excitation method which demonstrates significant improvements in count-rate performance. When applied to X-ray fluorescence analysis, measurement times are reduced by at least a factor of four.

DESCRIPTION OF METHOD

The effect of pile-up rejection at high rates is illustrated in Fig. 1 where we have plotted the calculated output versus input counting rate for the case of a continuous, random excitation source. We assume that each input pulse generates a dead-time during which any additional event is rejected. Each of the curves represents a different peaking time τ_p of the Gaussian shaped amplifier output pulse. We assume a dead time $\tau_d = 3\tau_p$ corresponding to that used in the pile-up rejector described in Ref. 1. The correspondence between these calculations and measured data supports the validity of our assumptions. It is apparent from the curves that there is a maximum output rate for each τ_p corresponding to less than 40% of the input rate. This decrease of output rate arises from fundamental statistical considerations and cannot be remedied except by somehow modifying the time distribution of input pulses.

The basis of the pulsed excitation method is the elimination of the necessity for pile-up rejection by ensuring that no additional pulses will reach the detector in the time interval required to process an event. As shown in Fig. 2, a transmission X-ray tube[2] with grid control supplies the excitation for the sample. Detection of an event by the system triggers a fast turn-off of the electron beam for a specified dead time determined by the amplifier time constant, thus eliminating the possibility of any signals interfering with the original pulse. Although this description emphasizes the use of X-ray tubes, the pulsed method could be adapted to other excitation modes (such as electron probes or heavy charged particles from accelerators).

The triode electron gun is designed for a cut-off voltage at -10 V, the ON pulse reaches its +100 V operating point in less than 0.2 μsec. The maximum current is limited to less than 2 mA at present due to space charge effects in the narrow angle beam. The uncertainty in the turn off time of the tube is estimated to be <0.5 μsec. This interval should be maintained as short as possible since pile-up events may occur at very high rates.

* This work was done under the auspices of the U.S. Atomic Energy Commission.

Considering the operation of the system in detail, we note several features:

i) The system output rate is equal to the input rate. This is obvious since the tube is on only when the amplifier system is able to accept a pulse for processing.

ii) At very high rates the arrival of events becomes essentially periodic. As the X-ray intensity is increased there is a large probability of detection of an event in a time interval short with respect to the amplifier time constants; the tube is on for only a small part of the total cycle time and the events appear periodic.

iii) The maximum count-rate is $\frac{1}{\tau_d}$ where τ_d is the dead time per pulse--this is a corallary to (ii).

iv) If we continue to increase the X-ray intensity after reaching this limit, no apparent increase in output will result, but multiple events will become more common during the time required to shut off the tube following the detection of an event. Thus, although the output rate remains the same, an increasing fraction constitutes pile-up.

RESULTS

With these preliminary ideas in mind it becomes possible to understand the input-output characteristics illustrated in Fig. 3. These data were obtained by allowing the Mo X-ray from the pulsed tube to strike an Fe target. The curves shown were taken with τ_d = 23, 41, and 79 μsec corresponding to amplifier peaking times of τ_p = 9, 17 and 35 μsec respectively. The measured output rate is restricted to only these pulses which did not undergo pile up at the high rates--this choice of representation was selected because the non-pile up output is a more valid measure of the capability of the system. Furthermore, the data for total output versus input counting rate are far less interesting, since the rates are equal to within 2% over the total range for all time constants. The $\frac{1}{\tau_d}$ input rate limits are indicated by the vertical lines. The periodic behavior of the output data at these limits is evident in the data of Fig. 4 which are curves taken directly from oscilloscope photographs. Observation of the data at still slower sweep speeds showed that the periodic structure is maintained coherently over 150 cycles at τ_d = 79 μsec.

The pile-up probability at high rates is evidenced by the decrease in output rate near the $\frac{1}{\tau_d}$ input counting rate limits in Fig. 3. Although the losses are minimal for the system shown, they could be reduced still further by designing an X-ray tube with faster turn-off characteristics.

