Title
RELATIVISTIC TWO-PHOTON EMISSION: LIFETIME OF THE 21SO STATE OF HELIUM-LIKE Kr34+

Author
Marrus, R.

Publication Date
1986

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Materials & Molecular Research Division

Submitted to Physical Review Letters

RELATIVISTIC TWO-PHOTON EMISSION: LIFETIME OF THE 2^1S_0 STATE OF HELIUM-LIKE Kr^{34+}

R. Marrus, V. San Vicente, P. Charles,
J.P. Briand, F. Bosch, D. Liesen, and I. Varga

January 1986

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

DISCLAIMER

RELATIVISTIC TWO-PHOTON EMISSION : LIFETIME OF THE $2\,^1S_0$ STATE OF HELIUM-LIKE Kr^{34+}*

Richard Marrus
Department of Physics and Lawrence Berkeley Laboratory
University of California, Berkeley, CA 94720

Victorino San Vicente, Pierre Charles and Jean Pierre Briand
Université P&M Curie and Institut Curie, Section de Physique et Chimie**
11 rue P&M Curie, 75231 Paris Cedex 05, France

*Fritz Bosch, Dieter Liesen and Imre Varga****
Gesellschaft für Schwerionenforschung mbH
6100 Darmstadt, Federal Republic of Germany

Abstract

The two-photon (2E1) decay 2^1S_0-1^1S_0 in the helium-like ion Kr^{34+} has been observed and the lifetime measured. Departures are measured for the first time from the predictions of the non-relativistic theory, and are sensitive to relativistic effects on the transition matrix elements. The measured lifetime is τ (measured) = 34.08 (.34) x 10^{-12} s. compared to τ (non-relativistic) = 31.15 (0.10) x 10^{-12} s.

Radiative decay of excited atomic and nuclear states by the simultaneous emission of two electric dipole photons (2E1 decay) is a relatively rare process that is usually observed only when decay by single-photon emission is rigorously forbidden (e.g. for 0→0 transitions) or highly inhibited by the angular momentum and parity selection rules as in the case of the $2S_{1/2}$-$1S_{1/2}$ transition in hydrogenic ions. To date a number of observations and measurements have been reported in atoms (1) and nuclei (2). The best tests of the theory have been made on hydrogenic ions in the $2S_{1/2}$ state. The most accurate calculation of the non-relativistic rate for

* *Experiment carried out at GANIL (Caen, France) and GSI (Darmstadt, FRG)*
** *Unité Associée au CNRS No UA 040771*
*** *Permanent address : Technical University Budapest, H-111 Budapest, Hungary*

these ions is due to Klarsfeld (3) which yields for the transition probability/sec. A (non-relativistic) = $8.22943Z^6 s.^{-1}$. Measurements have been made (1) on hydrogenic ions through Z=18 and are all in good agreement with this calculation.

Recently, Parpia and Johnson (4) and Goldman and Drake (5) have made calculations of the 2E1 decay rate using a relativistic formulation. Their work indicates that deviations $\geq 1\%$ from the non-relativistic rate should exist for $Z \geq 20$. In this paper we report a measurement of the 2E1 transition $2^1S_0 \rightarrow 1^1S_0$ in the helium-like ion Kr^{34+}. This measurement shows for the first time departures from the predictions of the non-relativistic theory. The result represents the first test of the relativistic theory of the 2E1 process and indeed is the first test of the relativistic contribution to the dipole matrix element.

The beam-foil time-of-flight method is employed in these measurements. The apparatus consists of two Si(Li) detectors located on opposite sides of a box through which the Kr^{34+} beam passes (see Fig.1). The positions of the exciting foil (200 $\mu g/cm^2$ carbon) and one of the detectors are fixed thoroughout the experiment. The second detector - collimator assembly is moved parallel to the ion beam by means of a precision translator with a positional accuracy $\sim$ 1 μm. This accuracy is important since the decay length of the 2^1S_0 state is about 2mm. The raw data consists of the ratio of the number of counts in the 2E1 spectrum of the moveable detector to the number of 2E1 counts in the fixed detector as a function of the separation of the two detectors. Thus, in this experiment, the fixed detector is used to normalize the count rate in the moveable detector. This differs from the conventional time-of-flight method where the integrated beam current in a Faraday cup is used for normalization and where the foil-detector separation is varied. The method used here has two important advantages over the conventional method:

1) Normalization is directly to the number of ions in the 2^1S_0 state.