Figure 5 shows the Fe X-ray spectrum produced at a counting rate of 12 K c/s with a pulse processing time of 79 μsec (Gaussian-shaped pulse with a peaking time of 35 μsec). The resolution achieved (192 eV) is only 6 eV higher than that measured at very low rates. For a pulse peaking at 9 μsec and processed in 21 μsec an output rate of 40 K c/s was recorded with an energy resolution of 260 eV. This is 7 eV higher than the resolution observed at low rates with this system. The excellent performance at high counting rates is attributed, at least in part, to the favorable conditions for a baseline restorer operating with regular pulses instead of random ones.

DISCUSSION

A comparison between conventional and pulsed excitation can be made as follows: For a Poisson arrival distribtuion, the output rate N_o from a system with a dead time τ_d and an input rate N_I is given by

$$N_o = N_I e^{-N_I \tau_d} .$$

The maximum output rate is $N_o = (e\ \tau_d)^{-1}$ and occurs at an input rate $N_I = (\tau_d)^{-1}$. This describes the situation in a spectrometer operated with a continuous excitation source.

For pulsed excitation, $N_o = N_I$ and the maximum output rate occurs at $N_o = N_I = (\tau_d)^{-1}$. Thus, the maximum output rate obtained in the pulsed mode is increased by a factor e over that of the continuous excitation with pile-up rejection assuming the dead times in both case are equivalent. However, this is not a valid comparison since the use of pile-up rejection with continuous excitation results in longer effective dead times for equivalent minimum pulse separations. This apparent anamoly arises because events separated by less than τ_p must both be rejected in order to prevent ambiguity. For a minimum separation of $2\tau_p$ the equivalent dead times are $\tau_d = 3\tau_p$ for continuous excitation and $\tau_d = 2\tau_p$ for pulsed X-ray excitation. A more realistic estimate for the output rate increase obtained with pulsed excitation is therefore $\frac{3}{2} e$ or ~ 4.

In addition to this increased rate capability there are several other features which recommend pulsed excitation. The elimination of the pile-up rejector greatly simplifies the electronics. The periodic structure of the arrival distribution at high rates facilitates certain electronic operations such as baseline restoration and pulse height analysis. The multichannel analyzer can be made to operate with zero losses of events. Calibration of the system for analytical applications is simplified since the duration and frequency of the grid pulse is a direct measure of the system dead time. Finally, the pulsed method uses only as much X-ray intensity as is required to achieve the given count rate. At the maximum counting rates the duty factor in the X-ray tube is a minimum, thus reducing power dissipation requirements and radiation hazards.

As a final illustration of the use of the present excitation method we show in Fig. 6 a spectrum obtained by exciting a freeze dried whole blood specimen with Mo characteristic $K_{\alpha,\beta}$ radiation. The data were taken at a rate of 20 K c/s for a period of 10 min. Peaks due to the presence of <1 ppm of trace impurities in the actual specimen are clearly visible. Analysis of the Pb peak indicates a statistical accuracy in the peak area of 0.12 ppm. The level of Pb impurity detected in this particular specimen is 0.2 ppm when referred to the original whole blood sample--a number typical for normal urban population. It is impressive that these results were obtained in a counting time less than 1/4 of that previously necessary without losses either through pile-up rejection or analyzer dead times.

ACKNOWLEDGMENT

We thank B. Jarrett, A. Jue, G. Skipper and W. Searles for their assistance in various phases of this work. We are indebted to R. Giauque for some of the analytical results presented here.

REFERENCES

1. D. A. Landis, F. S. Goulding, R. H. Pehl and J. T. Walton, IEEE Trans. Nucl. Sci. Vol. NS-18,No. 1, pp. 115-124 (1971).

2. J. M. Jaklevic, R. D. Giauque, D. F. Malone, and W. L. Searles, "Small X-ray Tubes for Energy Dispersive Analysis Using Semiconductor Spectrometers", presented at the 20th Annual Denver X-ray Conference, Aug. 11-13, 1971, Denver,Colorado. Lawrence Berkeley Laboratory Report Number LBL-10.

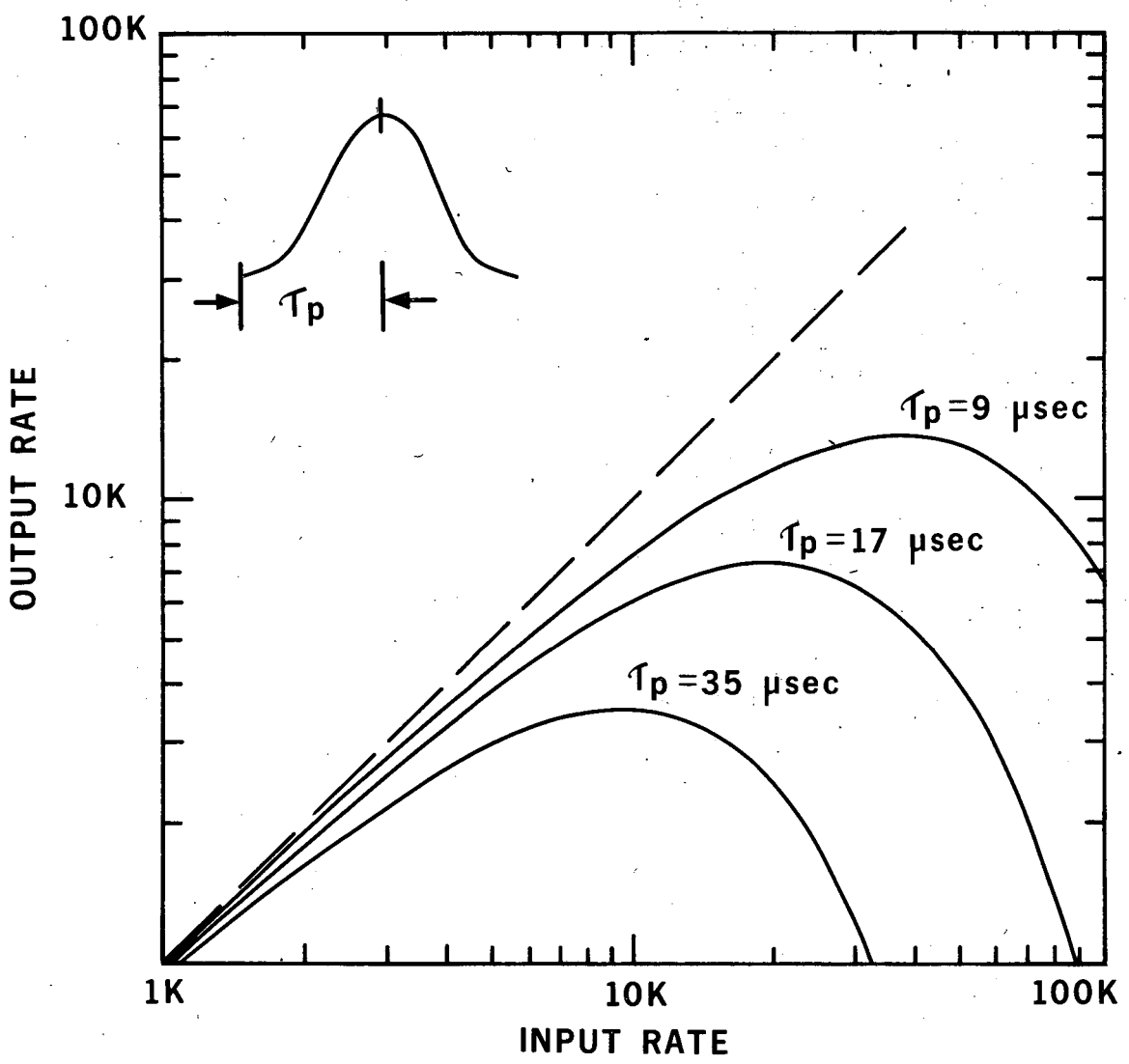


Fig. 1. Output versus input counting rate for continuous excitation with pile-up rejection.