2) Changes in the shape and state of the exciting foil during the experiment are of no consequence.

A first attempt to observe the 2^1S_0 decay was made with the 35 MeV/A krypton beam obtained from the GANIL accelerator (Caen, France). This experiment

succeeded in observing the decay and in obtaining a preliminary value for the lifetime. However this beam contained a relatively large ratio of $Kr^{35+}/Kr^{34+} \approx 0.25$. This is important since the decay $2S_{1/2} \rightarrow 1S_{1/2}$ in Kr^{35+} is primarily by 2E1 with a spectrum that is essentially indistinguishable from the 2^1S_0 spectrum under study here. This contamination precludes an accurate lifetime measurement. To surmount this problem an experiment was performed with the 18.9 MeV/A krypton beam from the UNILAC accelerator (GSI Darmstadt).

The krypton beam is prepared in the following way. The 18.9 MeV/A krypton beam is pre-stripped by a thick carbon foil producing a distribution of Kr ions in highly-charged states. A dispersing magnet selects only the Kr^{34+} charge state which is then deflected into our beam line. This beam is then excited by a $\sim$ 200 $\mu g/cm^2$ C foil which produces the Kr^{34+} ions in the 2^1S_0 state. This procedure insures a high ratio of Kr^{34+}/Kr^{35+} ions in the beam. Charge fraction measurements (6) made with the Kr beam on various thickness C foils show that at this energy $N(Kr^{34+})/N(Kr^{35+}) \geq 90{:}1$. Even with this ratio, there is still a small number of $2S_{1/2}$ counts under the 2E1 spectrum. These are measured and subtracted out by a procedure described later in this paper.

The lifetime measurement requires an accurate measurement of the decay length ($\sim$ 2 mm in this experiment) and the beam velocity. The accuracy of the decay length is insured by establishing the position reference relative to a fixed detector rather than to the exciting foil. Using a precision translator to move the second detector and taking care to align the detector translation axis parallel to the beam axis, the error in the decay length from mechanical and alignment errors is $\leq 0.05\%$. The beam velocity is measured by a time-of-flight system installed after the first stripping foil but before the exciting foil. The system consists of two detectors separated by about 16 m that detect arrival pulses from the beam bunches. The system yields for the beam energy a value of 18.934 MeV/A. The correction to the energy resulting from passage through the exciting foil is based on the energy loss tables by Ziegler (7) for 19 MeV/A ^{84}Kr on carbon. The result is $\Delta E = -5.1$ MeV (7).

Hence, the beam velocity after the foil is $\beta = v/c = .19842$ (29).

The raw data consists of spectra taken at different detector positions. A sample spectrum used in the decay length measurement is shown in Fig. 2. In evidence is the two photon spectrum. Analysis of the peak (M1+M2) shows that $n=2 \rightarrow n=1$ transitions in Kr^{35+} and Kr^{34+} are also present and finally there is a fairly uniform background at $E \geq 19$ KeV. The electronics was calibrated before and after the run and was found to be stable to ≤ 20 eV. For analysis purposes, the data under the continuous 2E1 spectrum was separated into four equally-spaced energy windows from 3.5 KeV to 12.38 KeV (Fig. 2). The first step was to subtract from each window counts arising from hydrogen-like ions in the beam and counts from ions in the 2^1S_0 state that arise from cascades from higher states. The number of counts from hydrogen-like ions is found by fitting the peak at 13.0 KeV (Fig. 2). In evidence are the $2^3S_1, 2^3P_2 \rightarrow 1^1S_0$ of Kr^{34+} and a relatively small peak arising from the single-photon M1 decay $2S_{1/2}-1S_{1/2}$ of Kr^{35+}. From the known branching ratio (8) 2E1/M1 = 1.96 for this decay and the measured window and detector efficiency, the number of 2E1 counts from hydrogen-like ions is unambiguously subtracted from the raw data. The number of counts from ions which have cascaded into the 2^1S_0 state can be unambiguously determined from the small observed peaks at 15.3 KeV and higher. These result from $np \rightarrow 1^1S_0$ transitions. From the known branching ratios $np \rightarrow 2^1S_0 / np \rightarrow 1^1S_0$ an unambiguous correction (upper limit) is made to the data for the cascading ions. This correction is small ($<10^{-4}$ of the decay length).