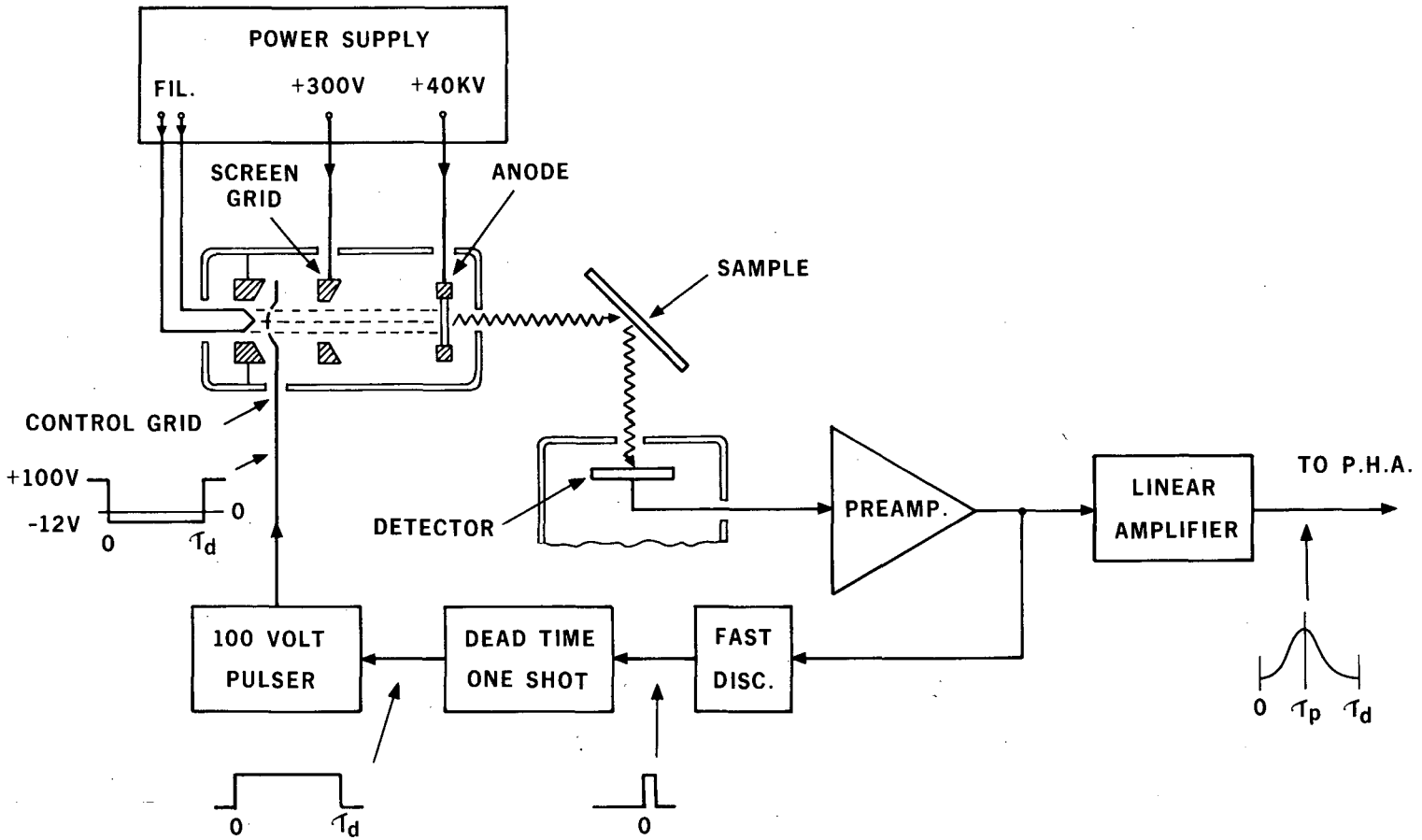


Fig. 2. Schematic of Pulsed X-ray System.

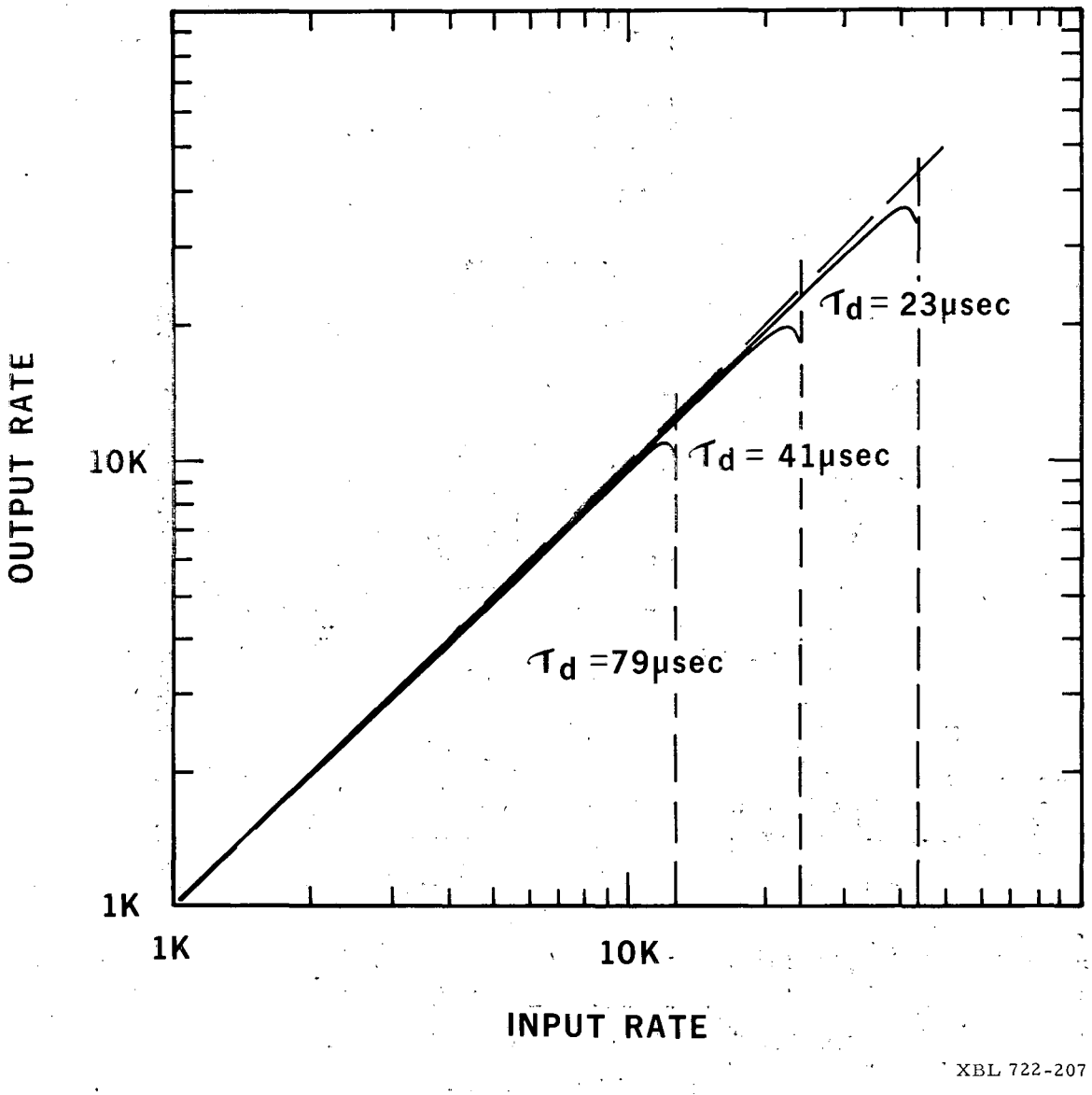
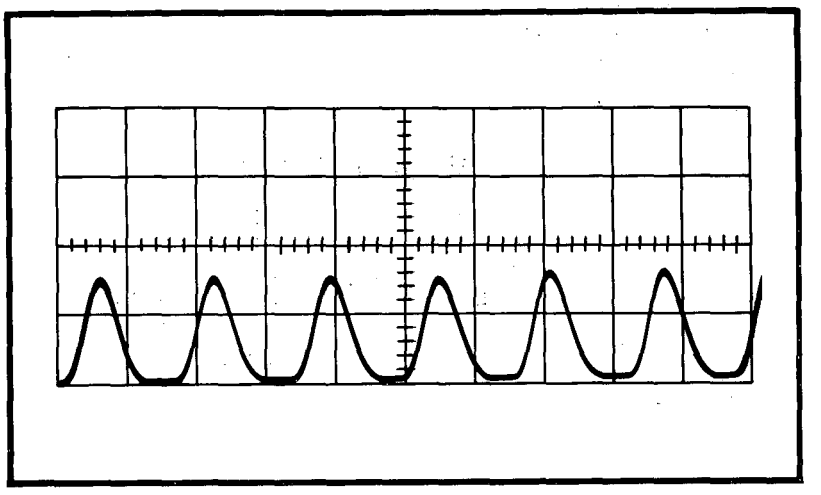