Background counts under the 2E1 spectrum were treated in two ways. First, a window was established at $\sim$ 22 KeV and the counts in this window are assumed to arise from background. It was then assumed that the background in each of the four 2E1 windows was proportional to the background with proportionality constant $K\alpha$ different for the windows (α=A,B,C,D) but is the same for all detector positions. The value of $K\alpha$ was then chosen so as to minimize χ^2 when all of the points are fit to a single exponential to yield a decay length. The second procedure used for fitting the data was to take the data corrected for hydrogen-like ions and

cascades in each of the four windows and then do a least squares fit to a single-exponential plus constant background. The two procedures yield results which differ by less than 1%. A sample decay curve obtained by this fitting procedure is shown in Figure 3.

Our final experimental result is $A_{exp}\ (2^1S_0) = 2.934\ (30) \times 10^{10}\ s.^{-1}$.

The error is mainly the statistical error associated with the fitting procedure. Errors associated with possible systematic effects are shown in Table I.

Our experimental result may be compared with theory in the following way. Non-relativistic variational calculations have been done (9) for $A_{NR}\ (2^1S_0)$ in the range Z=2-10. We can fit these results with a Z-expansion of the form (10)

$$A_{NR}\ (2^1S_0, Z) = 16.45886\ Z^6 - 79.62\ Z^5 + 169.77\ Z^4 - 175.63\ Z^3\ (s.^{-1}). \qquad \{1\}$$

The leading term is twice the hydrogenic $2S_{1/2}$-$1S_{1/2}$ rate and is known exactly. The other three coefficients are obtained from a least squares fit to the variational calculations in the range $4 \leq Z \leq 10$. The maximum discrepancy between {1} and the variational calculations is 0.4% which may be taken as an upper limit to the error at Z=36. For Z=36 we thus obtain $A_{NR}\ (2^1S_0,\ Z=36) = 3.13(1) \times 10^{10}\ s.^{-1}$ to be compared with our experimental value $A_{exp}\ (2^1S_0) = 2.935\ (30) \times 10^{10}\ s.^{-1}$. The leading (in Z^{-1}) relativistic correction has been determined by Parpia and Johnson (4) and by Goldman and Drake (5). Using eq. (4.7) of Goldman and Drake, the decay rate equation {1} becomes

$$A_{rel}\ (2^1S_0) = 16.45886\ Z^6 \left(\frac{1 + 3.9448\ (Z\alpha)^2 - 2.040\ (Z\alpha)^4}{1 + 4.6019\ (Z\alpha)^2} \right)$$

$$- 79.62\ Z^5\ \{1 + O\ (Z\alpha)^2\} + 169.77\ Z^4\ \{1 + O\ (Z\alpha)^2\} - 175.63\ Z^3\ \{1 + O\ (Z\alpha)^2\}\ s.^{-1} \qquad \{2\}$$

where the terms $O\ (Z\alpha)^2$ are as yet uncalculated. We thus obtain for the relativistic decay rate

$$A_{rel} = 2.979\ (35) \times 10^{10}\ s.^{-1}.$$

We note the following : 1) Relativistic effects decrease the decay rate, ever though

they increase the $2^1S - 1^1S$ energy separation. Thus this measurement is primarily sensitive to the relativistic effects on the transition matrix elements.

2) Our measurement is in good agreement with the result using the leading order correction. However, the error in the theoretical value is due mainly to the uncalculated $O(Z\alpha)^2$ correction to the Z^5 term. Since the theoretical error is of the same order as the experimental error, it will be necessary to calculate the higher order term in order to confirm the agreement.

We wish to express our appreciation to the operating crews and technical staff of both the UNILAC and GANIL accelerators for their outstanding efforts in providing the high-quality beams necessary for the success of these experiments.

This research was supported jointly by Chemical Sciences Division of the U.S. Department of Energy under contract DE-AC03 76SF00098, by NSF grant INT 83-13622, and by NATO grant No 252180.

REFERENCES

1. H. Gould and R. Marrus, Phys. Rev. A28, 2001 (1983) and references contained therein.
2. J. Schirmer, D. Habs, R. Kroth, N. Kwong, D. Schwalm, M. Zirnbauer, and C. Broude, Phys. Rev. Lett. 53, 1897 (1984).
3. S. Klarsfeld, Phys. Lett. 30A, 382 (1969).
4. F.A. Parpia and W.R. Johnson, Phys. Rev. A26, 1142 (RC) (1982).
5. S.P. Goldman and G.W.F. Drake, Phys. Rev. A25, 2877 (1982).
6. D. Liesen, Unpublished work and private communication.
7. J.F. Ziegler, Handbook of Stopping Cross Sections for Energetic ions in all Elements, Pergamon Press, New York, 1980.
8. W.R. Johnson, Phys. Rev. Lett. 29, 1123 (1972).
9. G.W.F. Drake, G.A. Victor and A. Dalgarno, Phys. Rev. 180, 25 (1969).
10. G.W.F. Drake, Private communication to R. Marrus, 1983.
We are grateful to Professor Drake for suggesting this approach and for providing the results of his own calculations.