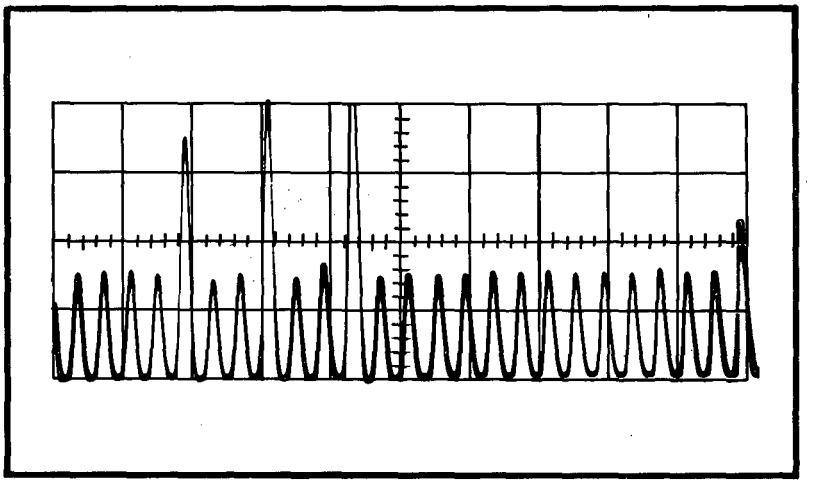


Fig. 3. Output versus input counting rate for pulsed system. Output rate is limited to non-pile up events. Vertical dashed lines correspond to $(\tau_d)^{-1}$.



t ⟶ 50 μs/cm



t ⟶ 200 μs/cm

Fig. 4. Oscilloscope photographs of amplifier output at 12 K c/s and τ_d = 79 μsec.

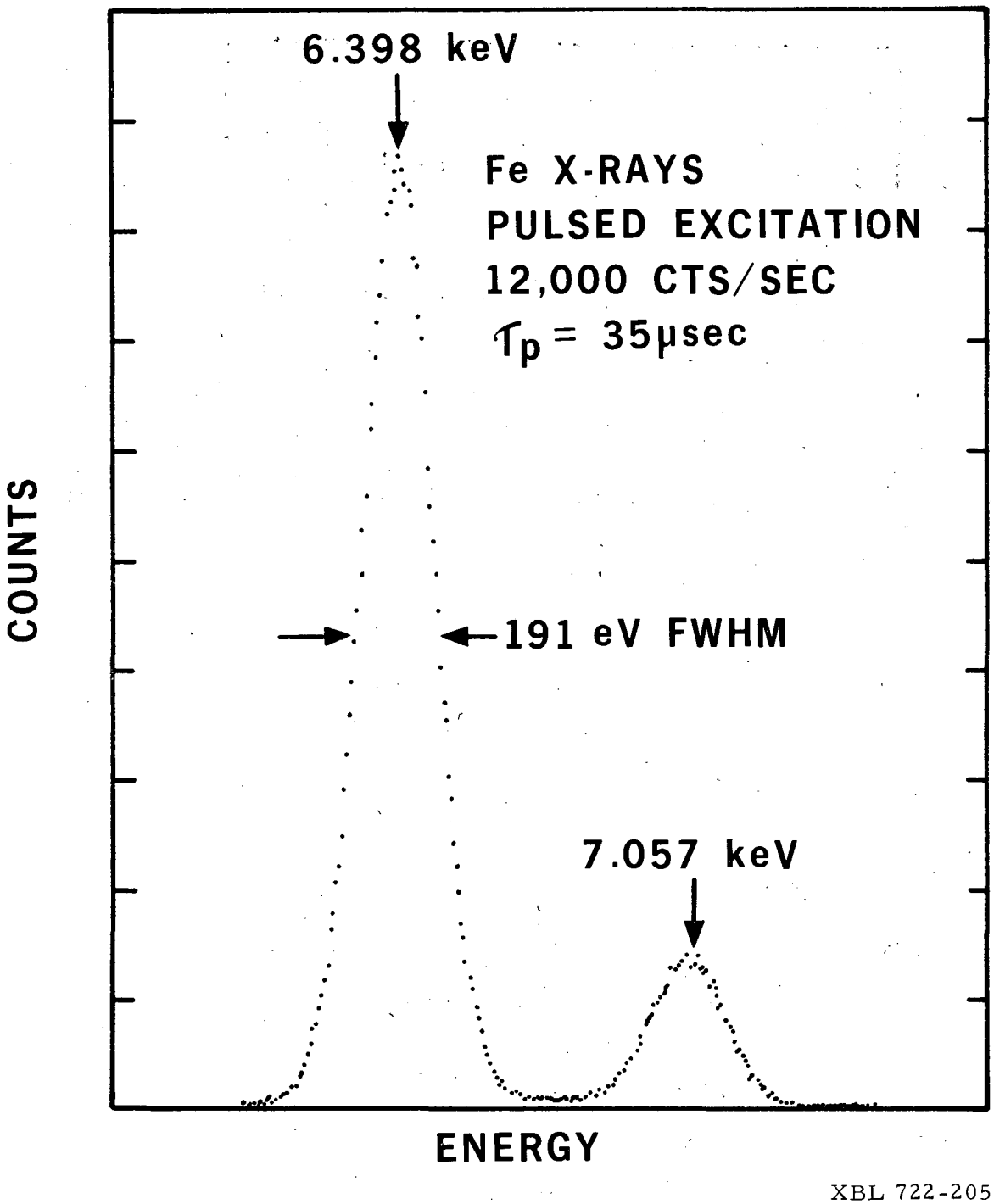


Fig. 5. Spectrum of Fe X rays taken at 12 K c/s.