TABLE I

Contributions to experimental error (%)

Beam velocity	0.1
Cascade correction	0.01
Geometry and misalignment	0.05
Hydrogenic $2S_{1/2}$ contamination	0.2
Dead time	0.1
Fitting error	0.9

Final result $\tau\,(2^1S_0) = 34.08(34) \times 10^{-12}$ s.

FIGURE CAPTIONS

Figure 1. Schematic of the apparatus.

Figure 2. Sample spectrum taken at near foil-detector separation. The "bins" labelled a,b,c,d were arbitrary selected for independent analysis of the 2E1 decay rate.

Figure 3. Sample decay curve obtained for window c (see Fig.2).

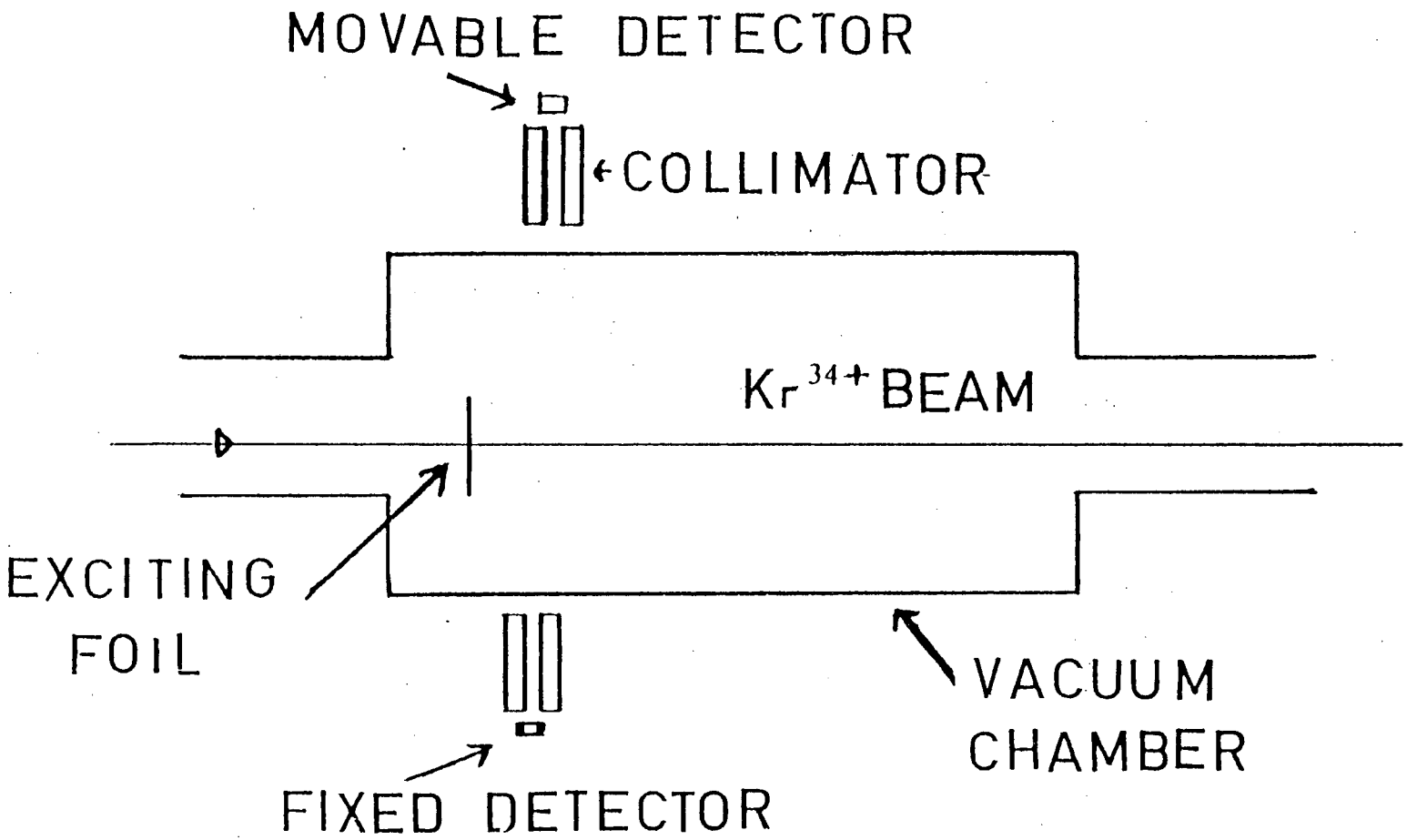

MOVABLE DETECTOR
COLLIMATOR
Kr 34+ BEAM
EXCITING FOIL
VACUUM CHAMBER
FIXED DETECTOR

FIGURE 1

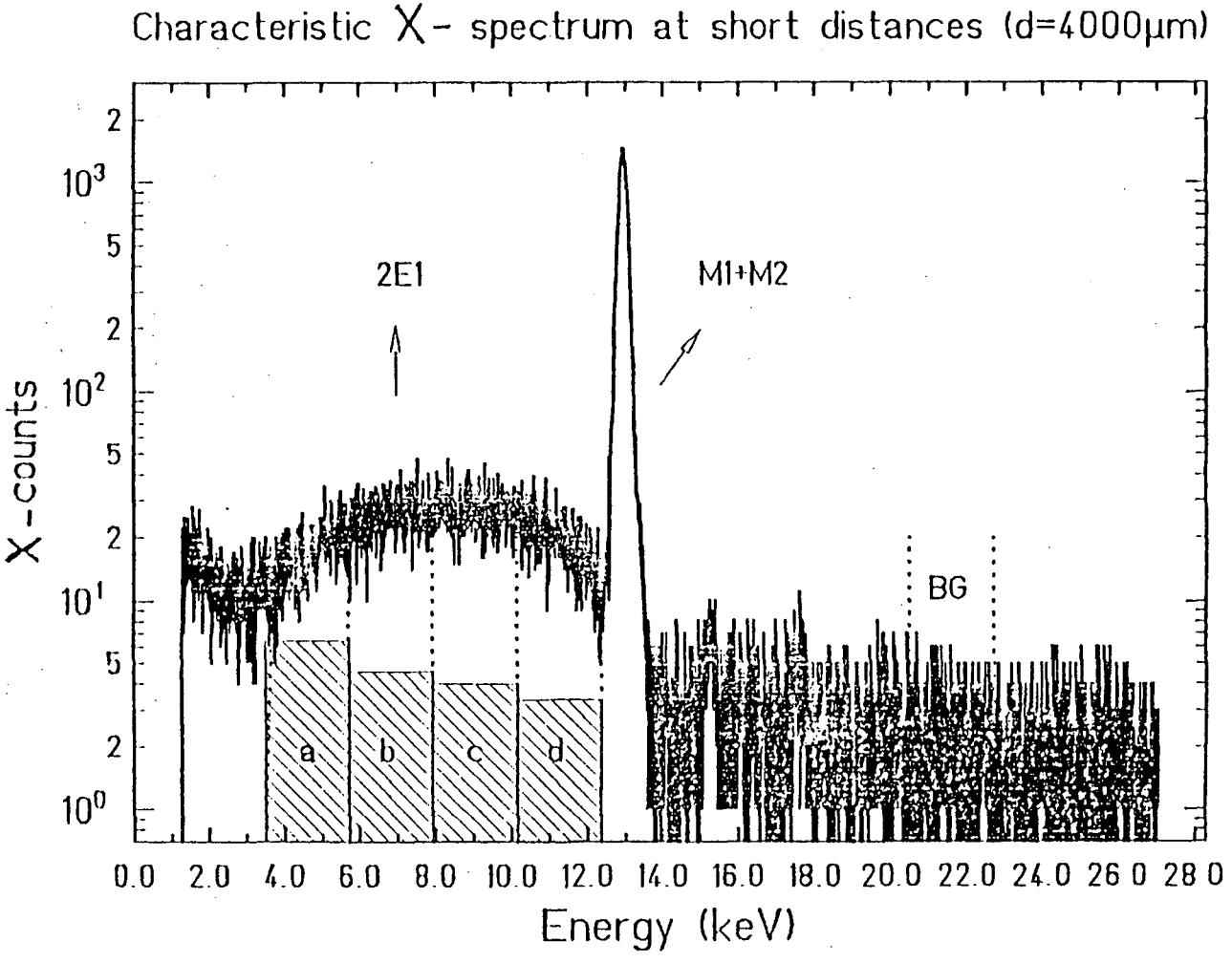
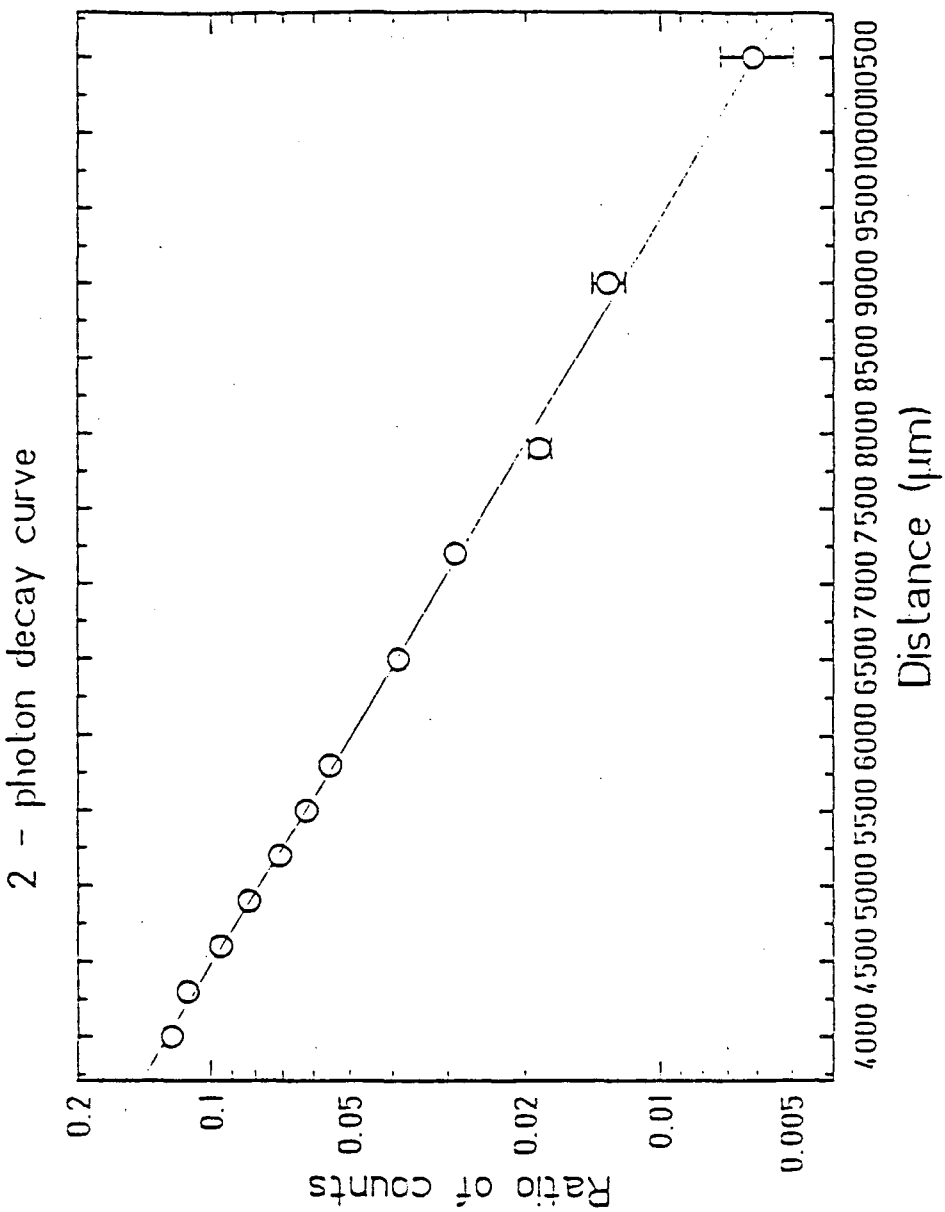

Characteristic X- spectrum at short distances (d=4000µm)
X-counts
Energy (keV)
2E1
M1+M2
BG
a
b
c
d
0.0 2.0 4.0 6.0 8.0 10.0 12.0 14.0 16.0 18.0 20.0 22.0 24.0 26.0 28.0

FIGURE 2

FIGURE 3


2 - photon decay curve
Ratio of counts
0.2
0.1
0.05
0.02
0.01
0.005
4000 4500 5000 5500 6000 6500 7000 7500 8000 8500 9000 9500 10000 10500
Distance (μm)

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