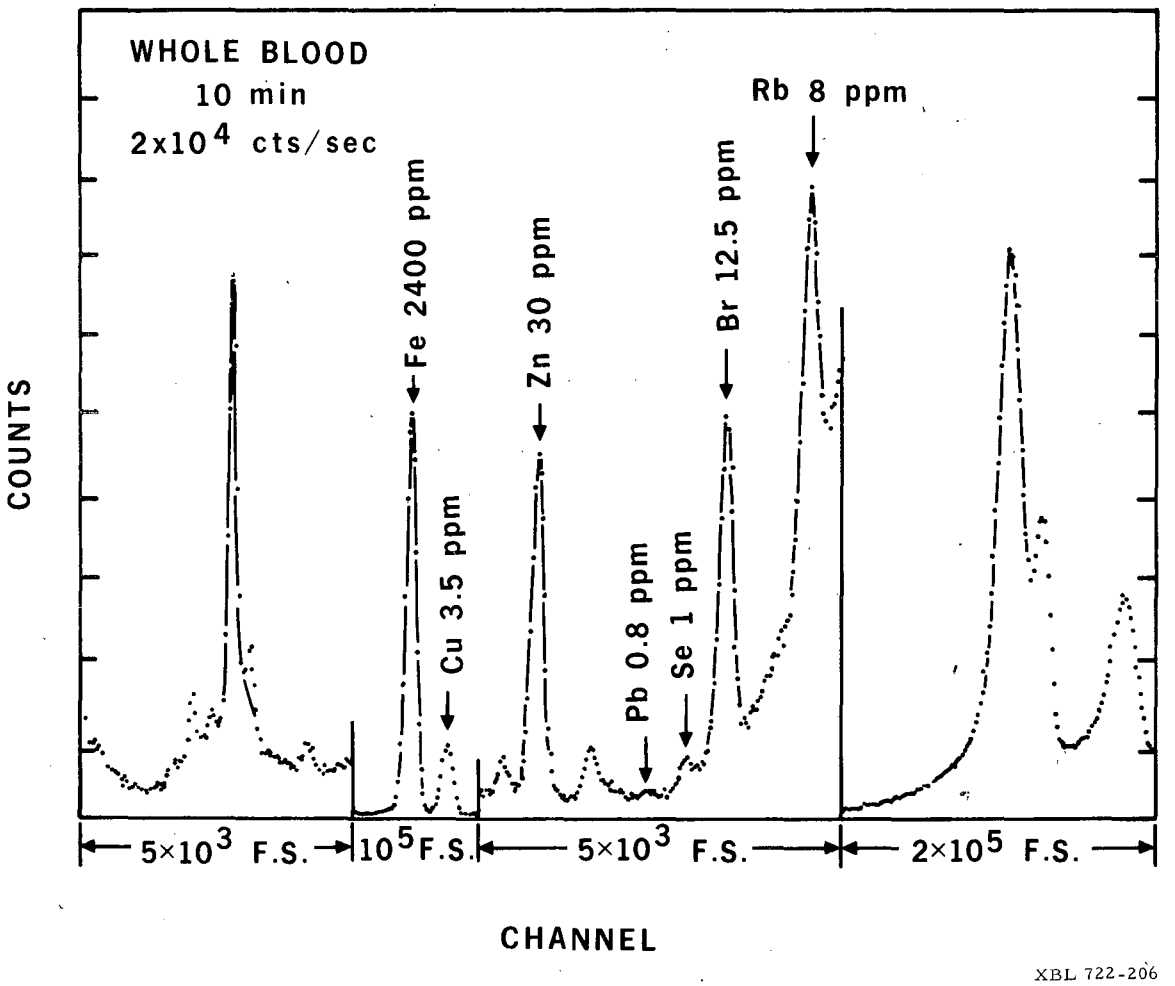


Fig. 6. Fluorescence spectrum of freeze dried whole blood taken at 20 K c/s with Mo excitation. Concentrations shown on figure refer to the actual sample as analyzed; levels referred to original blood are approximately five times less.

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720